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Exceedance of Critical Loads and Levels

Spatial and Temporal Interpretation of Elements in Landscape Sensitive to Atmospheric Pollutants

Report of a workshop held in Vienna, Austria under the Convention on Long Range Transboundary Air Pollution, 22-24 November 1995

Edited by M. Knoflacher, J. Schneider and G. Soja

Organised by the Austrian Research Centre Seibersdorf and the Federal Environment Agency

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Workshop Summary

A. Introduction

The workshop „Exceedance of Critical Loads and Levels: Spatial and Temporal Interpretation of Elements in Landscape Sensitive to Atmospheric Pollutants" was held in Vienna from 22-24 November 1995. The workshop was organised by the Austrian Research Centre of Seibersdorf in collaboration with the Austrian Federal Environment Agency (FEA) and the International Institute for Applied System Analysis (IIASA).

The workshop was attended by more than 55 scientists from 17 different countries.

The aim of the workshop was to provide input to the mapping exercises carried out within the Convention on Long Range Transboundary Air Pollution.

The main topics were

- to re-evaluate the procedure of deriving universal, consistent Critical Loads and Levels and mapping guidelines from discrete experiments carried out under defined conditions in the field,
- to provide understanding of the uncertainties influencing the result of mapping procedures and
- to work out principles for the interpretation of exceedances of Critical Levels and Loads.

B. Results and Recommendations

The results are grouped into two main topics:

1. Issues Concerning Critical Levels of Ozone

The generalisation of results obtained in field or open-top chamber experiments with only a few species under certain conditions to standardised rules for European - wide assessment of ozone effects on vegetation is a difficult task. This generalisation will always result in a certain degree of limitation, which should be kept in mind and be made explicit to users of the AOT40 concept.

Anyhow, the currently used concept of Critical Levels is commonly agreed to be a powerful tool for the assessment of ozone effects within the Convention. The use of the concept should include continuous re-evaluations and updating according to growing scientific knowledge.

1.1 Level I Approach

Till now there are a number of uncertainties to the deviation of level I critical levels. Especially concerning forest and seminatural ecosystems, there is still a number of limitations. It should be emphasised that the exceedance of critical levels can not be directly translated into a reduction of biomass production but rather be assumed as an indication for a growing risk of ozone-mediated damage.

Measurement profiles and model calculation show large vertical gradients in AOT40 values close to the ground. For crops and other low vegetation, this will introduce a substantial error,

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because the mapped values of AOT40 based on measurements made at a height of 3 - 4 m will differ from those at the crop envelope.

Research Recommendations:

- Available data and models should be used to quantify this effect and to develop a methodology which is suitable for mapping.
- Effect orientated ozone-exposure experiments should be extended to different species

1.2 Level II Approach

It is recognised that the development of level II approach based on the real effects of ozone needs new experimental work.

Nevertheless, it should be possible at present to make some progress by incorporating modifying factors. These include factors which influence the sensitivity of the vegetation like atmospheric conductivity; soil moisture deficit; vapour pressure deficit; phenological state; and factors influencing stomatal conductance. Stomatal conductans is of eminent importance to assess the potential ozone uptake by plants. In combination with micrometeorological models, this could lead to the calculation of absorbed doses.

Results have been reported covering a range of ozone interactions, e.g. with genotypic differences, water supply, growth stage, altitude and CO_2. The sensitivity of different species (including species of natural and semi-natural vegetation) has been investigated.

There is growing evidence of significant latitudinal and altitudinal variations in the susceptibility of vegetation to ozone. These effects should be incorporated into a level II approach of critical levels of ozone.

Research Recommendation:

- Analysis of modifying factors:
  - receptor sensitivity, genotypic differences
  - temporal and growth stage-dependent sensitivity
  - environmental and meteorological fators determing actual ozone uptake
  - differences in plant sensitivity depending on latitudes and altitudes
- Efforts should be strengthened to quantify the effects of the above mentioned factors to allow their incorporation into level II mapping.

2. Issues concerning Mapping in general

2.1 Scaling

At present data and maps obtained with different spatial resolution are used within the framework of the Convention. Combining sets of data with different resolution might lead to systematic errors. Calculations of exceedances of critical loads very often result in higher exceedances if high resolution deposition data are used instead of deposition data with a coarse resolution (e.g. 150 km x 150 km). Additional, aggregation rules substantially influence the outcome of European critical load maps derived from high resolution national data.

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Data used for national purposes have to be distinguished from data obtained by the groups within the Convention for integrated assessments.

For National Focal Points it is recommended to harmonise their mapping activities with neighbouring countries to yield homogenous maps over Europe.

2.2 Uncertainty Analysis

The uncertainties of models and maps should be addressed in more detail and have to be made explicit. More emphasis should be laid on uncertainty analysis in future.

2.3 Interpretation of Exceedances of Critical Loads and Levels

Concerning critical levels for ozone, level I approach is at present not suitable for predicting biomass reduction or accurate yield loss of crops over the whole of Europe. Anyhow, critcal levels should not be exceeded in order to protect vegetation.

Within a level II approach yield reductions could be approximated.

Dynamic models are believed to be a valuable tool to assess effects of long term depositions exceeding critical loads. However, further scientific research is needed to meet this goal and to evaluate the actual data requirements. The linking of dynamic models with biological models might be useful and is recommended for further investigations.

It is recognised that it is necessary to link damage assessments with exceedance of critical loads and levels. However, this linking has to be done very carefully to avoid misinterpretations.

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Key Elements In Ozone Risk Analysis

J. Fuhrer

Swiss Federal Research Station for Agricultural Chemistry and Environmental Hygiene[1]
Schwarzenburgstrasse 155, CH-3097 Liebefeld-Bern, Switzerland

Abstract

Ozone risk analysis can be separated into different elements. The first element consists of the identification of the assessment endpoint(s), i.e. the attribute(s) selected to characterize adverse effects, such as reductions in plant growth or yield. Further elements include the characterization of the environment, based on the continuous measurements of environmental parameters and of ozone concentrations. In order to relate measured ozone concentrations to effects, exposure indices must be identified which capture most efficiently the relevant information of exposure patterns. Based on experiments with crop and tree species, exposure indices should give greater weight to higher concentrations, and should accumulate concentrations over time. Within the UN/ECE, the exposure index AOT40 was selected, and exposure-response relationships were constructed. Modifying factors, such as light, temperature, soil water availability, or others, can be included in risk assessments, provided that their influence on selected plant responses to ozone is known in quantitative terms. Integrated risk assessments comprise tools to display the degree of risk, expressed as exceedance of critical levels or probability of adverse effects, in time or in space. In the latter case, mapping procedures are needed to spatially interpolate between measuring points, to account for altitude effects and local influences, and to integrate information. The present overview shows that significant gaps in knowledge exist concerning key elements such as critical levels, modifying factors, and protocols to validate exceedance maps, in particular for receptor categories 'forest trees' and 'fruit trees'.

1. Introduction

Setting critical levels for ozone aims at defining those levels of exposure above which adverse effects on sensitive receptors may occur. Critical levels can thus be used as the basis for the development of efficient strategies to reduce precursor emissions. The success of emission control measures can be assessed by the degree of reduction in exceedance, i.e. the difference between the actual level and the critical level of ozone.

Critical levels for ozone have been established in recent years (Fuhrer & Achermann, 1994), and maps showing the spatial distribution and the extent of exceedance have been produced both at the national and international level. As such maps become available, two questions must be raised:

1. Are there negative effects on plants in areas where the critical level is exceeded?
2. How certain can we be that the prediction of negative impacts is correct?

What is needed to answer these questions is called *Ecological Risk Analysis*, which can be defined as "process that evaluates the likelihood that adverse effects occur or are occurring as

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a result of exposure to one or more stressors". In this context, "stressors must have (1) the ability to cause adverse effects and (2) to co-occur with or contact an ecological component long enough and at sufficient concentrations to cause the identified adverse effect" (USEPA, 1992). Adverse effects can be detected as changes at any level of biological organization, ranging from molecular to the ecosystem level. According to Guderian *et al.* (1985) adverse effects can be classified into injury and damage. Injury includes all measurable effects that do not influence yield or biomass, and damage includes all effects that reduce the intended value. In the context of ozone risk analysis, Tingey *et al.* (1990) considered adverse effects as a loss in (i) economic production, (ii) ecological structure, (iii) genetic resources, and (iv) cultural values. In the framework of the UN/ECE Convention on Long-Range Transboundary Air Pollution (LRTAP), and as the basis for the revised Air Quality Guidelines of the World Health Organization (WHO), the main adverse effects presently considered are:

- loss of economic yield (crops)
- reduction in biomass or growth (trees)
- change in the relative species composition (semi-natural vegetation)

The choice of specific adverse effects is mainly based on the effects of ozone observed in experiments on the various receptors, and on the ecological and economic importance of changes in these parameters. The term 'effect' implies that a particular experimental treatment causes a change in the selected parameter, as compared to the control treatment which should reflect the unpolluted reference. Hence, not only the choice of the assessment endpoint, but also the choice of the reference is an important determinant for the outcome of the risk assessment. For ozone, there is considerable difficulty in defining the 'unpolluted' reference; it is likely that the reference varies with latitude and altitude.

The following discussion is based on a conceptual model for ozone risk analysis (Fig. 1), which is composed of physico-chemical components, including ozone exposure, and biological components, including ozone uptake, ozone effects, and the influence of modifying factors on both uptake of ozone and effect. Physico-chemical and biological components are separated by the boundary between outside and inside of leaves, i.e. the leaf surface, and the flux across the boundary is controlled by stomatal conductance.

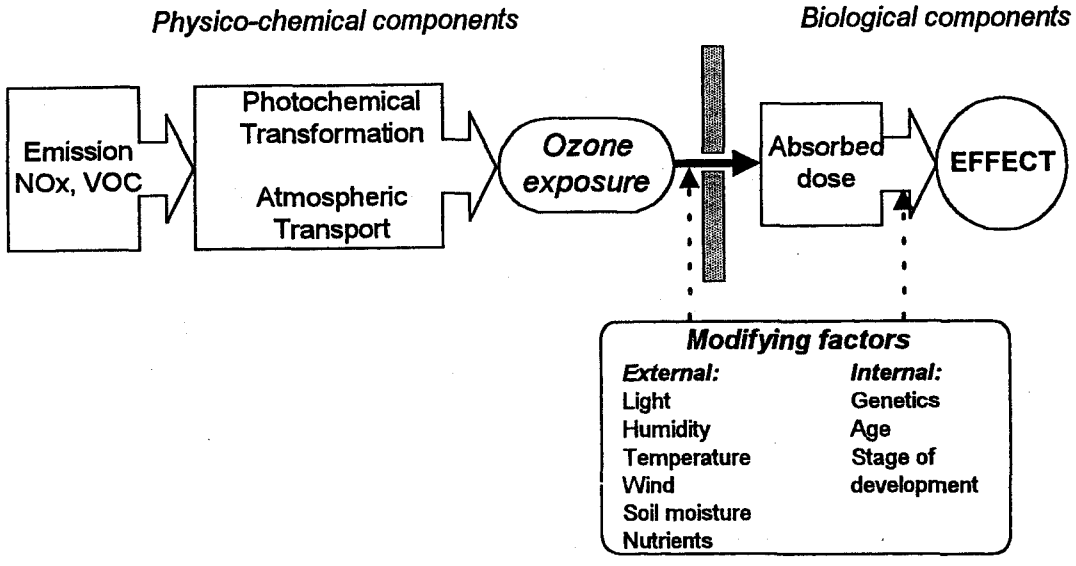


Fig. 1: Conceptual model for ozone risk analysis (after Fuhrer, 1993)

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2. The relationship between ozone exposure and ozone effects

In order to define the critical level for a particular pollutant-receptor combination, exposure-response information is essential. This can be obtained experimentally by using different exposure systems, or by field observation (Sanders *et al.*, 1995). In order to construct exposure-response relationships it is necessary to characterize ozone exposure by using a single statistical index, which efficiently captures those components of exposure which are biologically most effective. Long-term exposure to ozone can lead to growth and yield reduction. The most suitable exposure indices to be related to long-term effects are cumulative, i.e. they integrate exposure over time. This was shown both for crops and for forest trees. For instance, the study by Dizengremel *et al.* (1994) revealed that ozone effects on photosynthesis and RUBISCO activity in loblolly pine was related to the cumulative exposure expressed as the sum of the hourly mean concentrations over one or two experimental seasons.

Previous air quality guidelines and critical levels for long-term effects have been based on mean concentrations over a fixed period of time, e.g. the arithmetic mean over the growing season (e.g. April to September) of the daily mean concentrations for a specific 7-h period (usually 09.00-16.00). The use of a mean concentration implicitly gives equal weight to all concentrations. However, experimental exposure-response studies with ozone suggest that this is not appropriate; it was shown that it is the intermittent exposure to higher concentrations which is most important in causing long-term effects (cf. Lefohn, 1992). This can be explained physiologically by the capacity of the plant to detoxify ozone and other oxidants; it is only when the concentration, or flux, of ozone exceeds this capacity that adverse effects result.

This phenomenon can be incorporated into the exposure index by using procedures for continuous or discontinuous weighting of concentrations (cf. Hogsett *et al.*, 1988). Figure 2 illustrates two possibilities: (a) The use of a discontinuous weighting procedure. This approach

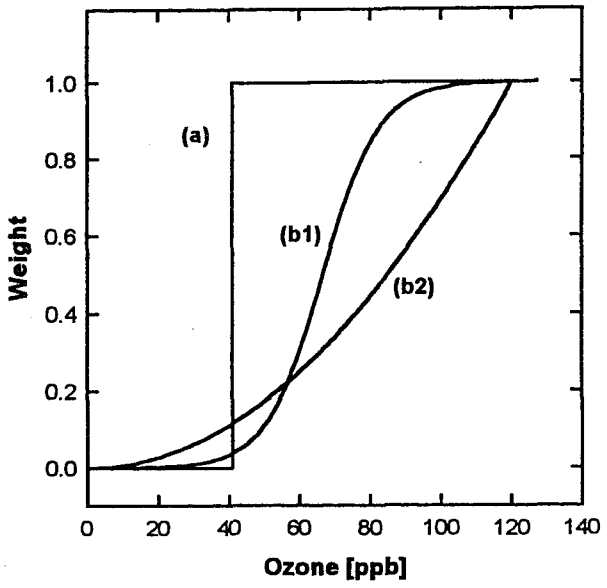


Fig. 2: Discontinuous and continuous weighting of ozone concentrations. Discontinuous weighting of concentrations depends on the choice of the cut-off concentration, i.e. 40 ppb. Continuous weighting depends on either a sigmoid weighting function (b1) or an allometric weighting function (b2).

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assigns the weight of 0 to all concentrations below a cut-off value and a weight of 1 to all concentrations above the cut-off. (b) The use of a continuous weighting function. The concentration can be weighted with a sigmoid function, i.e. a multiplicative weighting factor between 0 and 1, which depends on concentration, is used for each hourly O_3 concentration (b1). Alternatively, by using an allometric weighting function the concentration is raised to a power (b2). There are sound biological reasons for believing that the latter procedure, using continuous weighting, gives in theory a better representation of exposure. An index based on a sigmoid function which has been used successfully to fit exposure-response data is referred to as W126 and considers concentrations above 30 ppb (Lefohn *et al.*, 1988). However, given the limited European exposure-response data and the difficulties of accurately mapping or modelling complex exposure indices, the former procedure which depends on the definition of a cut-off concentration is more practical. In order to calculate a cumulative exposure index, such as the AOT (accumulated over threshold), the positive differences between the actual hourly mean concentration and the cut-off concentration are summed for the exposure period of interest. The calculation of the cumulative exposure index with a cut-off concentration of 40 ppb (AOT40) is illustrated in Figure 3. The shaded area represents the cumulative exposure. An alternative cumulative-type index often used in the US is the sum of concentrations equal or higher than a cut-off, for instance 60 ppb (SUM06) (Lefohn & Foley, 1992). As opposed to the calculation of an AOT, a SUM index considers the total hourly concentrations instead of the difference between the cut-off and the measured value. Hence, the AOT index focuses more directly on the portion of exposure which is thought to be biologically effective.

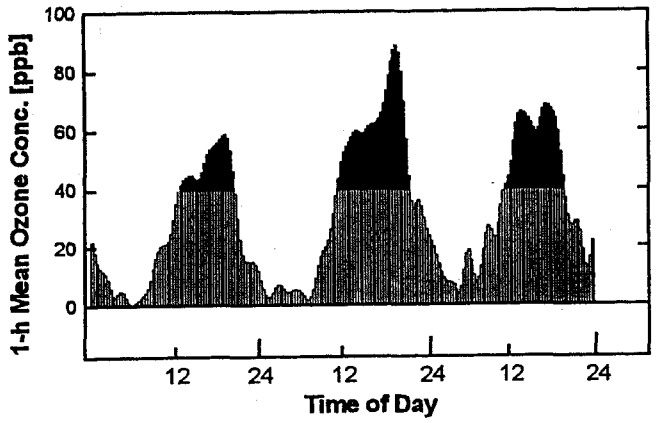


Fig. 3: Example of a typical 3-day time course of hourly mean ozone concentrations, with the fractions marked in black which are used to calculate the AOT40 (concentrations above 40 ppb during daylight hours).

This concept was adopted at the UN/ECE workshop at Egham in 1992, when a threshold concentration of 40 ppb was tentatively suggested (Ashmore & Wilson, 1993). The exposure index AOT40 is expressed in units of ppb.h or ppm.h. Subsequent statistical analysis of yield data from European open-top chamber experiments has demonstrated that the use of this threshold generally provides better linear fits to exposure-response data than the use of higher thresholds (Fuhrer, 1994). A linear exposure-response relationship provides a sounder statistical basis to define critical levels corresponding to a specific effect than does other types of exposure-response relationships (Sanders *et al.*, 1995). The use of 40 ppb as the threshold has been favoured over lower threshold concentrations because in Europe it broadly corresponds to the boundary between mean concentrations in areas with low and high frequencies of photochemical episodes. However, the choice of this threshold does not imply that concentrations below 40 ppb have no effect. In fact, there is evidence for a lack of a threshold for effects of

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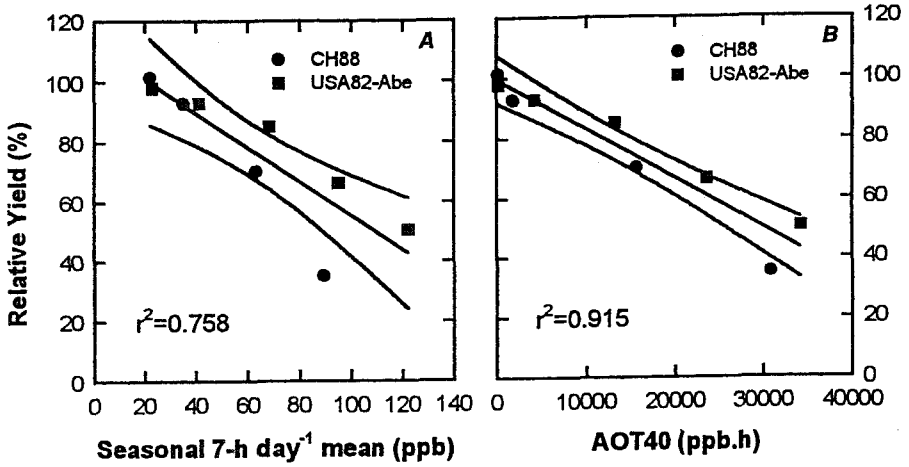


Fig. 4: Comparison of exposure indices. Exposure-response relationship (with 95% confidence interval) for the effect of ozone on grain yield of wheat using the seasonal 7-h day^{-1} mean concentration (A) or the AOT40 calculated for the exposure period of 87 days (CH88) or 55 days (USA82-Abe) (B).

ozone on wheat, and even reductions of ozone in the range below values currently found at the most remote locations can stimulate relative grain yield (not shown). Hence, the threshold concentration does not present a threshold for effects, but rather a cut-off concentration. Because of the increase in the proportion of background ozone with increasing altitude, the use of a cut-off concentration of 40 ppb may not be appropriate for higher elevations.

Based on a typical exposure duration, the AOT40 for crops is calculated over 3 months (e.g. May to July) and for forest trees over 6 months (April-September). In the case of agricultural crops, the index is calculated with concentrations during daylight hours ($\geq 50 Wm^{-2}$), whereas in the case of forest trees all hourly concentrations are considered because of ozone uptake during the night may occur. Recently, Matyssek *et al.* (1995) reported that in *Betula pendula* ozone exposure during the night was equally effective in reducing plant biomass and root to shoot biomass as was daytime exposure using the same concentration. However, it remains to be tested whether ozone uptake by single leaves or needles, as measured in fumigation chambers or gas exchange cuvettes, properly reflects nighttime ozone fluxes to forests.

It has been difficult to compare the performance of the AOT40 index with indices based on mean concentrations because of the lack of variability in the length of exposure between experiments. The situation was improved by including in the analysis data from the NCLAN (U.S. National Crop Loss Assessment Network) experiments at Argonne. Fig. 4 shows a comparison between the exposure-response relationships for the Swiss experiment in 1988 (duration=87 days) (Fuhrer *et al.*, 1989) and the 1982 Argonne experiment with the cultivar Abe (duration=55 days) (Kress *et al.*, 1985). The comparison demonstrates the improvement in r^2 achieved by using the AOT40 index instead of the seasonal 7-h day^{-1} mean. This result is supported by earlier data comparisons (e.g. Fuhrer, 1994).

In most cases, the performance of the AOT40 index has been evaluated by retrospective data analysis. However, the selected experiments were not designed to compare different exposure indices. This limitation could only be overcome by new experimentation. A few experiments designed to test different components of exposure have been carried out recently. For instance, Musselman *et al.* (1994) investigated the importance of peak concentrations by comparing treatments with different exposure profiles, i.e. treatments with constant concentrations vs.

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treatments with peak concentrations but with the same total exposure. Using bean plants, they observed stronger effects of the treatment with the peaks (Fig. 5). Mixtures of perennial ryegrass and white clover were used to investigate the importance of the pattern of exposure for effects on yield and species composition (Nussbaum *et al.*, 1995). Treatments with a constant increase in ozone and treatments with an episodic increase in ozone were compared with control and ambient ozone levels. The results suggested that AOT40 is a suitable predictor for changes in species composition, independent of the exposure regime, but to describe changes in total yield, AOTs with higher cut-offs are more efficient. This suggested that effects on the relatively more tolerant grass (*Lolium perenne*), which contributed the largest portion to total yield, were mainly related to the occurrence of peak concentrations of more than 100 ppb.

According to the concept in Fig. 1, effects of ozone depend on ozone uptake, and it has been suggested that ozone exposure indices should be replaced by flux estimates (Grünhage *et al.*,

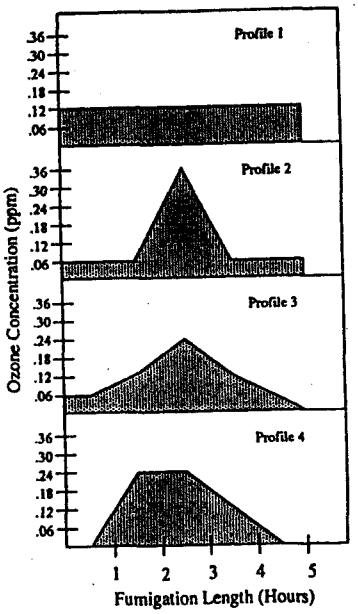


Fig. 5A: Different exposure regimes to test the importance of peak concentrations

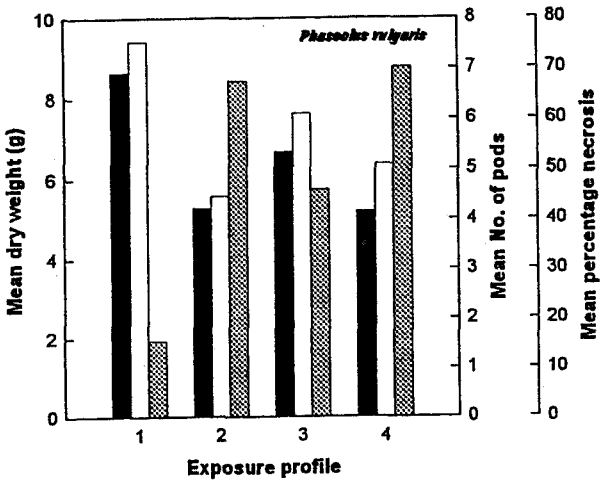


Fig. 5B: Effect of different exposure regimes on mean dry weight (filled bar), mean number of pods (open bar) and mean percentage necrosis (grey bar) of beans (Musselman *et al.*, 1994).

1993). In open-top chambers, ozone flux measurements have been carried out (Grandjean Grimm & Fuhrer, 1992) and it is possible to relate the mean ozone flux density, or mean ozone uptake, to AOT40. The data in Fig. 6 indicate a direct relationship between the two parameters suggesting that AOT40 can be used as a surrogate for ozone flux. However, this may only be true for the situation in open-top chambers with constantly high turbulence due to ventilation. Under ambient conditions, ozone flux is controlled by atmospheric and boundary layer resistance, and high ozone concentrations may or may not be associated with high ozone flux. It has been estimated that on average ozone flux in open-top chambers is about 20% higher than in the open field (Grünhage & Jäger, 1994), which is consistent with measurements over barley presented by Pleijel *et al.* (1994). Although the importance of this difference has been recognized, to date the concept of using ozone flux instead of concentration-based exposure indices could not be applied to critical levels because of a lack of quantitative relationships between ozone flux and long-term ozone effects under field conditions.

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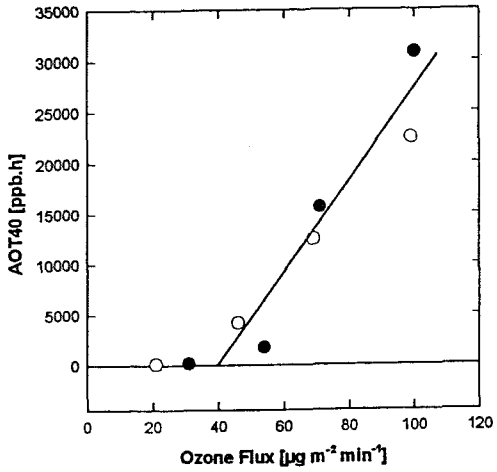


Fig. 6: Relationship between the growing season mean ozone flux density in open-top chambers with wheat and AOT40

Therefore, it seems to be more appropriate to use a correction factor to account for the difference between open-top chamber and ambient conditions, and to apply the correction to the AOT40 value. Similarly, there is need for transfer functions to relate the AOT40 determined in the open-top chamber experiments to AOT40 values measured routinely at a standard height of 3-5 m above ground. Because of deposition processes, a steep concentration gradient exists near the canopy, and the concentration at 3-5 m may be much larger than the concentration near the ground (Pleijel *et al.*, 1995b). Unless transfer functions are included, the direct application of AOT40 values obtained from monitoring networks may lead to a significant overestimation of potential effects.

3. The data base to derive critical levels

Crops

Based on the suggestion made at the Egham UN/ECE Workshop, results from recent European OTC experiments with a limited number of agricultural crop species were used to test the relationship between the cumulative exposure index using a threshold of 40 ppb (AOT40) and grain or seed yield. Indices with other cut-offs, such as AOT30 and AOT60, were included for comparisons. The most comprehensive data base was compiled for spring wheat. In an initial analysis, data from eight experiments were used (Fuhrer, 1994). These experiments represented a range of site conditions with locations from southern Sweden to Switzerland, and from both lower and higher elevations. No common cultivar was used across the different experiments, and the duration of exposure varied between experiments with an upper limit of about 90 days. For each experiment and each exposure index, the absolute yield at 0 ppb.h was determined by linear regression. In a second step, the yield at 0 ppb.h was used as the reference (100%) to calculate relative yields for each treatment. The data from all experiments were then combined and subjected to linear regression analysis. Based on the resulting regression equation, the AOT corresponding to different levels of relative yield reductions were calculated. For a 10%-reduction in grain, the corresponding AOT40 value was 5300 ppb.h, the corresponding AOT30 and AOT60 values were 6800 ppb.h and 2700 ppb.h, respectively. The coefficient of determination was not different when using AOT30 (r^2=0.91) or AOT40 (r^2=0.91), but it was lower when using AOT50 (r^2=0.88) and much lower when using AOT60 (r^2=0.79). This shows that concentrations between 40 and 60 ppb make an important contribution to the observed change in yield and should not be ignored. Following this initial analysis additional data sets were added. These included data for three cultivars of winter wheat tested

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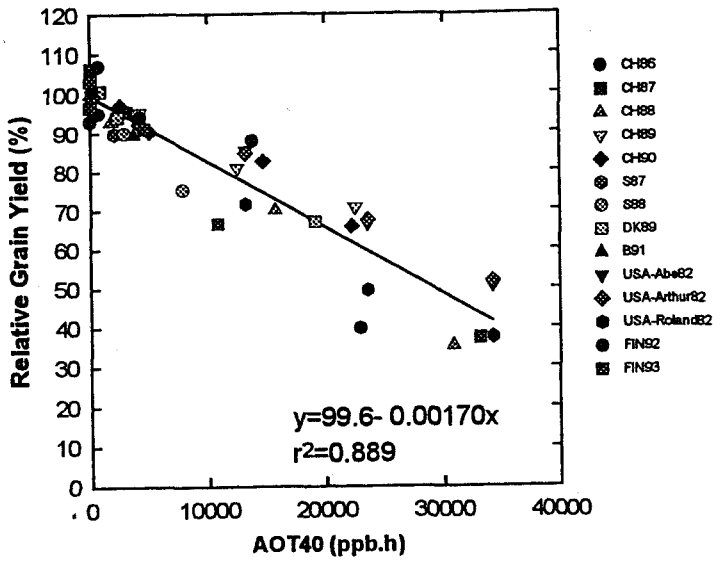


Fig. 7: Extended exposure-response relationship for ozone expressed as AOT40 and relative grain yield of wheat.

at Argonne (USA) in 1982, and more data from Finnish, Swedish, and Swiss experiments. The overall exposure-response relationship between AOT40 and grain yield of spring wheat for the combined data is shown in Figure 7. The data were fitted by using a linear model, and the regression equation was only marginally changed by the introduction of the additional seven data sets. The AOT40 corresponding to 10% yield reduction is 5800 ppb.h. Hence, using the AOT40 proved to be a robust exposure index for grain yield of wheat. This may not imply that relationships with other plant traits would be equally good; Pleijel *et al.* (1995a) found that changes in total plant biomass are better related to an AOT with a lower cut-off (AOT10), whereas the yield of grass-clover mixtures was best related to an AOT with a very high cut-off (AOT110) (Nussbaum *et al.*, 1995).

Forest trees

The data base for forest trees is much more limited than the data base for crops. Fig. 8 shows data for beech (*Fagus silvatica*) and Norway spruce (*Picea abies*) presented at the Bern workshop. Adopting the procedure used for the wheat data, relative changes in biomass after one or more years of exposure are plotted against AOT40 for the corresponding period. The plots show the large variability in the data, as compared with the data for wheat. Using the regressions shown in both plots, it is only possible to estimate with great uncertainty the AOT40 corresponding to a change in biomass by 10%. This value could be used as the critical level for one growing season. However, it must be emphasized that the results included in this analysis were obtained in open-top chambers with phenotypes from lower elevations. Hence, extrapolation to field conditions is difficult, and the use of the data to predict the risk for higher elevation forest must be questioned. Furthermore, the data are almost exclusively from experiments with seedlings and scaling up to mature trees is difficult. Clearly, more data analysis, including data for characteristics other than biomass (e.g. photosynthesis, chlorophyll content), and the development of methods for scaling up from seedling to mature trees are necessary before a critical level can be set with confidence.

Semi-natural vegetation

In the case of mixtures of species, the endpoint of the assessment is not total yield, because ozone effects on the relative species composition are more important and of higher ecological significance. Since there are very limited exposure-response data available for herbaceous plant communities, and these tend to relate to relatively short-term experiments on artificial mixtures, it is

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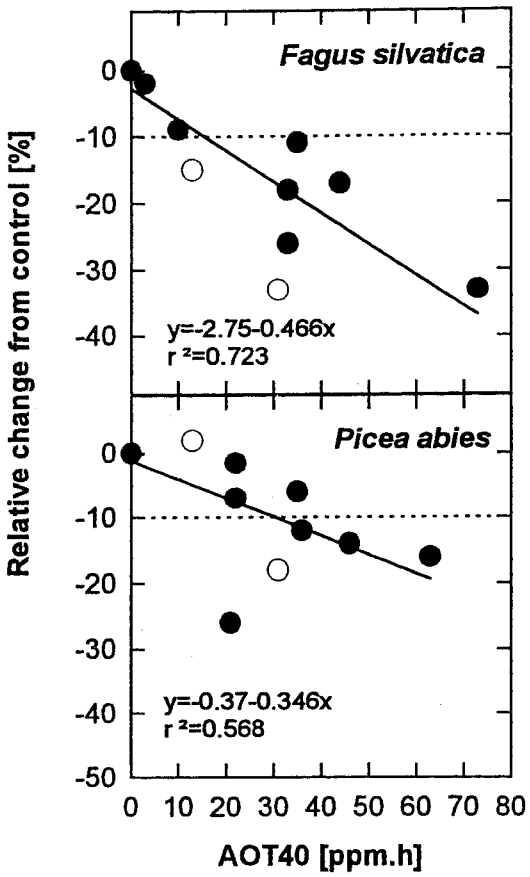


Fig. 8: Exposure-response relation-ships for beech and Norway spruce based on data from Braun & Flückiger (1994) (O) and Küppers *et al.* (1994) (●).

not possible to derive critical level values directly. However, there is evidence of significant changes in species composition at values of AOT40 similar to the critical levels set for agricultural crops. As in the case of crops, it is possible to calculate from the exposure-response relationship the ozone exposure corresponding to a given effect. Selecting a reduction by 10% in the proportion by biomass of the sensitive species as the criterion, the equivalent ozone exposure was 6.3 ppm.h for data obtained with managed pasture (Fuhrer *et al.*, 1994), and 5.7 ppm.h for data obtained with cut swards (Ashmore & Ainsworth, 1995). An experiment using calcareous grassland, rather than acidic grassland species, gave a value of 7.8 ppm (Ashmore *et al.*, 1995). All these values are very similar, and also are comparable to the value of 5.3 ppm.h for a 10% loss in yield in wheat.

Fruit trees

There are many crop species which have not been investigated experimentally in Europe, or for which exposure-response relationships have not been analyzed. An important lack of data concerns the Mediterranean region. Exceptions are studies with peach trees (*Prunus persica*) in central Italy (Paolacci *et al.*, 1995) and with watermelon (*Citrullus lanatus*) at the Ebro delta in Spain (Reinert *et al.*, 1992). Statistical analysis of the data for peach trees revealed significant linear relationships between the AOT40 and some but not all plant traits. Examples are shown in Fig. 9. A 10% change in economically important quality parameters, such as fruit flesh firmness, corresponded to an AOT40 below the value set for grain yield of wheat (Badiani *et al.*, 1996). Additional data supporting these preliminary observations are necessary before critical levels for fruit trees can be set with confidence. Importantly, a decision must be taken as to what assessment endpoint should be used instead of biomass, which is used for forest trees.

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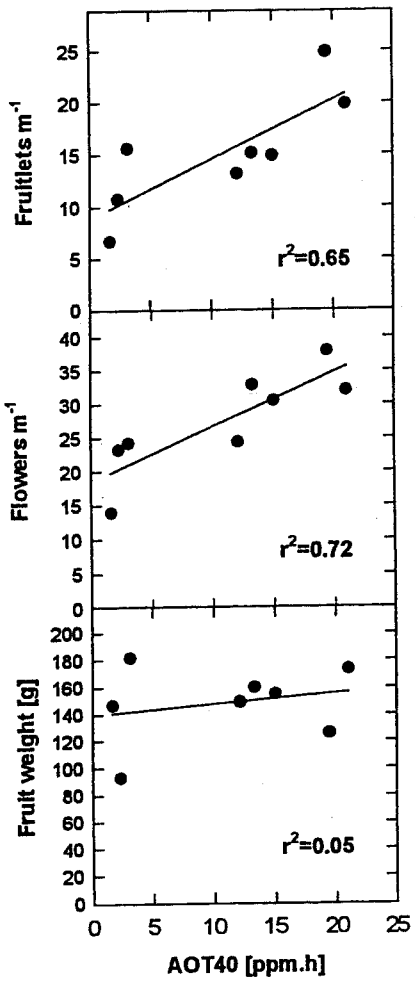


Fig. 9: Exposure-response relationships for selected parameters of peach trees (after Badiani *et al.*,1996)

4. Modifying factors

The effect of ozone on plants depends not only on the exposure pattern, but also on the interaction with environmental factors (see Fig. 1). Most importantly, factors that influence the stomatal conductance can alter the flux of ozone to the leaf interior. It was suggested to introduce factors for different environmental properties to modify long-term critical levels according to the expected interaction (=modifying factors). In order to be useful for mapping purposes, these factors must be estimated using simple procedures and environmental data widely available.

Soil moisture is among the factors which are of first priority to be considered, because effects of ozone under well-watered conditions differ from those under conditions of limited soil moisture content (SMC). This can be demonstrated by comparing exposure-response functions under different soil moisture conditions. In Fig. 10, an example for wheat is used to illustrate the effect of reduced irrigation on the response to increasing ozone levels (expressed as the slope of the regression).

A simple procedure to derive a modifying factor for SMC was presented recently (Fuhrer, 1995). This procedure, which applies to wheat, is based on (a) an estimate of the mean available soil water content (SMC) during 2 months before harvest (e.g. June and July), (b) an

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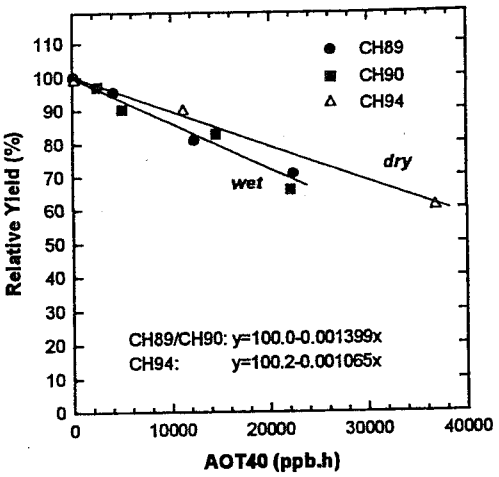


Fig. 10: Exposure-response relationship for grain yield of wheat exposed in OTCs under irrigated (wet) or non-irrigated (dry) conditions (data from Fuhrer *et al.*, 1992, and Nussbaum *et al.*, 1995).

empirical relationship between mean SMC and relative grain yield, and (c) the assumption that with decreasing SMC the sensitivity to ozone changes proportionally to grain yield. SMC can be estimated in various ways; it was proposed that a simple soil water balance approach be used which depends on precipitation and potential evapotranspiration as main input parameters. Based on the assumption stated under (c), 'relative yield' can be replaced by f_{water}, the modifying factor for SMC. In a first approach, soil characteristics were not considered, but the procedure includes a 'drying factor' which could be adjusted to different soils according to their water retention capacity. In cases where irrigation is used to supplement natural precipitation, the amount of irrigation must be entered into the calculation. It is thus possible, that by using irrigation the estimated ozone effect becomes larger than without irrigation.

Plant developmental stage is among the 'internal' factors which should be considered as modifying factor. There are indications that ozone affects final yield relatively more during flowering and grain filling, as compared with earlier or later developmental stages (Pleijel H., pers. comm.). This may be due to increased sensitivity, or because these stages are most important for final yield. As an example, ozone patterns during 1989 and 1990 at two Swiss sites (Oeschberg and Zimmerwald) are shown in Fig. 11. Maximum hourly concentrations were relatively low during the most critical stages, and much higher during grain maturation until harvest. In 1990, on the other hand, ozone levels were higher during the same period. It seems important to account for such differences in the co-occurrence of high ozone levels and high plant sensitivity and to introduce a modifying factor which gives highest weight to concentrations during the most sensitive stages. However, up to now such a phenological weighting function for wheat has not been defined.

4. Integrated risk characterization

In order to obtain a picture of the spatial distribution of ozone risk, the information concerning ozone exposure and plant sensitivity must be combined (Fig. 12). Using a geographical information system (GIS), data for ozone exposure, expressed as AOT40, can be included by applying spatial interpolation procedures using data from monitoring sites or outputs from photochemical models. The GIS can also contain data on land use or species abundance in each grid. For the species present in a grid, exposure-response function can be entered. By combining ozone exposure with the exposure-response function, the potential effect on the selected species can be calculated and plotted. In order to consider the effect of modifying factors, their

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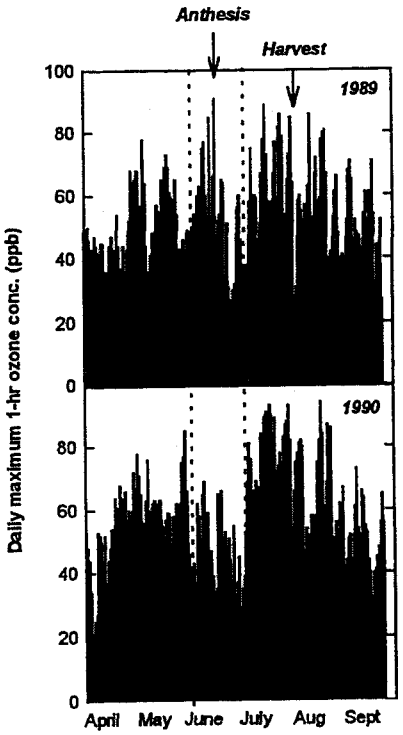


Fig. 11: Daily maximum ozone concentration during the experimental periods of 1989 and 1990 (CH89 and CH90). Dashed lines indicate the critical period with respect to ozone effects on final grain yield, e.g. from anthesis to grain filling. This period was about between mid June and mid July in both years. In 1989 this period was characterized by an AOT40 of 2033 ppb.h, whereas in 1990 the AOT40 for the same period was only 1609 ppb.h.

interaction with the effect of ozone must be quantified. For instance, f_{water} as the modifier for SMC can be entered as a function of precipitation, radiation, and temperature. These environmental variables can be included in the GIS for selected years. The outcome of the GIS-based calculation is a map of the selected assessment endpoint, for instance a map of potential yield loss in wheat (Fig. 13). According to the example for Switzerland, ozone risk is lowest in the central part of the country, and increases with elevation. Highest risk at low elevation sites occurred in the southern part (Ticino), whereas in the Valais, ozone effects are

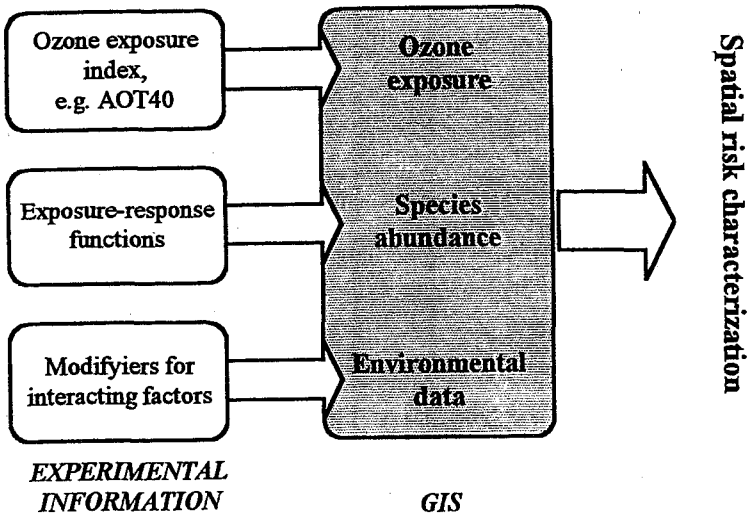


Fig. 12: Integrated risk characterization using a GIS-based approach.

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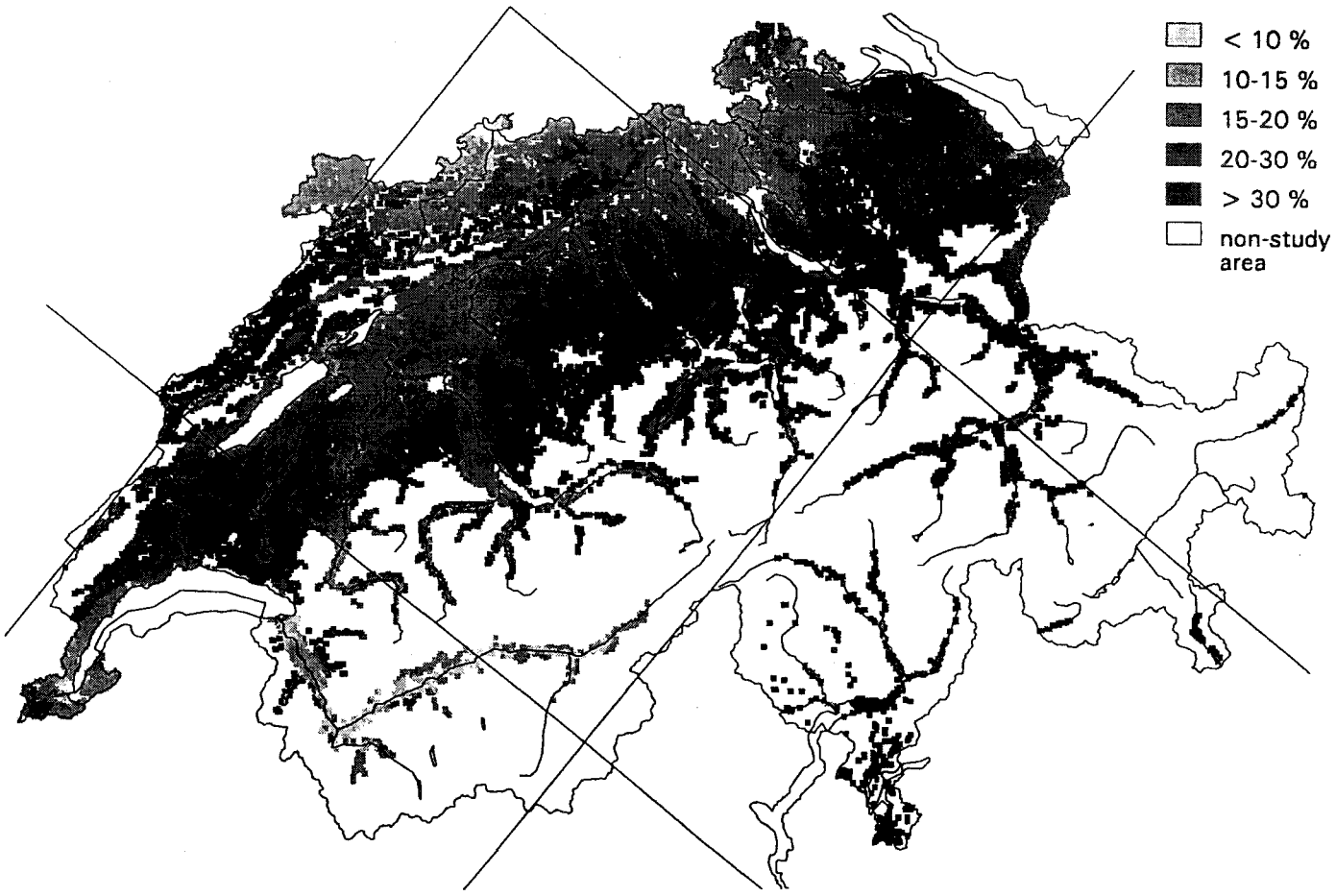


Fig. 13: Potential yield loss in wheat calculated for wheat growing areas of Switzerland in 1994. The calculation is based on maps for AOT40 and f_{water}, and the exposure-response relationship shown in Fig. 7 (from Fuhrer, 1995).

strongly reduced by low SMC. The same type of maps could be produced at the European scale, and it could be combined with maps for other receptors, such as other crops species, forests, or semi-natural vegetation. Before this is possible, the procedure to combine the different pieces of receptor-specific information must be developed.

5. Validation

The maps for integrated risk characterization are very useful and provide a illustration of the spatial distribution and extent of ozone risk. However, the questions raised in the Introduction remains: are there real effects in areas where effects should occur according to the maps, and how certain is the prediction? It is of utmost importance that maps of spatial risk distribution are being validated. However, validation is not easy. It requires experimental systems which can be used in the field and which provide ambient exposure and controls. Within the UN/ECE International Cooperative Programme 'Crops and Non-Wood Vegetation (ICP-Crops) indicator plants are exposed at a range of sites across Europe and ethylene diurea (EDU), a protective substance, is being applied as control treatment. Results show that ozone injury occurs at many locations, but effects of ozone on growth and yield (chronic effects) are more difficult to detect with this experimental technique (see Benton *et al.*, this volume). An alternative approach is to use clones differing in ozone sensitivity. A system with white clover clones developed in the USA is based on the yield ratio between a sensitive clone (NC-S) and a resistant clone (NC-R) (Heagle *et al.*, 1994). The ratio in dry matter yield between NC-S and NC-R is related to the ozone level, as shown in open-top chamber experiments and by field exposure. Results from a preliminary experiment conducted in southern Switzerland supported the po-

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tential use of the system in Europe. The yield ratio determined by three consecutive harvests averaged 0.73. This implies that under well-watered conditions the yield of the sensitive clone was reduced by 27% compared with the resistant clone, thus supporting the result of the calculation of potential yield loss for wheat. As shown in Fig. 13, the potential yield loss is predicted to be between 15 and 20% if the correction for SMC is taken into account. Eliminating the interacting effect of SMC, potential yield loss would be estimated at >30% (data for 1994). Hence, the use of a white clover indicator system can be used to validate the prediction of yield loss in selected areas.

Validation would also be extremely important in the case of the prediction of potential growth reductions in forest trees. However, this will be much more difficult than for crops, and possibilities of using biochemical or physiological markers instead of growth measurements should be investigated. In this case the assessment endpoint should be reconsidered, i.e. the critical level for forest trees should be based on the parameter used for validation.

6. Conclusions

This overview shows that ozone risk analysis is based on key elements, such as critical levels, modifying factors etc. However, to date the amount of information regarding these key elements differs between different receptor categories. The following summary is an attempt to identify the largest gaps in knowledge (Table 1). The picture shows that amount of information concerning crops is by far larger than for the other receptors. In the case of forest trees, the most important gaps are concerned with exposure-response relationships, modifying factors, and verification of risks. Also, the exposure index (AOT40 for 24 hours per day, expressed for one single season) should be critically re-assessed. For semi-natural vegetation, verification of risk and modifying factors are lacking. Finally, for fruit trees, very little information exists for all key elements. On the other hand, the procedures to map exceedance of critical levels, and for integrated risk analysis is available for all receptors, but it must decided, how to combine the information for the different receptors in one single assessment. One possibility would be to adopt the procedure used to map critical loads and critical load exceedances. This possibility should be tested in the near future. Regarding the main questions raised in the Introduction, the present summary suggests that (1) in the case of crops, growth and yield reductions are very likely to occur in areas where the critical level is exceeded. Effects on semi-natural vegetation

Table 1

Key elements for ozone risk analysis: present state of knowledge

Element	Crops (long-term)	Forest trees	Semi-natural Vegetation	Fruit trees
Endpoint(s)				
Exposure index				
Exposure-response data				
Modifying factors				
Receptor-specific mapping				
Integrated mapping				
Verification of risk				

Information:	
High	
Medium	
Low	
Missing	

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are also quite likely to occur, but the main difficulty lies in the extrapolation of data from short-term experiments to long-term changes in the botanical composition. In the case of forest trees, growth reductions may occur in sensitive species, such as beech, but this prediction is still highly uncertain. Furthermore, growth reduction may not be the appropriate endpoint for the assessment. For fruit trees, the few data suggest that effects on important plant characteristics may occur, but the degree of uncertainty is still extremely high.

7. Acknowledgments

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Ambient Ozone and Adverse Crop Response: An Evaluation of North American and European Data as They Relate to Exposure Indices and Critical Levels [1]

A.H. Legge [a], L. Grünhage [b], M. Nosal [c], H.-J. Jäger [b] and S.V. Krupa [d]

[a] Biosphere Solutions, Calgary, Alberta, Canada
[b] Institute for Plant Ecology, University of Giessen, Giessen, Germany
[c] Department of Mathematics and Statistics, University of Calgary, Calgary, Alberta, Canada
[d] Department of Plant Pathology, University of Minnesota, St. Paul, Minnesota, USA

Abstract

This paper provides an overview of present ambient critical levels, objectives, guidelines, etc., for ozone and a brief review of the current research efforts in the U.S. and in Europe (UN-ECE, Economic Commission for Europe) to relate O_3 exposure statistics to adverse crop yield responses. These studies are based on retrospective statistical analyses of data from the U.S. NCLAN (National Crop Loss Assessment Network) and the EOTCP (European Open-Top Chambers Programme). In these investigations, a sigmoidal O_3 concentration weighting function and/or different types of summation techniques have been used. For example, in the U.S., from a regulatory perspective, one of the descriptors of O_3 exposure is SUM06 (sum of all hourly concentrations above 60 ppb). Based on this statistic, it was predicted that a three month growing season SUM06 value of 26.4 ppm·hours of O_3 would protect 50% of the crops from yield losses of $>10\%$. In contrast, in the UN-ECE studies, an exposure index or critical level of AOT40 (Accumulated exposure Over Threshold of 40 ppb) was selected as the best predictor of adverse crop response and has been used to predict yield losses in spring wheat, green beans and clover.

Most recently we have also statistically analyzed both the NCLAN and the EOTCP data retrospectively. The results showed that the cumulative frequency of occurrence of intermediate or moderate hourly O_3 concentrations is important in crop yield loss response. While our NCLAN data analysis showed that the cumulative frequency of occurrence of hourly O_3 concentrations between 50 and 87 ppb is the best predictor of crop response in the U.S., the corresponding results from EOTCP showed a range of 35 to 60 ppb as being important in Europe. There is a debate at the present time as to whether peak or moderate O_3 concentrations are critical in causing crop yield reductions. The resolution of this issue is related to O_3 concentrations that occur when both the atmosphere and the phytosphere are sufficiently conductive for optimal O_3 flux (exposure potential) and uptake. In this context, summary results from a number of independent studies are reviewed to further underscore the significance of moderate O_3 concentrations.

In addition, results on the relevance of moderate or intermediate O_3 concentrations are explained in this paper with the use of an independent hybrid, statistical-mechanistic (micro-meteorological) approach. Here the concept includes the importance of ambient O_3 concentrations as they relate to the atmospheric conductivity and the physiological and

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functional properties of the plant. Further, we provide the numerical equations for applying this approach in O_3 measurement networks, the results of which, can be coupled with crop specific biology by using descriptors such as the "growing degree days." It is concluded that these types of efforts will be very valuable in significantly reducing the current controversy associated with the presently popular single point, statistical cause-effect models and the scaled uncertainties consequently associated with regional scale assessments of ambient O_3-induced crop loss through the application of such models.

Introduction

Ambient ozone is an all pervasive phytotoxic air pollutant (SCHÖNBEIN, 1844; for a review see e.g. KRUPA and MANNING, 1988). The processes that contribute to surface level O_3 at non-urban locations are: (a) transport of O_3 formed in the stratosphere into the free troposphere and subsequent transport down into the planetary boundary layer (PBL); (b) photochemical O_3 formation within the free troposphere and the clean PBL; (c) photochemical O_3 formation within the polluted PBL, especially during the passage of warm high pressure systems; and (d) O_3 formation within single or superimposed plumes (ALTSHULLER, 1986).

Background concentrations of O_3 observed at a number of so-called clean locations showed average daily 1 hour maxima of ~20-60 ppb (SINGH et al., 1977, 1978). To explain the behavior of O_3 in remote locations, SINGH et al. (1977, 1978) developed a schematic representation of variations in O_3 concentration by season. According to this scheme, natural O_3 formation is expected to be at a maximum during early spring. The changes in O_3 concentrations due to both local O_3 production and transport from urban centers result from photochemical phenomena, leading to maxima in summer and early fall. These conclusions of SINGH et al. (1977, 1978) have been supported by the findings of OLTMANS and KOMHYR (1976), LOGAN (1985) and more recently by the limited data presented by LEGGE et al. (1991).

As with other climatic variables, because of all the aforementioned considerations, ambient O_3 concentrations clearly vary in time (inter- and intra-annual, seasonal and diurnal) and space (longitudinal, latitudinal and elevational) (LEGGE and KRUPA, 1990). Because of these and other considerations such as crop biology and sensitivity, the relationships between O_3 exposure and crop response are inherently stochastic in their nature (KRUPA and TENG, 1982).

Traditionally plant responses to ambient O_3 and other air pollutant exposures are classified as acute and chronic. Acute responses involve rapid changes in the physiological and biochemical processes of the plant (SCHULTE-HOSTEDE et al., 1988). Under appropriate conditions, such changes frequently lead to the production of symptoms of foliar injury (KRUPA and MANNING, 1988). On broadleafed plants, symptoms of O_3 injury can include upper surface chlorosis, bronzing, flecking, bleaching, stippling and bifacial (both surfaces) necrosis. Acute responses are considered to be induced by relatively high hourly O_3 concentrations from a few to several hours on a given day or on recurring days, with the injury symptoms developing within a few to several days after such exposures. Most recent evidence suggests that acute responses appear to be due to moderately high, but not the highest hourly O_3 concentrations (TONNEIJCK and BUGTER, 1991; GRÜNHAGE et al., 1993; KRUPA et al., 1993). Most likely, this phenomenon is due to stomatal closure by the

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plant at high O_3 concentrations (U.S. EPA, 1986) as a protective reaction to drought stress, since the entry of O_3 into the leaves is primarily through the stomata (RUNECKLES, 1991). In addition, most recently GRÜNHAGE and JÄGER (1994a), GRÜNHAGE et al. (1994a) and GRÜNHAGE et al. (1996) showed that maximum amount of transfer (flux) of O_3 from the atmosphere to plant canopies (Exposure potential, *E*) occurs during periods of day when O_3 concentrations are in the intermediate range and not at peak or at the highest diurnal concentration (Table 1 and Figure 1). This subject is discussed further in a later section of this paper.

Tab. 1: Frequency distributions of atmospheric ozone exposure potential $E(O_3)$ versus O_3 concentration during the growing seasons (May - September) 1989-94 for daylight hours (global radiation ≥ 50 W m^{-2}). [a]

Range, ppb O_3	*Range, ppb m s^{-1} $E(O_3)$* [b]				
	≤1	*>1 - 2*	*>2 - 3*	*>3 - 4*	*> 4*
Beltsville, Maryland					
≤50	3956	871	122	10	2
>50 - 70	863	978	338	89	14
>70 - 90	403	668	326	116	24
>90 - 120	75	246	181	79	38
>120	4	16	14	3	2
Bondville, Illinois					
≤50	2836	1927	740	224	109
>50 - 70	595	1029	755	425	350
>70 - 90	137	342	293	173	189
>90 - 120	5	45	45	12	29
>120	0	0	0	0	3
Perkinstown, Wisconsin					
≤50	6861	1178	62	1	0
>50 - 70	674	715	174	24	2
>70 - 90	31	63	38	10	1
>90 - 120	0	1	0	1	0
>120	0	0	0	0	0
Georgia Station, Georgia					
≤50	3240	709	43	0	0
>50 - 70	930	956	169	14	1
>70 - 90	290	320	95	8	1
>90 - 120	56	67	28	4	0
>120	1	0	0	0	0

[a] Braunschweig, Germany data are not included here, because the original model (Figure 6) was developed based on those data.

[b] For details regarding the computation of $E(O_3)$, see the section on hybrid statistical-mechanistic method.

(Source: GRÜNHAGE et al., 1996)

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In contrast to acute responses, chronic effects are due to the exposures to O_3 for weeks, months, whole growing seasons and entire life cycles. Chronic exposures involve relatively low hourly O_3 concentrations, with periodic, intermittent, highly variable, comparatively higher O_3 concentrations. While acute responses may or may not lead to growth and yield reductions, chronic responses can lead to foliar chlorosis, retarded growth, foliar abscission, retarded flowering, flower abscission, reduced biomass and economic yield (KICKERT and KRUPA, 1991). Chronic effects are particularly important since growth and biomass effects can occur without attendant visible foliar injury.

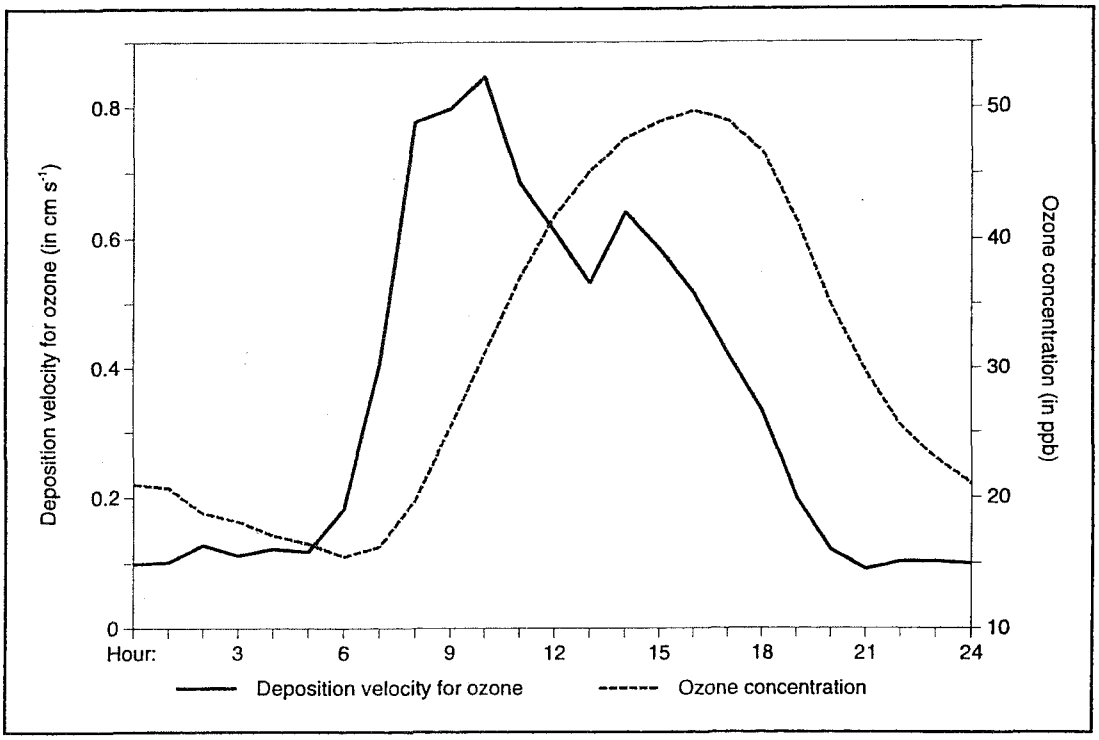


Fig. 1: Mean diurnal variations of O_3 deposition velocities and O_3 concentrations. Composite figure modified from Figures 3 and 7 of GRÜNHAGE et al. (1994a).

Critical levels versus the ambient air quality objectives/standards

In the efforts to assess the present and potential adverse impacts of air quality (e.g., O_3) on the environment (e.g., crops), scientists within the UN-ECE have applied the concept of critical levels. In contrast, in North America ambient air quality objectives (Canada) or standards (U.S.) have been in use.

Critical levels are defined as concentrations of pollutants in the atmosphere above which direct adverse effects on receptors, such as plants, ecosystems or materials, may occur according to present knowledge (UN-ECE, 1988; BULL, 1991; TEMANORD, 1994). A critical level is thus based solely upon the best available scientific knowledge and understanding. In contrast, ambient air quality objectives/standards in North America, while based upon the best available scientific knowledge and understanding, are balanced against social, economic and political considerations in the regulatory process. An additional difference between critical levels and ambient air quality objectives/standards is the

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fact that the critical levels approach explicitly assumes the existence of a "threshold" concentration above which there is a directly measurable adverse effect on sensitive components of the ecosystem. Ambient air quality objectives/standards do not assume the existence of a "threshold". Although one can debate the validity of utilizing the notion of a "threshold" concentration (WOODWELL, 1975; GRIGAL, 1991), the idea of such a "threshold" is fundamental to the application of the concept of critical levels. Another difference between critical levels and ambient air quality objectives/standards relates to how they are applied. Ambient air quality objectives/standards provide regulators with a measure of air quality for select air contaminants for the purpose of compliance with a legislation (IUAPPA, 1988). Critical levels are not used for compliance purposes. They are used as an integral part of pollutant emission control/abatement strategies. In this case, initially the geographic distribution of critical levels for selected sensitive ecosystem receptors is mapped. These data form the basis for testing emission control/abatement strategies with the objective of formulating a emission strategy that will reduce the ambient concentrations of the air contaminant of concern to the critical level or below.

In spite of the fact that there are major differences between the UN-ECE critical levels approach and the ambient air quality objectives/standards in North America, it is clear that the critical levels concept to environmental protection has merits. The most positive feature is that the critical levels philosophy addresses scientific issues in a proactive fashion and is not encumbered by social, economic and political considerations. This is in strong contrast to the situation encountered in the process of evaluating, revising and setting ambient air quality objectives/standards in North America. Here, social, economic and political considerations have a major impact on the scientific agenda. Nevertheless, it should be recognized that a strong and unbiased scientific foundation is the only way to reduce uncertainty in ensuring the protection of the environment from the adverse effects of air contaminants.

There is a long and somewhat convoluted history behind the critical levels approach to gaseous air contaminants, since that approach has evolved from the concept of critical and/or target loads relevant to acidic deposition. The critical levels approach is still in evolution. It has been recognized, for example, that the respective critical levels and critical loads for gaseous pollutants and acidic deposition must be addressed together, rather than separately. For additional details on these issues the reader is referred to: critical levels - NILSSON (1988); UN-ECE (1988); SVERDRUP et al. (1990); BULL (1991); GRIGAL (1991); ASHMORE and WILSON (1992); CAPE (1993); ASHMORE (1994); TEMANORD (1994); FUHRER and ACHERMANN (1994); RAITIO and KILPONEN (1994); and ASHENDEN et al. (1995); and ambient air quality objectives/standards - IUAPPA (1988).

Table 2 provides a comparative summary of the O_3 exposure values or concentrations as they relate to the critical level or ambient air quality objectives, standards and other regulations.

Although in the following sections of this paper no effort is made to provide a comparative analysis of the scientific basis for the specific values presented in Table 2 (this will constitute a major independent effort), emphasis is directed to an assessment of the current research on establishing cause (ambient O_3 exposure)-effect (adverse crop yield response) relationships.

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Tab. 2: Comparative summary of the O_3 concentrations **(ppb)** as they relate to the critical levels or ambient air quality objectives, standards and other regulations from select governing bodies.

Nation(s)	*Type of approach*	*Exposure duration (hours, as appropriate)*							
		0.5	*1.0*	*2.0*	*4.0*	*8.0*	*24.0*	*Seasonal*	*Annual*
Canada [a]	Objective								
(A) Max desirable		---	50	---	---	---	15	---	---
(B) Max acceptable		---	80	---	---	---	25	---	15
(C) Max tolerable		---	150	---	---	---	---	---	---
U.S. [b]	Standard	---	120 [c]	---	---	---	---	---	---
FRG [d]	Guideline								
(A) Sensitive		160	80	55	45	35	---	---	---
(B) Intermediate		240	160	120	95	80	---	---	---
(C) Less sensitive		400	240	200	185	160	---	---	---
Europe	Guideline	100 [e, f]	---	---	---	---	32.5 [e, f]	30 [e]	---
UN-ECE [g]	Critical level	Short-term (provisional) AOT40: 700 ppb·h accumulated over 3 consecutive days Long-term AOT40: 5300 ppb·h (10% yield loss) accumulated over 3 consecutive months							

[a] IUAPPA (1988)
[b] IUAPPA (1988)
[c] The standard is attained when the expected number of days per calendar year with a maximum hourly average concentration above 120 ppb is ≤ 1.
[d] VDI (1989)
[e] WHO (1987)
[f] EWG (1992)
[g] FUHRER and ACHERMANN (1994)

Current approaches to establishing cause-effect relationships: Statistical methods

According to JORDAN et al. (1988) the lack of correlation between vegetation exposure statistics which have been used in modeling and ambient air measurements in the U.S. has posed a major problem for those trying to assess the effects of ambient O_3 exposures. Any satisfactory cause-effect model must be general and universally applicable, if the results of the application of such a model will be used in a regulatory context on a broad scale. Such a goal can only be achieved if the model can satisfy the dynamics of the atmospheric and the corresponding plant physiological processes (KICKERT and KRUPA, 1991). However, most current statistical approaches to explain artificial O_3 exposure (primarily from open-top chamber treatments)-crop response relationships have not performed consistently between study sites, years and crops (ENVIRONMENTAL POLLUTION, 1988; HECK et al., 1988). This is because, among other things most such studies have used single point (one season end yield measurement) models (KICKERT and KRUPA, 1991). These types of efforts cannot account for feedback mechanisms among stress, repair and compensation by the plant receptor. In contrast, mechanistic-process or multi-point models can overcome most

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of the stated difficulties, but are generally hard to implement on a regional scale past the study plot level (KRUPA and TENG, 1982). In any efforts to overcome this difficulty, currently there are approaches available that deserve consideration, which involve hybrid statistical-mechanistic models (an example is described further in a later section of this paper).

Independent of the aforementioned brief discussion, numerous investigators have used statistical techniques to force relationships between cause (O_3 exposure) and effect (crop yield) (KICKERT and KRUPA, 1991). Most popular among these O_3 exposure statistics are: (a) SUM06(XX) = sum of all hourly O_3 concentrations above 60 (XX) ppb during the crop growing season (HOGSETT et al., 1995), and (b) W126 = a sigmoidal weighting function of hourly O_3 concentrations with an inflection point at 67 ppb and giving equal weight to values above 100 ppb (LEFOHN and RUNECKLES, 1987); both these exposure statistics have been used in the U.S.; and (c) AOT40 = Accumulated exposure Over a Threshold of 40 ppb, this statistic has been used in the UN-ECE studies (FUHRER and ACHERMANN, 1994).

According to HOGSETT et al. (1995), in the U.S. SUM06 values were determined from percentile distributions of average predicted relative yield losses of all crops previously computed by TINGEY et al. (1991) (Figure 2A). A three month SUM06 of 26.4 ppm·hours was the exposure index that would protect 50% of the crops from yield losses >10%. Another type of comparison was also made using an average predicted relative total biomass loss for all crops in the NCLAN (National Crop Loss Assessment Network) (LEE et al., 1988; TINGEY et al., 1991). This computation of the average exposure-response across all species is a means to predict the degree of protection afforded at various concentrations of O_3. A SUM06 value of 39.7 ppm·hours is predicted to protect 75% of the crops from 10% yield loss (LEE et al., 1988).

In comparison to the U.S. NCLAN results, according to the latest report by the UN-ECE-Agriculture Working Group (refer to FUHRER and ACHERMANN, 1994), the analyses presented by some of the UN-ECE scientists indicated that use of the threshold concentration of 40 ppb recommended by the UN-ECE Egham (U.K.) workshop in 1992 gave a good fit to experimental data. The use of higher threshold concentrations clearly gave a poor fit to the data. Overall, a threshold of 30 ppb gave correlation coefficients similar to those for 40 ppb. However, because 30 ppb is within the range of natural background concentrations, it was decided that the threshold concentration of 40 ppb should be retained. It was recommended that the index be referred to as the AOT40, to provide consistency between European and North American nomenclature.

Before deciding on the actual critical level for this exposure index, the group considered whether it should use wheat, bean or pasture data, or a combination of them, for its derivation. Although the bean data suggested a lower critical level value, it showed the poorest correlation between O_3 exposure and yield, partly because of yield stimulation in low O_3 treatments, while the pasture data was drawn from only one country. In contrast, the spring wheat relationship was the most comprehensive, covering five growing seasons, four countries and four cultivars. It therefore provided a much broader scientific basis on which to set the critical level. The O_3-AOT40-crop response relationship for spring wheat is illustrated in Figure 2B.

Good statistical relationships necessarily do not mean good cause-effect relationships (SNEDECOR and COCHRAN, 1978), unless they can explain the meaning in the context of the

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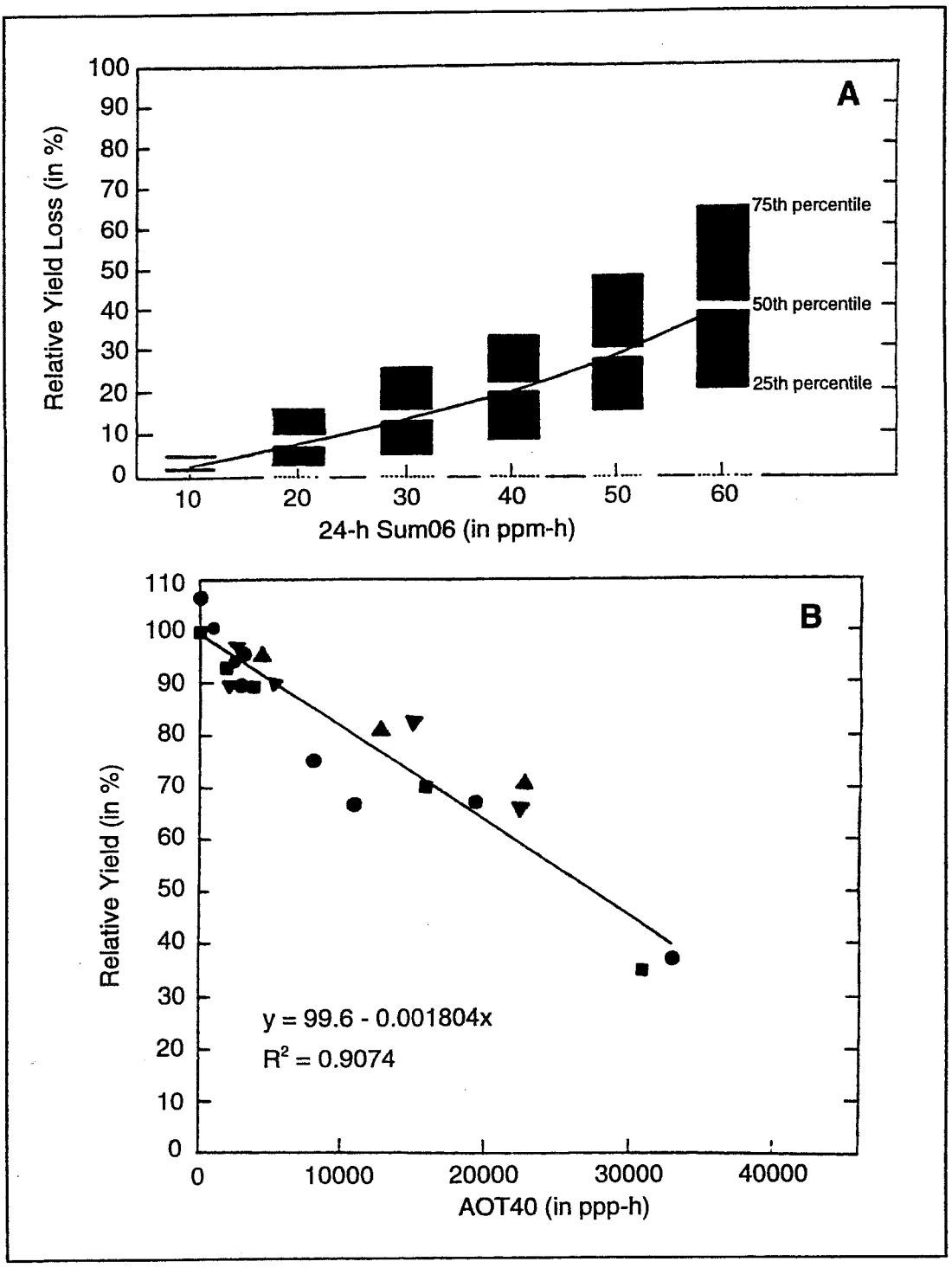


Fig. 2: **(A)** Box plot showing the biomass predictions from Weibull and linear exposure response models that relate to O_3 exposure characterized by the 24-hour SUM06 statistic using data from 31 crop studies of the U.S. NCLAN (Source: HOGSETT et al., 1995). **(B)** Relationship between relative grain yield of spring wheat and O_3 exposure expressed as AOT40 (Source: FUHRER, 1994).

properties or the behavior of the atmosphere and plant biology in this case. Nevertheless, both in the U.S. and in the UN-ECE, statistical definitions of the dynamics and stochasticity of ambient O_3 exposures have been largely ignored by summation and weighting functions. SUMXX and AOTXX are different types of summation techniques of hourly O_3 concentrations that negate the feedbacks among stress, repair and compensation. Weighting

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functions such as W126 differentially weight the mid-level concentrations, but fail to do so, with stationary or equal weighting of all high concentrations (>100 ppb). At the end, neither numerical definition considers the concept of probability: namely the right concentration of O_3 occurring at the right time of the day when both atmospheric and plant phytospheric conductivities are in conformity with each other (for details of this subject, see later sections of this paper).

Recently BÖHM et al. (1991, 1995) identified 17 characteristic patterns of O_3 exposure regimes in the western United States (Figure 3). Examination of these data by the authors using Bray-Curtis ordination analysis indicated that univariate statistics, such as 7-hour mean, 24-hour mean, SUM06 and SUM08, were unable to identify for up to 39% of variance in the data matrix. More studies of this type are needed to test the relevance of some of these exposure indices to ambient O_3 measurements, particularly in the context of the earlier statement from JORDAN et al. (1988).

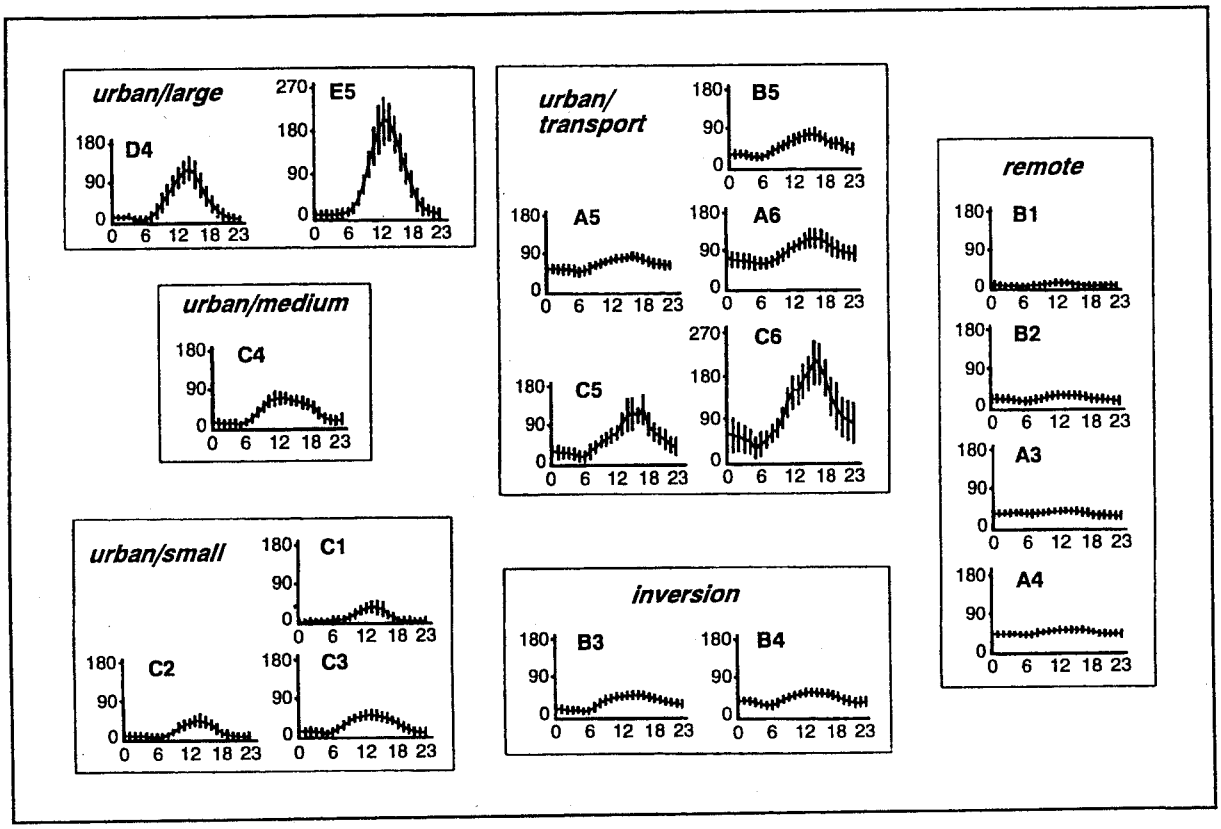


Fig. 3: Seventeen characteristic patterns of hourly O_3 concentrations (ppb) as determined by cluster analysis (Source: BÖHM et al., 1995).

Nevertheless, the numerous efforts in establishing O_3 exposure-crop response relationships in the U.S. have relied heavily on open-top chamber experiments involving above ambient, artificial O_3 exposures (refer to ENVIRONMENTAL POLLUTION, 1988; HECK et al., 1988). LEFOHN et al. (1989) described some of the difficulties and artifacts associated with the past experimental implementation of these enhanced O_3 treatments in the U.S. NCLAN and some aspects of the lack of relevance of such artificial exposures to the ambient exposure dynamics.

In contrast to the SUM06 or W126 approach in the U.S., UN-ECE scientists have used the

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AOT40 method (Figure 2B). A recent Scandinavian report (TEMANORD, 1994) concluded that in Scandinavia (defined as Sweden, Norway, Denmark and Finland) yield of spring wheat can be affected by O_3 levels below 40 ppb. According to the authors of that report, "A method to make a scientific sound translation of results from experiments in open-top chambers into the real field situation is now requested as the next step, in order to receive realistic estimations of O_3 induced crop loss all over Europe".

Ambient O_3 standards, objectives and guidelines are designed to control surface level O_3 concentrations and prevent adverse effects on human health and welfare. Although the application of exposure indices such as SUM06 and W126 and the critical level concept of AOT40 have relied heavily on crop response to artificially enhanced, above ambient O_3 levels, the critical question is: Are present day ambient O_3 exposures causing reductions in crop yield? This is a key issue in formulating emission standards and controls for the precursor pollutants of O_3 (NO_x and volatile hydrocarbons). To address this important question and in the absence of any other extensive data sets, KRUPA et al. (1994, 1995) and LEGGE et al. (1995) performed retrospective statistical analyses of the relationships between ambient O_3 exposures and cases of yield reductions in NF (non-filtered) compared to CF (charcoal filtered) open-top chamber treatments in the U.S. NCLAN and EOTCP (European Open-Top Chambers Programme) studies.

(A) *Summary results of the retrospective analysis of the NCLAN data*

According to KRUPA et al. (1994), within the U.S. NCLAN open-top chamber data, some 77% of the 73 crop harvests examined, showed **no** statistically significant yield differences between NF (non-filtered open-top chamber) and AA (chamberless ambient air) treatments (no easily discernable chamber effects on yield). Among these cases eight examples showed statistically significant yield reductions in NF compared to the CF (charcoal filtered open-top chamber) treatment. However, in these eight examples, one set of yield data proved to be abnormal compared to the other data for the same crop species and was therefore, deleted in the final analysis (total n = 7, see Table 3).

Tab. 3: Geographic locations, years and crops with yield in NF significantly lower than CF open-top chamber treatment in NCLAN (National Crop Loss Assessment Network) and EOTCP (European Open-Top Chambers Programme). [a]

NCLAN			*EOTCP*		
Location	*Year*	*Crop*	*Location*	*Year*	*Crop*
Ithaca, NY	1980	Kidney bean	*Belgium*	*1991*	*Wheat*
Ithaca, NY	1982	Wheat	*Denmark*	*1991*	*Wheat*
Ithaca, NY	1983	Wheat	*England (Imperial College)*	*1989*	*Bean*
Raleigh, NC	1983	Soybean	*Switzerland*	*1987*	*Wheat*
Raleigh, NC	1984	Clover: Yield No. 2 Clover: Yield No. 4	*Germany (Essen)*	*1988*	*Oats cv. Nova*
Raleigh, NC	1985	Clover: Yield No. 2			

[a] NF = non-filtered and CF = charcoal filtered treatments
(Source: KRUPA et al., 1994; LEGGE et al., 1995)

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Nevertheless, Table 4 provides summary O_3 statistics for the initial eight cases exhibiting yield losses in NF compared to CF treatments. A further examination of the summary statistics provided for the NCLAN data in Table 4 with the corresponding cases without yield loss showed that the mean, median and the various percentiles were all higher ($\geq$ 3 x) in the former in contrast to the latter scenario (KRUPA et al., 1994).

Tab. 4: Comparative summary statistics of ambient hourly O_3 concentrations **(ppb)** between the U.S. NCLAN (National Crop Loss Assessment Network) and EOTCP (European Open-Top Chambers Programme) for cases where crop yield of NF/CF[a] was < 1.0 (see also Table 3).

Statistic	*NCLAN (combined values)* [b]	*EOTCP (Range)*
Mean	36.94	16.61 - 29.79
S.D.	23.05	11.02 - 19.70
Median	35.00	14.00 - 27.00
1st quartile	19.00	5.00 - 20.00
3rd quartile	52.00	23.00 - 38.00

[a] NF = non-filtered and CF = charcoal filtered open-top chamber treatments
[b] Combined value from 8 cases exhibiting significantly lower yield in the NF compared to the CF open-top chamber treatment.
(Source: LEGGE et al., 1995)

The combined frequency distribution function (CDF) of hourly O_3 concentrations for the cases with yield loss in NF clearly separated from the corresponding distribution with no yield loss, at O_3 concentrations >47 ppb (Figure 4A and Table 5). Univariate linear regressions which were performed for the sake of simplicity between various O_3 exposure parameters and percent yield losses in NF showed that the cumulative frequency of occurrence of O_3 concentrations between 50 and 87 ppb was the best predictor (percent adjusted R^2 = 71.2 and p = 0.011) (Table 6). This analysis also showed that the frequency distribution of hourly concentrations up to 87 ppb O_3 represented a critical point, since the addition of the frequency distributions of >87 ppb O_3 did not improve the R^2 values. In fact as the frequency of hourly O_3 concentrations included in the regression moved toward 50-100 ppb, the R^2 value decreased substantially and the p value increased inversely (KRUPA et al., 1994).

(B) *Summary results of the retrospective analysis of the EOTCP data*

According to LEGGE et al. (1995), a comparison of NF (non-filtered open-top chamber) and AA (chamberless ambient air) crop yields to test for possible chamber effects was not possible with the EOTCP data due to a lack of consistent AA data. Nevertheless, the statistical t test of pair-wise comparisons between crop yield in NF and CF open-top chamber treatments for some 31 crop harvests within the EOTCP database, showed seven instances where the yield in the NF treatment was significantly lower than the corresponding yield in the CF treatment. Based on the previous results of KRUPA et al. (1994) with NCLAN data, preliminary linear regression analysis was attempted between the cumulative frequency distribution functions of hourly ambient O_3 concentrations at or

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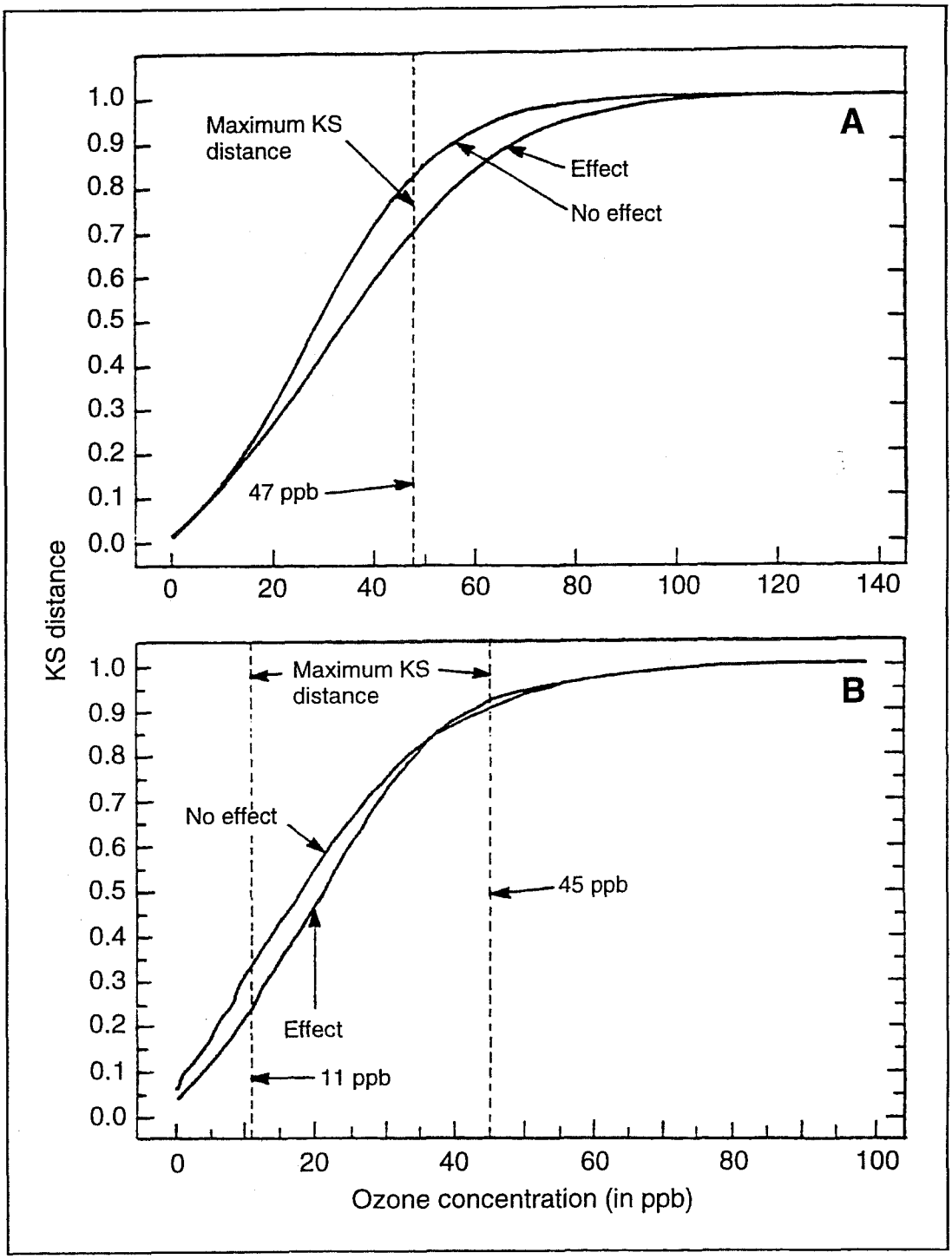


Fig. 4: Results of Kolmogorov-Smirnov least distance test (GIBBONS, 1976) to determine the differences between the cumulative ambient hourly O_3 frequency distributions for cases with yield reductions in NF vs. CF and cases without such an effect. **(A)** Results from the analysis of the U.S. NCLAN data (KRUPA et al., 1994) and **(B)** results from the analysis of the EOTCP data (LEGGE et al., 1995). Note the unimodal behavior of the NCLAN data and the bimodal behavior of the EOTCP data. Because of the scale used for plotting, the bimodal nature of the EOTCP data may not be readily obvious (see Table 5 for details).

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Tab. 5: Summary results of the test of significance of the differences in whole and conditional distribution functions in NCLAN and EOTCP respectively.

(A) NCLAN (National Crop Loss Assessment Network, US)			
Characteristic	0-87 ppb **(Figure 4A)**	Significance (*p*)	Conclusion
Point of minimum KS distance[a] **(ppb)**/value of the distance **(D^+)**	87/0	---	---
Point of maximum KS distance **(ppb)**/value of the distance **(D^+)**	47/0.117	0.0000	The O_3 CDF[b] for the **effects** and the **no effects** scenarios clearly separated from each other at concentrations above 47 ppb (unimodal distribution, Figure 4A)

(B) EOTCP (European Open-Top Chambers Programme)				
Characteristic	0-34 ppb **(Figure 4B)**	35-60 ppb **(Figure 4B)**	Significance (*p*)	Conclusion
Point of minimum KS distance **(ppb)**/value of the distance **(D^+)**	34/0	---	---	---
Point of maximum KS distance **(ppb)**/value of the distance **(D^+)**	11/0.097	---	0.0000	The O_3 CDF for 0-34 ppb for the **no effects** scenario is significantly larger than for the effects scenario (bimodal distribution, Figure 4B)
Point of minimum KS distance **(ppb)**/value of the distance **(D^+)**	---	60/0	---	---
Point of maximum KS distance **(ppb)**/value of the distance **(D^+)**	---	45/0.020	0.0000	The O_3 CDF for 35-60 ppb for the **effects** scenario is significantly larger than for the no effects scenario (bimodal distribution, Figure 4B)

[a] Kolmogorov-Smirnov distance (GIBBONS, 1976)
[b] CDF = **C**umulative frequency **D**istribution **F**unction
(Source: LEGGE et al. unpublished)

above 50 ppb and NF/CF (<1.0) crop yield. Such an effort was not very successful. Continued regression analysis using the entire spectrum of measured hourly ambient O_3 concentrations showed that the cumulative frequency distributions of hourly O_3 concentrations $\geq$35 ppb were the best predictors of adverse crop yield response in EOTCP (Table 6). Initially there were two clear, outlier data points in this regression. Therefore, these two cases were omitted from this and all additional efforts. Thus, the final "n" value was five cases (Table 3). Table 4 provides a comparison of the summary statistics of the ambient O_3

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concentrations for the five specific cases in EOTCP and the combined values for negative crop effects provided previously by KRUPA et al. (1994) from the U.S. NCLAN data. The mean, median, and 3rd quartile values of hourly ambient O_3 concentrations were all lower in the EOTCP compared to the U.S. NCLAN. Yet, there were crop yield reductions in NF compared to the CF treatment in both programs.

To examine the relatively low EOTCP O_3 exposure summary statistics better, a comparison of the O_3 summary statistics was made between cases in EOTCP with crop yield reductions in the NF compared to the CF treatment and corresponding cases with no differences between the two treatments (similar locations and crops, but different years) (LEGGE et al., 1995) This comparison did not provide any readily visible differences between the two sets of cases. Missing here is a clarification of any possible differences in the frequency distributions of exposure concentrations between the two types of differing cases of response. Therefore, Kolmogorov-Smirnov non-parametric least distance test (GIBBONS, 1976) was used to examine the differences in the O_3 exposure dynamics. The results showed that the two cumulative frequency distributions merged at 34 ppb and separated at concentrations ≥ 35 ppb (Figure 4B). This supports the results presented in Table 6 showing that the cumulative frequency distribution function of hourly O_3 concentrations ≥ 35 ppb (CDF35) was the best predictor of adverse crop yield response. With regard to the data presented in Figure 4B, it is important to note that the frequency of occurrence of hourly ambient O_3 concentrations > 60 ppb was relatively low at all study sites. The spatial and temporal variabilities in the observed daily patterns of ambient air concentrations of O_3 are most likely governed by the complex interactions involving: (a) proximity or relatively closely clustered urban centers within Europe and their pollutant emissions; (b) local patterns of photochemical O_3 production; (c) patterns of long-range transport; and (d) types and patterns of atmospheric O_3 scavenging reactions. Nevertheless, additional fine mesh (decrements or increments by 1 ppb from the 35 ppb O_3) regression analyses were performed between the cumulative frequency densities of O_3 at and above various concentrations on either side of 35 ppb and crop response NF/CF $= < 1.0$ (refer to Table 6). The results showed a decline in the adjusted R^2 values and a corresponding increase in p values on either side of 35 ppb O_3. Again, it can be concluded from these results that the cumulative frequency of occurrence of ambient O_3 concentrations ≥ 35 ppb is the best predictor of NF/CF crop response < 1.0 in EOTCP.

From the previous results based on NCLAN and EOTCP data, the application of cumulative O_3 frequency distribution functions shows the importance of moderate hourly concentrations in inducing adverse crop yield response. However, it does not negate the importance of peak hourly O_3 concentrations, if such levels occur during periods when both optimal atmospheric and plant conductivities coincide. The relative importance of peak versus moderate O_3 concentrations in causing yield reduction is presently a major area of controversy (MUSSELMAN et al., 1994; KRUPA et al., 1995; NUSSBAUM et al., 1995). Nevertheless, it is important to note that as O_3 concentrations reach high levels, the corresponding frequency of their exposure potential [$E(O_3)$] decline because of the nature of atmospheric conditions (GRÜNHAGE and JÄGER, 1994*a*; KRUPA et al., 1995; Table 1 in this paper, as an example).

As opposed to the summation (SUM06 and AOT40) and weighting (W126) functions, CDF (Cumulative frequency Distribution Function) does include a probability for the spatial and temporal variabilities in the occurrences of hourly O_3 concentrations. However, it does not include a time element, particularly in the context of varying crop responses to O_3 with varying physiological growth stages (Kickert & Krupa, 1991). Effort is presently in progress to address this issue. Most importantly, the CDF approach allows a comparison of results

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from different experiments with clearly differing durations.

Tab. 6: Summary results of linear regression analyses between NF/CF open-top chamber yield <1.0 in the NCLAN (National Crop Loss Assessment Network) and EOTCP (European Open-Top Chambers Programme) and the frequency density of hourly O_3 concentrations of varying ranges.

NCLAN			*EOTCP*		
Range, ppb O_3	*% Adj. R^2*	*p*	Range, ppb O_3[a]	*% Adj. R^2*	*p*
50-80	57.0	0.030	*>30*	*56.0*	*0.090*
50-81	57.3	0.030	*>31*	*66.1*	*0.059*
50-82	58.9	0.027	*>32*	*74.2*	*0.038*
50-83	61.0	0.023	*>33*	*78.7*	*0.029*
50-84	65.0	0.018	*>34*	*79.8*	*0.026*
50-85	63.9	0.019	*>35*	*81.5*	*0.023*
50-86	69.1	0.013	*>36*	*72.5*	*0.043*
50-87	**71.2**	**0.011**	*>37*	*70.2*	*0.048*
50-88	68.8	0.013	*>38*	*66.6*	*0.058*
50-89	62.9	0.020	*>39*	*69.1*	*0.051*
50-90	.57.9	0.029	*>40*	*61.2*	*0.074*
			>41	*63.9*	*0.066*
			>42	*60.7*	*0.075*

[a] The upper bound in the range is **60 ppb** and only 2.5-5.0% of the observations were above this value. See also Table 5 for conclusions from the EOTCP data analysis.

(Source: KRUPA et al., 1994; LEGGE et al., 1995).

KRUPA et al. (1994, 1995) and LEGGE et al. (1995) used simple, univariate, linear regressions in establishing cause-effect relationships for easy understanding in a regulatory framework. It is clearly recognized that the frequency distributions of ambient hourly O_3 concentrations do not exhibit a normal distribution and by nature crop response follows an asymptotic behavior. Under these conditions, use of a linear regression is statistically inappropriate. Therefore, in the context of scientific correctness, efforts are underway using non-linear regressions to apportion the importance of the **C**umulative frequency **D**istribution **F**unctions (CDFs) of all measured O_3 values versus CDFs of daily periods supporting high O_3 uptake by the crop (Figure 1A and Table 1). Such efforts, however, are limited by the single point plant response (season-end yield, with no time series measurements of growth dynamics) assessments. Nevertheless, this is a good start in ambient O_3 exposure-crop yield response analysis.

An alternative approach to establishing cause-effect realtionships: A hybrid statistical-mechanistic method

The application of O_3 exposure-plant response relationships to ambient conditions based on open-top chamber experiments has led to controversial and unsatisfactory results (KRUPA et al., 1995). Because of the air circulation conditions inside the chambers, it can be assumed that the pollutant gas fumigation concentrations reflect the concentrations at the

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plant envelope[2] (GRÜNHAGE and JÄGER, 1994a). In contrast, under ambient conditions it is physically impossible to measure the O_3 concentrations at the envelope of the phytosphere. In fact under field conditions, O_3 concentrations are normally measured at a standard measurement height (e.g., 3.5 or 5.0 m; Figure 5).

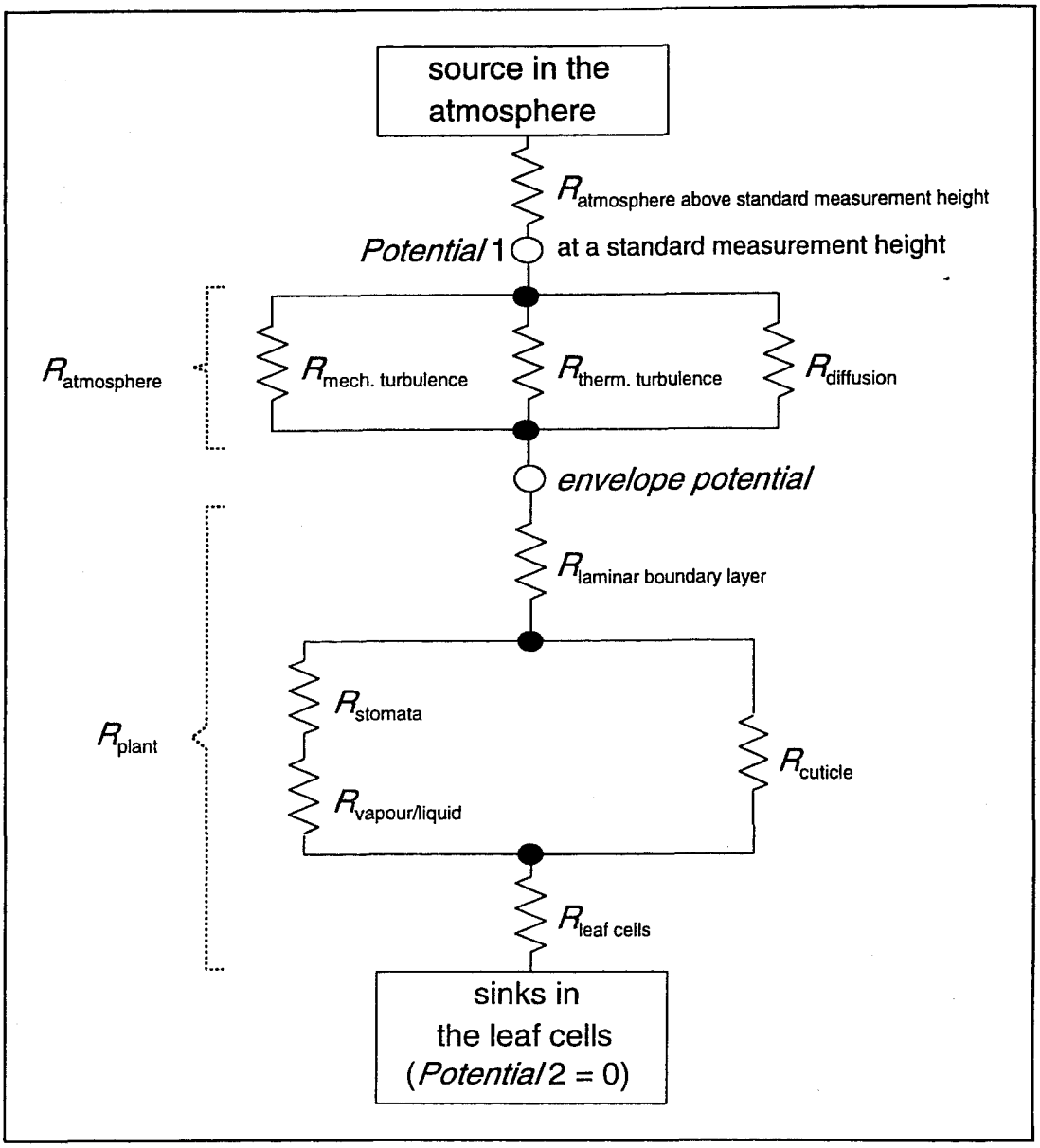


Fig. 5: A schematic representation of the resistance analogue of the O_3 flux densities to plants (modified from: GRÜNHAGE and JÄGER, 1994a).

Micrometeorological measurements of the exchange of O_3 between the atmosphere and the phytosphere show that there is a sharp decline in the O_3 concentration profile above the canopy (e.g., ETLING, 1987). Therefore, it is necessary to transform the O_3 concentrations measured at a standard measurement height into concentrations at the plant envelope

[2] The upper surface of the laminar boundary layer.

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(envelope potentials, for details see the later parts of this section) according to:

$$\textit{envelope potential} = a \cdot \textit{potential}_{\text{at a standard measurement height}}$$

where, a = the transformation factor

The envelope concentrations themselves are determined by both $R_{atmosphere}$ and R_{plant} (Figure 5). Therefore, the transformation factor depends on the turbulence of the atmosphere and on the biological activity of the plants. Such transformation factors can be deduced from micrometeorological flux measurements. From these types of measurements of the O_3 exchange between the atmosphere and a permanent grassland ecosystem, GRÜNHAGE and JÄGER (1994a) derived transformation factors with a median value of 0.8.

As an example, Table 7 provides observed mean yield of spring wheat relative to its yield in 1984, the year with the highest mean yield during the last 20 years, for the federal state of Hessia, Germany. In addition, the table shows estimates of yield losses due to O_3 using the UN-ECE exposure-response model for spring wheat (FUHRER, 1994) and estimates from O_3 concentrations measured at a height of 3.5 m above ground and transformed to the envelope potential. The observed high yield losses in 1987 and 1989 coincided with a low O_3 burden (Table 7) and therefore, was indicative of the influence of the physical climate on biomass production. For the years with a high O_3 burden (1992, 1994 and 1995) the application of the UN-ECE exposure-response model led to an overestimation of O_3-induced yield losses. Using a factor of 0.8 to transform the measured O_3 concentrations to the concentrations at the plant envelope resulted in more conservative or realistic yield loss estimates.

Tab. 7: Mean yield losses in spring wheat in Hessen, Germany.

Year	$\bar{x}$ *yield loss (Hessen)* [a]	$\bar{x}$ *potential yield loss (UN-ECE)* [b]	$\bar{x}$ *potential yield loss (e.p)* [c]
1985	17.1	0.7	-0.3
1986	10.6	8.5	3.7
1987	25.4	1.2	0.1
1988	17.1	4.5	1.3
1989	33.7	5.0	1.3
1990	20.8	10.0	4.3
1991	1.0	1.3	0.2
1992	7.5	23.4	10.7
1993	14.5	15.1	7.1
1994	14.0	23.2	11.4
1995	17.4	32.3	14.7

[a] Observed % mean yield losses in Hessen relative to the yield in 1984 (Hessisches Statistisches Landesamt).

[b] Potential % mean yield losses using the UN-ECE exposure-response model (FUHRER, 1994) with regard to the **O_3 burden** in 1984.

[c] Potential % mean yield losses using the UN-ECE exposure-response relationship (Fuhrer, 1994) with regard to the **O_3 burden** in 1984 taking into account a **factor of 0.8** to transform the O_3 concentration measured at a height of 3.5 m to the plant envelope potential (*e.p*) (GRÜNHAGE et al., 1994b, updated).

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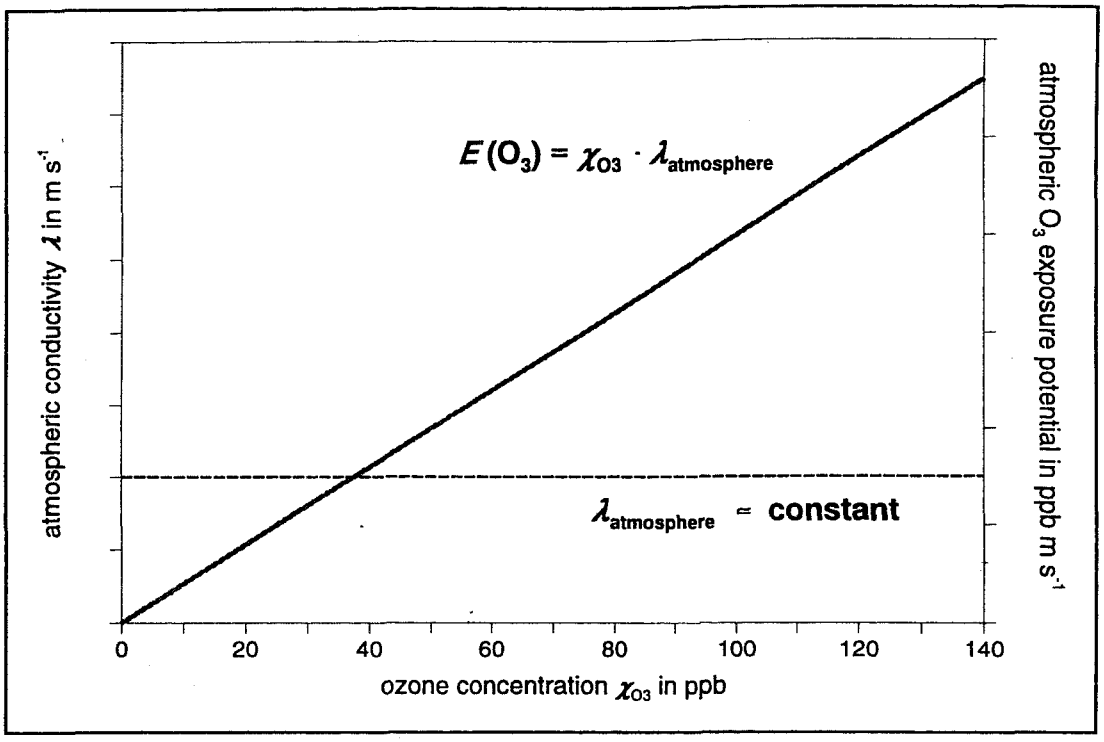


Fig. 6: A schematic representation of the relationship between atmospheric conductivity and O_3 concentrations in an open-top chamber environment (Source: KRUPA et al., 1995).

On the other hand, in contrast to the ambient environment, under ***artificial*** chamber conditions, since the air-flow is virtually constant, the exchange of O_3 between the chamber atmosphere and the plant canopy will be relatively uniform (Figure 6). Under these conditions, up to a point, increasing O_3 concentrations in the chamber atmosphere will result in proportionate or disproportionate increases in crop response (ENVIRONMENTAL POLLUTION, 1988; HECK et al., 1988). Based on such controlled exposure experiments in artificial chamber environments, MUSSELMAN et al. (1994) and NUSSBAUM et al. (1995) concluded that higher hourly average concentrations have the potential to cause disproportionately greater injury or damage than mid- and low-level hourly average values.

(A) There are several studies under ambient conditions and as previously discussed, a re-assessment of U.S. NCLAN and EOTCP results which indicate that O_3 exposure conditions in open-top chambers do not reflect the ambient exposure conditions:

- TONNEIJCK and BUGTER (1991) found a tendency for a reduced sensitivity of tobacco cv. Bel-W3 plants to relatively high O_3 concentrations in the Netherlands.

- KRUPA et al. (1993) found that sums of all hourly concentrations >40 ppb (SUM40) and >60 ppb (SUM60) jointly proved to be the best predictors of median value of foliar injury on tobacco cv. Bel-W3 at two geographic locations in northeast USA. Neither SUM40 nor SUM60 was effective when used alone, although SUM40 was more important than SUM60. Sum of all hourly concentrations strictly >70 ppb or >80 ppb were not as important in explaining the cause-effect relationship.

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- The results of a combination of measured and modeled frequency distributions of differing percentages of leaf area injured on tobacco cv. Bel-W3 at various ambient concentrations of O_3, converted to their corresponding flux densities, showed that the ambient O_3 concentrations between 50 and 90 ppb are likely to have the most effect and may be more important than concentrations >90 ppb (GRÜNHAGE and JÄGER, 1994b).

(B) Several studies show that O_3 uptake and/or foliar injury or yield reduction due to O_3 are highest at times when or where the ambient O_3 concentrations are moderate:

- GRÜNHAGE et al. (1994a), SCHMITT et al. (1995): Above two grassland ecosystems the diurnal pattern of O_3 deposition velocities reached peak values before noon, whereas the diurnal pattern of O_3 concentrations attained peak values during the afternoon.

- GRULKE cited by SKELLY (1995): The data of GRULKE showed that trees of giant sequoia responded to low (ambient) values of O_3, not just high ones.

- SHOWMAN (1991): Field observations indicated that foliar injury in O_3 sensitive species was widespread in a moist year with low O_3 levels, whereas injury in a dry year with high O_3 concentrations were rare.

- WIESER and HAVRANEK (1993): The diurnal pattern of O_3 flux densities from the air to the needles of Norway spruce peak in the late morning, whereas the diurnal pattern of O_3 concentrations peak in the afternoon.

- WIESER and HAVRANEK (1995): When ambient O_3 concentrations were highest, O_3 flux to the needles of European larch tended to be restricted by narrowing of the stomatal opening.

(C) OJANPERÄ et al. (1994) observed in open-top chamber experiments that a 40 ppb threshold did not provide a significant correlation with changes in the grain yield of spring wheat, while the inclusion of all O_3 concentrations below 40 ppb provided an R^2 value of 0.93.

(D) KRUPA et al. (1994) concluded by developing a statistical relationship between ambient O_3 concentrations and yield reductions in appropriate NF compared to CF open-top chamber treatments in the U.S. NCLAN that the frequency of occurrence of mid-range O_3 concentrations was important in causing adverse chronic effects on crops.

(E) LEGGE et al. (1995) in their retrospective analysis of the data from the European Open-Top Chambers Programme found that the cumulative frequency distribution of O_3 concentrations between 35 ppb and 60 ppb provided the best explanation for O_3 induced crop yield reductions.

In the context of all the aforementioned observations, the exchange of gases between the atmosphere and the phytosphere is controlled by both the exchange properties of the atmosphere and the sink properties of the plants (Figure 5). The main pathway of entry of O_3 into the plant is through their stomata (RUNECKLES, 1991). The mechanism of transport between the upper surface of the laminar boundary layer of the leaf exterior and the interior

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leaf tissue is diffusion. The laminar boundary layer is predominantly induced by the properties of the plant (e.g., hairs on the epidermis) or the canopy. At the upper surface of the laminar boundary layer the mean horizontal wind velocity becomes zero. Therefore, this surface represents a sensible boundary between the phytosphere and the atmosphere. It is referred to as the ***envelope*** of the plant/soil system. The O_3 concentration at this surface is the ***envelope potential*** (GRÜNHAGE and JÄGER, 1994a; GRÜNHAGE et al., 1995).

Toxicologically relevant are fluxes of ozone into the plant rather than concentrations (DÄMMGEN et al., 1993). It is generally agreed that the amount of O_3 diffusion into the leaf depends on the difference between the O_3 concentration at the envelope of the plant and the O_3 concentration inside the leaf cells. It is convenient to assume that there are sinks in the plant which reduce the O_3 concentration in the leaf tissue to zero (LAISK et al., 1989; WANG et al., 1995). The absorbed dose $F(O_3)$[3] can then be described by the following equation:

$$F(O_3) = -\frac{\textit{envelope potential} - \textit{potential 2}}{R_{plant}} = -\frac{\textit{envelope potential}}{R_{plant}}$$

where, *potential* 2 = O_3 concentration inside the leaf

If an O_3 molecule which diffuses into the leaf is not replaced directly by another one, the absorbed dose will decrease. Assuming a constant R_{plant} the relatively constant air circulation inside an open-top chamber will lead to a constant O_3 uptake at a given exposure concentration most of the time.

Under ambient conditions, the replacement of an O_3 molecule which diffuses into the leaf by another one, depends on the exchange properties of the atmosphere. The resistance of the atmosphere $R_{atmosphere}$ describes the turbulence of the atmosphere that is driven by both mechanical and thermal forces. The latter intensifies the mechanically induced turbulence during periods of atmospheric heating during daylight hours (unstable atmospheric stratification), whereas it weakens mechanically induced turbulence during cooling periods especially during the night (stable atmospheric stratification). The transport by diffusion in the atmosphere can be neglected under turbulent conditions. In a chamberless environment the absorbed dose $F(O_3)$ can then be described by the following equation:

$$F(O_3) = -\frac{\textit{potential 1} - \textit{potential 2}}{R_{atmosphere} + R_{plant}} = -\frac{\textit{potential 1}}{R_{atmosphere} + R_{plant}}$$

where, *potential* 1 = O_3 concentration at a standard height above the canopy
potential 2 = O_3 concentration inside the leaf

Under the assumption that R_{plant} becomes zero, the ratio of the O_3 concentration at a measurement height above the canopy (***potential*** 1 or χ_{O_3}) and $R_{atmosphere}$ or the product of χ_{O_3} and the atmospheric conductivity $\lambda_{atmosphere}$ reflects the maximum possible absorbed dose under the atmospheric conditions at any given moment. Therefore, this product was defined by GRÜNHAGE and JÄGER (1994a) as the O_3 exposure potential of the atmosphere $E(O_3)$ for plants:

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[3] In the micrometeorology literature, it is conventional to describe uptake with negative values in comparison to emissions which are assigned positive values.

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$$E(O_3) = \chi_{O_3} \cdot \lambda_{atmosphere}$$

where, χ_{O_3} = O_3 concentration at the measurement height above the canopy
$\lambda_{atmosphere}$ = conductivity of the air column

Micrometeorological studies show that O_3 uptake depends on stomatal as well as on atmospheric conductivities (e.g., MONTEITH and UNSWORTH, 1990; GRÜNHAGE and JÄGER, 1994a). It is well known that stomatal conductivity is influenced by the water status of the plant/soil system as well as by the water vapor pressure deficit of the atmosphere (***VPD***). If the plant/soil system has a restricted water balance, high atmospheric water vapor pressure deficits will result in a closure of the stomata. Evaluations of German and U.S. data sets by GRÜNHAGE and JÄGER (1994a) and GRÜNHAGE et al. (1996) showed that both in Germany and in the U.S. high O_3 concentrations coincide normally with high air temperatures and therefore, high water vapor deficits. Similarly high O_3 concentrations often coincide with low to moderate horizontal wind velocities. It is apparent that under these conditions high O_3 concentrations occur during moderate to low atmospheric conductivities (Figure 7). The isolines in Figure 7 represent combinations of χ_{O_3} and $\lambda_{atmosphere}$ resulting in similar $E(O_3)$. For example, following the isoline $E(O_3)$ = 4.5 ppb m s^{-1} O_3, one can deduce that e.g., 120 ppb O_3 and 60 ppb O_3 can result in the same amount of uptake assuming that the conductivity of the plant is equal during both situations.

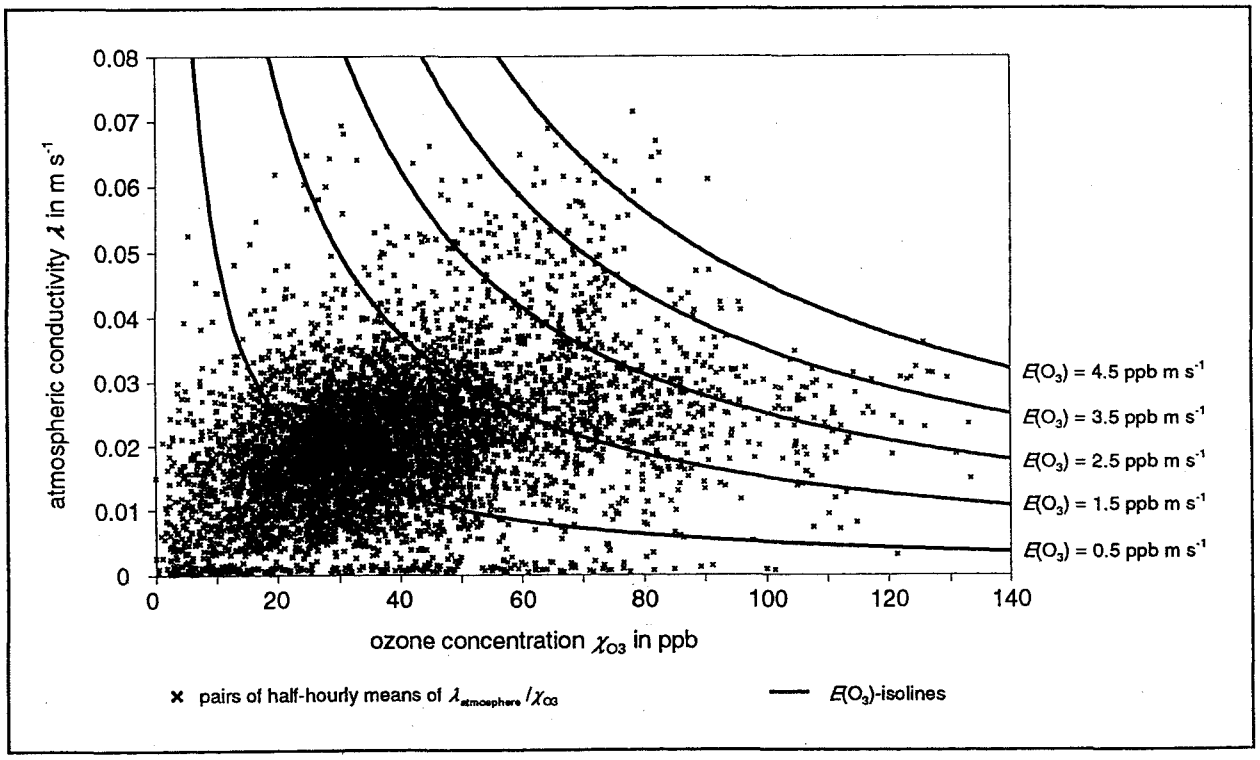


Fig. 7: Relationship between atmospheric conductivities and ambient O_3 concentrations (growing seasons, May - September, 1990 and 1991, global radiation ≥ 50 W m^{-2}, study location - Braunschweig, Germany (Source: GRÜNHAGE and JÄGER, 1994a).

Therefore, under chamberless conditions dose-effect relationships can only be established if both the exchange properties of the atmosphere and the sink properties of the leaves are taken into account. In the literature several approaches for calculating $R_{atmosphere}$ and R_{plant} have been

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published (BALDOCCHI et al., 1987). A very simple approach for calculating the atmospheric conductivity $\lambda_{atmosphere}$ was developed by GRÜNHAGE and JÄGER (1992, 1994*b*) for use in the air quality monitoring networks in Germany (Table 8). This model reflects that the atmospheric turbulence is driven by both mechanical and thermal forces. The model uses weighted mean horizontal wind velocities u measured at a height of 10 m. The weighting factors are based on global radiation G and air temperature T in order to account for atmospheric stratification. In the model, atmospheric instability at a time n is attributed to the increase in temperature between the previous data set (n-1) and the current data set (n).

Tab. 8: Model for calculating the conductivity of the atmosphere using weighted mean horizontal wind velocities[a].

$$\lambda_{atmosphere} = u(n) \cdot [a + \{T(n) - T(n-1)\} \cdot b]$$

Four cases are distinguished:

(I)	$G = 0$ W m^{-2}	:	a = 0.0025	$b = 0$ K^{-1}
(II)	0 W m^{-2} $< G \leq$ 200 W m^{-2}	:	a = 0.0045	$b = 0$ K^{-1}
(III)	$G >$ 200 W m^{-2} and $[T(n) - T(n-1)] \leq 0$ K	:	a = 0.0080	$b = 0$ K^{-1}
(IVa)	For data sets with a resolution in time of 0.5 hour $G >$ 200 W m^{-2} and $[T(n) - T(n-1)] > 0$ K	:	a = 0.0080	$b = 0.004$ K^{-1}
(IVb)	For data sets with a resolution in time of 1 hour $G >$ 200 W m^{-2} and $[T(n) - T(n-1)] > 0$ K	:	a = 0.0080	$b = 0.002$ K^{-1}

[a] u = mean horizontal wind velocity at 10 m; G = global radiation at 2 m; T = air temperature at 2 m. (Source: Grünhage & Jäger, 1994b).

KÖRNER et al. (1995) developed an approach for calculating leaf conductivities λ_{leaf} for O_3 during the vegetative period as a function of radiation, water vapor pressure deficit of the atmosphere and time of day. The model reflects the fact that the importance of radiation and water vapor pressure deficit of the atmosphere change during the day. During the early phase in the morning, the leaf conductivity is dominated by radiation. On sunny days, during the late morning the control is taken over by the pressure deficit. In the late afternoon additional factors are important (KÖRNER and COCHRANE, 1985). As an example, Table 9 summarizes the calculation procedure used for *Picea abies*.

From these models, O_3 uptake can be calculated by means of O_3 concentration, horizontal wind velocity, relative humidity, air temperature and radiation, which are normally measured by most air quality monitoring networks, using the following equation:

$$F(O_3) = -\chi_{O_3} \cdot \left[\frac{1}{\lambda_{atmosphere}} + \frac{1}{\lambda_{leaf}} \right]^{-1}$$

where, $\lambda_{atmosphere}$ = conductivity of the atmosphere (e.g. Table 8 in this paper)
λ_{leaf} = conductivity of the "big leaf" (e.g. Table 9 in this paper)

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Tab. 9: Model for calculating leaf conductivities for O_3

First step: The maximum stomatal conductance g_{max} (in mmol $H_2O \cdot m^{-2}$ needle surface area $\cdot s^{-1}$) for a plant species is weighted by empirical species dependend monthly weighting factors resulting a potential leaf conductance g_{pot} (in mmol $m^{-2} s^{-1}$).

$$g_{pot}(H_2O) = g_{max}(H_2O) \cdot f(\text{month})$$

e.g., *Picea abies*: $g_{max}(H_2O)$ = 55 mmol $H_2O \cdot m^{-2}$ needle surface area $\cdot s^{-1}$

f (April) = 0.82 f (May) = 0.91 f (June) = 1.0 f (July) = 1.0
f (August) = 0.91 f (September) = 0.82 f (October) = 0.73

Second step: Consideration of the effect of radiation on the stomatal conductance resulting in a species dependend radiation weighted potential leaf conductance $g_{radiation}$ (in mmol $m^{-2} s^{-1}$).

$$g_{radiation}(H_2O) = \frac{g_{pot}(H_2O) \cdot a \cdot (radiation + b)}{g_{pot}(H_2O) + a \cdot (radiation + b)}$$

e.g., *Picea abies*: a = 2.25 b = 0.333 *radiation* in μmol $m^{-2} s^{-1}$

Third step: Consideration of the effect of water vapor pressure deficit on the stomatal conductance resulting in a species dependend *VPD* weighted potential leaf conductance g_{VPD} (in mmol $m^{-2} s^{-1}$).

if $VPD < a$ **then:** $g_{VPD}(H_2O) = g_{radiation}(H_2O)$

if $a \leq VPD \leq b$ **then:** $g_{VPD}(H_2O) = g_{radiation}(H_2O) \cdot [(1 + 10c) - (c \cdot VPD)]$

if $VPD > b$ **then:** $g_{VPD}(H_2O) = g_{radiation}(H_2O) \cdot [(1 + 10c) - (c \cdot b)]$

e.g., *Picea abies*: a = 10 mPa Pa^{-1} b = 40 mPa Pa^{-1} c = 0.029

Fourth step: Consideration of the time of day on the stomatal conductance resulting in a species dependend daytime weighted potential leaf conductance g_{leaf} (in mmol $m^{-2} s^{-1}$).

before 15.00 h: $g_{leaf}(H_2O) = g_{VPD}(H_2O)$
after 15.00 h: $g_{leaf}(H_2O) = g_{VPD}(H_2O) \cdot f(\text{daytime})$

e.g., *Picea abies*:
if *VPD* (15.00-16.00) < 12 mPa Pa^{-1} then f (time) = 1

if 12 mPa $Pa^{-1} \leq$ *VPD* (15.00-16.00) $\leq$ 20 mPa Pa^{-1}
then f (time) = 15^h-16^h 0.88; 16^h-17^h 0.75; 17^h-18^h 0.63; 18^h-19^h 0.5; 19^h-20^h 0.4

if *VPD* (15.00-16.00) > 20 mPa Pa^{-1}
then f (time) = 15^h-16^h 0.90; 16^h-17^h 0.80; 17^h-18^h 0.70; 18^h-19^h 0.6; 19^h-20^h 0.5

Fifth step: Conversion of the stomatal conductance for water vapor into the stomatal conductance for ozone. [a]

$$g_{leaf}(O_3) = \frac{D_{O3}}{D_{H2O}} \cdot g_{leaf}(H_2O) = 0.613 \cdot g_{leaf}(H_2O)$$

where, D_x = diffusion coefficient

[a] $g_{leaf}(O_3)$ must be transformed in the dimension m s^{-1} resulting $\lambda_{leaf}(O_3)$.
(Source: KÖRNER et al., 1995)

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A logical step beyond this calculation is to apply the results to geographically resolved crop specific biology using descriptors such as the "growing degree days" (GDD). Growing degree days are calculated by using the following equation, for example:

$$GDD = \Sigma (\bar{T} - T_b)$$

where, $\bar{T}$ = average daily temperature

T_b = a base temperature or the highest temperature at which no growth occur, this beeing dependent upon crop in question

Σ indicates the difference between $\bar{T}$ and T_b, summed cumulatively day by day.

These types of efforts will be very valuable in significantly reducing the current controversy associated with the presently popular statistical cause-effect models and the scaled uncertainties consequently associated with regional scale assessments of ambient O_3-induced crop loss through the application of such models.

Conclusions

The relationships between ambient O_3 exposure and crop response are inherently stochastic in their nature. These are reflected by the temporal and spatial variabilities in the occurrences of ambient O_3 concentrations and in the behavior of crop biology and sensitivity to O_3. Nevertheless, because of necessity, numerous investigators have relied on the results from artificial, above ambient O_3 exposures and season-end, single point statistical models to establish cause-effect relationships. Most popular among these are the definitions of O_3 exposures through the use of different O_3 concentration summation techniques such as SUM06 (XX) and AOT40 (XX) that do not contain a time element nor feedbacks among plant stress, repair and compensation.

Although the use of cumulative frequency distributions of hourly ambient O_3 concentrations described in this paper also does not contain a time element, it does account for whole growing season O_3 exposure dynamics and for the corresponding spatial variability. A major conclusion drawn here is the importance of moderate or intermediate O_3 concentrations in causing adverse yield responses. A number of independent studies support this conclusion. The reason for this is that moderate O_3 concentrations frequently occur during periods of the day when both the atmosphere and the crops are very conducive for O_3 flux and uptake respectively. The micrometeorological approach described in this paper offers the rationale and the value of applying such an approach. While this overall conclusion does not negate the importance of peak hourly O_3 concentrations if they occur at the right time of the day, it is important to note that as ambient hourly O_3 concentrations reach peak concentrations, their frequency of occurrences decline and so do the properties of atmospheric (O_3 flux) and plant (uptake) conductance. Most importantly, the significance of the frequency of occurrences of moderate or intermediate hourly O_3 concentrations explain the observed ambient O_3-induced crop yield reductions during some years at some study sites in the U.S. and in Europe.

In a final analysis, any explanation of ambient O_3 exposure-crop response relationships should be broadly applicable. Such a goal can be accomplished by coupling the type of

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micrometeorological approach described in this paper with multi-point biological response measurements that include specific crop biology under chamberless, open-field environments.

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Address of the authors:

Dr. Allan H. Legge, Biosphere Solutions, 1601 - 11th Avenue N.W., Calgary, Alberta T2N 1H1, Canada

Dr. Ludger Grünhage, Prof. Dr. Hans-Jürgen Jäger, Institute for Plant Ecology, University of Gießen, Heinrich-Buff-Ring 38, D-35392 Gießen

Dr. Miloslav Nosal, Department of Mathematics and Statistics, University of Calgary, Calgary, Alberta T2N 1N4, Canada

Prof. Dr. Sagar V. Krupa, Department of Plant Pathology, University of Minesota, 495 Borlaug Hall, 1991 Buford Circle, St. Paul, Minnesota 55108-6030, USA

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Mapping Relative Potential Sensitivity of Vegetation to Ozone in Europe: A Preliminary Analysis

Emberson, L.D.[1], Kuylenstierna, J.C.I.[2], Ashmore, M.R.[1], Cambridge, H.M.[2] and Cinderby, S.[2].

[1]Imperial College Centre for Environmental Technology, 48 Prince's Gdns, London, SW7 2PE
[2]Stockholm Environment Institute at York, University of York, York, YO1 5YW

Abstract

At present, **Critical Levels** for ozone distinguish between crops and trees. However, within these groups a single value is used for all species, under all environmental conditions with no distinction in temporal variability in sensitivity throughout the growing season. This paper describes a methodology to evaluate and map potential relative sensitivity of selected species to ozone. The methodology estimates potential ozone uptake using stomatal conductance as a surrogate. As other factors, such as atmospheric conductance and detoxification capacity are known to modify the response of species to ozone, the methodology evaluates a "potential" sensitivity by relating stomatal conductance to potential ozone uptake. Variations in potential sensitivity over the course of the growing season are incorporated using knowledge of changing stomatal conductance with leaf age. The influences of prevailing environmental conditions in modifying uptake are included through the use of species-specific stomatal conductance relationships with four environmental variables: light, temperature, vapour pressure deficit and soil moisture deficit. A preliminary analysis of sensitivity is described by applying the methodology to three oak species (*Quercus petraea*, *Q. robur* and *Q. ilex*) and Scots pine (*Pinus sylvestris*). This analysis uses a single environmental variable, soil moisture deficit, to determine sensitivity and indicate the type of mapping output that can be achieved on a European scale. The resulting map, grouping potential sensitivity classes over Europe, can be used alongside current ozone critical level maps to highlight areas that may require additional attention during policy evaluation.

Keywords: Stomatal conductance, critical levels, ozone, mapping, *Quercus petraea, Quercus robur, Quercus ilex, Pinus sylvestris*, soil moisture deficit.

1 Introduction

Critical levels of ozone set at the Bern workshop (Fuhrer and Acherman, 1994) provide the basis for Level I assessment of ozone impacts on a European scale. Ideally, Level II mapping, incorporating temporal and spatial variations in sensitivity, would be adopted (see Ashmore and Emberson, this volume). However, the limited exposure-response data available place serious constraints on the development of such an approach. There are also significant practical limitations to developing appropriate mapping procedures.

In view of this, we propose an alternative approach which aims to map potential relative sensitivity to ozone, based on indirect estimates of relative ozone uptake. The methodology uses stomatal conductance as a surrogate as, under most conditions, ozone flux is primarily limited by leaf conductance. Reich (1987) demonstrated the correlation between inherent leaf conductance and effects of ozone on photosynthesis. The term "potential" is used as other factors, such as the level of anti-oxidants within the plant, may significantly influence sensitivity. Grunhage and Jager (1994) have also shown how atmospheric conductance can, under certain situations, limit the transport of ozone from the atmosphere to the leaf surface. This is especially important for short vegetation such as crops and certain semi-natural vegetation types.

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For many species, stomatal conductance (g_s) is better documented than ozone responses, both in terms of species-specific behaviour during the course of the growing season and modification of conductance by environmental variables. Therefore, an assessment of sensitivity based on stomatal conductance makes available a large source of information from relevant literature **and** allows the inclusion of both a temporal and spatial aspect for different receptors.

This paper is divided into two sections. The first describes the methodology used to assess relative potential sensitivity of different species to ozone whilst the second illustrates an application of the model with four selected species using digital geographic information on a European scale. This example represents a **preliminary** analysis of sensitivity as only one environmental variable (soil moisture deficit) is included. This exercise is designed to indicate the type of mapping output that can be expected upon the completion of the project.

A similar approach to our methodology has been used by Korner *et al* (1995) for assessing pollutant uptake to SO_2 for two species, *Pinus sylvestris* and *Picea abies*. Legge *et al* (this volume) applied Korners method to ozone uptake, and showed how it could be combined with a model of atmospheric conductance (Grunhage and Jager, 1994). However, neither paper considers how the respective methods may be used on a wider European scale to aid in the setting of critical levels and subsequent policy formulation. Our approach introduces a method which can be used on a European-wide basis through the employment of a Geographical Information System (GIS). A number of important European species (13 forest trees, 11 crop species and 3 semi-natural vegetation types) are being studied within the project, and this extends the applicability of this technique to the current critical level problem.

2 Methodology

The model follows three sequential steps in which stomatal conductance is used as a surrogate for sensitivity:

1) Assessment of species-specific relative sensitivity.
2) Assessment of temporal variability of sensitivity.
3) Assessment of sensitivity of species after modification by environmental variables.

2.1 Species-specific sensitivity

Maximum stomatal conductance (g_{max}) varies between species and can therefore be used as a primary indication of relative potential sensitivity of different species. g_{max} occurs in healthy plants under optimal conditions and is generally attained by young fully developed leaves. The use of g_{max} values as opposed to the average g_s values, ensures comparability between species. Maximum stomatal conductance is used instead of canopy conductance, as there are far fewer data available describing canopy conductance values,and as the upper leaves of the canopy tend to have the highest values of g_s, and therefore the highest potential uptake. The upper canopy also contributes most in terms of total carbon gain due to higher photosynthetic rates produced by high levels of incident radiation (Reich, 1993).

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2.2 Temporal variability in sensitivity

Maximal stomatal conductance (g_{max}) will occur only for a particular period in the growing season of a species. The potential stomatal conductance (g_{pot}) estimates the extent of deviation from g_{max} that is solely attributable to leaf age. This is difficult to evaluate, especially when using field data over the course of a year, as g_s may be modified by environmental pre-conditioning.

g_{pot} is rarely directly retrievable from the literature, and therefore the following methods are used to estimate as accurately as possible g_{pot} for individual species. Defining g_{pot} involves identifying the time in the growing season when g_{max} occurs, usually when the first annual flush of leaves or needles are just fully mature. From this point over the course of the growing season g_s will be declining as a function of leaf age, until the potential g_{max} value of a one year old leaf or needle is reached. This approach is modified accordingly for different vegetation types; for example, g_{pot} for deciduous tree species and crops is set to zero for those months during which leaves are absent. The growing season length may be mapped from species-specific relationships between temperature and bud burst (including chill days which are required by certain species for the breaking of dormancy) and leaf senescence.

2.3 Effects of environment on sensitivity

The "actual" stomatal conductance (g_{act}) represents modification of g_{pot} by environmental variables. There are four environmental variables that are commonly recognised as the most important in affecting stomatal conductance (eg. Jones, 1993):

Light
Temperature
Vapour pressure deficit (VPD)
Soil moisture deficit (SMD)

Each of the relationships between g_s and environmental variables are species-specific and therefore need to be extracted from relevant literature sources. For certain species this information may not be documented and in such cases generic relationships will be substituted to provide the best possible estimate.

To evaluate the modification by the environment of g_{pot} the relationships with the environmental variables are combined within a multiplicative model. The use of a multiplicative model assumes that environmental factors act independently of each other. Although this is probably an oversimplification of realistic stomatal functioning, it has been shown to provide a means of estimating stomatal conductance to a high degree of accuracy (Jarvis 1976).

The effects of the prevailing environmental conditions on g_s are evaluated separately to give functions of g_s with light, temperature, VPD and SMD ($f_{(light)}$, $f_{(temp)}$, $f_{(VPD)}$ and $f_{(SMD)}$). Each of these functions represents relative g, on a scale from 0 (no stomatal conductance) to 1 (full stomatal conductance or maximum potential stomatal conductance). These functions are then multiplied together to give a function for f_{act} as shown in equation 1.

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$$f_{act} = f_{(light)} \cdot f_{(temp)} \cdot f_{(VPD)} \cdot f_{(SMD)} \quad [1]$$

f_{act} is then used to modify g_{pot} to give a value of g_{act} (in mmol m^{-2} s^{-1}):

$$g_{act} = g_{pot} \cdot f_{act} \quad [2]$$

g_{act} therefore represents the actual stomatal conductance taking into account g_{max}, the time component (g_{pot}) and modifications to stomatal conductance by prevailing environmental conditions. g_{act} is a measure of the relative potential sensitivity to ozone, as a higher uptake will lead to an increase in the potential for damage. This allows the representation of sensitivity classes based on g_{act} which can be mapped over Europe to indicate areas of differential potential sensitivity to ozone. The geographic representation of sensitivity in this project uses monthly time-steps for environmental variables.

3 Mapping potential relative sensitivity to ozone: A Preliminary analysis

In this section the model described above is applied to create a map of potential relative sensitivity for areas containing four selected species: *Quercus petraea*, *Quercus robur*, *Quercus ilex* and *Pinus sylvestris*. Tree species have been chosen as they will be well coupled to the atmosphere, and hence the importance of the atmospheric conductance component in limiting potential uptake is reduced (Pleijel, 1995). Considered together, the species cover most climatic areas of Europe. This is an exercise to illustrate the use of the model and is **not** the final product of the project. This example looks into the variation of sensitivity using one variable alone, Soil Moisture Deficit (SMD), and creates a sensitivity map for one illustrative month. Other environmental factors are considered to be optimal for stomatal conductance and so relative g for these variables equals 1.0. For this example two geographic datasets are required: the distribution of vegetation and the soil moisture deficit for a given month.

3.1 Receptor Mapping

A digital dataset has been produced that shows the distribution of 82 different vegetation and land use types in Europe (excluding the former USSR and Turkey). The data for the map derive from a number of sources. The distribution of forest is taken from the European Space Agency forest map (ESA, 1994) and different vegetation types (including dominant tree species) are derived from the Land Use map of Europe (FAO-Cartographia, 1980). The map gives a general picture of the distribution of dominant tree speceis including *Q.ilex*, *P.sylvestris* and an oak category considered to be *Q.robur* and/or *Q.petraea*. The accuracy of the distributions has been assessed by comparison with other sources and is considered a reasonable overview. The combined distribution of the four tree species is shown in Figure 1.

3.2 Mapping Soil Moisture Deficit (SMD)

Various methods have been devised to calculate SMD (Penman, 1963; Thornthwaite and Mather, 1957; Grindley, 1969, 1970). The SMD in Europe has here been calculated using the Penman approach elaborated by Grindley. Monthly precipitation (P) and temperature have been digitised from the Climatic Atlas of Europe (WMO-Cartographia, 1970) and potential

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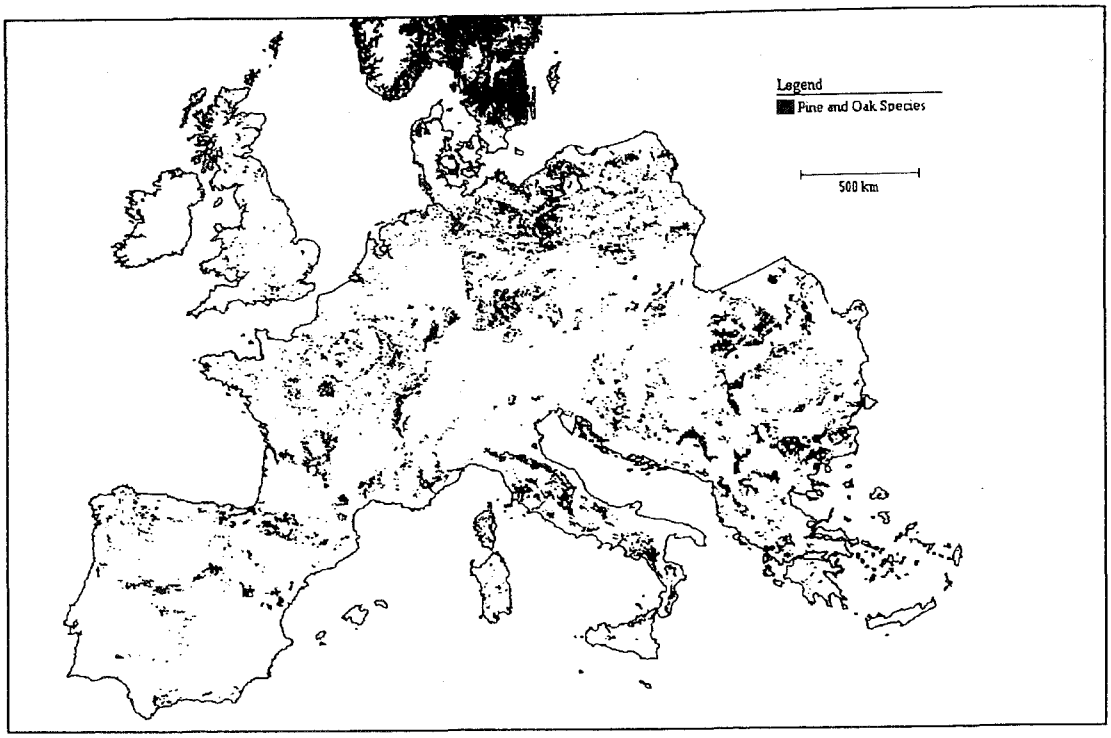


Figure 1: Distribution of *Quercus robur*, *Quercus petraea* , *Quercus ilex* and *Pinus sylvestris* over Europe

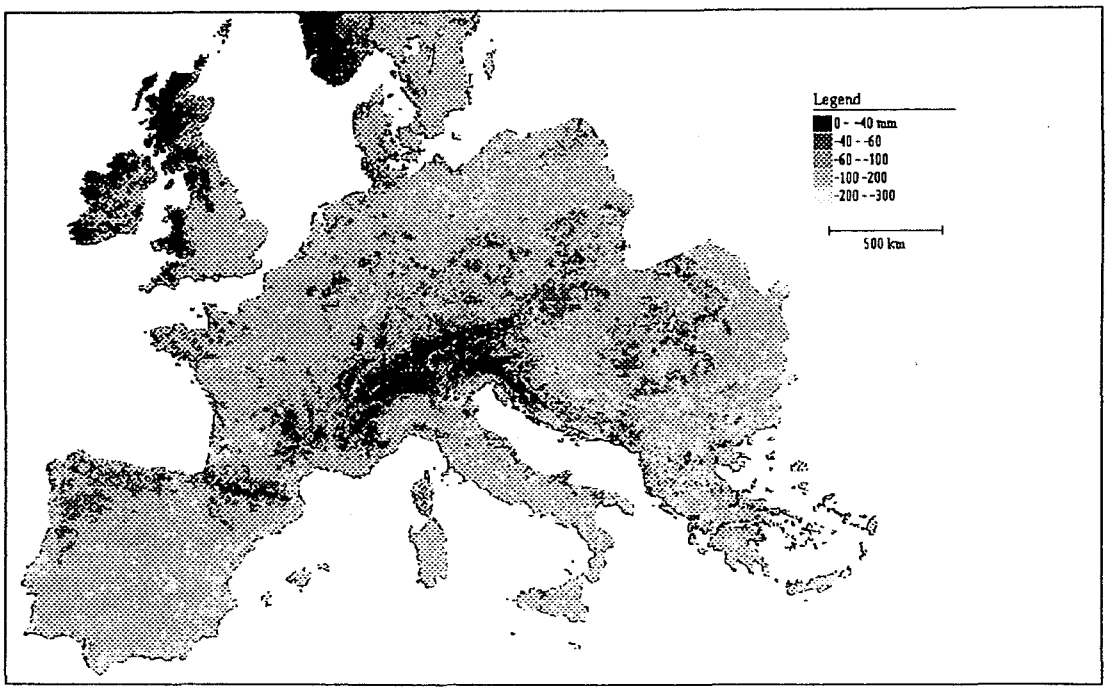


Figure 2: Soil Moisture deficit over Europe for September

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evapotranspiration (PE) for each month calculated using the Thornthwaite method. Values of actual SMD (in mm) are read from tables against values of accumulated P-PE (potential SMD). The tables refer to a given "root constant" (Penman, 1963) which is a measure of the easily available soil water that is transpired at the potential rate. After this point plant water uptake becomes more difficult and actual evapotranspiration becomes less than PE thereby slowing down the increase in SMD until the permanent wilting point is reached. At this point no further evapotranspiration is considered to occur as the plant has consumed all available soil water. The water potential relationship with g (Figure 3) refers to the predawn leaf water potential (in MPa) which is assumed to be equivalent to the soil water potential (Fitter and Hay, 1993). SMD is transformed into MPa using a soil water release curve (Pearcy *et al*; 1989). This is acheived by assuming that soil water potential at field capacity is equivalent to approximately 0 MPa and 0 mm SMD. Permanent wilting point (defined for trees by Penman (1963) as 250 mm SMD) is equal to the predawn leaf water potential at which minimum relative g is achieved (see Figure 3). The relationships are soil-type dependent but for this analysis the soil water release curve for sandy-loam was used to represent all soil types. The resultant SMD in September is shown in Figure 2.

There are uncertainties in SMD calculations which derive both from the calculation of PE and from the drying curve used, which differs in the Thornthwaite and Penman methods. Methods are being improved as the project progresses. Digital datasets for monthly irradiance, VPD and temperature are still being produced, therefore, within this analysis, they are assumed to be optimal for stomatal conductance.

3.3 Evaluation of sensitivity classes

The following section illustrates the manipulation within the methodology of the g_s data for the four selected species. This data has been extracted from an extensive literature search. Extraction of each g_s parameter (g_{max}, g_{pot} etc..) from relevant literature sources was acheived using a set of criteria to ensure that a true representative value is obtained. g_{max} values for the four selected species are displayed in Table 1, both as the extracted g_{max} range and the corresponding average value.

Table 1 g_{max} values for selected species.

Species	Average g_{max} mmol m^{-2} s^{-1} (g_{max} range)		g_{pot} (mmol $m^{-2}s^{-1}$) for September (Relative g)
Pinus sylvestris	325 (288 to 373)		283 (0.87)
Quercus petraea	250 (220 to 320)	270 (220 to 320)	270 (1.0)
Quercus robur	300 (280 to 320)		270 (1.0)
Quercus ilex	350 (310 to 420)		287 (0.82)

NB. All g_s measurements are for conductance to water vapour and refer to projected leaf area.

[a]Troeng and Linder (1982); Korner *et al* (1995); Whitehead *et al* (1984); Helkvist *et al* (1980); Kuppers and Shulze (1985)
[b]Breda *et al* (1993a); Breda *et al* (1993b); Vivin *et al* (1993); Epron *et al* (1993b)
[c]Breda *et al* (1993b); Epron *et al* 1993b); Vivin *et al* (1993)
[d]Acherar *et al* (1992); Epron and Dreyer (1990); Sala and Tenhunen (1994)

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The estimation of g_{pot} was achieved for individual species using the various methods outlined previously. *Pinus sylvestris* g_{pot} was extracted directly from the literature using values given in Korner *et al* (1995). *Quercus petraea* and *Quercus robur* g_{pot} were deduced from data in the literature that described g_s towards the end of the growing season under optimal conditions (Epron *et al*, 1993b). g_{pot} for *Quercus ilex* was estimated assuming a generic relationship with *Quercus coccifera* (an oak which exists under similar climatic conditions to *Quercus ilex*) for which g_{pot} was described directly by Caldwell (1986). September g_{pot} values are shown in Table 1 represented both as mmol $m^{-2}s^{-1}$ and as relative g (in brackets). As stated previously, for this analysis only soil moisture deficit is used as an environmental variable. The relationships in Figure 3 show each species' g_s relationship with predawn leaf water potential taken from the following literature sources.

Graph A *Quercus robur*:	Breda *et al* (1993b); Vivin *et al* (1993); Epron *et al* (1993)
Graph B *Quercus petraea*:	Breda *et al* (1993a); Epron and Dreyer (1990); Epron and Dreyer (1993a); Epron and Dreyer (1993b); Vivin *et al* (1993)
Graph C *Quercus ilex*:	Epron and Dreyer (1990); Acherar *et al* (1992); Sala and Tenhunen (1994)
Graph D *Pinus sylvestris*:	Ng (1979)

The species-specific values of g_{act} calculated from g_{pot} and the relative g for a given SMD value have been grouped into three sensitivity classes shown in Table 2. From this the sensitivity map is produced, as shown in Figure 4. For this example Figure 4 uses a 1 x 1° longitude latitude grid square to allow clear presentation of the sensitivity classes. These are assigned by allocating to the whole square the sensitivity class that is most frequent in that square. Since the values of g_{pot} for the four species were very similar, the geographical variation in sensitivity is almost entirely due to variation in SMD in this case. The potential sensitivity is therefore lowest in southern Europe.

Table 2 Allocation of values of g_{act} to sensitivity classes

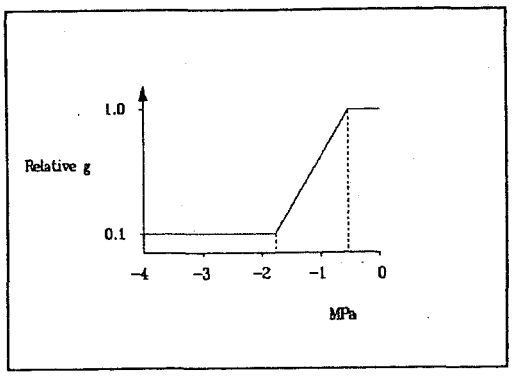
	Sensitivity class	g_{act} mmol $m^{-2}s^{-1}$
(Least sensitive)	1	0 to 100
	2	100 to 200
(Most sensitive)	3	200 to 300

4 Conclusions

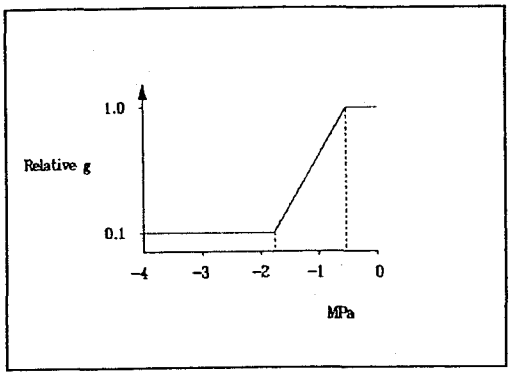
This paper describes a method to show the differential sensitivity of vegetation to ozone concentrations across Europe. This illustrates the fact that, although current critical levels assume equal sensitivity of forest trees across Europe during the growing season, there are likely to be differences which will have implications for policy development. For example, if the map in Figure 4 is representative of true vegetation sensitivity then the highest sensitivity is found in areas with lower ozone exposures in northern Europe and lower sensitivity is encountered in central Europe and the Mediterranean region, where ozone exposures are higher.

This paper describes only a preliminary stage in the development of the GIS model, which will be expanded to include other environmental variables, and the effect of atmospheric

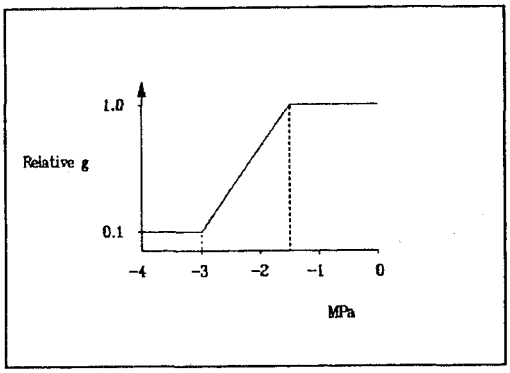
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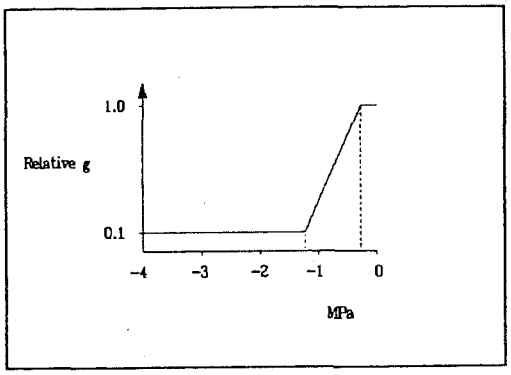
Graph B *Quercus robur*



Graph A *Quercus petraea*



Graph C *Quercus ilex*



Graph D *Pinus sylvestris*

Figure 3: g_s relationships of the four selected species with predawn leaf water potential (Mpa)

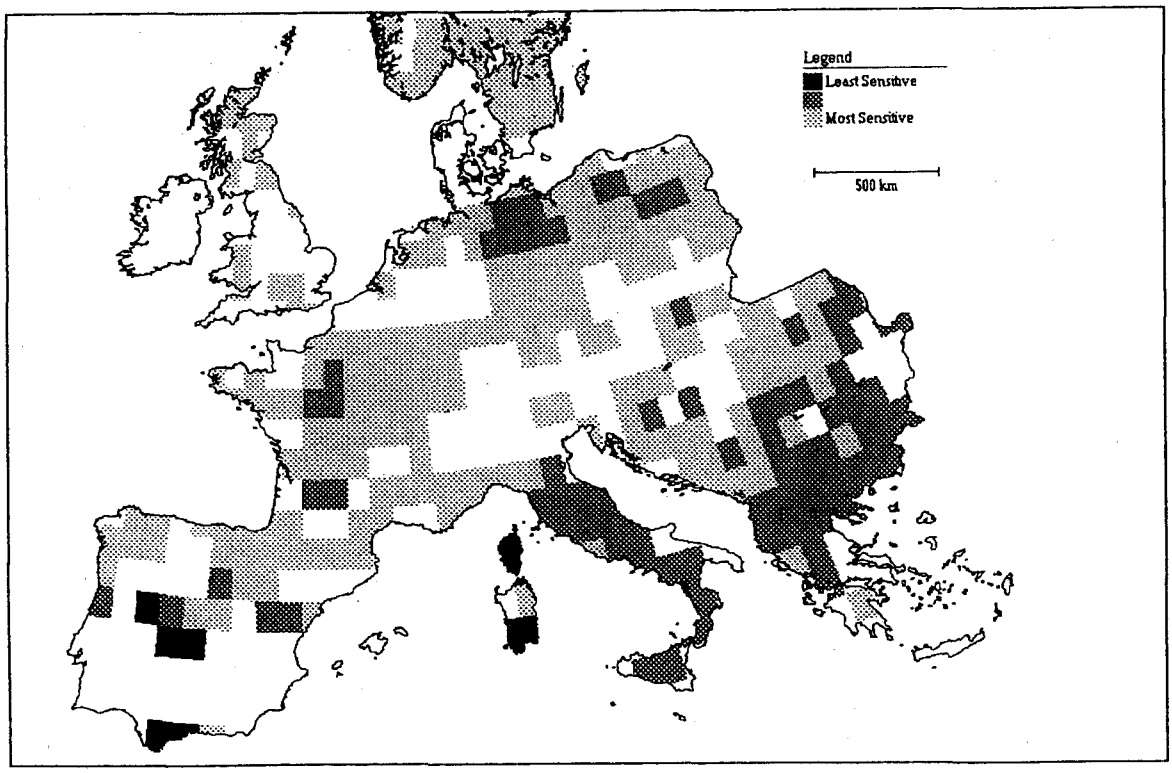


Figure 4: The mode relative potential sensitivity of the selected species in 1° x 1° longitude-latitude grids to ozone over Europe for September.

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conductance. Inevitably, implementation of the model on a European scale has required many simplifications of which the most important may be the need to use monthly means of climatic variables. This does not allow the investigation of daily changes in stomatal uptake in relation with diurnal fluctuations in ozone exposure. The availability of data describing stomatal behaviour also limits the application of the methodology to a few select species. These limitations, and the application of the sensitivity maps within the critical levels framework, are more fully considered by Ashmore and Emberson (this volume).

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DEFINING AND MAPPING CRITICAL LEVELS OF OZONE IN EUROPE

M.R.Ashmore & L.D.Emberson

Imperial College Centre for Environmental Technology
48 Prince's Gardens, London SW7 2PE, U.K.

Abstract

The critical levels of ozone set at the Bern workshop provide the basis for Level I assessment of ozone impacts on a European scale. The biological uncertainty in setting these values is substantial, but attention should also be paid to the subsequent evaluation and mapping of critical level exceedences, since this involves significant uncertainties important in influencing policy evaluation. Ideally, Level II mapping, incorporating temporal and spatial variations in sensitivity, would be adopted. However, the limited exposure-response data available place serious constraints on the development of such an approach. Currently, there are also significant practical limitations to developing appropriate mapping procedures on a European scale. In view of this, methods of mapping relative sensitivity to ozone may provide a more practical goal which will assist in evaluating the limitations of the current Level I approach. We outline here a method, currently under development, which aims to map potential relative sensitivity to ozone, based on indirect estimates of ozone uptake.

Introduction

Over the past fifteen years, a substantial body of evidence has accumulated to demonstrate that current concentrations of ozone in many parts of Europe can cause adverse effects on crop yield, tree growth and vitality, and the composition of natural communities. The need to prevent, minimise or reduce these impacts of ozone on vegetation will be an important consideration in the negotiations within UNECE of new protocols relating to the control of VOCs and NOx over the next few years.

Within these negotiations, the key element in translating scientific understanding into policy objectives will be the concept of a 'critical level', defined as "the concentration of a pollutant in the atmosphere above which direct adverse effects on receptors may occur according to current knowledge". The current critical levels for ozone were set following UNECE workshops at Egham (Ashmore & Wilson, 1994) and Bern (Fuhrer & Achermann, 1994). These critical levels use the concept of cumulative exposure above 40ppb (AOT40) as their basis.

The critical level concept is designed as a policy tool; hence it has to be sufficiently simple and robust for it to be applied in assessment models and policy evaluation. Therefore, in

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setting critical level values, it is inevitable that much of the detail of scientific knowledge cannot be incorporated. However, there is clearly a danger in making the approach too simple, in that the omission of key elements of scientific understanding may lead to the adoption of policies which are less effective in reducing the impact of ozone.

Specifically, the critical levels currently defined are Level I values, that is there is a single value to be applied, for example, to all crop species in all climates in all soils. To satisfy the precautionary principle from which the critical level approach is derived, values are based on the most sensitive known receptor and conditions for which data of the required quality are available. Thus application of the Level I approach may in some cases mean that the potential damage to vegetation is overestimated. It is therefore important to consider the feasibility of moving towards a Level II approach, in which the effects of environmental factors, as well as variations in species sensitivity, are incorporated into the analysis.

In practical terms, it is also important to consider not only how critical levels are defined, but also how they are actually applied in policy assessment, and indeed by whom. The key elements in the dialogue between effects scientists, mappers and modellers, and policy-makers, are the maps which show the geographical distribution of areas with ozone exposures in excess of the relevant critical level. If these groups work in isolation there is a danger that errors or misjudgments will be made - in particular that the mapping procedures adopted may not properly represent our understanding of the real biological effects as summarised in setting a critical level.

This paper will aim to critically evaluate whether current mapping procedures, based on the critical level values set at Bern, will provide a meaningful basis for a risk-based evaluation of policies to reduce ambient ozone exposures on a European scale. Our focus is not specifically on the uncertainties in the biological basis for critical levels of ozone, which are covered elsewhere (e.g. Ashmore 1994; Sanders et al., in press) and in other papers at this workshop, but on difficulties at the interface between the effects and mapping communities. In particular, we will draw on experience in mapping critical levels exceedence in the UK (UKCLAG, 1996; UKPORG, in prep.) and from a current joint project on mapping relative potential sensitivity to ozone on a European scale, which is being carried out in collaboration with the Stockholm Environment Institute (SEI).

There are two sections to this paper. The first, will consider the uncertainties associated with the current Level I approach, while the second will consider the difficulties associated with developing a Level II approach and introduce a proposed method for moving towards an assessment of potential relative sensitivity to ozone.

Current Level I Mapping

Uncertainty in mapping receptor exceedence

Having defined a critical level value for agricultural crops and for forests, areas of critical exceedence need to be identified; this entails an overlay within a GIS of interpolated or modelled AOT40 values and of appropriate receptors. In the UK, a network of 17 rural measurement stations is available from which to estimate the national concentration field. However, each of these stations is influenced by local effects (e.g. altitude, NOx emissions,

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coastal influences) which mean that they may not be representative of general exposures in the area of the country in which they are located. A methodology has been developed to take account of the different diurnal cycles at different altitudes (UKPORG, 1994), but other factors have still to be incorporated into the methodology; in particular, NOx emissions which will significantly alter the exposure field over most of southern Britain.

Thus significant uncertainties exist in the mapping of ozone exposure; but do these affect the assessment of critical level exceedence ? In the UK, most of the arable agriculture is situated in the East Anglian region. However, the interpolated maps of AOT40 show a large area in this region where the critical level is not exceeded. The precise boundaries of this area will significantly affect the assessment of impacts on arable production because of its high intensity in the region. Unfortunately, no monitoring station exists within this 'hole' of low ozone exposure, and the two closest stations are both influenced by local NOx emissions. Hence, because of the co-occurrence of the receptor and areas of exposure uncertainty, the national proportion of arable agriculture experiencing exceedences of the critical level is very uncertain, and changes to monitoring station location will need to be made. This example also illustrates the importance of local spatial variation in ozone exposure and receptor location which will need to be considered further when evaluating ozone control strategies based on the EMEP model, which operates on 150km grid squares.

Measurement position

The critical levels of ozone set at the Bern workshop were derived from experimental data on crops and young trees exposed in open-top chambers. In these chambers, there is forced ventilation of air to ensure uniform mixing of pollutants and to minimise ingress of ambient air. Ozone concentrations are typically measured at a location immediately above the crop or adjacent to the trees.

In contrast, measurements of ozone in national networks are typically made at an inlet height of 3-5m, while models of ozone normally predict mean concentrations in the well-mixed boundary layer. However, these values may not be comparable to those at the surface of the vegetation to which the critical levels actually apply.

Analysis of measurement data over a barley field in Sweden (Pleijel et al., in press) shows substantial gradients of AOT40 over a short vertical distance. Ozone concentrations were measured at 9m, 5m, 2m and 1m above the ground over a crop which was 0.7m tall. The results show a decrease in AOT40 from about 2000ppb.h at 9m to 800 ppb.h at crop height. This indicates that for crops the effect of measurement height may be substantial, potentially resulting in a considerable error in the estimation of critical level exceedence. This issue has also been addressed using deposition models. Tuovinen & Laurila (1993) calculated seasonal AOT40 values at 0.1m compared with those measured at 5m using a deposition model. The calculated value at 0.1m was 2550 ppb.h, compared with 6450 ppb.h at 5m, and 8550 ppb.h at 50m, the height in the boundary layer used in the EMEP model. This was despite the fact that the differences calculated in seasonal mean concentrations are quite small. This is because at the location in Finland for which the calculations were carried out, a substantial part of the annual AOT40 is made up of frequent exposure to relatively low concentrations; thus a small decrease in mean concentrations can have a large impact on AOT40 values.

Thus, there is the potential for very substantial overestimates of the extent of critical level

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exceedence for low-growing vegetation such as grassland, moorland and arable crops. For estimates based on measurement networks, studies suggest an error of around 100%, while for modelling results the discrepancy could be even greater. However, the studies were from relatively northerly locations, where AOT40 is dominated by chronic exposure to relatively low concentrations. The effect at more southerly locations where episodic exposure is more important may be smaller in size, and it would be unwise to apply any corrections until this issue has been investigated further. For forests and other aerodynamically rougher vegetation, this issue is of less concern and measurement based AOT40 maps should provide a relatively accurate indication of the exposure experienced at the surface of mature forests.

Night-time exposure

The crops group at the Bern workshop defined the critical level on the basis of daytime exposures only. This is because the flux of ozone into the leaf is controlled by stomatal uptake; since the stomata in almost all situations close in the absence of light, this flux essentially becomes zero during the night. Hence any ozone exposure during the night is likely to have no biological impact and is ignored in determining exceedence of the critical level. However, a very different approach was taken by the forests group. Here it was argued that there is some evidence of stomatal uptake at night in certain tree species. While others argued that this was not the case in most species and situations, it was decided that on a precautionary principle, the ozone critical level should also consider night-time exposure.

In considering the validity of the approach taken at Bern, there are two important points to bear in mind. Firstly the experimental studies used to derive critical levels mainly used fumigation patterns in which exposure to ozone occurred mostly during daylight hours. This was either through the design of the experiment, or because the study was a filtration experiment in which most of the ozone exposure occurred during the day. Secondly, there is only limited evidence of significant nighttime stomatal uptake of ozone. Furthermore, it is likely that many species, including most broadleaved species, will show no such stomatal uptake, and field studies of ozone deposition to forest canopies provide little evidence of any significant nighttime stomatal uptake.

Thus, there are reasons to doubt the 24 hour basis for calculating ozone critical levels for forests, especially as the experiments used to derive the critical level do not in general involve nighttime fumigation. The implications of using a 24 hour basis rather than daylight hours have been examined for the UK. There is at every site a significant nighttime component to the seasonal AOT40, and at four sites the mean AOT40 is above the critical level when calculated on a 24 hour basis, but not when calculated on a daylight hours basis (UKPORG, in prep.). This suggests that the map of critical level exceedence for the UK would be substantially altered if only daylight hours were considered. The proportion of total AOT40 occurring during daylight hours varies substantially between sites, from 63% to 94%. In general terms, it is the more remote and high elevation sites which show the lowest proportion in daylight, and hence for which the uncertainty over nighttime uptake is most significant.

Annual variation

A major issue of discussion at Bern was the years for which the AOT40 value should be

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calculated for comparison with the critical level. In the group considering annual crops, it was agreed that the aim should be to prevent a 10% yield loss in the worst year, rather than as an average over several years. However, the AOT40 clearly also should refer to recent years and thus, as a compromise, it was agreed that the worst year in the last five should be used. However, this decision does not necessarily resolve some of the problems which are induced by the fact that ozone concentrations can vary dramatically from year to year.

A key issue here is that it is not just the overall ozone levels which vary from year to year; the spatial pattern of ozone exposure may also be quite different. Putting the issue another way, the year in the last five which is highest at one site may not be the same as that at another site. Hence it is difficult to identify for Europe as a whole, rather than a specific site, which is the worst year in the last five. From a biological effects perspective, this is not a problem in that for each site one would simply select the highest AOT40, and then interpolate to create a worst case scenario. However, there may be difficulties here with the mapping and modelling approaches which require a coherent set of input data for a common time interval. Furthermore, from a statistical standpoint, mapping the AOT40 averaged over several years has considerable attractions, since the data are more robust and should better represent the average spatial distribution over Europe.

The growing season

The definition of the period to be used in calculating the cumulative exposure above 40ppb is critical. If the period is too long, an artificially high AOT40 value will be determined which includes periods when the vegetation is not actually present or is taking up little ozone. If it is too short, then too low a value will be calculated and periods when the vegetation is experiencing significant impacts of ozone may be omitted.

For the biologist, the easiest solution is to calculate the AOT40 value over the growing season, although this does not take into account the possibility that the sensitivity of vegetation to ozone may vary over the growing season. However, a fixed growing season was adopted at the Bern workshop because of a perceived need in the mapping community for consistent and readily defined time periods. For crops, the period of interest is largely defined by the local cropping patterns, which are also climatically influenced. It should also be remembered that the impact of ozone may be greatest during certain specific periods of crop development (eg. Younglove *et al*, 1994). Thus, the timing of episodes may influence the impact of ozone significantly, especially at those sites where intermittent episodic exposure makes up the bulk of the AOT40.

Furthermore, when the AOT40 is applied to European arable agriculture in general, and not just to spring wheat, the situation becomes more complex. The three months when the most sensitive crop is in the ground clearly will vary greatly, and a variable growing season for calculation of AOT40 values clearly needs to be incorporated into the assessment procedures. One possible approach would be to develop twelve monthly sets of modelled or mapped AOT40 values over Europe. It should then be possible for each area of Europe to identify the three months of interest, and compare the mapped or modelled AOT40 for those three months with the critical level. Whichever approach is adopted, a more detailed analysis of cropping patterns in relation to seasonal ozone patterns is needed to assess the significance of this factor for agriculture.

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For forests, the situation is a little different, given the longer fixed growing season of six months which was adopted. For deciduous trees, producing a new set of leaves each year, a particular concern is how much of the seasonal AOT40 occurs in the period of April and May before budburst. This will depend on species, location and climate, but could be particularly significant in northern Europe and at high altitude. In the case of evergreen species, the situation is more complex. Budburst produces a new set of needles which are likely to be more sensitive to ozone, but also increases the ozone flux to the older needles. Thus, the inclusion of AOT 40 values before budburst is still of concern. Nonetheless, significant ozone flux to older, overwintered needles can occur in warmer days in March, and the possibility of additional significant exposure in this period, especially at the more remote sites, should not be excluded.

These issues are of particular relevance to the Mediterranean region, where the active growing seasons of crops, forests and natural vegetation are quite different from those elsewhere in Europe. It is thus essential that this time factor be properly incorporated if meaningful assessments of ozone critical exceedence are to be made on a European scale.

Natural vegetation

At the Bern workshop, critical levels were set for annual crops and for forest trees. However, no critical level value was set for natural vegetation, such as grassland, heathland, maquis, and moorland. Such vegetation occupies a substantial area of Europe; furthermore the distinction between systems managed for production and yield and those which are primarily of value for nature conservation is unclear, especially in the case of forests and grasslands.

If critical levels are not assigned to such vegetation, then this may lead to important omissions in the mapping of critical levels exceedence. It is therefore important that we develop methods to allow critical levels to be assigned to this category of vegetation. However, this raises a number of important issues since it is unclear what criteria should be used to define a critical level for such vegetation types. For example, ozone may impact by shifting the competitive balance between species rather than reducing total production (Fuhrer et al., 1994; Ashmore & Ainsworth, 1995). Should then the critical level be set to prevent the loss from any site of any species ? Should some species be given greater value than others ? Should the aim be to maintain a particular type of community ? These are issues which are essentially not only matters of scientific evaluation but also of societal and political judgement.

Level II Mapping

Implicit in some of the points made above is that there is spatial and temporal variation in sensitivity to ozone across Europe which needs to be considered when developing policy based on critical levels. In theory, the Level II approach would allow the control policies to be focused on those parts of Europe where the impact of ozone is likely to be greatest. However, to implement such an approach, some means of identifying these areas is required. A complete Level II approach would require different critical level values to be set, depending on a range of factors known to affect ozone responses. In practice, three major types of factors would need to be included:-

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Spatial receptor variability

Species, cultivars and ecotypes vary in ozone sensitivity; although some rankings of species have been published there is still much uncertainty in these on the basis of direct empirical evidence. Certain species characteristics are known to influence sensitivity to ozone injury, and a comprehensive description of these factors can be found in Tingey and Taylor (1982). Primarily, receptor sensitivity is determined by characteristics that will affect both the potential for ozone uptake and the detoxification capacity of the plant. Uptake is controlled principally by stomatal conductance, although atmospheric conductance (influenced by vegetation roughness) and boundary layer conductance (influenced by leaf morphology) will also affect ozone uptake. The capability of a plant to deal with an absorbed ozone dose is controlled by inherent genetic factors. These determine factors such as species' stomatal behaviour to ozone stress, detoxification capacity through anti-oxidative or enzymatic pathways and the ability to repair injured tissue. These detoxification mechanisms can be rapidly induced by ozone exposure (Bender *et al*, 1994).

Spatial environmental variability

The environment across Europe is highly variable with sharp contrasts from boreal to Mediterranean climates. Prevailing environmental conditions have been shown to alter species sensitivity both through modification of the uptake process, and through environmental stress reducing a plant's ability to detoxify ozone.

Temporal variability

The environmental conditions experienced in any one area will vary considerably through the course of a year. Furthermore the temporal variations in environmental factors which have a strong influence on ozone uptake, such as soil moisture deficit and vapour pressure deficit, are often associated with variation in ozone concentrations. In terms of ozone impact on vegetation this fluctuation is critical in determining receptor response, but phenological variation in sensitivity may also be important. The sensitive period of the vegetation, as defined by the growing season, is both species and location specific. However, species sensitivity will not be constant over the course of this period, since stomatal conductance changes with leaf age and since sensitivity to damage varies with developmental stage.

Hence, the sensitivity of vegetation to ozone varies in both space and time. The locations which current mapping procedures indicate show the greatest critical level exceedence may have crops or forest species present which are, for example, less sensitive than those used to set the current critical levels. Equally, ozone exceedence at these locations may be calculated for a time interval when the local vegetation is not at its most sensitive. This spatial and temporal variation is driven both by variation in the inherent sensitivity of the vegetation, and by spatial and temporal variation in environmental conditions. We emphasise here that a Level II approach needs to contain a temporal element as well as a spatial element; the importance of the time factor has, we believe, not been given sufficient attention to date.

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Requirements for the Implementation of Level II

Two main criteria are thus required in order to achieve a comprehensive Level II assessment of ozone damage on a European scale. These are (a) a comprehensive experimental data set and (b) a mapping capability combined with information detailing species location, environmental variables and other modifying factors, the latter at a well defined temporal as well as spatial resolution.

European research programmes over the last 15 years have greatly increased our knowledge of exposure-response relationships to ozone. However, the data concentrate on a few specific species covering a limited geographical range, and information describing exposure-response relationships under different combinations of modifying factors is very limited. To establish such relationships from experimental work will be both costly and time-consuming and hence is probably outside the range of the timescale of present negotiations. However, in the longer-term such work is essential to the development of a Level II approach.

In terms of receptor mapping, it would obviously not be feasible to map every vegetation type over Europe, and thus some means of selecting the major species to be mapped needs to be devised. The fact that detailed digitised maps describing relative abundance of receptors, and maps describing key environmental variables, do not exist at a European scale at present adds further complications to the reality of achieving a Level II map. Finally, inclusion of the time factor, even at relatively low resolution, will substantially increase the complexity of the mapping process.

An alternative approach

Thus, at present it seems unlikely that a fully comprehensive Level II approach can be achieved within any reasonable time-scale. However, it may be possible to move towards such an approach by using a more limited, or more subjective, assessment of relative potential sensitivity in conjunction with the Level I maps.

A methodology for assessing potential relative sensitivity of selected European vegetation types is currently being devised within a collaborative project between Imperial College (IC) and the Stockholm Environment Institute at York (SEIY). The methodology is based on the principle that ozone flux is one major determinant of sensitivity and thus could be of importance in identifying areas of Europe with relatively high sensitivity to ozone, and the time of year when sensitivity is greatest.

Flux as an exposure index

Many authors have recognised the advantages of using ozone flux, rather than ambient ozone concentration, as a measure of dose in dose-response fumigation studies (Fuhrer *et al.* 1993, Fowler and Cape 1982, Leuning *et al.* 1979). Ozone flux density represents the gas phase transport of ozone from its source in the atmosphere to a sink within the plant. This is dependent upon both the conductance of the atmosphere and plant, and the concentration gradient between the ambient air and the deposition sites within the leaf. This can be described by the following simple equation:

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$$FO_3 = G \times [O_3]$$

Where FO_3 is the ozone flux
G is the conductance of the flux pathway or deposition velocity
and $[O_3]$ is the ozone concentration at a standard height

It is widely recognised that, under most conditions, flux is primarily limited by the stomatal conductance to ozone (Fuhrer, 1993). The extent to which stomatal behaviour dictates different plant species' responses to ozone has been highlighted by Reich (1987), whose study showed how differences in ozone uptake and subsequent response could be predicted from differences in the inherent leaf diffusive conductance.

Therefore, it may be feasible to use stomatal conductance as a surrogate for the potential sensitivity of vegetation, and to add to this a separate element describing variation in atmospheric conductance. This type of approach has also been proposed and developed by Korner (1995) and Legge *et al* (this volume). This is the objective of the current collaborative research project between ourselves and SEIY. The methodology developed for this project is discussed below in terms of its similarity to a Level II approach. The applications of a preliminary version of the model to four tree species is illustrated in Emberson *et al* (this volume). This aims to detail how components that determine plant response to ozone, by affecting the potential for uptake, can be integrated and manipulated within a workable methodology which can be implemented within a GIS framework. This should also help to highlight the problems faced in moving to a Level II approach, or defining relative sensitivity.

A Methodology to Assess Relative Potential Sensitivity

The methodology used to assess sensitivity is based on a simple model which can be divided into the following three sequential steps:

- Assessment of relative species sensitivity under optimal conditions.
- Assessment of temporal variability of species sensitivity with developmental stage.
- Assessment of species sensitivity after modification by prevailing environmental conditions.

Receptor sensitivity and mapping

Various attempts have been made to assess relative sensitivities of different species. These assessments have generally used data obtained from fumigation experiments and ranked species by their performance in exposure-response studies. This approach has two important limitations. Firstly, only a relatively small experimental database exists and secondly, the conditions under which experiments are carried out are not standard and therefore not entirely comparable. Factors such as ozone exposure pattern, prevailing environmental conditions and variable developmental stages will all affect plant response and may produce

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misleading results when comparing species sensitivity. Furthermore, species' relative sensitivity to the acute exposures commonly used may differ from those to longer term chronic exposure.

The approach of using a surrogate for sensitivity, stomatal conductance, makes available a large source of information from relevant literature. However, still there is a problem with standardising the data. This has been overcome by devising a method to extract species maximum stomatal conductance (g_{max}) values from the literature. The use of g_{max} ensures that stomatal conductance is measured under optimal conditions and at a point in the leaf life span comparable across species. g_{max} represents a reference stomatal conductance value for individual species, which is then modified within the model by phenological and environmental factors.

A further problem associated with receptor variability is choosing which species are to be mapped. Selection within the project has been decided on three factors: (1) ecological and economic importance, (2) stomatal conductance data availability and mapping capabilities and (3) importance within existing ozone exposure-response literature. On this basis eleven agricultural crops, thirteen forest trees and three semi-natural vegetation types will be included in the mapping process.

Temporal variation

It is widely recognised that species sensitivity is not only determined by the length of the growing season, but also by other plant factors that influence susceptibility to ozone during this period. Fumigation studies can identify stages of increased sensitivity. For example, recent work by Pleijel (pers.comm.) has shown wheat to be more sensitive to exposure after, as opposed to before, anthesis. However, a far more comprehensive response data set would be needed to incorporate phenological variation in sensitivity into a Level II approach.

Stomatal conductance changes with leaf age, and this is recognised to be a species-specific phenomenon. Therefore, the potential maximum stomatal conductance throughout the growing season can be evaluated for certain species and incorporated in the model. This is a potential stomatal conductance as it is recognised that, in reality, other environmental factors will influence the time-course of stomatal conductance.

Environmental factors

The fact that environmental variability alters plant response is recognised, but there appears to be no easy means by which this can be incorporated into a Level II approach. This is probably because environmental conditions will affect both a plant's potential for ozone uptake and its susceptibility to ozone flux. The most extensively studied environmental variable is soil moisture deficit, which has been reported to be protective against ozone impacts on crops such as cotton (Heagle *et al*, 1988; Tingey and Hogsett, 1985), although other studies (eg. Gruters *et al* 1995) have found no such protective response. A further complication is the action of ozone itself, either directly or indirectly, affecting stomatal behaviour.

The approach used within the model focuses on the extensive literature which exists describing the role that the environment plays in modifying stomatal conductance. The

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predominant environmental variables considered to elicit responses in stomatal conductance are: irradiance, temperature, vapour pressure deficit and soil moisture deficit. The stomatal response to each of these variables is species-specific, although generic trends may exist for species groups. These relationships have been identified from an extensive literature search and are used to modify the value of potential stomatal conductance, using a multiplicative model.

A key difficulty in applying this approach is the time-resolution of the data. Given the capability of the GIS to be used, and the availability of the European wide data sets, it is only possible to use monthly mean values for each of the key parameters, whether obtained directly or indirectly, as in the case of soil moisture deficit. However, variation between and within days, for irradiance and vapour pressure deficit in particular, will be large and linked to variation in ozone concentration; for example days with high ozone concentration tend to have high vapour pressure deficits, leading to reduced stomatal conductance and ozone uptake (Grunhage and Jager, 1994). Although the effects of such variation could in theory be developed within the model this would place very large data demands on the GIS, and initially at least it is best addressed on a site-specific basis. Hence, the model based on monthly data can only provide a semi-quantitative assessment of relative sensitivity over different parts of Europe.

Atmospheric conductance

An important component of ozone flux is atmospheric conductance, especially over short vegetation where there is a reduced coupling of the canopy-atmosphere interface. This has important implications when choosing an ozone descriptor. If ozone concentration is measured at a standard height, and atmospheric conductance limits the transport of ozone to the canopy surface, the actual ambient concentration around the plant will not be the concentration measured.

The model incorporates an atmospheric component for short vegetation such as crops and grasslands, using a standard equation to calculate the effects of controlling variables such as wind speed, irradiance and vegetation height. The same issue of the time resolution arises here as for the estimation of the effects of environmental variables on stomatal conductance. Although stomatal and atmospheric terms can be added at any instant in time to determine total conductance, this is not possible for monthly means. Hence, the atmospheric term will be taken into account alongside the stomatal term in assigning relative potential sensitivity, but only in a semi-quantitative manner.

Conclusions

In summary, the model offers the potential of mapping spatial and temporal variation in ozone flux over Europe for a limited number of species. However, it only allows potential sensitivity to be estimated, as the detoxification potential of individual plants to ozone is not included. The major constraint to the application of the methodology is data availability. It should be noted that this includes not only data on stomatal responses; GIS data on both a regional and temporal scale also limit the manner in which the model can be used.

These limitations mean that the method cannot be used to derive and map Level II critical

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level values. However, the maps produced will indicate the potential scale of variation in sensitivity to ozone which exists over Europe and indicate how the spatial variation in sensitivity varies from month to month. Hence the significance of these factors for the evaluation of ozone impacts can be considered, and by placing them alongside critical level exceedence maps, some consideration of receptor sensitivity may be incorporated in the development of pollution control strategies.

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SURFACE OZONE AT THE CZECH REGIONAL STATION KOŠ

PEKÁREK Jaroslav, VÁ_A Milan

Czech Hydrometeorological Institute,Observatory Košetice

CZ-394 22

Abstract. The surface ozone monitoring is implemented within the framework of international programmes GAW/WMO and EMEP at the regional station Košetice (Czech Hydrometeorological Institute). The results of the surface ozone measurements in the period 1992 - 1994 are presented. Mean annual concentration of surface ozone at the observatory Košetice (about 75 mg/m^3) is together with the data from similarly located German stations the highest within the EMEP network. The air quality standard for the concentrations of surface ozone (8-hours' moving average of 160 mg/m^3) is surpassed quite often, especially during the summer period.

1. Basic information about the station

- Latitudinal and longitudinal co-ordinates : N 49°35', E 15°05'

- Elevation above sea-level : 534 m

- Basic climate characteristics (Period 1961-1990)

  Mean annual temperature : 7.1 °C

  Total annual precipitation : 621 mm

  Prevailing wind direction : western

  Number of snow days per year : 60 to 100

  Mean hours of sunshine per year : 1 800

2. Measurement of surface ozone

The measurement of the surface ozone concentrations at a regional level within the scope of the EMEP programme was started at the station Košetice in the year 1989. The Measurement was performed with the automatic analyzer Dasibi (hour means), when both stations have become a part of the AIM Czech network (in the course of the year 1992) the measurement with the automatic analyzer Thermoelectron M 49 (half-hour means) was applied. The measurement with the analyzer Dasibi was affected with a number of failures and for this reason the results obtained in the years 1992-1994 were used for evaluation.

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3. Evaluation and conclusion

- Mean annual concentrations of surface ozone at the observatory Košetice (about 75 mg/m3) are together with the data from similarly located German stations the highest within the EMEP network. Higher values are recorded only at mountain stations. [2].

- Very similar annual and monthly characteristics as at the observatory Košetice were recorded also at the other Czech regional station Svratouch [3].

- Mean annual and monthly concentrations at regional stations are always conspicuously higher than in impacted areas [4].

- Surface ozone concentrations have evident annual variation with maxima during spring months. In June and sometimes even in July a decrease takes place which is caused by the onset of the so-called European continental monsoon bringing an increased cloudiness and decrease of solar radiation. In very warm summers as e.g. in 1992 or 1994, we register a second maximum in July and August (Figure 1). Minimal values are registered in winter [3].

- Diurnal variation of surface ozone concentrations is typical with the minimum at about 6 a.m. and the maximum between 2 and 4 p.m. (Figure 2). Daily amplitude increases with the intensity of total radiation [3].

- Direct dependence of surface ozone concentrations on sunshine duration and intensity of total radiation is presented (Figure 5 and Figure 6) [3].

- The concentrations exceeding air quality standard are measured at a regional level (8-hours' moving average higher than 160 mg/m^3 and short-term critical level AOT40 > 700 ppb.h) at some episodal situations (Table 1 and Table 2) . These situations happened during very warm summer days with a clear sky and high intensity of solar radiation.

- The daily concentrations of nitrogen oxides in all days, when
the air quality standard of the surface ozone concentrations was surpassed, was lower than 15 mg/m3.

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- The course of surface ozone concentration during the episode with the up to now highest concentrations at the turn of July and August, 1994 is presented (Figures 11 and 12).

Table 1. Number of exceeds of 8-hours' moving average concentrations of surface ozone at the observatory Košetice in 1992 - 1995.

limit	number of exceed			
$\mu g/m^3$	1992	1993	1994	1995*
200	1	1	0	0
180	6	1	6	1
160	14	9	16	9
140	49	28	40	29

* January - September

Table 2. Number of days contained in at least one three-days period where short-term critical level (AOT40) exceeds 700 ppb.h in 1992-1994.

	Apr	May	Jun	Jul	Aug	Sep	total
1992	7	21	13	15	18	8	82
1993	11	14	7	4	10	0	46
1994	5	2	12	24	10	0	53

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5. Figures

FIGURE 1 **Surface ozone and nitrogen dioxide (monthly means) and total radiation (monthly sums) in the period 1992 - 1994.**

FIGURE 2 **Mean diurnal concentrations of surface ozone in the period 1992 - 1994.**

FIGURE 3 **Mean diurnal concentrations of surface ozone and total radiation in the warm (April - September) period 1992 - 1994.**

FIGURE 4 **Mean diurnal concentrations of surface ozone and total radiation in the cold (October - March) period 1992 - 1994.**

FIGURE 5 **Mean diurnal concentrations of surface ozone by various sunshine duration in the cold period 1992 - 1994.**

FIGURE 6 **Mean diurnal concentrations of surface ozone by various sunshine duration in the warm period 1992 - 1994.**

FIGURE 7 **Mean diurnal concentrations of surface ozone and mean diurnal sums of total radiation in the cold period 1992 - 1994 in the days without sunshine.**

FIGURE 8 **Mean diurnal concentrations of surface ozone and mean diurnal sums of total radiation in the warm period 1992 - 1994 by 8 and more hour sunshine.**

FIGURE 9 **Cumulative distributions of surface ozone concentrations in the cold period 1992 - 1994 according to the sunshine duration.**

FIGURE 10 **Cumulative distributions of surface ozone concentrations in the warm period 1992 - 1994 according to the sunshine duration.**

FIGURE 11 **Surface ozone concentrations, episode 20.VII.1994 - 11.VIII.1994. Air quality standard are 160 mg/m^3 (8-hour moving average).**

FIGURE 12 **Surface ozone concentrations , episode 5.VII.1994 - 7.VII.1994.**

FIGURE 13 **Surface ozone concentrations, episode 25.VII.1995 - 28.VII.1995.**

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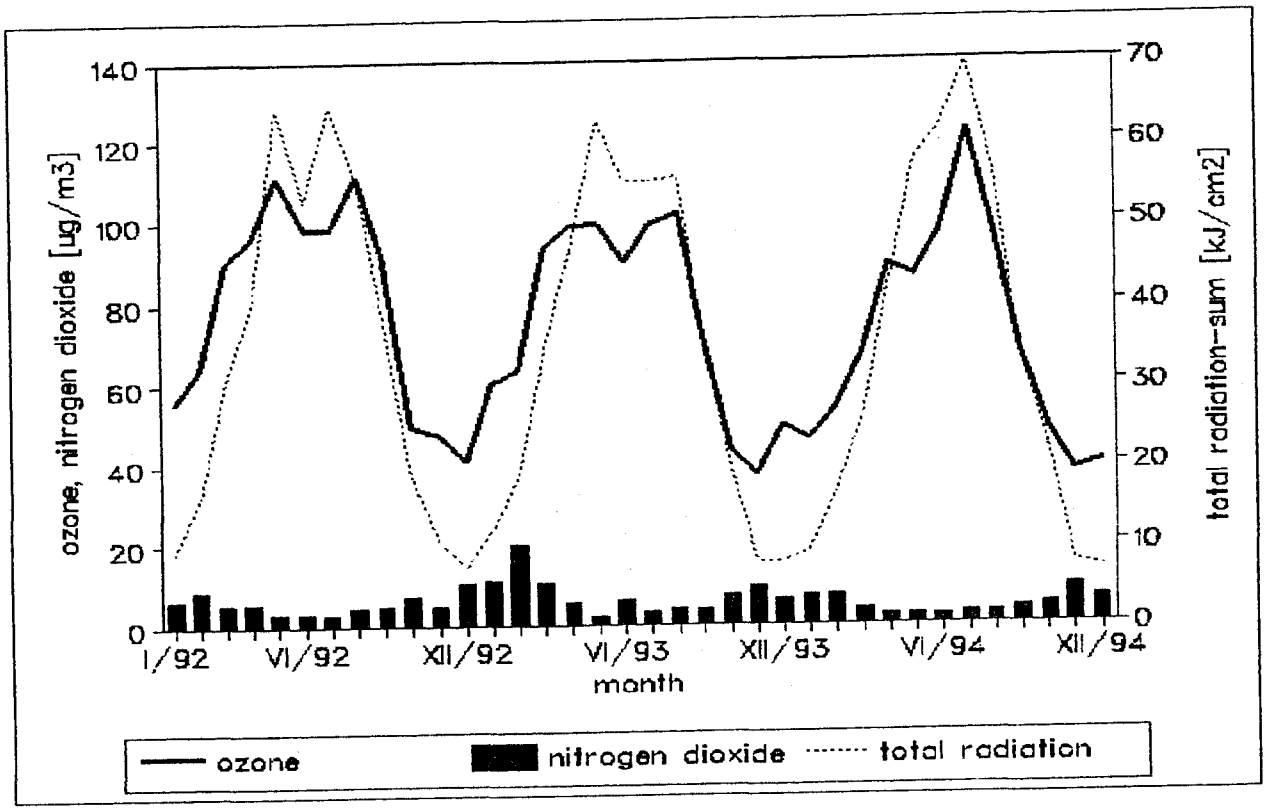


FIGURE 1 Surface ozone and nitrogen dioxide (monthly means) and total radiation (monthly sums) in the period 1992 - 1994.

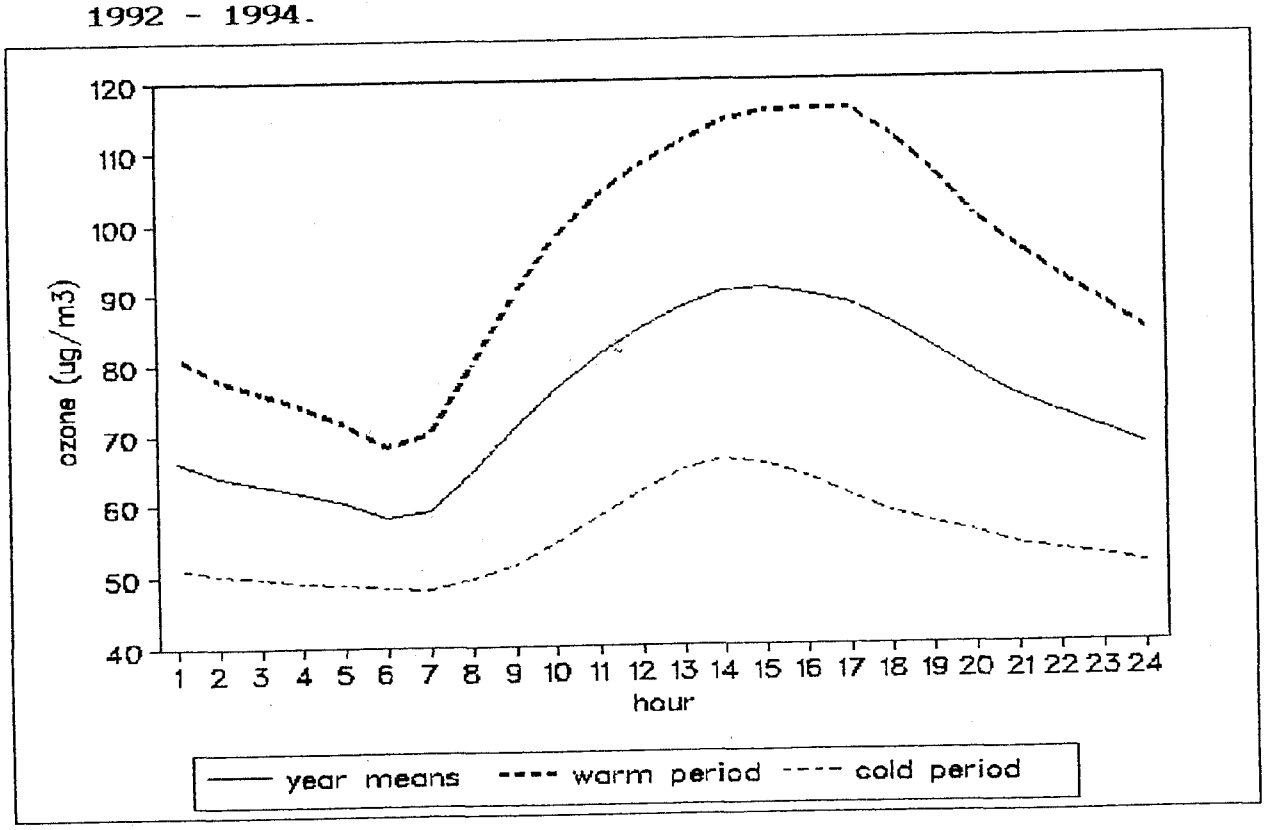


FIGURE 2 Mean diurnal concentrations of surface ozone in the period 1992 - 1994.

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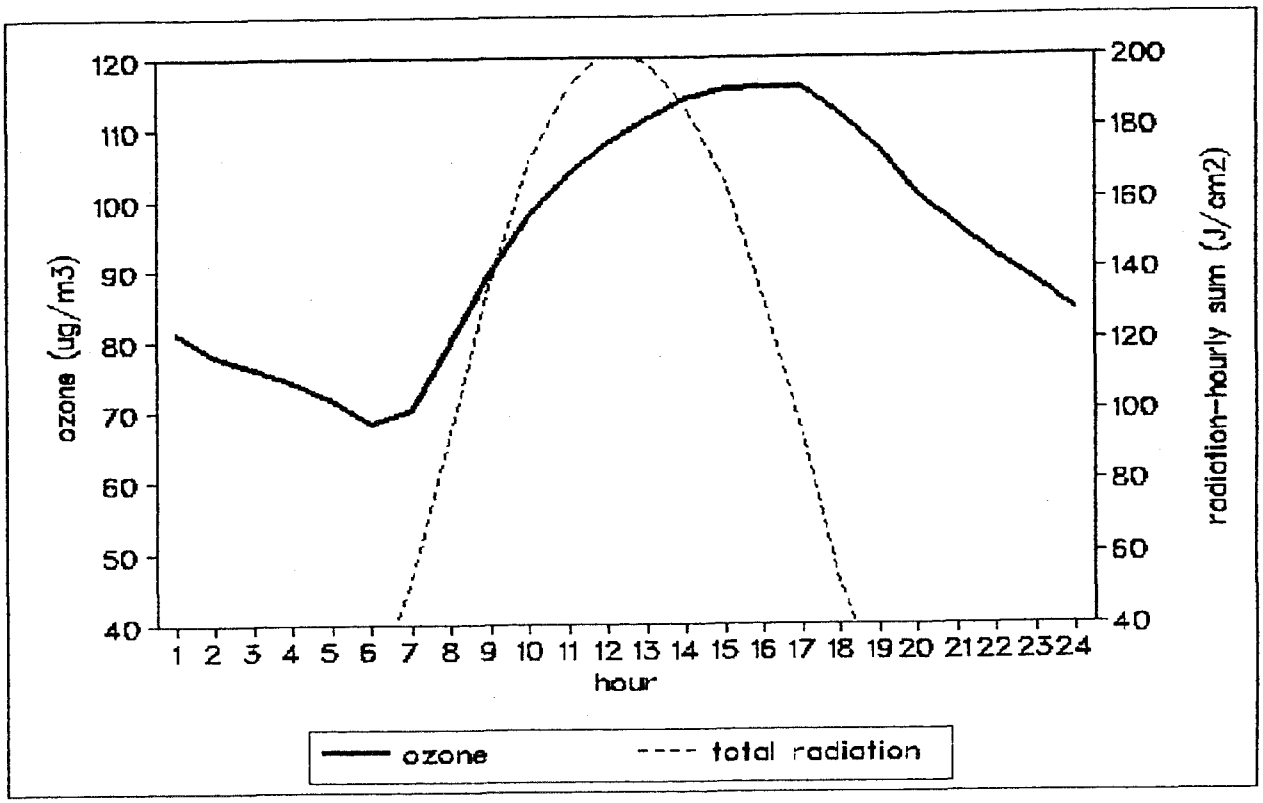


FIGURE 3 Mean diurnal concentrations of surface ozone and total radiation in the warm (April - September) period 1992 - 1994.

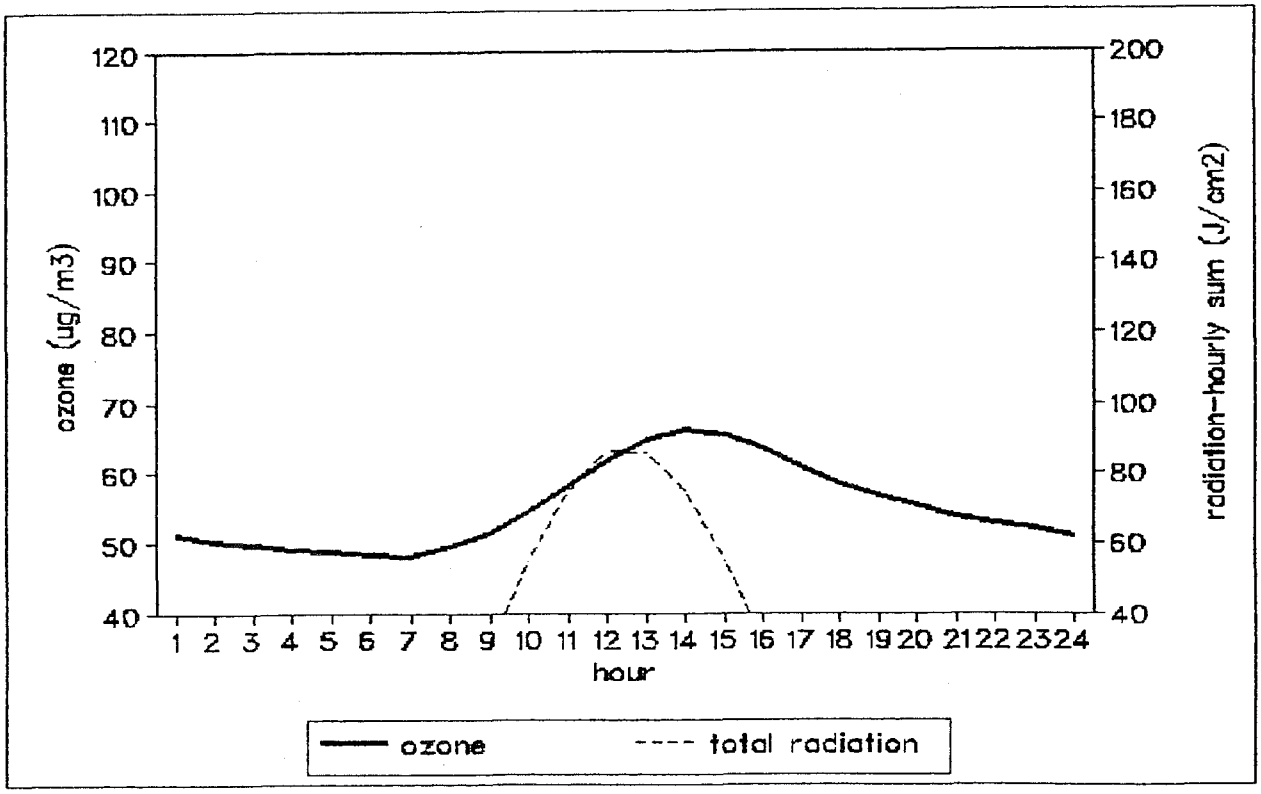


FIGURE 4 Mean diurnal concentrations of surface ozone and total radiation in the cold (October - March) period 1992 - 1994.

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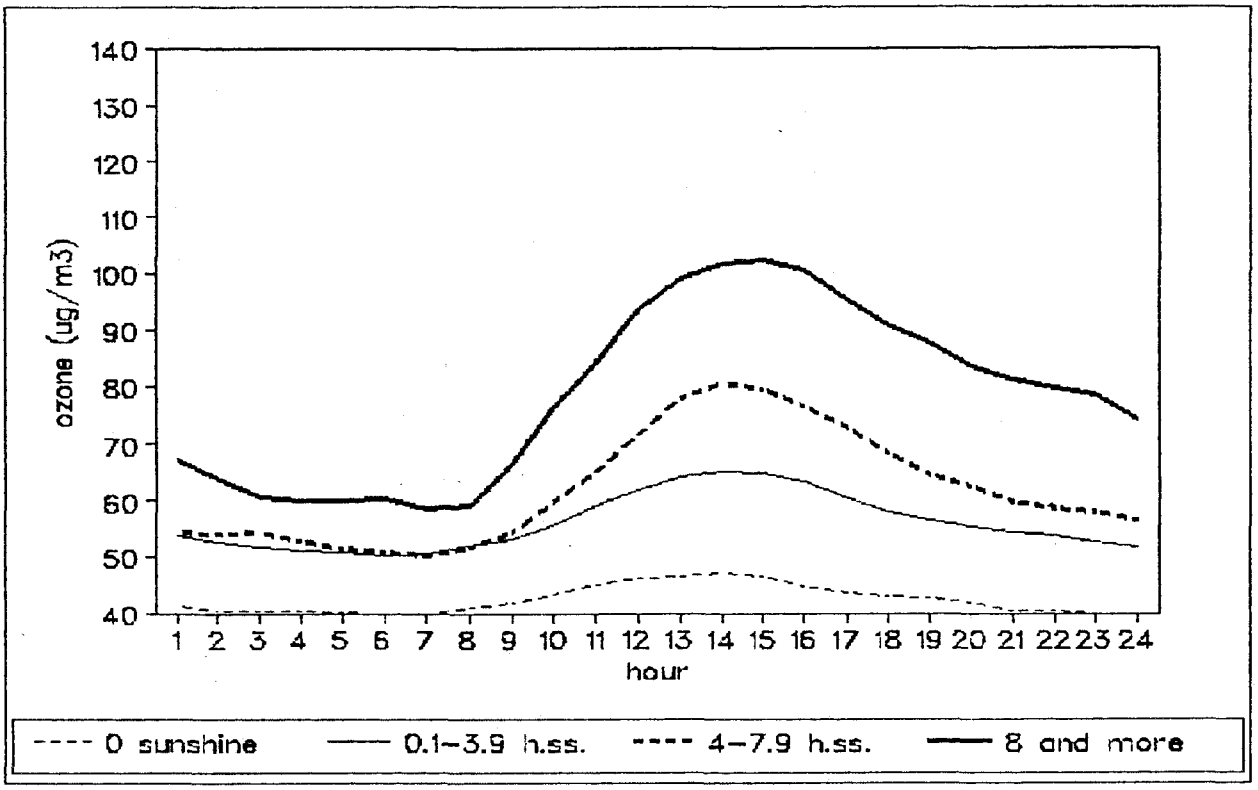


FIGURE 5 Mean diurnal concentrations of surface ozone by various sunshine duration in the cold period 1992 - 1994.

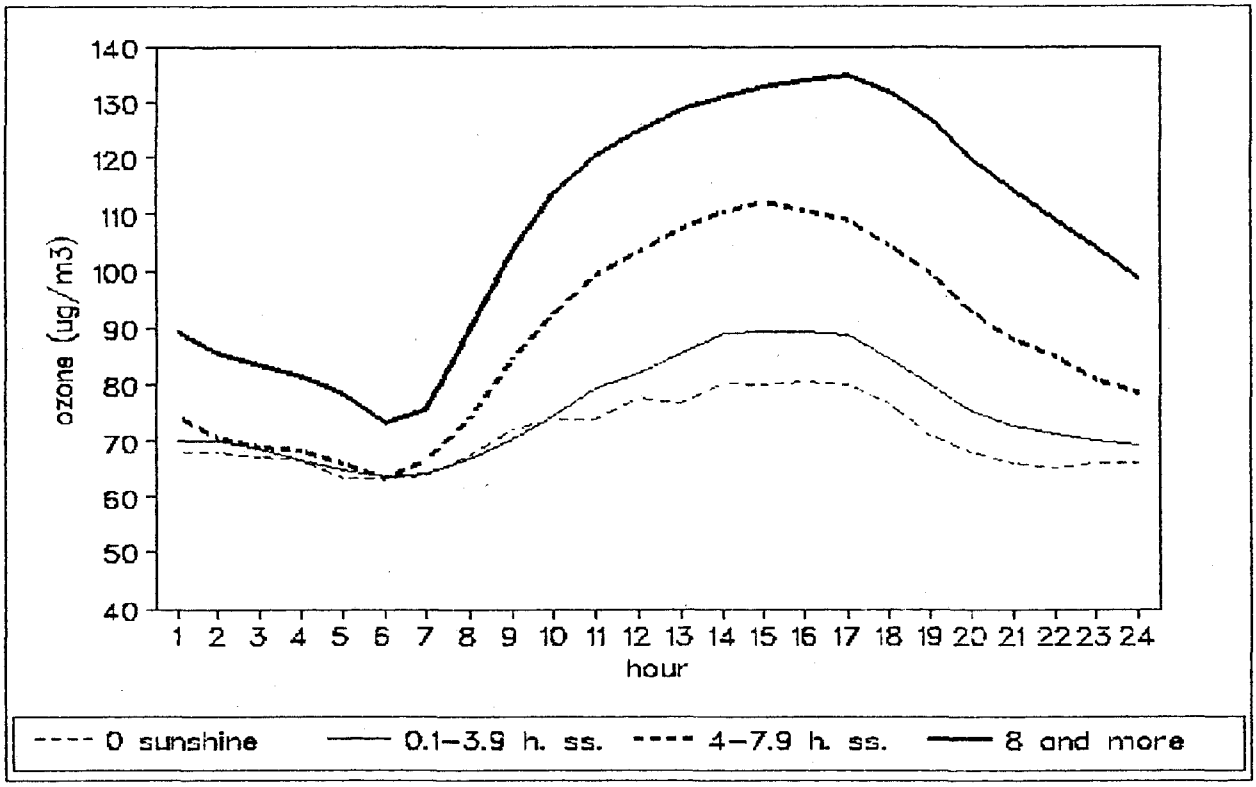


FIGURE 6 Mean diurnal concentrations of surface ozone by various sunshine duration in the warm period 1992 - 1994.

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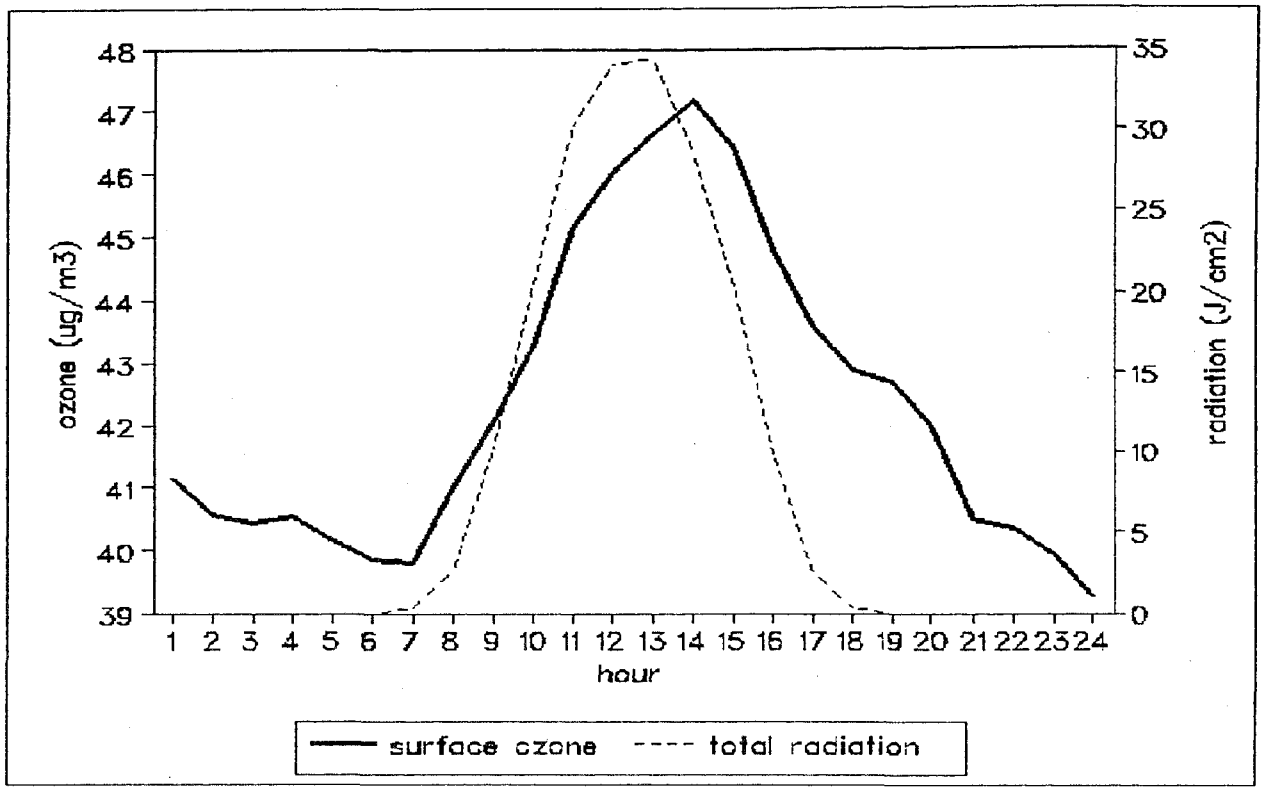


FIGURE 7 Mean diurnal concentrations of surface ozone and mean diurnal sums of total radiation in the cold period 1992 - 1994 in the days without sunshine.

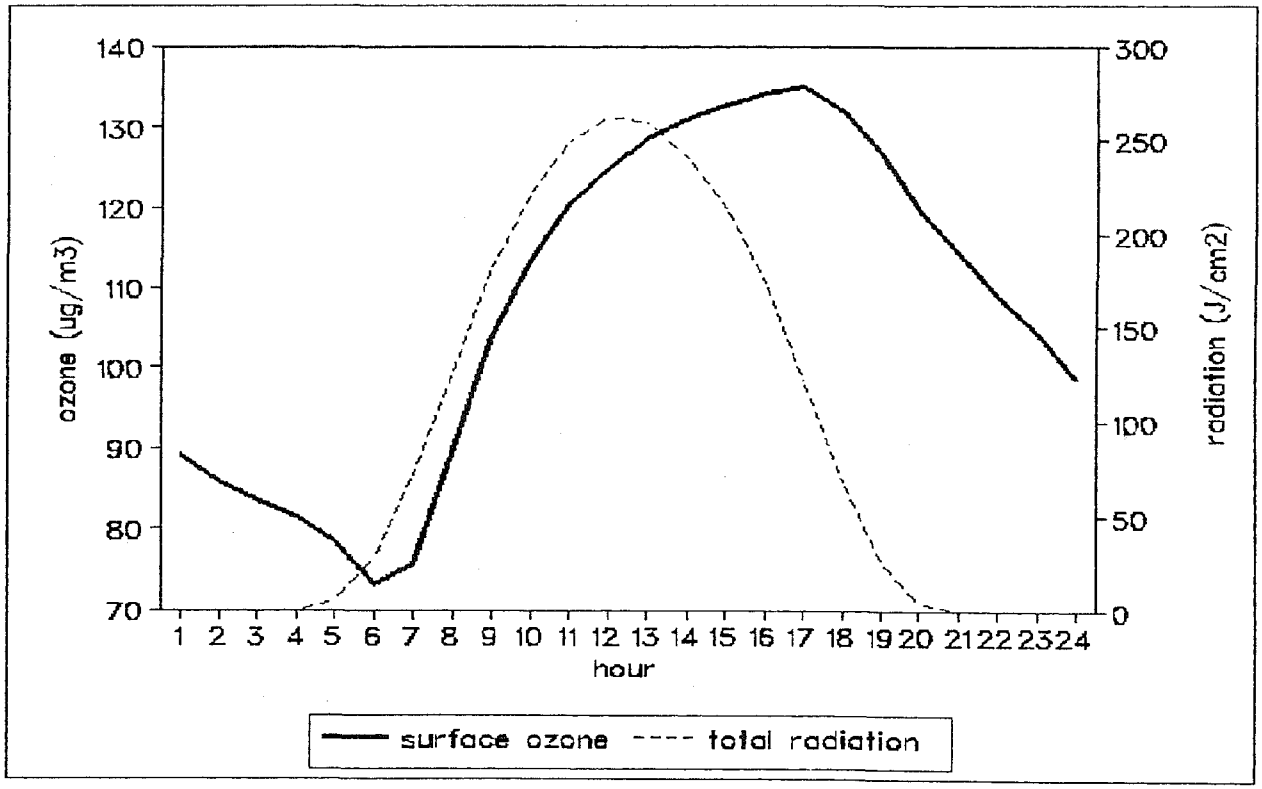


FIGURE 8 Mean diurnal concentrations of surface ozone and mean diurnal sums of total radiation in the warm period 1992 - 1994 by 8 and more hour sunshine.

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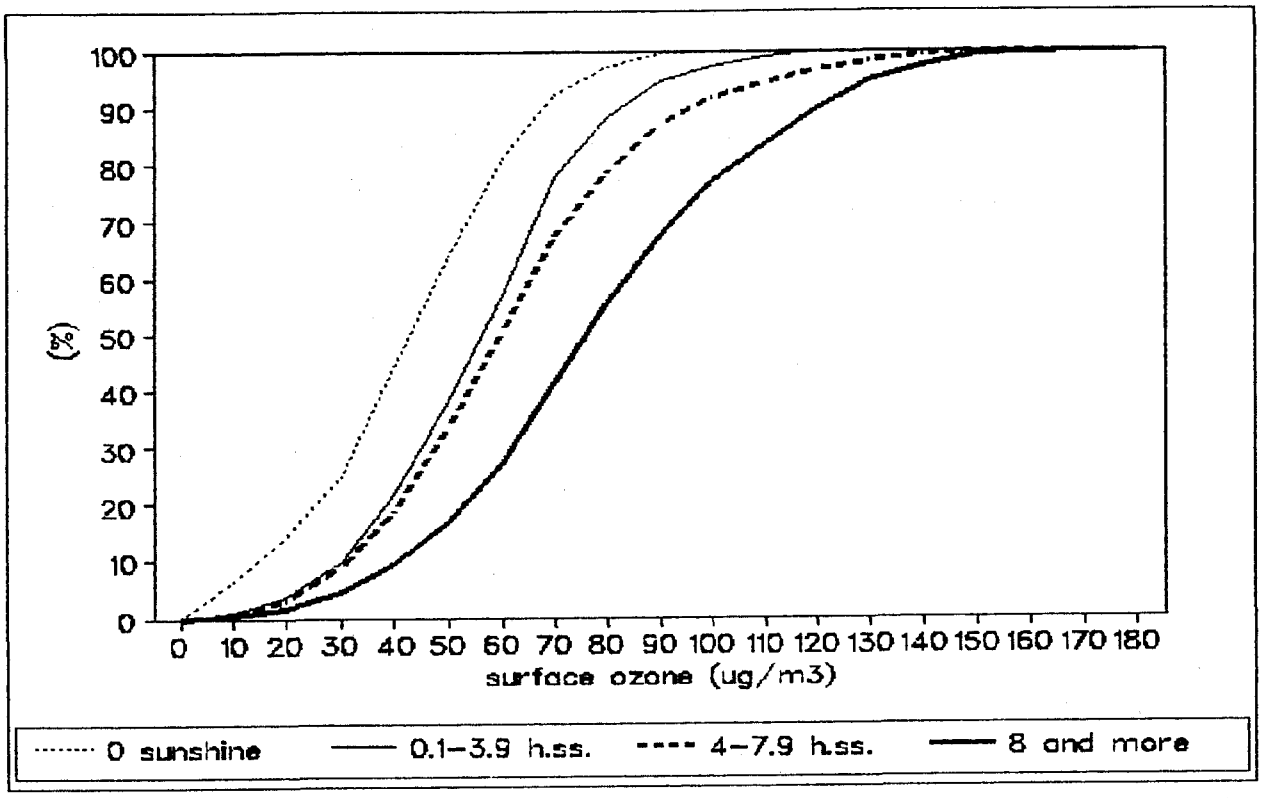


FIGURE 9 Cumulative distributions of surface ozone concentrations in the cold period 1992 - 1994 according to the sunshine duration.

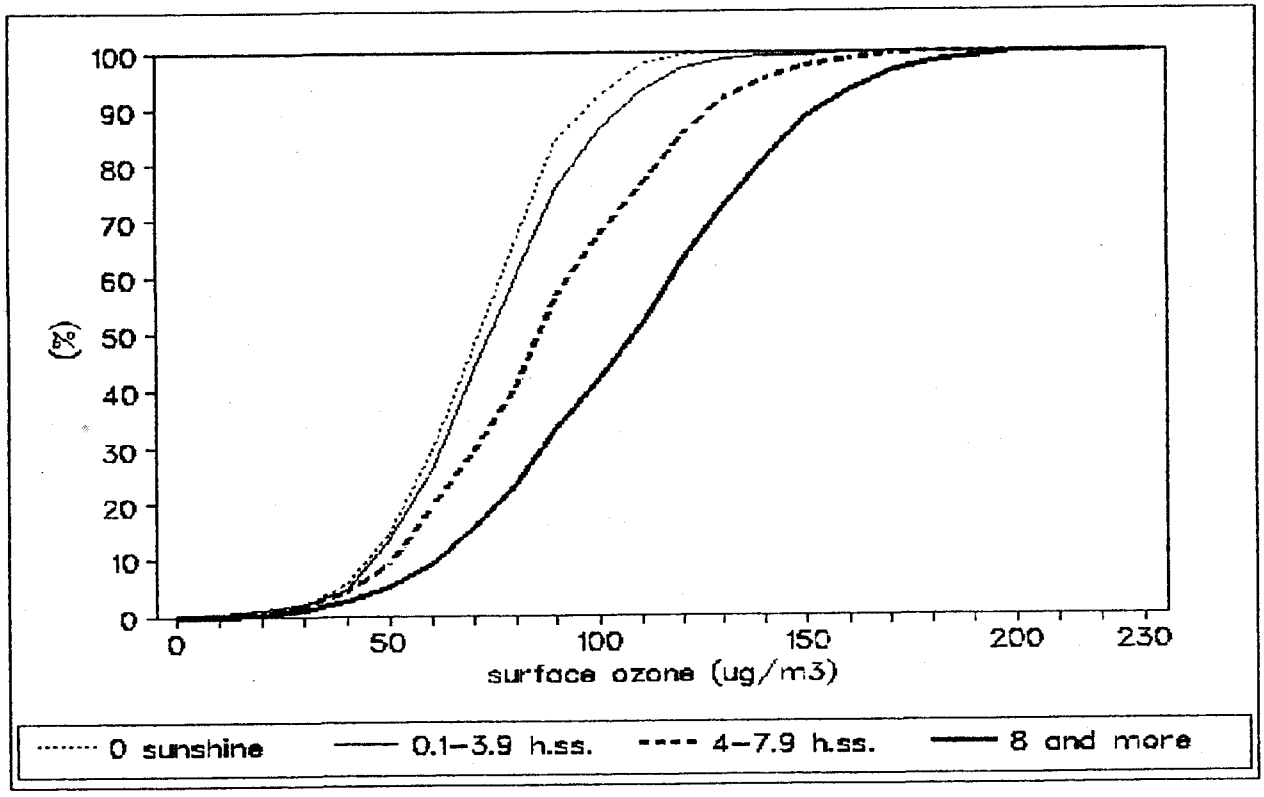


FIGURE 10 Cumulative distributions of surface ozone concentrations in the warm period 1992 - 1994 according to the sunshine duration.

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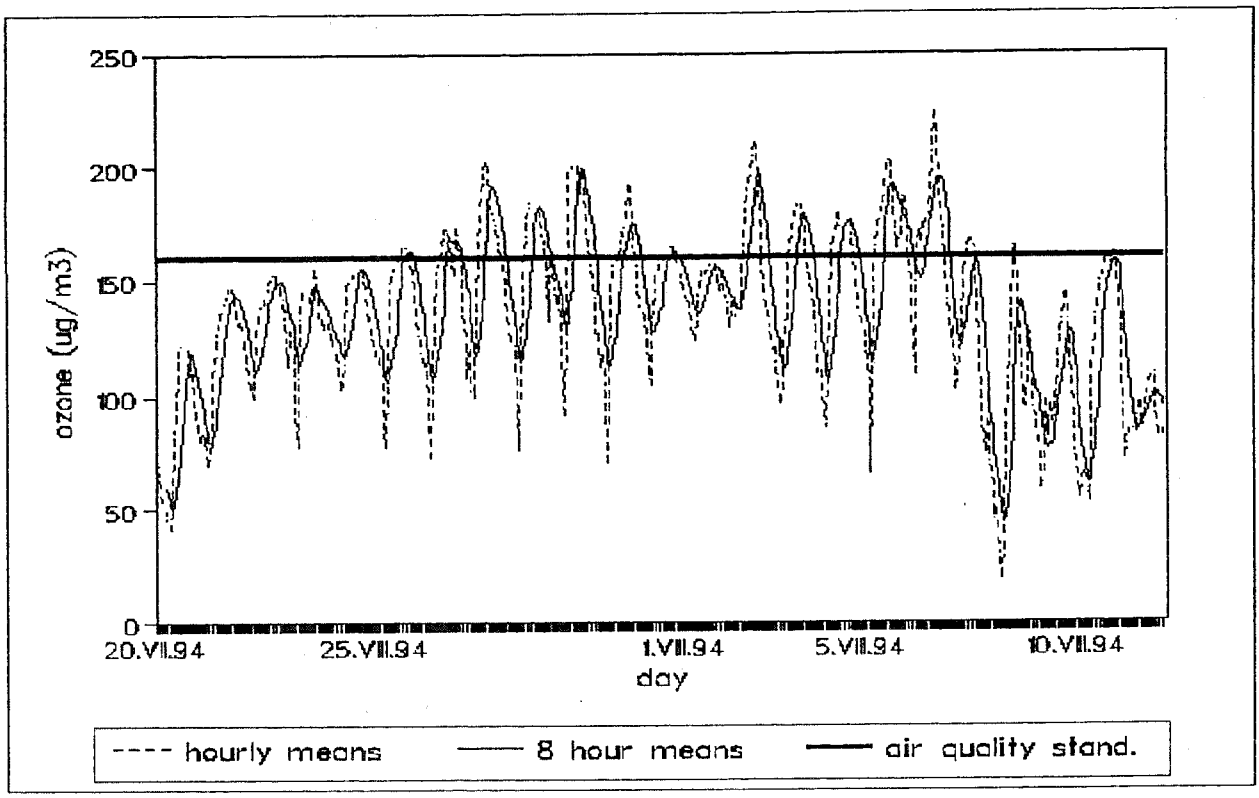


FIGURE 11 Surface ozone concentrations, episode 20.VII.1994 - 11.VIII.1994. Air quality standard are 160 µg/m3 (8-hour moving average).

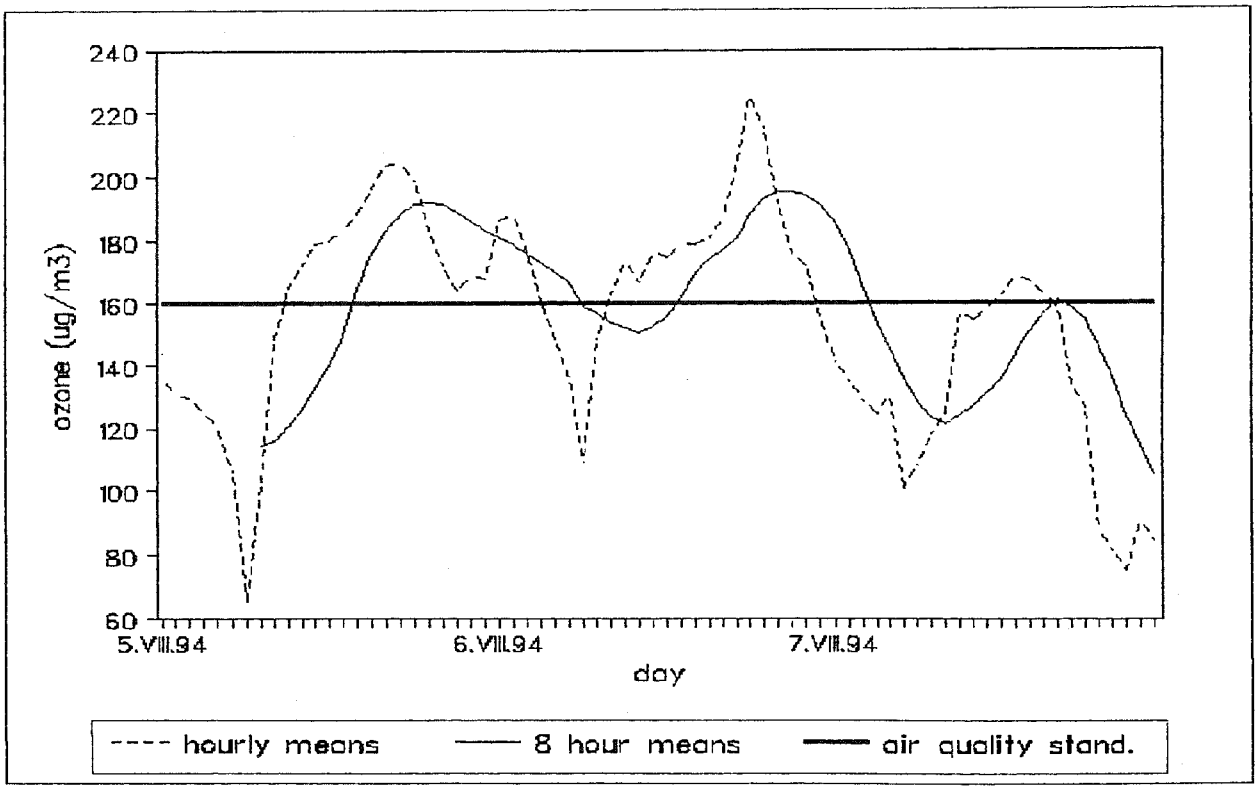


FIGURE 12 Surface ozone concentrations , episode 5.VII.1994 - 7.VII.1994.

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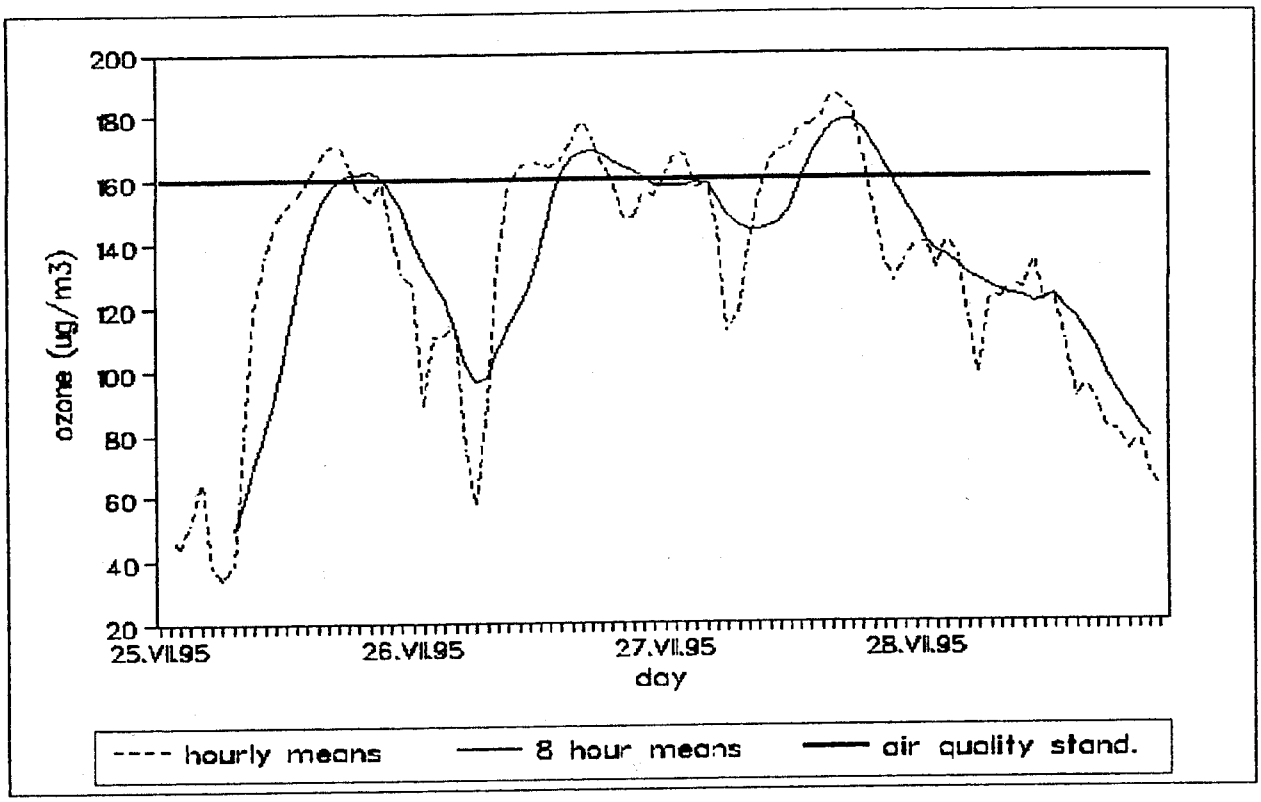


FIGURE 13 Surface ozone concentrations, episode 25.VII.1995 - 28.VII.1995.

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Mapping of the Accumulated Ozone Exposure and Exceedances of Critical Levels in Austria

Loibl W. and Schneider J. *

Austrian Research Center Seibersdorf, Dept. Environmental Planning/SU, A-2444 Seibersdorf, Austria;
*: Federal Environmental Agency, Spittelauer Laende 5, A - 1090 Vienna, Austria

Abstract

High tropospheric ozone concentrations especially during the summer months are a major environmental problem in most parts of Europe. Exposure to elevated ozone levels can cause negative effects on human health and vegetation. Critical Levels for ozone exposure (defined as AOT40 values over certain growing periods, see *Critical Levels for Ozone, Fuhrer and Achermann (1994)*) were set up within the Convention on Long Range Transboundary Air Pollution to evaluate the effects of ozone on vegetation. If the actual exposure exceeds the Critical Level, adverse effects of ozone exposure on vegetation can occur. To map the *spatial* distribution of ozone exposure, the measurements carried out at discrete points have to be 'interpolated' using more or less sophisticated models. This interpolation is not easy especially in mountainous regions because the ozone concentration (and therefore the AOT40 value) is highly dependent on several parameters as relative altitude, distance from sources of ozone precursor substances, etc. To overcome these problems, a special daytime- and elevation-dependence function was developed to calculate ozone concentrations in complex terrain (*Loibl W. et al., 1991, 1992, 1993, 1994b*). The spatial distribution of AOT40 values at a resolution of 2,5 x 2,5 km was calculated starting from the ozone measurements conducted at approximately 120 monitoring sites located throughout the federal territory of Austria. First, the measured ozone concentrations were interpolated over the whole territory. These calculations were carried out for every hour (i.e. every one hour mean). In a second step the AOT40 was determined for each grid point by summing up the calculated ozone concentration depending on the receptor type (forest and agricultural areas) corresponding to the recommendations of the Bern Workshop (*Fuhrer and Achermann, 1994*).
These calculations show dramatic exceedances of Critical Levels for ozone for forest ecosystems and crops, respectively, in most regions of Austria.

1. Introduction

The Critical Levels approach was adopted by the UN/ECE to control emissions of gaseous pollutants. The aim of this effect-orientated approach is to specifically protect areas and ecosystems exposed to evelated levels of air pollutants.
Within this process the actual (accumulated) ozone exposure is mapped as an AOT40 value and compared to the value of the Critical Level. If the actual exposure exceeds the Critical Level in a specific region, measures to amend the ozone exposures via reduction of emissions of ozone precursors are necessary.
Mapping the spatial accumulated ozone exposure in a country like Austria with complex terrain is not an easy task. In mountainous regions complex transport phenomena and chemistry lead to large variations ot the ozone concentration within short distances between areas of different altitudes in a daytime specificpattern. To estimate the spatial ozone distribution in mountainous regions a complex algorithm was developed *(Loibl W. et al.,*

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1991, 1992, 1993, 1994b). It is based on data from ozone monitoring stations and a function that considers the elevation dependence of the diurnal ozone concentration variations.

2 Ozone formation in mountainous regions and model assumptions

Ozone formation processes are well known since the early 1950's. Peak levels of ozone usually occur when there are

- considerable background concentrations (mainly caused by long range horizontal transport) and
- local photochemical ozone formation due to high solar radiation and high ozone precursor concentration.

Nocturnal decreases are caused by chemical consumption and dry deposition of ozone, enhanced in cases when a temperature inversion is formed.

In mountainous anthropogenic emission sources of ozone precursors (mainly VOCs and NOx) are usually located at the bottom of larg valleys and basins. Vertical mixing induced by thermal turbulence causes vertical mixing both of ozone and its precursors causes rather homogenuous vertical ozone distribution during daytime, whereas during night strong vertical concentration gradients form (high concentrations at high altitudes, low concentrations in valleys). At the bottoms of valleys and basins, at night ozone is depleted by dry deposition and chemical consumption, mainly by NO, a process inforced in case of termperature inversions. These processes leads to a bell-shaped daily concentration variation. On the other hand, at mountain-tops low nocturnal ozone deplection occurs due to low rates of dry deposition and relatively strong air exchange with the free troposphere. (*Broder. et al., 1985, Puxbaum et al, 1991*).
Therefore the diurnal variations of ozone concentrations shows a characteristic pattern at different altitudes:
In valleys and plains the diurnal variation of ozone concentration is bell-shaped, with a maximum during afternoon hours. In contrast, the ozone concentrations stay at a higher level and usually do not vary as much during night and day hours at higher altitudes.
Additional theoretical considerations as well as empirical investigations lead to the conclusion that ozone concentrations depend not on the absolute altitude above sea level, but the relative altitude above the nearest valley ground.(*Rau G. et al. 1991, Loibl W. et. al. 1991, 1993).*
It is evident that the diurnal variation of ozone concentration depends on many additional factors like emission density of ozone precursors, solar radiation, atmospheric convection, wind speed and others. Anyhow, starting from monitoring data as a basic input to map the spatial pattern of ozone levels, a realistic picture of the true situation can be obtained.

3 The Model

The algorithm to interpolate ozone concentrations can be described as a statistical model similar to the spatial application of a regression model which is used to describe the overall spatial trend as regression surface. Residuals resulting from local influences (which are causing deviations from the spatial trend) are interpolated and added to the regression surface as regional deviations.
The elevation dependence of the ozone concentration is changing continuously during the day. Spatial ozone concentration pattern can be generated applying a statistical model (adapted to general daytime specific ozone concentration trends) together with a digital elevation model to derive a statistical ozone concentration surface. The deviations from the results of the

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statistical model at the monitoring sites are interpolated and included as deviation surface to the ozone concentration surface. The result is the expected spatial ozone concentration pattern including the regional deviations.

The following overview describes shortly the steps of generating ozone concentration maps.

- **Calculating standard ozone concentrations using the elevation/time-dependence function**
  A statistical function was approximated to estimate standard ozone concentrations depending on relative altitude and day-time. The results of the function are daytime-specific elevation-dependence curves of the standard ozone concentration depending on relative altitude.
- **Fit of the elevation/time-dependence curves to the day-specific ozone situation**
  Because of different day-specific ozone situations, adaptations of the elevation-dependence curves have to be made. The results are 'expected ozone concentrations' for a specific day and specific day-time, depending on altitude.
- **Calculating the 'expected day-specific hourly ozone concentration'-distribution**
  The 'expected day-specific spatial ozone concentration' pattern, reflecting the measurements, is calculated using the adapted elevation dependence curves and the digital elevation model. The results are presented as grided data with the spatial resolution of the elevation model.
- **Calculating the local deviations of the measurements from the elevation-dependence curves**
  Measurements at the monitoring stations deviate from the 'expected day-specific ozone concentration', because of local and meteorological influences. The (recalculated) deviations of the measurements from the expected concentration represent the remaining deviations due to local influences to the ozone concentration.
- **Calculating the regional deviations by spatial interpolation of the local deviations**
  Assuming the monitoring stations are situated at representative locations, local deviations reflect not only the ozone situation at the monitoring sites but also the ozone situation within the region surrounding these sites. Spatial interpolation of these site-specific deviations generates a spatial deviation pattern as grid model, representing regional influences.
- **Generating ozone concentration maps**
  Maps of the spatial ozone concentration pattern considering the regional influences are generated for every hour by summing up the grid of the expected ozone concentration pattern and the grid of the regional deviations which includes the regional influences to the ozone concentration.

Within the following chapters a more detail description is given.

4. The model details

4.1. The daytime and elevation dependence function

To describe the diurnal variation of the ozone concentration depending on daytime and elevation, an analytical function was approximated by non-linear estimation, that reflects these influences. The function was approximated using scatter-plots of the average of half-hour mean values of ozone measurements against the relative altitude of 100 to 120 monitoring stations throughout Austria. The average ozone concentration (per half hour) was calculated for several ozone episodes of the years from 1991 to 1993. The relative elevation is defined as the

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difference between the absolute elevation of the monitoring station and the point of the lowest elevation, with optimal results within a radius of 5 km distance.

The function, describing the ozone concentration, originally consists of 2 exponential sub-equations *a* and *b*. *b* describes the general influence of altitude as a logarithm-dependence while *a* and *b* are describing the continuous variation of the influence of day-time. *(Loibl W. et al. 1992, published in English as Loibl W. et al., 1994a).*
The following equation (1) shows the last version of the model with resolved sub-equations and all single parameters. *(Loibl W. et al., 1993).*

$$Ostd_{(hrel,t)} = (a_1 + a_2 \cdot e^{-(t-a_3)^2 \cdot a_4}) \cdot \ln\left(\frac{b_5 \cdot h_{rel}}{100} + b_1 + b_2 \cdot e^{-(t-b_3)^2 \cdot b_4}\right) - \frac{c_1}{1-(t-c_2)} \cdot \frac{h_{rel}}{c_3} \qquad (1)$$

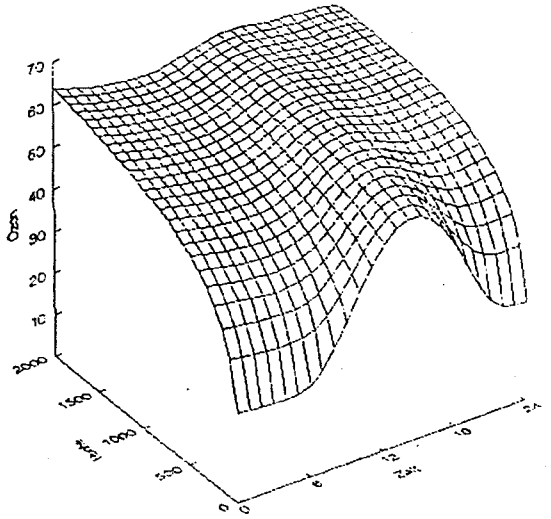
with O_{std}=standard-ozon-concentration, h_{rel}=relative altitude , t=day-time,
a_x, b_x, c_x = parameters of the function and e= exponent

The function results in a 2-dimensional trend surface of the standard ozone concentration against elevation and daytime (fig.1):

- Along the y-axis, curves per time interval show the ozone concentration gradient dependent on rising elevation - this elevation dependence curves of the ozone concentration are of main interest for the application and day specific adaptation of the model
- Along the x-axis, curves show different concentrations at different elevation levels during the day: bell-shaped in low regions, line-shaped with stable concentration at higher levels in elevated regions

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Fig.1: Trend surface of the day-time- and elevation-dependence function of ozone



The results of the function, used as curves of ozone concentration (along the y-axis) against relative altitude (along the x-axis) represent the expected ozone concentrations at given relative altitudes for every half hour. Because of general day-specific and local influences the measured ozone concentration deviate from the modelled ones. In order to separate the locally caused deviations from general day-specific influences which lead to country-wide effects on the ozone concentration pattern, these country-wide effects have to be integrated into the elevation dependence curves.

This step is done using a curve-fitting procedure. The procedure uses the mean deviations of the measurements from the calculated standard ozone concentration data within 3 elevation ranges and is shifting the elevation dependence curve considering these elevation range specific deviations. This curve fitting is included into the approach since 1993. *(LOIBL W. et. al. 1993)* As advantage of this modification, the adapted elevation dependence curves allow to estimate day-specific ozone concentration patterns not only for episodes with high ozone concentrations, but also concentration patterns during episodes with moderate or even low ozone concentrations during early spring and late summer days.

4.2 Calculation of the expected day-specific ozone concentration distribution

The calculation of the day-specific ozone concentration distribution is performed using the adapted ozone concentration curve and a digital elevation model (DEM) of the relative altitude. The used elevation model has a spatial grid-point resolution of 1 km x 1 km. A DEM of relative altitudes was created by calculating the elevation difference between every point of the actual DEM and the DEM point of the minimum elevation within a specified search radius (here 5 km).
The expected ozone concentration is calculated separately for all DEM - grid-cells by including the grid-cell-specific altitude and the day-time into the equation. The result is a day- and day-time specific ozone concentrations surface considering the elevation dependence with the same spatial resolution as the DEM.

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4.3 Adding the local deviations from the monitored half-hour mean values to the expected day-specific ozone concentration

General day-specific and local influences can lead to discrepancies between measured and modelled ozone concentrations. After adaptation of the elevation- dependence curves the ozone concentration pattern includes the day-specific country-wide influences. The remaining deviations of the actual ozone measurements from the expected ozone concentrations reflect local influences which lead to 'atypical' ozone concentrations.
Local influences effect the ozone concentration in the vicinity of the monitoring sites. The pattern of the regional deviation from the expected concentration can be calculated by interpolation of the deviations at the monitoring sites. In this procedure it was assumed that the influence of the effects which lead to deviations is decreasing with growing distance to this monitoring site, while the influence of the effects of other neighbouring monitoring sites is increasing with decreasing distance to this monitoring sites. The result of this interpolation is a (statistical) deviation surface which covers the region to be investigated.
The spatial interpolation is performed by the inverse distance-weighting algorithm (IDW) - a linear interpolation algorithm where the influence of data points are depending on the distance. The influence of one data point on another declines with distance from the point being estimated. The IDW- algorithm will always produce an interpolation surface, where the calculated grid-cell-values are between those observed values of the neighbouring monitoring sites. *(see Loibl W. et al., 1993a)*

The deviations turned out to be elevation-dependent due to higher local influences within the valleys than in mountainous regions: The lower the relative elevation is, the higher the deviations are scattering around the ozone concentration curves. The symmetry of the scattered deviations along the x-axis, which represents zero deviation from the expected elevation dependent ozone concentration, shows that the function is realistic - there are no systematic deviations in plus or minus direction *(Loibl W. et al. 1993)*. An interpolation of the deviations without eliminating the elevation-specific effects may lead to implausible results in special cases inside regions with high elevation differences. Therefore a sub-step within this deviation-interpolation has to be included to correct the elevation dependence of the deviations. The details of the correction can be read in the above cited report.

4.4 Combining the spatial day-specific ozone concentration with the regional deviation from the expected concentration

The unification of the ozone concentration surface with the deviation surface is performed by a simple cell-by-cell addition of the values of the two grids produced by the model and the deviation interpolation. The resulting map shows the spatial patterns of the elevation-dependent ozone concentration including the regional deviations from the expected values.
Because of the curve-fitting algorithm, the elevation-dependence curves can be automatically adapted to the day-specific elevation-dependence of the ozone concentration for every random hour and day, at least during the period from early spring to late autumn.

5 Comments on the validity of the results

The validity of the function can be easily tested by creating scatter-plots with deviations of the measurements from the model results against the relative altitude. The deviations will usually scatter symmetrically along the x-axis from low to high elevated areas. *(ref. to Loibl W. et al.(1993), fig.6)*.

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At the monitoring sites the calculated ozone concentration will certainly be the same as the measured ones because of the inclusion of the deviations. (The linear interpolation of the deviations does not allow differences to the original data at the monitoring sites. Only if the spatial resolution of the elevation model is low and more than one monitoring stations are situated inside one grid-cell-element, will the results inside this cell be a mean value of all monitoring sites situated there).
To test the spatial representativeness of the results for regions in-between the monitoring stations, ozone concentration maps can be calculated with and without the exclusion of several monitoring sites. Spatial representativeness is proved if both maps show the same or nearly the same results. (This test is only significant if there are no monitoring stations excluded within regions of great influence to the ozone formation process. If one excludes these stations, there will certainly be a difference between such two test maps).
Tests with monitoring data show just little deviations - up to a few ppb during hours of high ozone concentration and usually below 10 ppb during early morning hours, when ozone concentration reacts more sensitively to anthropogenic influences. The exclusion of monitoring stations with typical 'town characteristics' (such as early morning ozone depressions) leads to significant deviations from the ozone concentration pattern calculated using the complete set of monitoring stations. The differences may range up to 30 ppb - the exclusion of monitoring stations in towns leads to an overestimation of ozone concentrations within the close region surrounding the respective town.*(ref. to Loibl W. et al., 1993, annex 3).*

6 AOT40-maps

Critical Level of ozone for crops and forest ecosystems were defined as accumulated exposure over a threshold of 40 ppb (AOT40). For forest areas the ozone exposure is accumulated during a period from April to September (24 hours a day). For crops the exposure between May and July at daylight-hours is taken into consideration. The calculations were performed stepwise for every one-hour mean and subsequently summed up for each grid-cell. The resulting maps were combined with the corresponding land-use maps (indicating areas with forests and agricultural utilisation, respectively).
Figure 2 shows

- AOT40 for forest areas 1994
- AOT40 for crops 1994.

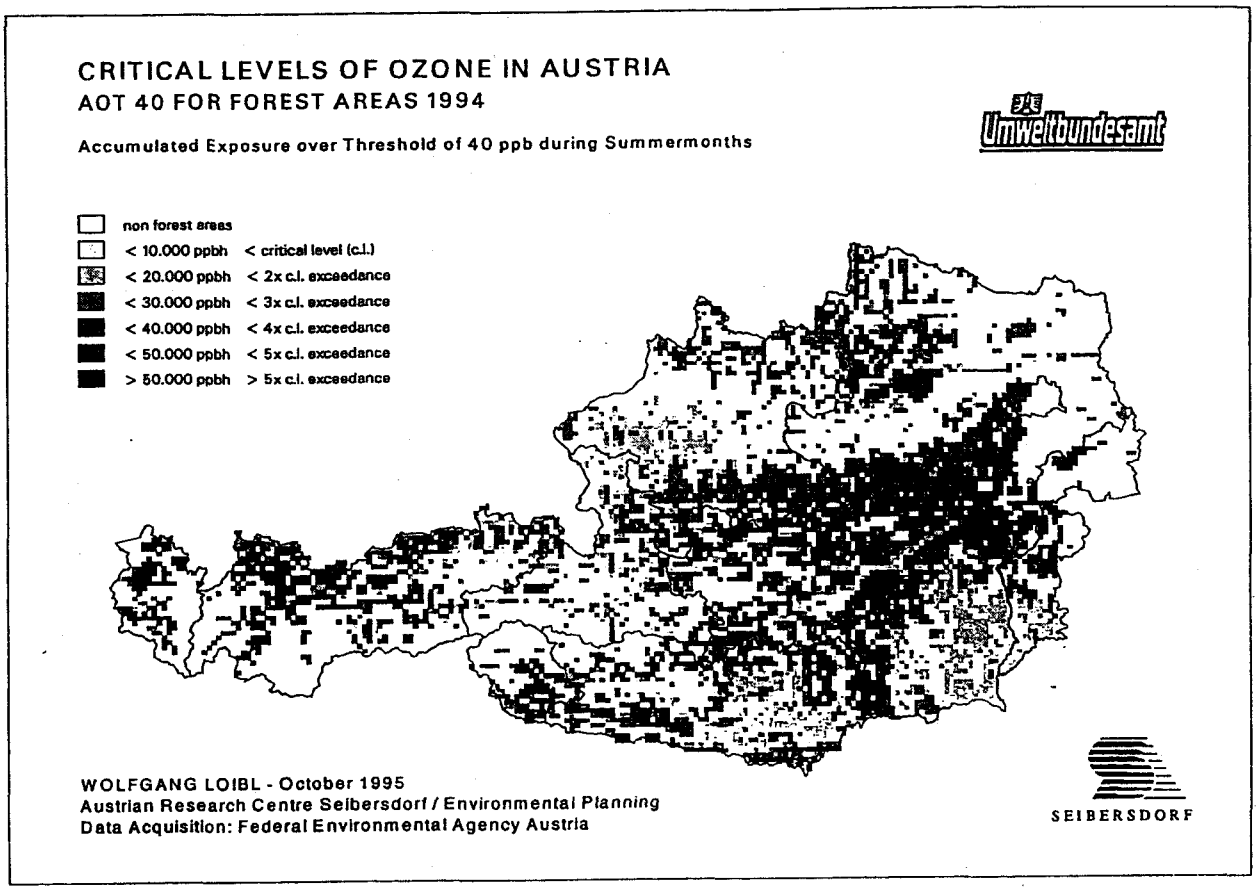
The distribution of the classes of ozone exposure reflects the Critical Level-values. For forest ecosystems, the AOT40 is shown in 10.000 ppbh steps, for Crops in steps of 5.300 ppbh.
It can be seen that the AOT40 values exceed the Critical Levels in most part of Austria.
For forest ecosystems, exceedances are highest in Alpine regions. This can be explained (partly) by the fact that ozone concentrations do not decline much during night in regions in high altitudes. The highest AOT40 values exceed 70.000 ppbh.
The Critical Levels for crops are exceeded in all agricultural areas throughout Austria. As an average the AOT40 values are about twice as high as the Critical Level.

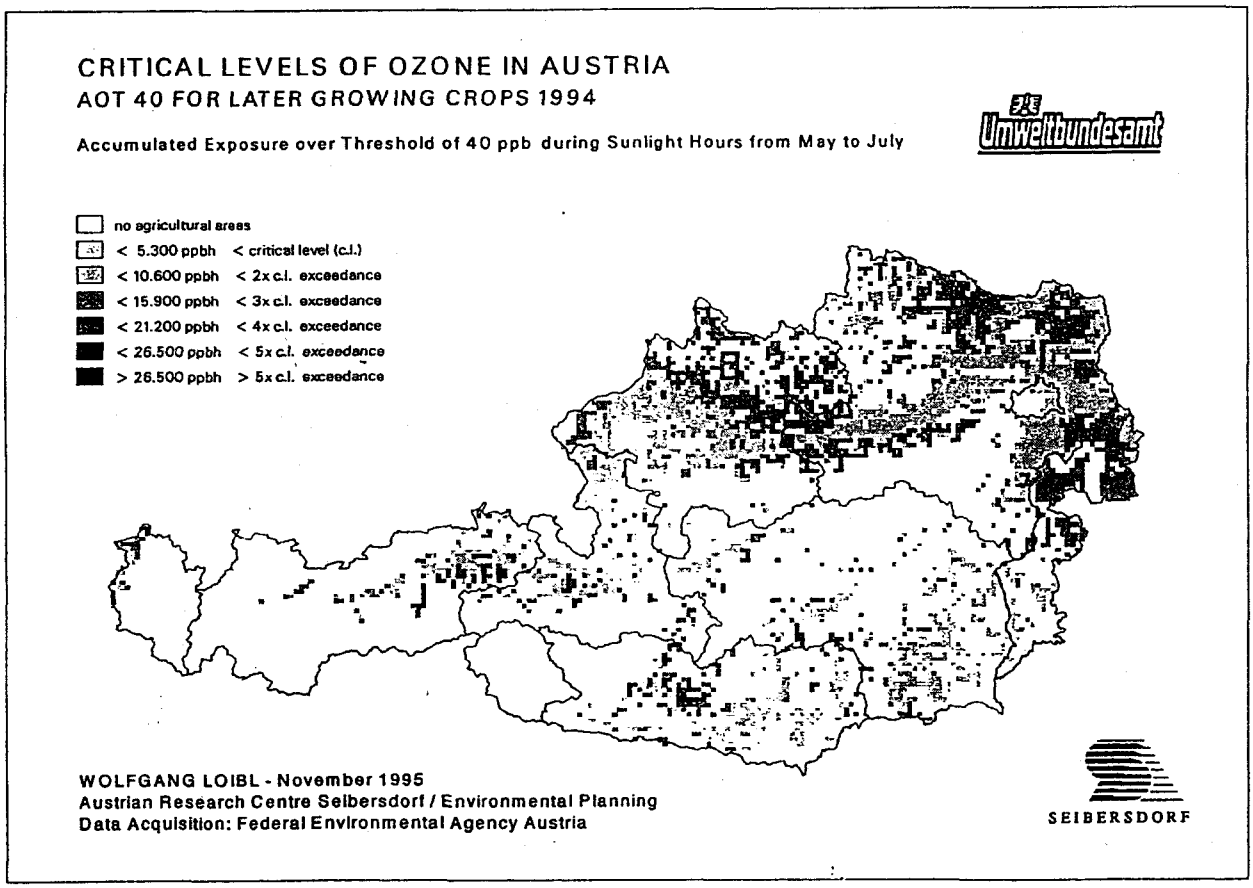
7 AOT40 measurements at certain monitoring sites

Additional to the spatial mapping, AOT40 values were calculated for the years of 1990 to 1995 for most of the approx. 120 ozone-monitoring stations. These calculations show that 1994 was a year with relatively high ozone levels. But generally it can be said that Critical Levels for ozone (for forests and crops) are exceeded at almost all monitoring stations in every year (for details see *Ozone in Austria - an overview* by W. Spangl in this volume).

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Fig. 2





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The Ozone Situation in Alpine Valleys

St. Smidt

Federal Forest Research Centre, A-1131 Vienna, Austria

Abstract

Alpine forest ecosystems are subject to numerous stressors, one of them being airborne pollutants. While the mean concentrations of SO_2 and NO_x significantly decrease with altitude a.s.l., or with the altitude above valley bottom, the ozone concentrations increase up to the timberline.

The danger which is imminent on Alpine forest ecosystems through higher ozone concentrations can be assessed with the help of effect-related limit concentrations of ozone input (AUSTRIAN ACADEMY OF SCIENCES, 1989) and in accordance with the Critical Levels of the UN-ECE (1994; "AOT40").

For both of the above-mentioned criteria, the evaluation shows frequent exceedances in Austrian forests, which implies a danger for *sensitive* plants. However, according to the current standard of knowledge the major Austrian tree species, spruce, appears to be less sensitive; there have not been any concrete indications to the high ozone sensitivity of larch specified under VDI Guideline 2310 either. It is well possible that there is a danger to pine and/or to deciduous trees. Nevertheless one can assume that forest ecosystems, especially those at altitudes above 1,000m a.s.l., are subject to a latent or dormant stress caused by the permanent ozone input, even if that additive stressor were effective only in case of high total stress. By now, there have been no concrete indications of *acute* danger for Alpine forest systems.

Introduction

Alpine forest ecosystems are exposed to natural stress factors which are intensified by air pollutants. Whereas the mean SO_2 and NO_2 concentrations in Alpine valleys decrease with increasing altitude a.s.l. and relative altitude above valley bottom, the ozone concentrations increase up to the timberline. Soil analyses imply that lead inputs increase with altitude (MUTSCH, 1995); the nitrogen and sulphur inputs do not show significant altitudinal trends in Alpine valleys; the input of protones frequently increases with altitude. According to the current standard of knowledge, photooxidants are, together with acid, nitrogen, and heavy metal inputs, among the most important pollutants found in Alpine valleys (SMIDT et al., 1995).

During the last two decades the global ozone concentrations at background stations increased by 1% to 3% annually; the development was not uniform, not even in Central Europe (SMIDT & GABLER, 1994). Since the beginning of the 80ies the annual ozone means have stagnated (cf. SMIDT & GABLER, 1994). Despite that stagnation of the long-term ozone values observed in the Alps it is striking that, compared to metering stations which had beed started in the middle of the last century and which had indicated annual ozone means significantly below 30ppb, a marked increase of the ozone concentration has been observed since the beginning of the 50ies. As a consequence, several values regarding the vegetation period mean were exceeded. The ozone concentrations in Alpine valleys show seasonal and daytime variations which depend on the valley-slope wind system as well as on weather conditions and photochemical reactions.

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From 1987-1993 the annual means from Austria, Bavaria and Switzerland varied between 19 and 132µg/m³, depending on the altitude, and increased significantly up to the timberline. Approaching the "middle troposphere" (approx. 3,000m) the mean concentrations frequently decrease again ("ozone belly" between 1,000 and 1,500m; PUXBAUM et al., 1991). The maximum daily means increased equally, while the maximum half-hour means did not show any altitudinal trend (SMIDT, 1996).

Criteria of assessment and metering stations

Criteria of assessment

To protect sensitive plants, the AUSTRIAN ACADEMY OF SCIENCES (1989) has formulated effect-related limits for ozone concentrations. The relatively high limit concentrations for half-hour and one-hour means indicate that short-term stress can be handled relatively well by the plants, while long-term inputs cause problems even if the concentrations are very low (like background stress).

According to new results (UN-ECE, 1994) accumulated ozone doses from a threshold concentration characterize stress better than input concentrations do. Contrary to agricultural plants, the Critical Level ("AOT40") for trees is calculated for both day and night, as it is assumed that the stomata of the trees are, or may be, open also during the night (Table 1).

Additional criteria of assessment, which will not be discussed in greater detail in that paper, include the Immissionsgrenzkonzentrationen/ maximum air pollution laid down by the VEREIN DEUTSCHER INGENIEURE (1987) and the WHO (1987) as well as the (legal) ozone limit values laid down in the SCHWEIZERISCHES DEPARTEMENT DES INNEREN (1985).

Table 1: Ozone limit values
*) sensitive plants
**) European forest trees (provisional; UN-ECE 1994), based on open top chamber experiments

	Mean value			
	1/2h	**8h**	**Vegetation period**	**Literature**
O_3-concentration	300µg/m³ *)	60µg/m³ *)	60µg/m³ *) April - October, 9.00 - 16.00	AUSTRIAN ACADEMY OF SCIENCES (1989)
O_3-dose	AOT40: "Accumulated exposure over a threshold of 40ppb" 10ppm*h (sum of 1h mean values > 40ppb over 6 months, e.g. April - September **)			UN-ECE 1994 WHO (1995)

In any case it is important to note that primarily the ozone flux to the interior of the leaves (the part of the ozone stress having a physiological impact) is decisive, not the actual concentration (dose), as the ozone uptake is influenced to a greater degree by the conductivity of the stomata than by the temporary ozone concentration. The vapour saturation deficit is to a high degree

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correlated with the ozone concentration; dry air, however, causes the closing of the stomata and therefore leads to a reduced uptake (WIESER & HAVRANEK, 1993).

Metering stations
For the evaluation under UN-ECE (1994) and the AUSTRIAN ACADEMY OF SCIENCES (1989), the ozone values from the period between 1991 and 1993 from 16 (partly Alpine) Austrian stations were used (SMIDT, 1994).
Alpine stations: Achenkirch/Tyrol (920m, 930m, 1280m, 1758m), Gaimberg/Tyrol (1250m), Gaisberg/Salzburg (1010m), Haunsberg/Salzburg (730m), Hochgößnitz/Styria (900m), Höfen/Tyrol (880m), Kreuzbergl/Carinthia (550m), Stolzalpe/Styria (1302m), Vorhegg/Carinthia (1020m).
Other stations: Nebelstein/Lower Austria (1012m), Ostrong/Lower Austria (570m), Schöneben/Upper Austria (920m), Steyregg/Upper Austria (335m).

Results

Annual means and vegetation period means

As the typical annual development of the ozone concentration has a maximum in spring/summer, the vegetation period means are significantly higher than the annual means (Figure 1). The lower the annual mean value, the bigger the difference compared to the vegetation period mean; for an annual mean between 20µg/m³ and 30µg/m³ that difference is on average approx. 30µg/m³, for an annual mean of 80µg/m³ approximately 10µg/m³.

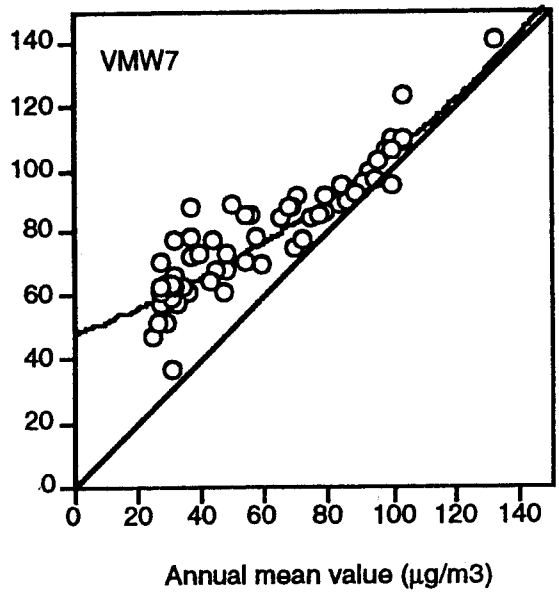
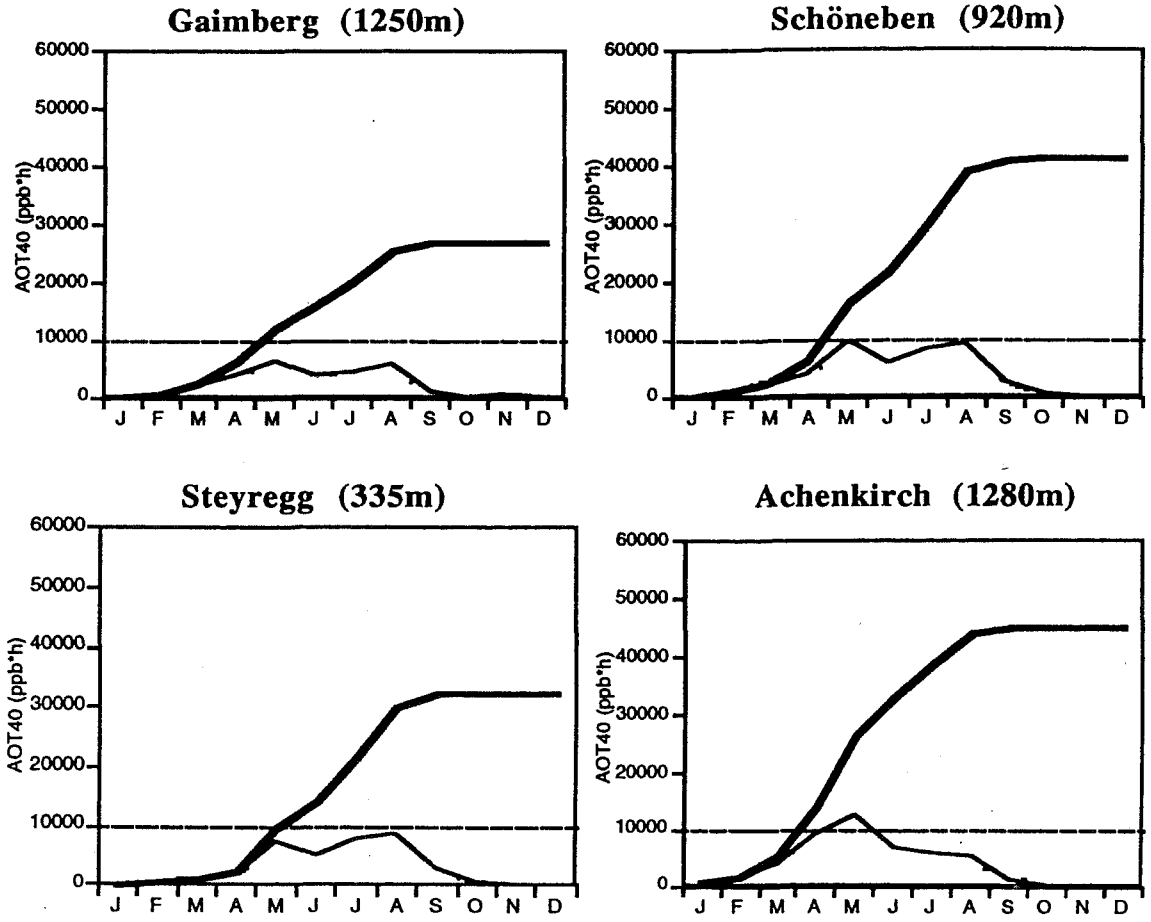


Figure 1: Ozone annual mean values vs. vegetation period mean values (VMW7; Austria, 1987-1993; µg/m³)

AOT40
At the examined stations the Critical Level of the UN-ECE (1994; 10ppm*h) was exceeded up to six-fold. At most of those stations that maximum was reached already in March or April. Even at Steyregg, where relatively high NO_x values are metered (annual mean values from 1991-1993: 29-40µg/m³), the AOT40 was exceeded within few weeks. For the stations of Gaimberg, Schöneben, Steyregg, and Achenkirch the developments are delienated graphically (Figure 2).

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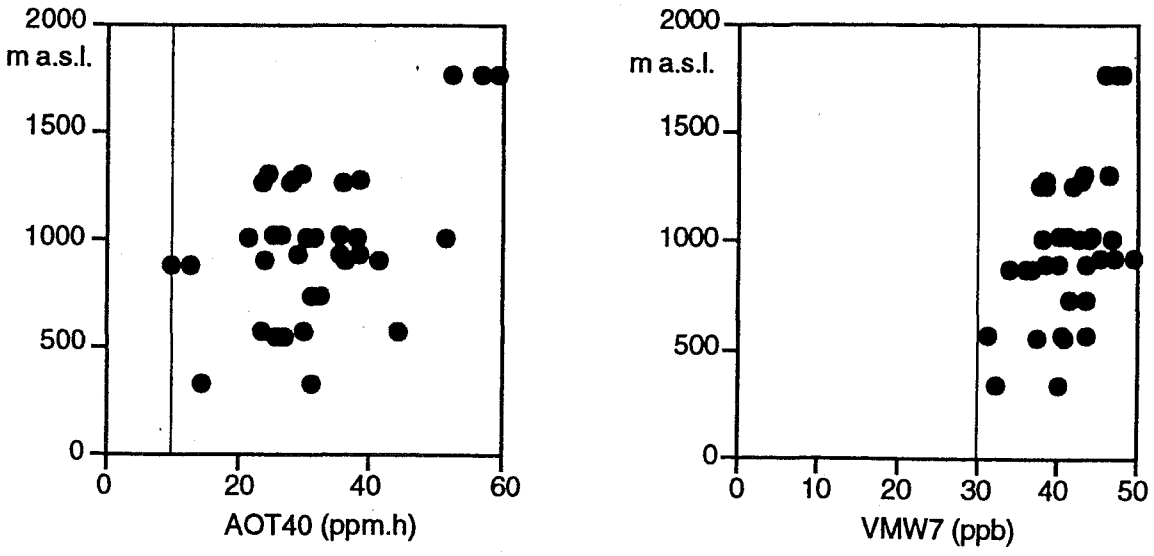
Figure 2: Monthly and accumulated AOT40 values metered at the stations of Gaimberg/Tyrol, Schöneben/Upper Austria, Steyregg/Upper Austria, and Achenkirch/Tyrol (based on metering values from 1992). Bold line: accumulated values. Broken line: monthly values.



Range of values and altitudinal correlation of VMW7 and AOT40

Figure 3 shows the increase of the AOT40 and VMW7 values with the altitude above sea level. Apart from one single exception, all values are higher than the limit values.

Figure 3: AOT40 and VMW7 values vs. altitude above sea level.



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Discussion

Relevance of the concentrations and doses to different plants

Depending on the applied assessment criterion (sensitivity of foliage or impact on photosynthesis) and the exposure conditions, different authors will differently assess the sensitivity of plants against ozone (for examples, see Table 2). As far as spruce is concerned, there are concrete indications that, together with acid deposition, ozone has an impact on plants on the physiological level (BLANK & LÜTZ, 1990; KRUPA & ARNDT, 1990); apparent reactions were also observed in connection with acid fog (SKEFFINGTON et al., 1985). However, there are no concrete indications to an acute endangering of that tree species through concentrations in the surroundings. (For that species one would probably have to assume a higher threshold value for the ozone dosis.) Deciduous trees seem to be more sensitive. The investigations referred to in Table 2 are mostly based on the exposure of young plants under controlled conditions; the classifications of the sensitivity of the different species by the VDI are based on short-term gassings of American provenances; many of the results therefore have only limited validity for forest ecosystems.

Tab.2: Ozone sensitivity of the major Austrian tree species with regard to the sensitivity of foliage and the impact on photosynthesis (simplified).

Tree species	Austria: Stocked area (Growing stock)	Sensitivity of foliage	Impact on photosynthesis	Literature
Conifers	*77.3% (82.4%)*		*less sensitive*	REICH 1987
Picea abies	61.8% (60.9%)	less sensitive	less sensitive	WIESER & HAVRANEK 1993 VDI 1987; LANDOLT & LÜTHY-KRAUSE 1991
Larix decidua	5.0% (6.9%)	very sensitive	less sensitive	VDI 1987 HAVRANEK & WIESER 1993
Abies alba	2.7% (4.7%)	less sensitive	less sensitive sensitive	GROSS 1987, SENSER et al. 1987; LANDOLT & LÜTHY-KRAUSE 1991 ARNDT & KAUFMANN 1985
Pinus sylvestris	6.4% #)	intermediate *) sensitive		VDI 1987 LANDOLT & LÜTHY-KRAUSE 1991
Pinus cembra		sensitive **)		SIMONS 1993

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Tree species	Austria: Stocked area (Growing stock)	Sensitivity of foliage	Impact on photosynthesis	Literature
Deciduous trees	*22.7%* *(17.6%)*		*intermediate *)*	REICH 1987
Fagus silvatica	9.8% (9.1%)	less sensitive (very) sensitive **)	 intermediate*) sensitive less sensitive	VDI 1987 REICH 1987 KRAUSE & PRINZ 1989 GUDERIAN et al. 1990 ARNDT & KAUFMANN 1985
Acer platanoides			sensitive **)	KRAUSE & PRINZ 1989
Betula pendula			sensitive	MATYSSEK et al, 1990
Quercus robur			sensitive **)	KRAUSE & PRINZ 1989

*) compared to herbaceous plants and conifers (deciduous trees resp.)
**) young trees
\# Total share of pine: 7.7% (9.8%)

In spite of the well-known phytotoxicity of ozone to forest trees and the great number of exceedances of effect-related limit values especially at higher elevations, there are no definite indications to an *acute* endangering of the Austrian forest ecosystems by ozone alone. However, a combination with other pollutants (esp. those including SO_2 and NO_2 were investigated) leads in most cases to synergistic effects. In Austria, no symptoms have been found on the needles or leaves of forest trees that can clearly be identified as a consequence of a higher ozone concentrations; in addition, the diagnosis is even more difficult as mistakes are easily possible in connection with other (biotic) damage (HANISCH & KILZ, 1990). One can start from the assumption that the forest ecosystems are subject to a permanent dormant stress through oxidants, which probably affects mainly the deciduous trees.

Apart from ozone, the nitrogen and heavy-metal inputs represent an additional potential stress factor among the airborne pollutants, at least if the input continues over the next years. An acute and directly harmful effect of organic compounds (PAN, chlorocarbons) which are partly involved in photochemical processes, can be practically excluded outside agglomerations; apparently also negative effects of acid inputs can be ruled out on carbonate-influenced soils of the Limestone Alps.

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ICP-CROPS AND CRITICAL LEVELS OF OZONE FOR INJURY DEVELOPMENT

J. Benton[1]., J. Fuhrer[2]., B.S Gimeno[3]., L. Skärby[4]., G. Balls[1]., D. Palmer-Brown[5]., C. Roadknight[5] and G. Sanders[1]

[1]) The ICP-Crops Coordination Centre, Department of Life Sciences, The Nottingham Trent University, Clifton Lane, Nottingham, NG11 8NS, UK.
[2]) FAC, CH-3097, Liebefeld-Bern, Switzerland.
[3]) CIEMAT-Ima 3B, Avda. Complutense 22, Madrid 28040, Spain.
[4]) IVL, Box 47086, S-40258 Göteborg, Sweden.
[5]) Department of Computing, The Nottingham Trent University, Burton Street, Nottingham, NG1 4BU, UK.

Abstract

Studies by the UN/ECE ICP-Crops in 1994 showed that visible injury to crops caused by ozone pollution was widespread across Europe indicating the extent of exceedance of the provisional short-term critical level (an AOT40 of 700 ppb.h accumulated during the daylight hours over three consecutive days). Injury was seen on a wide range of crops such as clover, bean, soybean, tomato and watermelon at ICP-Crops sites throughout Europe with the exception of Finland. Mean daily maximum ozone concentrations for individual clover experiments ranged from 40 ppb in Finland to 84 ppb in Switzerland. Examination of the daily ozone concentrations before the onset of injury indicated that injury usually developed following episodes where concentrations exceeded 700 ppb.h. However, in some cases, injury did not develop and possible explanations for this are being considered by the ICP-Crops. Analysis of ICP-Crops data using artificial neural networks (ANNs) indicated that lower ozone concentrations, following a high ozone episode, coincided with the appearance of visible injury. It is believed that ANN analysis of more comprehensive data sets obtained by the ICP-Crops in 1995 will improve the short-term critical level for visible injury (Level I and Level II).

Introduction

The role of the United Nations Economic Commission for Europe International Cooperative Programme on effects of air pollution and other stresses on crops and non-wood plants (UN/ECE ICP-Crops) is to monitor and document the effects of ambient air pollution on crop and more recently, non-wood plant species, throughout Europe. Since its initiation, 22 Parties of the Convention on Long-range Transboundary Air Pollution (LRTAP Convention) have participated in the programme. A large database now exists for ozone pollution and the visible injury and yield/biomass losses that it causes in sensitive crop species. The crops of primary interest to the ICP-Crops are from the *Leguminosae* family and include *Trifolium subterraneum* L. (subterranean clover), *Trifolium repens* L. (white clover) and *Phaseolus vulgaris* L. (green bean). Other species being studied include *Glycine max* (soybean), *Lycopersicon esculentum* (tomato),

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Citrullus lanatus (watermelon), *Nicotiana tabacum* (tobacco), *Malva sylvestris* L. (common mallow), *Cirsium arvense* (L.) scop. (creeping thistle) and *Plantago major* L. (great plantain). Characteristic injury symptoms such as bronzing, chlorosis and necrosis of leaf foliage have been widely reported throughout Europe on these and other ozone sensitive species (Sanders and Benton, 1995).

Two critical levels for ozone were established at the UN/ECE Critical Levels for Ozone Workshop held in Bern Switzerland (Fuhrer and Achermann, 1994) . These were (i) a long-term critical level which would cause reductions in biomass or yield if exceeded and (ii) a short-term critical level which, when exceeded, would cause visible injury to develop (Sanders *et al*, 1994).

Long-term critical level	-	an AOT40* of 5300 ppb.h accumulated during daylight hours over a three month period.
Short-term critical level	-	an AOT40 of 700 ppb.h accumulated during daylight hours over 3 consecutive days.

* accumulation of the hourly means of ozone where the mean exceeds 40 ppb.

The long-term critical level for ozone was derived from analysis of spring wheat (*Triticum aestivum*) data (Fuhrer, 1994) which were published as part of the results of the European Open-Top Chamber Project (EOTC) (Jäger *et al*, 1993). The provisional short-term critical level of ozone for visible injury was established using data collected from ambient air field observations by the ICP-Crops and The Netherlands Biomonitoring Network.

It is recognised that the use of a single critical level for a gaseous pollutant (Level I) is rather simplistic as it does not consider variations in the sensitivity of individual receptors to pollutants or the influence of environmental factors such as relative humidity and temperature on receptor response (Level II). Thus, it has been acknowledged that further research is required to improve the assessment of the geographical distribution of critical level exceedances in Europe (Sanders *et al*, in press) and that critical levels for ozone need to be defined which take environmental conditions into consideration (Gimeno *et al*, in press).

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This paper presents some of the data from the 1994 ICP-Crops experiments which demonstrate the extent of exceedance of the short-term critical level for visible injury. In addition, the methodology and preliminary results are described of a computer modelling investigation into the factors which may modify the response of a receptor to ozone and the onset of visible injury.

Experimental methodology of the ICP-Crops

Each year, participants conduct a series of coordinated experiments across Europe to determine the effects of ambient ozone on crops. The Coordination Centre provides a comprehensive experimental protocol, seed and other equipment to ensure that experiments are standardised as far as possible (UN/ECE, 1994a). Participating countries in 1994 were Austria, Belgium, Denmark, Finland, France, Germany, Greece, Hungary, Italy, Latvia, The Netherlands, Poland, Russian Federation, Spain, Sweden, Switzerland and the United Kingdom.

Ambient air experiments to determine the extent of visible injury on sensitive crops: T.subterraneum cv. Geraldton (Subterranean clover) and T. repens cv. Menna (white clover) :

Forty pots containing three *T.subterraneum* or *T. repens* clover seedlings were used for each experiment at each experimental site. Twenty pots were treated with 100 ml 150ppm ethylene diurea (EDU) at emergence of the first trifoliate leaf and then after at regular intervals in order to establish alterations in biomass due to ozone exposure (data not presented). EDU is a chemical which is effective at protecting plants from ozone injury and can be applied to the foliage or to the roots (Carnahan *et al*, 1978). For example, a soil application of EDU to *Phaseolus vulgaris* was shown to provide complete protection against visible injury caused by exposure to ozone (Bergmann *et al*, 1993). The remaining twenty pots were treated with 100 ml of distilled water and used to determine the extent of visible injury. All pots were placed outside in ambient conditions. The pots were watered by a self-watering system placed below ground. Plants were examined daily for the appearance of visible injury and assessed 28 and 56 days after emergence of the first trifoliate leaf for the number of ozone injured leaves and the total number of leaves. From these data, the percentage of injured leaves could be calculated per pot. The leaves were removed from the plants at 28 days and their dry weight determined (data not shown). Plants were harvested again 56 days after emergence and the total plant dry weight determined (data not shown).

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Throughout the growing period, climatic conditions (temperature, relative humidity, daylight hours, rainfall) and pollutant levels (ozone, sulphur dioxide and oxides of nitrogen) were recorded continuously.

P. vulgaris cv. Lit, cv. Stella, cv. Groffy (green bean):

Twenty bean plants of each cultivar (one per pot) were exposed to ambient conditions for approximately 100 days. Plants were assessed daily for the onset of ozone injury and the percentage of ozone injured leaves determined. Pollution and climate conditions were monitored throughout the growing season.

G. max cv. Ceresia (soybean), L. esculentum cv. Tiny Tim (tomato), B. napus cv. Comet (oilseed rape) and Phaseolus vulgaris cv. Lit (green bean):

Ten plants (one per pot) were placed in ambient ozone conditions for 56 days (tomato and soybean), 70 days (green bean) or 96 days (oilseed rape) and assessed daily for the appearance of ozone injury. Pollution and climatic conditions were monitored throughout the experiment.

C. lanatus cv. Sugar Baby (watermelon), N. tabacum cv. Bel-W3, Bel-C, Bel-B (tobacco):

Extensive field surveys were conducted in Valencian and Catalan Autonomous Communities in eastern Spain to evaluate the extent of ozone injury on tobacco and watermelon plants. In total, 20 sites were used in the tobacco survey and several commercial fields in Valencia and Tarragona were visited to determine the amount of injury on watermelon.

Analysis of data by the use of artificial neural networks (ANN's):

The theory of artificial neural networks:

Over the last decade, significant advances have been made in pattern recognition and computational learning and this includes the development of artificial neural networks (ANNs). ANNs perform functions which are similar to some brain functions (Kothari and Heekuck, 1993). They do not rely on a program containing step by step procedures in order to function but actually 'learn' by studying patterns within data sets which is very much like the process of the human brain. They can also deal with 'noisy' or incomplete images or patterns which is another key feature of the brain (Kothari and Heekuck, 1993). Similar to biological systems, neural networks

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consist of many, connected, simple processing elements called neurons or nodes (Figure 1). They are ideal to use for analysis of data obtained from experiments where many parameters have an influence on the response. For example, the extent of ozone injury may be modified by factors such as relative humidity, temperature and photosynthetically active radiation (PAR) as these influence stomatal conductance (Kappen and Haeger, 1991) and thus the flux of ozone into the plant (Kersteins *et al*, 1992).

There are several different forms of ANN and those used in this work learn the data by a process of error minimisation. Initially the network must be trained with numerous items of data (the training set) enabling it to detect patterns within the data. The input layer receives data which represents the causal agents of an effect whereas the neurons in the output layer contain data which represents the 'actual' effect. From the input data, the network calculates a predicted output value and compares this with the 'actual' value entered into the output layer. The network then makes modifications which relate the predicted output to the 'actual' output. It does this by continually modifying the strength of the connections between the input and output neurons until the learning process is complete and the network can generate output which is a sufficiently accurate and meaningful interpretation of the input for all the training examples. The network can be tested by entering new data (independently selected) in to the input neurons and then comparing the predicted output response with an 'actual' response. In addition, the strength of the connections of the network may be analysed to indicate the relative importance of parameters entered into the input neurons.

The artificial neural network approach has been used in many scientific situations. For example, ANNs have been used to model complex data sets obtained from closed chamber experiments investigating the effects of different microclimates, ozone doses and exposure times on the extent of injury using *T. subterraneum* (Balls *et al*, in press). This approach has also been used in the taxonomic identification of plankton (*Ceratium* spp) (Simpson *et al*, 1992).

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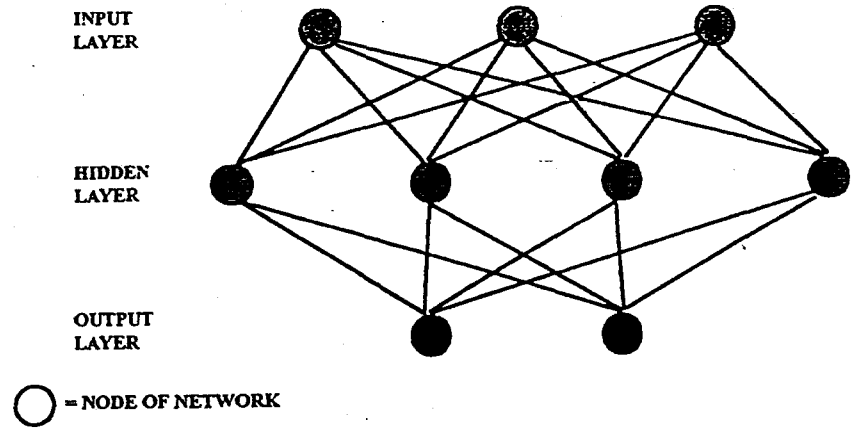


Figure 1. A simplified model of a neural network structure (adapted from Kothari and Heekuck, 1993).

Analysis of the ICP-Crops data using ANNs

Analysis of the ICP-Crops data requires intensive processing and thus a 64 processor parallel computer system (CNAPS) has been employed for all ANN computations. This system is ideal for neural network applications because of its increased speed in handling sets of data where each data element is operated in a similar way. All of the active processors in the CNAPS can execute the same instruction simultaneously thus, performing an operation on an entire block of data in one step. This is known as a SIMD architecture (Single Instruction, Multiple Data).

ANN models were developed using data from sites where the date of onset of ozone injury, hourly ozone concentrations, and climatic conditions were recorded for *P. vulgaris*, *T. repens* and *T. subterraneum* experiments. These were Austria, Belgium, Italy, The Netherlands, Sweden, Switzerland and the United Kingdom. ANN's were used to elucidate why injury occurred after one 3 or 5 day ozone episode but not after another. The ozone parameters entered into the network were the 7 hour mean, the daylight AOT40 and the daily maximum ozone concentration. The day after emergence on which injury occurred and whether injury occurred before or after the first harvest were also entered. All values were normalised so that the largest value for each input was 1 and the lowest value for each input was 0. The intermediate values were linearly transformed. The 3 or 5 day period before the onset of ozone injury was given an output of 0.9 and the 3 or 5 day period preceding the absence of injury was given a value of 0.1 (it is better not

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to operate at the extremes of the range). The 3- and 5-day networks were trained using 24 and 18 complete data sets respectively. These data sets contain data for both the input and output neurons. Following training, a network configuration was discovered that would predict whether leaf injury would appear or not given certain inputs. The 3- and 5-day models were tested with 8 and 6 further data sets respectively, previously unknown to the network, to see whether the model could correctly predict whether injury would occur.

Results and discussion

The extent and onset of visible injury on crops throughout Europe.

Visible injury due to ambient ozone conditions was widespread during the 1994 growing season (Figure 2) indicating that the short-term critical level for visible injury was exceeded widely across Europe. A wide range of ozone concentrations were recorded at ICP-Crops sites in 1994. For example, the mean daily maximum ozone concentrations measured during the first clover (*T.subterraneum* or *T. repens*) experiment (usually June and July) in Finland and Switzerland were 40 ppb and 84 ppb respectively. The range of mean daily maximum ozone concentrations at *P.vulgaris* sites was from 40 ppb in Finland to 70 ppb in Italy (Naples). Many crop species were susceptible to ambient ozone pollution (Table 1). The variability in response emphasises the need for a clearly defined short-term critical level for visible injury for each species or group of species (Level II).

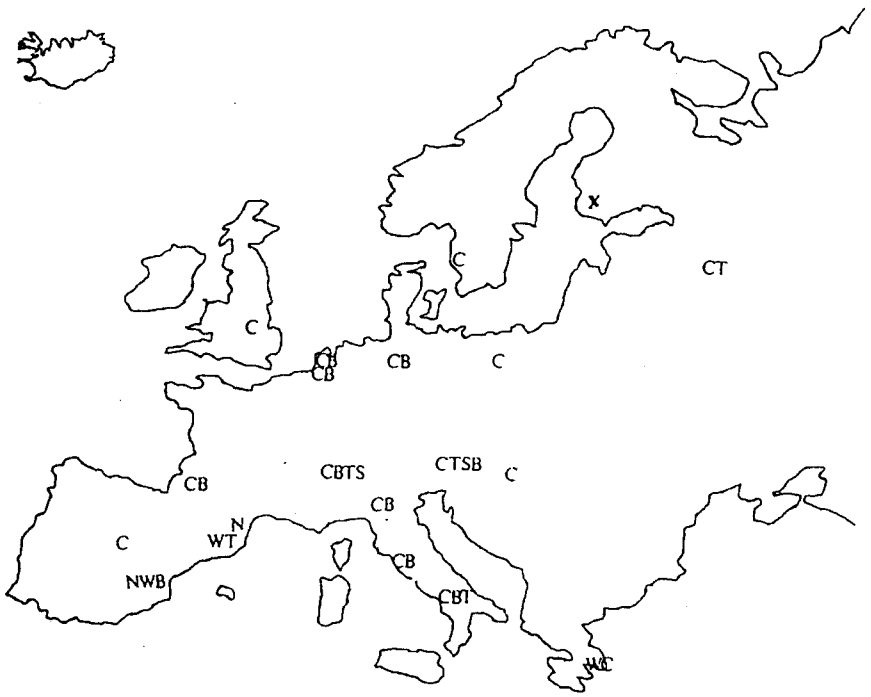


Figure 2. Map showing the ICP-Crops sites where visible injury was seen on crop species in 1994 (C - clover; B - beans, T - tomato; S - soybean, N - tobacco and W - watermelon, X - no injury).

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Table 1. Ozone injury on different crops species from selected ICP-Crop sites (*T.subterraneum* data is from the 2nd harvest of the first experiment)

Country	Crop species	% injured leaves (per pot or plant)	Growing period AOT40 (ppb.h)
Austria	*P. vulgaris cv. Lit*	19.5	13129
	T. subterraneum	7.4	6434
	G. max	11.6	13129
	L. esculentum	16.6	13129
The Netherlands	*T. subterraneum*	7.5	4604
Sweden (Ostad)	*T.subterraneum*	45.5	3064
Switzerland	*T.subterraneum*	34.5	8012
	P. vulgaris cv. Lit	32.1	5125
	G.max	2.0	15950

The short-term critical level for visible injury was investigated further using data from sites where the onset date of visible injury was recorded and where ozone concentrations were monitored hourly. Daily AOT40 values for the growing season of the crop were plotted and compared with the date that injury was first identified (for example, Fig. 3). Thus, it was possible to see if injury occurred after a three day episode where the AOT40 was equal to, or greater than, 700 ppb.h. Visible injury was first seen on *T. subterraneum* clover in Belgium on July 4th (experiment 1) and 25th (experiment 2) after three days where ozone concentrations were greater than 700 ppb.h (Fig. 3a). Similarly, injury occurred on July 5th and Aug 1st after high three day ozone episodes at the experimental site in The Netherlands (Fig.3b). However, injury also occurred on September 20th in The Netherlands even though the AOT40 did not exceed 700 ppb.h in the preceding 3 days. This latter effect was also seen at a site in Austria (Fig. 3c) and in Sweden (Fig. 3d) and this variability suggests that the provisional short-term critical level for clover may require revision following further data analysis To substantiate this, open top chamber studies in Sweden by Pihl Karlsson *et al* (1995) have shown that 50% of clover leaves are injured before the AOT40 is exceeded and it has been suggested by results from the 'Clover Sweden' project that an AOT 'cut off' value of 40 ppb is too high for clover grown in Swedish conditions (Pihl Karlsson *et al*, in press). In contrast to this, there was a large ozone episode at the end of June in the United Kingdom (Fig. 4) but injury failed to develop on *T. subterraneum* until July 13th after the 1st harvest.

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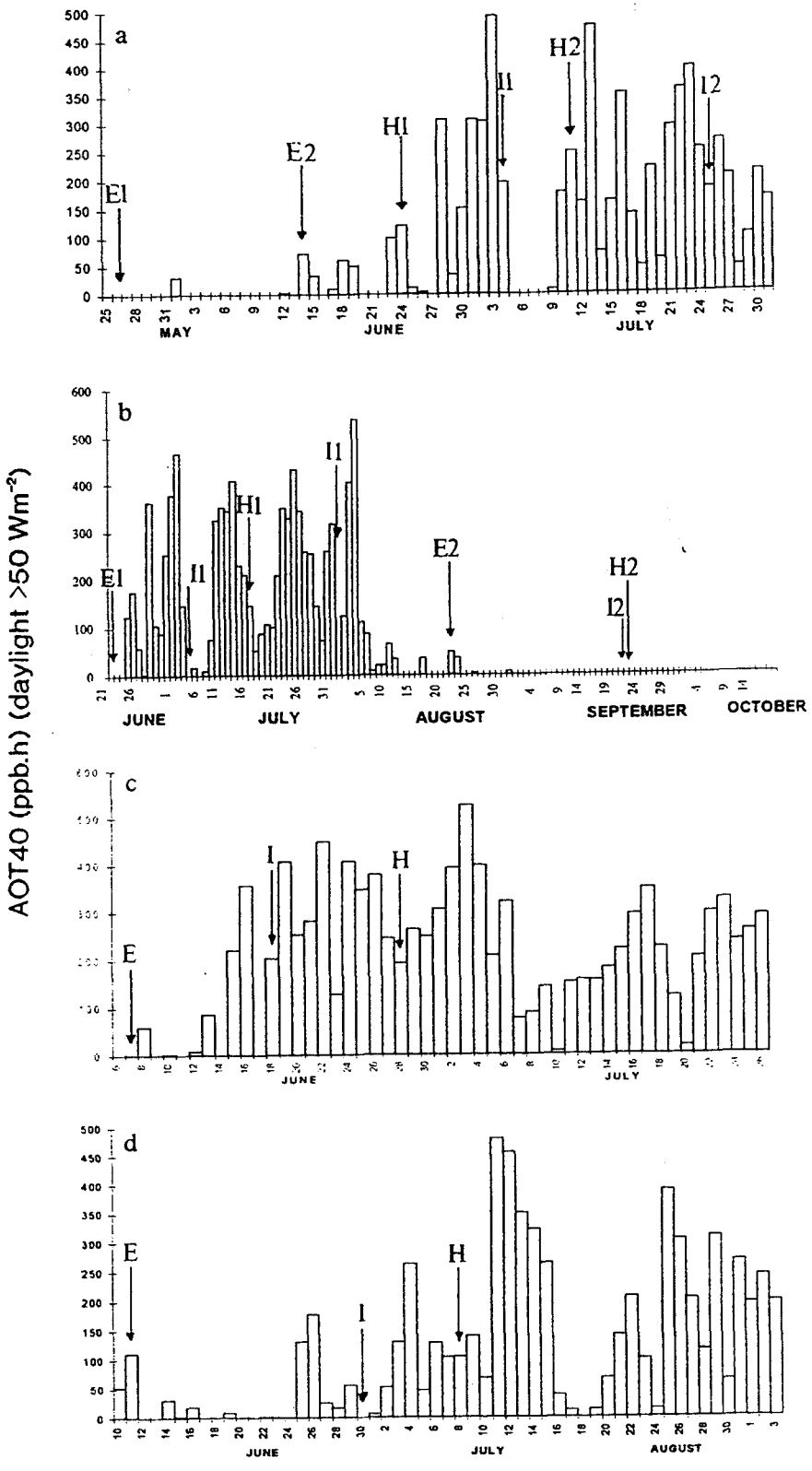


Figure 3. Daily AOT40 values and the onset of injury on *T. subterraneum* clover in (a) Belgium and (b) The Netherlands (c) Austria and (d) Sweden) (E - emergence of 1st trifoliate ; I - injury and H - harvest; 1 - experiment number 1; 2 - experiment number 2).

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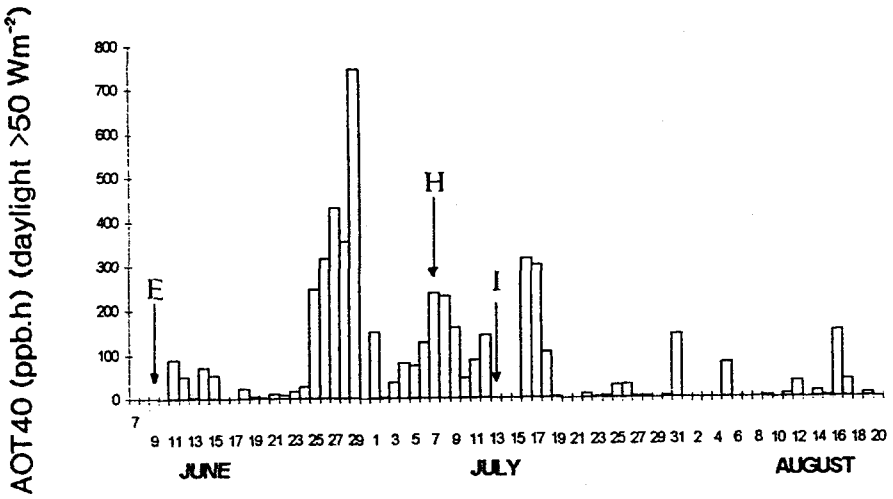


Figure 4. Daily AOT40 values and the onset of ozone injury on *T. subterraneum* clover in The United Kingdom (E - emergence of 1st trifoliate, I - injury and H - harvest)

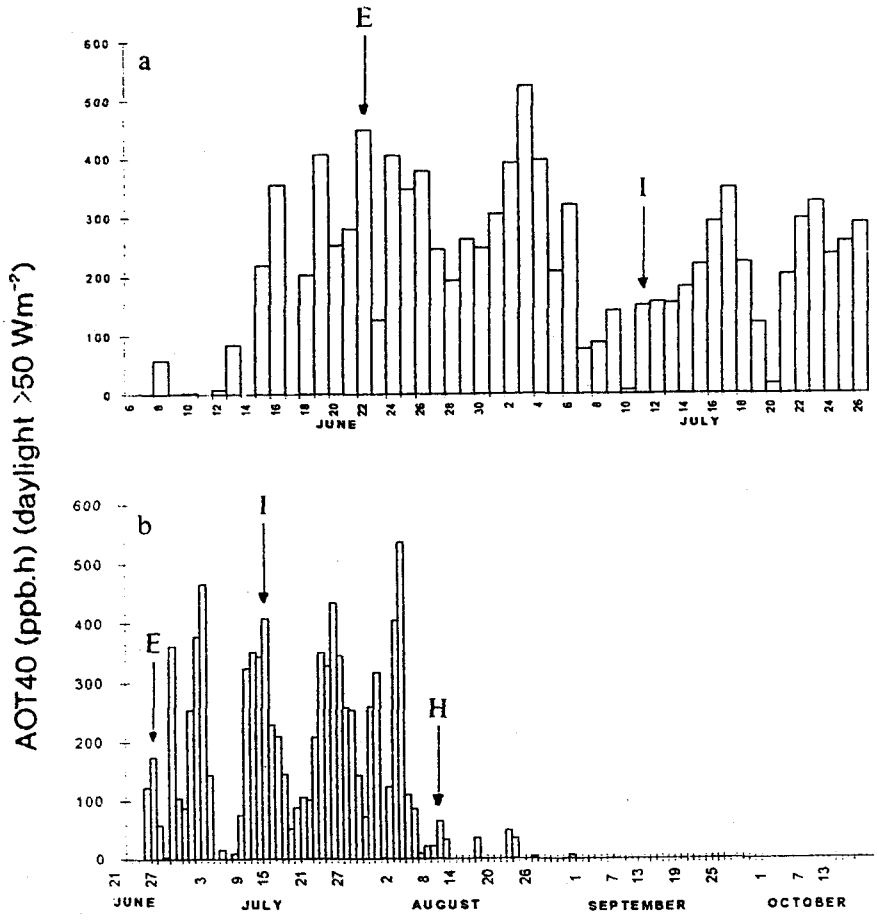


Figure 5. Daily AOT40 values and the onset of ozone injury on *P. vulgaris* foliage in (a) Austria and (b) The Netherlands (E - emergence of 1st trifoliate, I - injury and H - harvest)

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Injury occurred on *P. vulgaris* foliage in Austria (Fig. 5a) and The Netherlands (Fig. 5b) after episodes where the AOT40 was above 700 ppb.h for 3 consecutive days although it is unclear which 3 days were responsible for causing injury. In summary, the data from the ambient air experiments of the ICP-Crops provides some support for the use of a short-term critical level for visible injury of 700 ppb.h. However, the inconsistencies in the data indicate that this value needs substantiating.

Analysis of data using ANNs:

The complexity of the information gained from the ICP-Crops ambient air experiments necessitated the use of a novel approach to data processing and modelling as two dimensional statistical methods are believed to be ineffective. ANNs were chosen for data analysis because it is thought these will determine the influence of multiple modifying factors such as climate on the critical levels of ozone for visible injury. In addition, equations from the networks may be extracted to justify and interpret the behaviour of the network. These equations may be used to predict critical levels in different physical and pollution climates since they constitute an explicit, fully-interpretable model of the data which can be related to scientific processes which are already understood or that are actively being researched. An example of equation extraction can be given for the networks which had been successfully trained, as they correctly predicted that injury did/did not occur for nearly all the test data sets and provided some useful information regarding the onset of injury. The analysis indicates that the sequence of ozone concentrations during an episode may be important to the onset of injury. The network trained with the 3-day data indicated that a day with low ozone concentration coincided with the appearance of ozone injury on the following day. For example, equation 1 has been extracted from the network and is a simplified form of the equation that represents the most important hidden nodes of the trained 3-day network. It indicates that ozone concentrations on day 2 and 3 are more important than those on day 1 and the negative sign at day 3 indicates the lower ozone concentration on the day before injury development. This was also indicated by part of the equation extracted from the 5-day network as day 5 (AOT40) is multiplied by -3 (Equation 2). This suggests that a lower ozone concentration may serve to accelerate the onset of ozone injury. The 5-day network also showed the positive influence of day number on the onset of injury confirming that injury observation occurs on older plants (Equation 2).

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Equation 1: day 2 (7h mean + max ozone) - day 3 (AOT40 + max ozone)

Equation 2: F(day 1 (AOT40 - 7h mean) + day 2 (AOT40) + day 4 (7h mean) - 3*day 5 (AOT40))
+ F(sum (no. of days after emergence for days 1 to 5) - day 2 (7h mean))

(F is a function with a minimum value 0 and maximum value 1 which increases with its input value).

These effects can be related to the results shown by Figure 3. For instance, injury to *T. subterraneum* clover in Belgium (Fig. 3a) and The Netherlands (Fig 3b) occurred following a day when the ozone concentration was lower than on previous days although this effect was not quite as clear with the *P.vulgaris* data (Fig. 5).

Results of the analysis of ICP-Crops data by ANNs has provided some useful information regarding the short-term critical level for visible injury. It has been indicated that day number, which can be related to leaf and plant age, influences the onset of injury and this is in agreement with the findings of others. For example, open-top chamber studies by Pihl Karlsson *et al* (1995) have shown that *T. subterraneum* leaves of up to seven days old can withstand injury even at high ozone concentrations. It is believed that the leaf is most ozone sensitive following the period of maximum expansion rate but before the period of maximum surface to volume ratio and full expansion involving the formation of the secondary cell wall and lignification. In addition, visible injury was more apparent on *C. lanatus* (watermelon) after the flowering and fruiting growth stage rather than in the early stages of plant development (Gimeno *et al*, 1995; UN/ECE, 1994b) which clearly indicates that younger plants are less sensitive to ozone pollution.

The network predicted that the timing and magnitude of ozone concentrations may be important for injury development as a day with a lower ozone concentration seemed to coincide with the expression of ozone injury. This effect has also been indicated by studies using closed chambers and *T. subterraneum* (Balls *et al*, in press). Data analysis using an ANN (Neuroshell 2, Ward Systems Group Inc., USA) gave predictions that plant sensitivity decreased as the time of ozone

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exposure increased and this was consistent with 'actual' experimental data.

Further studies incorporating the use of ANNs have shown that other factors are important for the onset of injury on clover plants and these need careful consideration when establishing and applying the Level II concept of the short-term critical level. ANN predicted injury and actual injury indicated that clover plants were more sensitive to ozone at lower PAR, lower temperatures and at higher humidities. For example, plants were more sensitive to ozone at 100µmol m^{-2} s^{-1} PAR, 20°C and 55% RH. as opposed to 350µmol m^{-2} s^{-1} PAR, 28°C and 28% RH (UN/ECE, 1995). This effect can be partly related to the stomatal response to humidity. Stomata close in response to increased transpiration and a reduction in guard cell turgor (Mott and Parkhurst, 1991) but if the humidity is high then the tendency is for the stomatal pore to remain open which allows the passage of ozone into the leaf tissue. Similar results were shown by Mortensen (1992) using growth chambers. The shoot dry weight of ozone treated *L.esculentum* was reduced to a greater extent at 90% RH than at 70% RH and ozone injury was reduced at higher light intensities. Thus, the results obtained from closed chamber experiments can be used to indicate how environmental conditions influence the extent of ozone injury seen in the field. Evidence for this was obtained by field surveys of *N.tabacum* grown in eastern Spain. Ozone injury was more prevalent in the coastal regions of Catalunya which had a higher relative humidity than regions further inland (Gimeno *et al*, in press).

Following the 1995 experimental season, much more data will be available from the ICP-Crops which will be used to verify the results and theories described above. As well as recording the actual date of onset of injury, participants have assessed the level of injury. This means that an actual value can be entered into the network and not just numbers corresponding to whether injury did or did not occur. Daily values for relative humidity and temperature and PAR will be entered into the network for the days preceding the onset of injury in addition to other parameters describing daily ozone concentrations. This will determine the relative importance of environmental factors as well as ozone concentrations for the development of ozone injury. In addition, further species have been examined and assessed for injury at regular intervals. These results will provide further information regarding 'stocks at risk' from ozone pollution and for the derivation of the short-term critical level for visible injury (Level I and Level II) and this

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information can be further used to verify the predictions made on exceedance maps.

Conclusion

Ozone injury was widespread across Europe in 1994 providing evidence that the critical level for visible injury was exceeded. Investigation of the AOT40 profiles for the days before the onset of injury indicated that, although injury usually developed after an AOT40 of 700 ppb.h, this was not always the case. Artificial neural networks have indicated that the plants may be responding to the pattern of daily ozone concentrations. A day with low ozone concentration may be necessary before the appearance of visible injury and the response to individual episodes may depend upon the previous ozone history of the plants. It is believed that following analysis of data from the 1995 season, the ICP-Crops will be in a position to provide a more 'robust' Level I short-term critical level together with suggestions for Level II.

Acknowledgements

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THE 1995 OZONE EXPERIMENT ON DURUM WHEAT (*Triticum durum* L.) IN VITERBO (CENTRAL ITALY): PRELIMINARY RESULTS ON YIELD AND GRAIN MINERAL NUTRITION

M. Badiani[1], F. Rossini[2], A.R. Paolacci[1], C. Perani[1], A. Porri[1], A. D'Annibale[1], G. Falesiedi[1], A. Maggini[1], G. Giovannozzi Sermanni[1], and R. Ambrogi[3]

[1] Dipartimento di Agrobiologia e Agrochimica, and [2] Dipartimento di Produzione Vegetale, Universita' di Viterbo, Via S. Camillo De Lellis, I-01100 Viterbo, Italy

[3] Ente Nazionale per l'Energia Elettrica (ENEL), SpA, Direzione Studi e Ricerche, Centro Ricerche Ambiente e Materiali, Via Rubattino, 54, I-20134 Milano, Italy

Abstract

Two cultivars of durum wheat (*Triticum durum* L.), namely Duilio, early maturing, and Neodur, late maturing, were grown in the field at a rural site in Central Italy and exposed to ozone (O_3) within a range of realistic partial pressures during the summer 1995. Exposure variants which, respect to ambient air O_3, ranged from 0.50 to 2.26 and from 0.62 to 1.50 in terms of hourly mean concentration and cumulative dose, respectively, were obtained by enclosing the plants in open-top field chambers. Negative correlations were found between increasing O_3 levels and canopy parameters related to productivity in the cv Duilio, whilst the cv. Neodur was unaffected. Differential O_3 effects on the grain mineral composition were also noticed for the two *T. durum* cultivars. The effects of the pollutant on the responsive plant parameter were described at the best by cumulative O_3 exposure indices rather than by hourly mean concentrations. Critical O_3 levels of 5412 and 5302 (nL L^{-1})*h were calculated for a 10% reduction of grain yield and total aboveground biomass, respectively, of the sensitive cv. Duilio.

Introduction

The Critical Level (CL) concept adopted by the Convention on Long-Range Transboundary Air Pollution of the United Nations Economic Commission for Europe (UN-ECE) is based on the principle that protection will be achieved when CL for ozone (O_3) are no longer exceeded. According to the UN-ECE definition (UN-ECE, 1988) CL means "the concentrations of pollutant in the atmosphere above which direct adverse effects on receptors, such as plants, ecosystems or materials, may occur according to present knowledge". Implicit in this definition is that the CL values need to be critically reassessed on a regular basis, using the most recent data available.

The UN-ECE CL for O_3 have been recently determined for agricultural crops, forest trees and natural/semi-natural plant communities, based on the most recent scientific findings (UN-ECE, 1994). However, it has been recognized that uncertainties exist about the sensitivity of the species from the Mediterranean region, although experimental evidence and records of visible injury confirm the existence of phytotoxic concentrations of ambient O_3 (Schenone, 1992; Schenone and Lorenzini, 1992).

The Italian electricity board (ENEL SpA) has promoted a research program since 1986 aimed at investigating the effects of air pollution on Italian agricultural and forest ecosystems. As part of this program, an open-top field chambers (OTC) facility has been built in a rural area near Viterbo, Central Italy, and dedicated to active fumigation experiments with O_3 on both herbaceous and tree crops (Paolacci et al., in press). The Viterbo OTC facility was used for evaluating the effects of realistic O_3 exposure on peach trees [*Prunus persica* (L.) Batsch; 1992] and for a preliminary experiment on durum wheat (*Triticum durum* L.; 1993). From the result obtained, it was suggested that, when submitted to realistic exposure regimes in OTC using standard agronomic practices, both *P. persica* and *T. durum* behaved as O_3-sensitive species. It was of concern that realistic above-ambient O_3 levels aggravated undesired or even phytotoxic effects already present in plant material growing in the ambient air of a rural site (Paolacci et al., in press). Furthermore, re-calculation of O_3 exposure parameters from the 1992 experiment

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allowed to propose O_3 CLs for those facets of peach tree biology found to be responsive to the pollutant (Badiani et al., in press)

During the spring and summer 1995, an O_3 fumigation experiment was performed on two *T. durum* cultivars differing in the timing of commercial maturity, namely Duilio, early, and Neodur, late. Besides, a replication of the 1992 experiment was performed on P. persica. Exposure variants were obtained by enclosing the plants in OTCs.

The present paper deals with a selection of the results obtained during the *T. durum* experiment.

Materials and Methods

Two varieties of *T. durum* were used for O_3 fumigation, namely Duilio, a popular, early maturing cv., and Neodur, a late cv. of recent introduction. The seeds of each cv. were sown in half of each experimental parcel (Paolacci et al., in press), so that the area enclosed in each OTC contained approximately the same number of individuals from each cv. At seedlings emergence, 1 m^2 subplots were chosen for each cv., both in the center of each OTC and in the non-chambered plant canopies used as the ambient air (AA) control. During the O_3 experiment, the whole of non-destructive and destructive measurements were performed on these subplots.

The experimental crop was managed according standard agronomic practices. At the time of enclosure in the OTCs, the cv. Duilio plants had just entered the stage of inflorescence emergence, whilst the cv. Neodur individuals were in the late-booting stage

Full details concerning the experimental set-up have been already reported (Paolacci et al., in press) and are summarized in Table 1. The 12 cylindrical OTCs used in the present experiments were of NCLAN-type (Heagle et al., 1973).

Table 1 - Protocol and timetable of events during the 1995 ozone fumigation experiment on durum wheat plants grown in open-top chambers (OTC) at Viterbo, central Italy.

Experimental site:	Viterbo, Central Italy (N42 26/E12 05; 308 m a.s.l.; 60,000 residents)
Soil:	Low humus sandy-clay, pH 6.50
Plant material:	*Triticum durum* L.
Cultivars:	Duilio, early maturing Neodur, late maturing
Sowing date:	17 November 1994
Seed density:	240 kg Ha^{-1}
Chambering date:	14 April 1995
Number of OTC used:	12
Air changes allowed by the OTC pumps:	2 - 2.5 per minute
Mean O_3 filtration efficiency in OTC:	69.3 ± 18.8%
Number of plants in each OTC:	3000 ca
Daily operation of OTC pumps (GMT):	08:00-18:30
Start of O_3 fumigation:	18 April 1995
End of O_3 fumigation:	26 June 1995
Days of effective fumigation:	66
Daily fumigation period (GMT):	09:00-17:00
Weekly fumigation frequency (days):	7
Harvest:	01 July 1995

The square-shaped O_3 waves used for experimental fumigation were generated by electric discharge in pure (>99.7%) dehumidified O_2. Due to technical problems with O_3 generation, the *a priori* regulation of the gas flux to the OTCs was only partially possible; thus, the dose received by each chamber was actually driven by the spontaneous distribution of O_3 inside the

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network of fumigation PTFE Teflon lines and was proportional to the distance between the individual chamber and the O_3 generating device.

Air samples were taken continuously at canopy height (40 cm) from all the OTCs and from ambient air (AA) according to a 10 min time-share basis and to a constant pre-set sequence. Ozone, NO, NO_2 and SO_2 partial pressures were measured. During the experiment, environmental variables, such as air temperature, humidity and solar radiation, were continuously monitored in two OTC and in AA at the same frequency adopted for air pollutants.

Ozone exposure indices (OEI) used to quantify the treatment variants, as well as the corresponding nomenclature, were adopted according to UN-ECE recommendations (Fuhrer, 1993; UN-ECE, 1994) and were as follows: the 8-hours (09:00-17:00 GMT) O_3 mean concentration (8-hMC, nL L^{-1}); the accumulated O_3 exposure over a threshold of 0, 30, 40, and 60 nL L^{-1} [AOT00, AOT30, AOT40, and AOT60, respectively, expressed as [(nL L^{-1})*h]; the number of hourly O_3 means exceeding 30, 40, and 60 nL L^{-1} (H30, H40, and H60, respectively). The OEI based on the O_3 exposure accumulation above, or exceedance of, a given threshold were calculated for daylight hours only, defined as those during which the mean global radiation was 50 W m^{-2} or higher (Ashmore and Fuhrer, 1994).

Results and Discussion

During the experiment, the temperature, relative humidity and light intensity regimes were the same in all the OTCs used (data not shown). However, in comparison with the open air, the presence of the OTC itself caused slight changes in the microclimate the plants were exposed to, as reported in Table 2.

Table 2 - Average (D-AVG) and maximal (D-MAX) divergencies from open air microclimate as recorded in open-top field chambers during the periods 18 April - 26 June 1995 at Viterbo, central Italy.

Microclimatic parameter	D-AVG	D-MAX
Air temperature (°C)(@*)	+ 1.64	+ 9.20
Relative humidity (%)(@)	+ 9.84	+ 42.50
Solar radiation (W m^{-2})(§)	-37.87	-454.00

(@) Data collected from two open-top field chambers
(*) Daylight hours only
(§) Data collected from one open-top field chamber

Table 3 shows the average and maximal (24-h basis) partial pressures observed in ambient air for the main gaseous pollutants during the experimental period. The mean efficiency of the OTC filtering systems is also reported. In this respect, it should be noted that the activated charcoal used for the filters ensured, on average, a 50% abatement of the major air pollutants.

Table 3 - Mean and maximal partial pressures in ambient air (AA, 24-h basis, nL per L of air) and OTC filtering efficiencies (percent ± sd) for the main gaseous pollutants recorded at Viterbo during the 1995 ozone experiment on durum wheat.

Pollutant	AA		Abatement in OTC
	Mean	Maximal	
O_3	33.90	70.00	- 69.38 ± 18.80
SO_2	3.33	47.00	- 35.53 ± 6.38
NO	1.41	82.00	- 53.03 ± 7.15
NO_2	6.99	64.00	- 51.73 ± 10.27

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The exposure variants obtained in the OTCs during the O_3 fumigation experiment are reported in Table 4. The plants chambered in one of the two control OTCs (FA2) faced a higher accumulated exposure to O_3 than one of the O_3-enriched treatment (L1), although it was still well below those which both L2-L10 and AA plants were submitted to. Also, Table 4 shows that the AA treatment was almost exactly in the middle of the exposure range for all the OEI considered but one, H30, in terms of which the above treatment was the top-ranking. In an attempt to explain similar discrepancies observed during previous experiments with the present OTC facility, Paolacci et al. (in press) imputed them to the fact that, in order to prevent an excessive rise in temperature within the chambers, the OTC pumps were kept on for a daily period which was about 20 % longer than the actual O_3 fumigation time (see Table 1). This caused the chambered plants to receive charcoal-filtered air during periods of the day in which the O_3 partial pressure in ambient air was frequently around 30-40 nL L^{-1} (data not shown).

Table 4 - Ozone exposure variants obtained for durum wheat grown in open-top field chambers at Viterbo, Central Italy, during the 1995 experiment. For acronyms denoting exposure indices see text. Units: 8-hMC, nL L^{-1}; H>, number of hours; AOT, (nL L^{-1})*h. FA, charcoal-filtered air; AA, ambient air.

O_3 levels	8-hMC	H>			AOT			
		30	40	60	00	30	40	60
FA1	23.7 ± 10.2	356	106	0	21067	2442	462	0
FA2	25.2 ± 10.7	303	132	0	21331	2904	898	0
L1	24.7 ± 10.0	330	145	0	20962	3062	700	0
L2	35.9 ± 11.9	528	370	53	28578	8593	3854	198
L3	44.5 ± 12.9	528	409	0	29040	8580	3788	0
L4	52.6 ± 13.5	541	475	92	32314	12038	6679	620
L5	57.0 ± 13.3	554	409	92	31112	10745	5834	594
L6	62.8 ± 15.2	581	515	198	36828	16540	11167	3841
L7	76.3 ± 21.2	554	515	198	37858	17068	11669	2930
L8	80.9 ± 24.2	434	401	291	38550	21571	17644	11121
L9	84.2 ± 23.7	568	502	422	46873	26096	20698	11431
L10	106.9 ± 35.3	581	528	449	50450	29489	23932	13741
AA	47.3 ± 15.8	620	462	132	33528	12566	6890	647

By considering only those OEI not using a cut-off concentration, the data in Table 4 allow to calculate that the exposure variants ranged from 0.50 to 2.26 in terms of 8-hMC, and from 0.62 to 1.50 in terms of AOT00, of the corresponding O_3 levels in AA. This leads to suggest that the present fumigation experiment provided the possibility of studying the effects of realistic O_3 regimes on the plant performance. The range of exposure variants was also remarkably comparable to those used within the U.S. National Crop Loss Assessment Network (NCLAN) and the European Open-Top Chambers project (EOTCP) networks (Hogsett et al., 1993; Unsworth and Geissler, 1993).

The plant parameters measured during the O_3 fumigation experiment on durum wheat were:

Whole plant: a) number of plants m^{-2}; b) number of ears m^{-2}; c) total aboveground biomass; d) harvest index; e) straw dry weight; f) ear dry weight; g) visual disease check;

Leaf: h) biometrics (foliar length and width, specific leaf weight and dry weight; i) net photosynthesis; j) dark respiration; k) chlorophyll *a* fluorescence; l) chlorophylls and carotenoids; m) water-soluble antioxidants; n) macro- and micronutrients (N, P, Ca, K, Mg, Fe, Mn, Zn);

Ear: o) number of spikelets per ear; p) number of grains per spikelet;

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Grain: q) yield; r) mean weight; s) dry weight; t) protein content; u) gluten components; v) macro- and micronutrients (N, P, Ca, K, Mg, Fe, Mn, Zn)

The present preliminary report concentrates on certain parameters directly related to grain yield and quality, namely those marked c), d), o)-q), s) and v) in the above list. In Table 5 these parameters are compared for the two durum wheat cultivars adopted; in order to minimize the interference due to possible differential effects of O_3, only the mean values obtained from plants grown in charcoal-filtered air (FA1 and FA2 in Table 4) are listed.

Table 5 - Yield and grain mineral nutrients in two cultivars of durum wheat grown in open-top field chambers supplied with charcoal-filtered air. FW, fresh weight; DW, dry weight.

Plant parameter	Duilio (early maturing)	Neodur (late maturing)
Total aboveground biomass [(ton FW) Ha^{-1}]	12.42 ± 5.76	12.06 ± 1.3
Harvest index (%)	49.31 ± 7.53	39.06 ± 0.2
Number of spikelets per ear	15.44 ± 0.14	15.93 ± 1.3
Number of grains per spikelet	1.49 ± 0.22	1.47 ± 0.11
Grain yield [(ton FW) Ha^{-1}]	5.14 ± 1.98	4.71 ± 0.53
Grain dry weight (% of FW)	88.15 ± 0.38	88.21 ± 0.33
Grain content of [mg (Kg DW)$^{-1}$]:		
nitrogen	18,000 ± 1860	24,900 ± 400
phosphorus	3,670 ± 250	3,450 ± 310
potassium	4,822 ± 550	4,821 ± 330
magnesium	925 ± 186	967 ± 44
calcium	158.48 ± 41.21	307.56 ± 5.57
iron	33.64 ± 4.32	42.62 ± 5.48
manganese	30.84 ± 0.81	28.91 ± 0.61
zinc	25.23 ± 3.87	31.29 ± 2.04

Table 5 reveals that the major canopy parameters related to yield, except harvest index which was higher in Duilio, did not differentiate the two cultivars. On the other hand, substantial differences were apparent as far as the kernel mineral composition was concerned. The grain of the late-maturing cultivar Neodur contained substantially higher amounts of N and Fe and about twice more calcium than Duilio.

In order to obtain exposure-response relationships, the results obtained for each of the plant parameter listed in Table 5 (data not shown) were submitted to regression analysis against each of the OEI reported in Table 4. Five different regression models were tested, i.e. linear, multiplicative, exponential, second order and third order polynomials. An O_3 effect was assumed to occur only for those plant parameters showing a statistically significant ($P = 0.05$) relationship with at least one of the OEI in Table 4. The results of such regression analysis are shown in Table 6. It appears that the effects of O_3 on the responsive plant parameter were described at the best by cumulative OEI rather than 8h-MC. In all cases except one, OEI based on the accumulation of hours above a given cut-off concentration appeared to be better predictors of the O_3 effects than their dose-based counterparts (AOT). For certain plant parameters, the form of the dose-response curve was found to be non-linear (Table 6 and Figures 1 and 2). Indeed, the analysis of the NCLAN and EOTCP large databases pointed to the relevance of cumulative exposure indices as predictors of plant responses to O_3 and to the occurrence of non-linear dose-response functions (Hogsett et al., 1993; Unsworth and Geissler, 1993; Fuhrer, 1994)

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Table 6 - Best fits among O_3 exposure indices (OEI, independent variable, x; acronyms as in Tab. 4) and O_3-responsive plant parameters (dependent variable, y) measured in *Triticum durum* L. cv Duilio and cv. Neodur plants submitted to ozone fumigation during the spring-summer 1995 at Viterbo, Central Italy. Acronyms for plant parameters are: TAB, total aboveground biomass; HI, harvest index; GY, grain yield; GMC, grain mean content of.... Regression models (RM) tested were; linear (L), $y = ax + b$; multiplicative (M), $y = bx^a$; exponential (E), $y = \exp^{(ax + b)}$; 2° order polynomial (2P), $y = a + bx + cx^2$; 3° order polynomial (3P), $y = a + bx + cx^2 + dx^3$. The ranking criterion was the degree-of-freedom adjusted coefficient of determination, R^2. *P*, probability level. $n = 11$.

Plant parameter	Best fitting OEI	RM	Equation	R^2	*P*
			cv. Duilio (early maturing)		
TAB	H60	L	$y = 1825 - 1.782x$	0.937	<0.001
HI			No correlation	===	===
GY	AOT60	L	$y = 752 - 0.0223x$	0.752	<0.001
GMC:					
- Nitrogen	H40	L	$y = 20818 + 1433x$	0.307	0.049
- Calcium	H30	L	$y = 60.0 + 44.437x$	0.539	0.009
- Manganese	H40	2P	$y = 47.3 - 15.6x + 2.4x^2$	0.674	0.031
			cv. Neodur (late maturing)		
TAB			No correlation	===	===
HI			No correlation	===	===
GY			No correlation	===	===
GMC					
- Potassium	H60	L	$y = 4908 - 24.6x$	0.355	0.050
- Iron	H40	M	$y = 44.7x^{(-0.217)}$	0.600	0.007

As far as yield-related parameters are concerned, dramatic differences were observed in the O_3 responsiveness of the two *T. durum* cvs. adopted (Table 6 and Figure 1). In terms of total aboveground biomass and of grain yield, reductions in the range 25-50% were observed for the cv. Duilio, whilst the cv. Neodur was substantially unaffected (Table 6 and Figure 1).

Clear differences among the two cvs. were also suggested by the results obtained for the grain mineral composition (Table 6 and Figure 2): increasing O_3 affected the N, Ca, and Mn contents in Duilio, causing them to increase, whilst the O_3-responsive nutrients in the Neodur grain were K and Fe, which were negatively correlated to O_3 levels. It is interesting to note that certain of the mineral nutrients found to be O_3-responsive, namely N, Ca, and Fe, were the same that differentiate the two cvs. also in plants grown under charcoal-filtered air (Table 5).

Since no previous data are available for *T. durum*, comparison of mineral nutrition results is only possible with *T. aestivum*. As contrasting results have been reported for the latter species (Fuhrer et al., 1990; Vandermeiren et al., 1992; Anguissola Scotti et al., 1994), the coincidence of the present data set with previously reported evidence was expected to be only sporadical, and indeed it was (data not shown). This could have been also depended on substantial differences in the protocols adopted for the O_3 treatment (Fuhrer et al., 1990; Vandermeiren et al., 1992; Anguissola Scotti et al., 1994).

The results reported here show for the first time that productivity performance and grain quality in *T. durum* can be affected by the presence of realistic O_3 levels. The form and slope of the dose-response function for grain yield in the cv Duilio (Figure 1) was found to overlap almost perfectly to that derived by the analysis of the EOTCP database on winter wheat and adopted by the UN-ECE for establishing O_3 CL for agricultural crops (Fuhrer, 1994; see

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below). This confirms once more the robustness of the latter and suggest its suitability for deriving O_3 CL also for crops grown under Mediterranean pedoclimatic conditions. As far as grain yield is concerned, a similar sensitivity to O_3 of *T. durum* and *T. aestivum* is also suggested. Furthermore, the vast divergence of effects observed on Duilio and Neodur (Table 6 and Figures 1 and 2) confirms the importance of varietal characteristics as a modifying factor in plants' responsiveness to O_3 (Level 2 approach *sensu* UN-ECE; Ashmore, 1994; Fuhrer, 1994). The ontogenetic, physiological, and biochemical bases of the differential sensitivity to O_3 exhibited by the two *T. durum* cultivars are currently being analyzed (Badiani et al., in preparation).

Finally, the criteria adopted by the UN-ECE were used for the calculation of O_3 CL regarding two *T. durum* canopy parameters related to productivity, namely total aboveground biomass and grain yield. These criteria are summarised below (UN-ECE, 1994):

1) the CL is expressed as O_3 cumulative dose and based on a cut-off concentration of 40 nL L^{-1} (AOT40);

2) linear functions should be used to fit AOT40 to the experimental data obtained from plants. This is because, unless functions of other degrees, the linear regression model allows to individuate the point where an O_3 effect starts, thus making it clear which O_3 level should be used as the reference (Fuhrer, 1994);

3) because of limitations in the experimental procedures available to test and verify a predicted difference in the response of <10% between two treatments, it was recommended to accept a deviation in grain yield (or in total biomass for forest trees) of 10% from the reference as the effect corresponding to the CL (CL10) (Fuhrer, 1994).

4) on the basis of available data, CL10 of 5300 [(nL L^{-1})*h] were proposed for the protection of European agricultural crops (Ashmore and Fuhrer, 1994).

The CL10 obtained for *T. durum* cv Duilio according to the UN-ECE protocols are shown in Table 7. It is shown that, as expected from the aforementioned similarity among dose-response functions, the O_3 CL10 obtained for grain yield in the cv. Duilio was almost identical to that proposed by the UN-ECE (UN-ECE, 1994). It is also suggested that aboveground biomass accumulation and grain yield are facets of *T. durum* biology which exhibit similar sensitivity to the presence of O_3.

Table 7 - Ozone critical levels causing a 10% change [CL10, (nL L^{-1})*h] in two O_3-responsive yield parameters of the sensitive *T. durum* cultivar Duilio. The CL10 are based on the accumulated O_3 exposure (daylight hours) over a threshold of 40 nL L^{-1} (independent variable, x) fitted to each of the O_3-responsive plant parameters (dependent variable, y) by means of linear regression. Acronyms for plant parameters, R^2, P, and n as in Table 7.

Plant parameter	Equation	R^2	P	**CL10**
TBA	y = 1855.791 - 0.035x	0.862	<0.001	**5302**
GY	y = 811.942 - 0.015x	0.701	<0.001	**5413**

Conclusions

In the present work, O_3 CL have been proposed aiming at the protection of certain O_3-sensitive biological features of durum wheat. The results obtained suggest that the CL10 set by UN-ECE for agricultural crops might be adequate to protect the most sensitive among the O_3-responsive plant parameters prone to impact directly the marketable yield of this species.

Many other crops and natural plant communities of ecological relevance in the Mediterranean basin, either perennial or herbaceous, await due consideration; several of these might well be highly susceptible to, and even heavily impacted by, the present-day O_3 levels within the area. The setting of reliable air quality standards at the regional level will only become feasible as far as suitable data sets amenable for the CL approach will become available.

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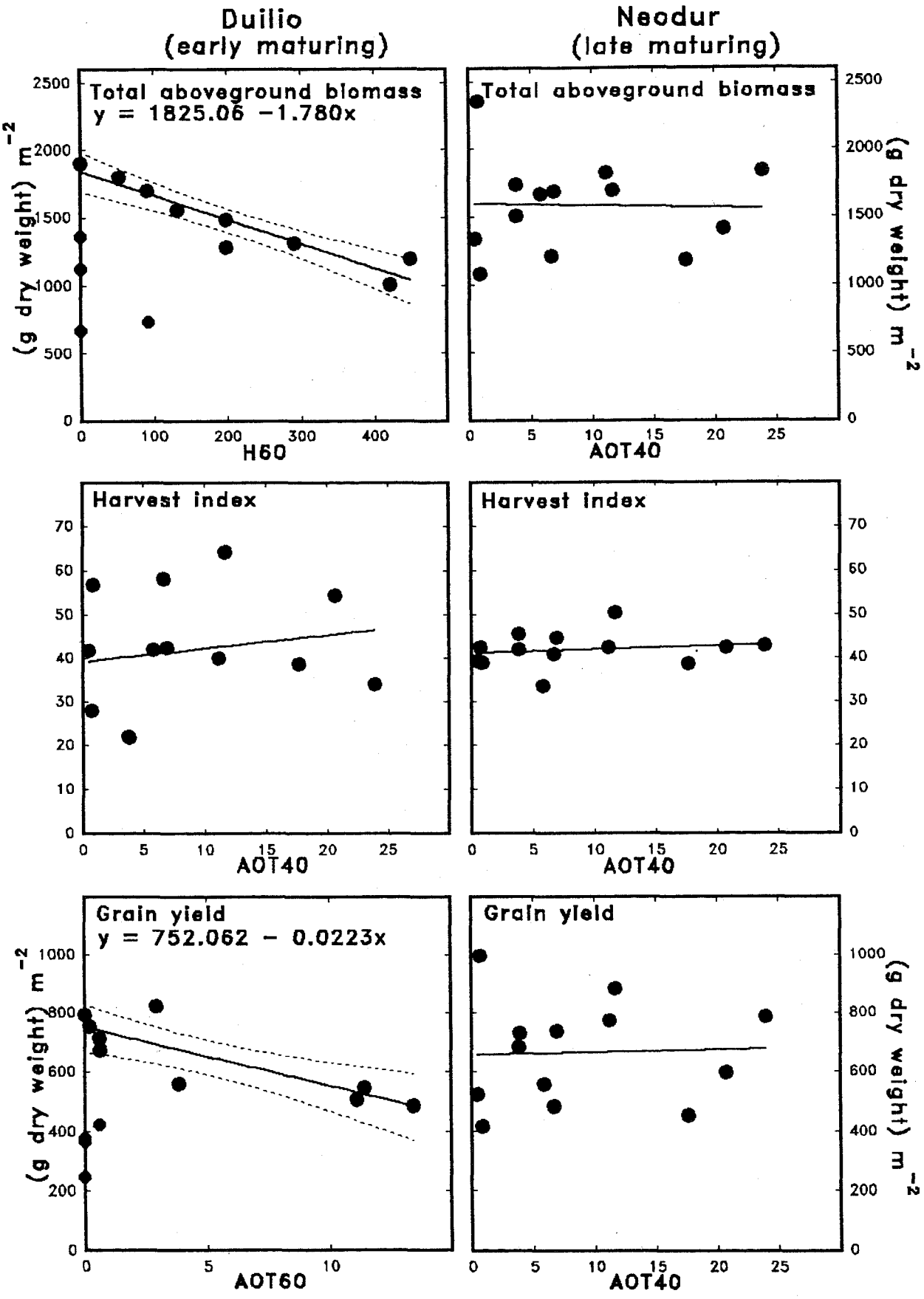


Figure 1 – Yield parameters in two cultivars of Triticum durum exposed to ozone fumigation in open-top chambers. For ozone exposure acronyms, see the text.

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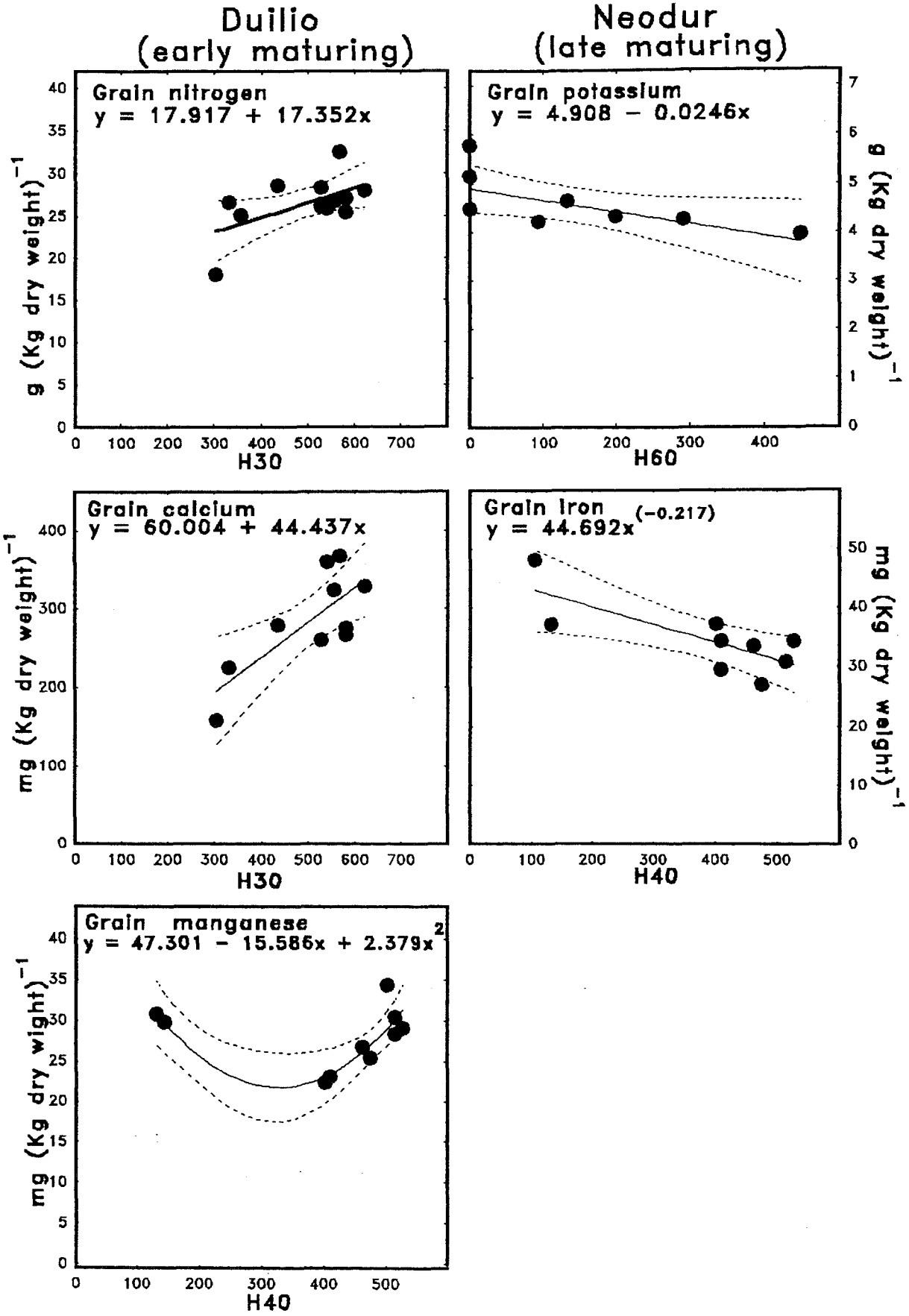


Figure 2 — Grain mineral nutrients in two cultivars on *Triticum durum* exposed to ozone fumigation in open-top chambers. For acronyms of ozone exposure, see the text.

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ASSESSMENT OF EDU PROTECTION FROM OZONE EXPOSURE ON THREE HORTICULTURAL CROPS

B. S. Gimeno[1], M. Mendoza[2], S. Sánchez[1], V. Bermejo[1]

1.- CIEMAT - IMA 3b. Avda. Complutense 22. 28040 Madrid. Spain.
2.- MIGJORN S.A. 43877 San Jaime de Enveija. Tarragona. Spain.

ABSTRACT

An assessment of O_3 effects on three different horticultural crops was performed during 1995: tomato (cv. Tiny Tim), bean (cv. Lit) and watermelon (cv. Sugar Baby). Plants from every species were either treated or non treated with EDU. Visible injury and plant fruit yield were recorded. Ozone visible injury was observed on the leaves of the three horticultural crops regardless of the EDU treatment. However, EDU did protect form visible injury on bean plants. EDU benefitial effects on yield were recorded for tomato and bean plants. EDU application did not induce any effect on visible injury or yield on watermelon plants.

1.- INTRODUCTION

Ozone-induced visible injury has been observed in a wide range of plant species in eastern Spain: watermelons, beans, soybeans, tomatoes, peanuts, potatoes, vines and Aleppo pine (Gimeno *et al.*, 1995a). Passive and active bioindication studies showed that ozone (O_3) phytotoxicity is widespread all over this region, from Catalunya to Andalucía, covering an area of at least 40.000 km^2 (Gimeno *et al.*, 1995b).

An experimental field with open top chambers (OTC) has been in operation at the Ebro Delta (Spain) since 1988. Detrimental effects of ambient levels of O_3 on the physiology and yield of different crops from the area have been reported (Bermejo *et al.*, 1993; Gimeno *et al.*, 1993).

The International Coperative Programe on effects of air pollution and other stresses on Crops and non-wood plants (ICP-Crops) has been in operation since 1988. It aims to provide quantitative information on crop and non-wood plants to air pollution in the UN/ECE area. Participants perform coordinated experiments each year. These experiments incorporate the use of the chemical protectant for ozone (Carnahan *et al.*, 1988), ethylenediurea (N-[2-(2-oxo-1-imidazolidinyl)-ethyl]-N-phenylurea; EDU). Participants record visible injury and biomass or yield of EDU-treated and untreated plants.

This paper presents the results from the experiments carried out at the Ebro Delta (Spain) in 1995, to assess the potential protection of EDU from O_3 exposure on three different horticultural crops, tomato, bean and watermelon.

2.- MATERIAL AND METHODS

Ozone-sensitive cultivars from three representative horticultural crops of eastern Spain were selected: tomato (cv. Tiny Tim), bean (cv. Lit) and watermelon (cv. Sugar Baby). After plant emergence, two sets of plants for each species were established at random; half of them

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were treated with EDU while the rest remained untreated. Two experiments were carried out with beans, while only one was carried out with tomatoes and watermelons, due to their longer vegetative cycle.

Since different growing mediums were used for each crop, a detailed description of the experimental protocol followed for every experiment is provided.

2.1.- First experiment with bean plants (from July 24th to September 27th)

Bean seeds (cv. Lit) were sown on the 19th of July. Two seeds per pot were sown in 6 cm of diameter pots. The substrate used was as follows: 40% peat (pH:3.5-4)+ 40% vermiculite + 20% perlite. 3kg/m^3 of a commercial slow-release fertilizer (Plantosan) were applied (NPK:20/15/15). The final soil mixture was adjusted to pH 6-6.5 adding carbonate.
Plants emerged on the 24th of July. On July 26th, plants were transplanted to 20 cm of diameter pots, only one plant per pot was left. The substrate composition was: 30% peat, 30% vermiculite, 30% perlite, applying 3kg/m^3 of Plantosan as a fertilizer.
Plants were irrigated daily from above and below ground using both droplet irrigation and wicks (two wicks per pot at 8-10 cm below ground surface) to prevent the plants from drought-stress at mid-day.

After plant emergence, two sets of plants were established at random. 20 plants were selected to be treated with EDU, while another set of 20 plants would remained untreated, acting as an experimental control. Increasing doses of EDU (100, 150, 200 and 250 ppm) were applied following plant development. 200 ml of EDU doses per pod were applied each time and were injected into the ground by means of a syringe. The same volume of water were applied on the same date to the Non- EDU treated plants. A detailed description of the schedule of EDU application is provided in Table 1.

Plants were fertilized on August 27th and 30th with a commercial fertilizer (Laibono, NPK:11/5/30) and also on September 1th,5th and 25th (Hakaphos naranja, NPK:15/5/30 + NH_3PO_4, NP:12/60). Insecticides and fungicides were used whenever necessary.

Visible injury was recorded on a regular basis, 20, 29, 36, 43, 57 and 65 days after plant emergence (DAE). The intensity of O_3-induced visible injury was assessed at the plant level and expressed as percentage of plants with ozone symptoms. In the last two sampling dates O_3 injury was also assessed at the leaf level using a five-grade scale (0 - no effect; IV - maximum O_3 injury), following the protocol described elsewhere (Reinert et al., 1992). The schedule for the evaluation of O_3-induced visible injury is presented in Table 1.

The final harvest was performed 74 and 79 DAE (October 6th and 11th), to enable the plants to reach to a uniform maturation stage, as presented in Table 1.

2.2.- Second Bean experiment (August 7th to October 20th)

Bean seeds (cv. Lit) were sown on the 2nd of August. Three seeds per pot were sown in 6 cm of diameter pots. The substrate used was as follows: 25% peat (pH:3.5-4)+ 25% vermiculite + 40% perlite and 10% sand. 3kg/m^3 of a commercial fertilizer (Plantosan) were applied. The pH of the mixture was corrected to 6-6.5 by adding carbonate.
Plants emerged the 7th of August. On August 16th, they were transplanted to 22 cm of diameter pots and were thinned on August 31st, keeping two plants per pot. The substrate

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composition was: 50% peat (pH 6-6.5), and 50% perlite, 3kg/m^3 of Plantosan fertilizer was applied. Thus, a more porous medium was used compared to the previous experiment, to prevent the plants from suffering from drought and heat stresses. Plants were irrigated daily from above and below ground using both droplet irrigation and water containers.

After plant emergence, two sets of plants were established at random. 10 plants were selected to be treated with EDU, while another set of 10 plants would remained untreated. Increasing doses of EDU (100, 150, 200 and 250 ppm) were applied following plant development. Since two plants per pot were kept, a double EDU dose per pot (400 ml) was used to mantain the same EDU dose per plant as in the first experiment. A detailed description of the schedule of EDU application is provided in Table 2.
Commercial plant fertilizers were applied on August 30th, September 5th (Laibono), September 11th, 15th (Hakaphos naranja), September 17th (NH_3PO_4), and September 22nd and 27th (Hakaphos naranja + NH_3PO_4).

Visible injury was recorded at 44, 52 and 59 DAE. The intensity of O_3-induced visible injury was assessed at the leaf level using a five-grade scale as mentioned in the previous section. The schedule for the evaluation of ozone-induced visible injury is presented in Table 2. The final harvest was performed at 74 DAE (October the 20th).

2.3.- Tomato experiment (April 13th to July 5th)

Tomato seeds (cv. Tiny Tim) were sown in a greenhouse on the 4th of April. Two seeds per pot were sown in 8 cm of diameter pots. The substrate used was as follows: 33% peat + 33% vermiculite + 12% perlite, 2.5 kg/m^3 of a commercial fertilizer (Plantosan) were applied (final pH ground mix 6-6.5).
Plants emerged the 13th of April and then they were transplanted to the field. On May the 3rd, plants were transplanted to 17 cm of diameter pots, only one plant per pot was left. The substrate composition was: 25% peat, 25% vermiculite, 25% perlite and 25% sand, applying 3 kg/m^3 of Plantosan as a fertilizer (final pH ground mix 6-6.5). Plants were irrigated on a daily bases from above ground using droplet irrigation.

After plant emergence, two sets of plants were established at random. 20 plants were selected to be treated with EDU, while another set of 20 plants would remained untreated, acting as an experimental control. EDU doses were applied following plant phenology as described for the first bean experiment. A detailed description of the schedule of EDU application is provided in Table 3.
Plant fertilizers were applied every two-three days in June (Hakaphos naranja, 1 gr/l+ NH_3PO_4, 1gr/l).

Sequential harvests were performed, following fruit maturation, after 83, 90, 96, 103, 114 and 127 DAE, as can be observed in Table 3.

2.4.- Watermelon experiment (March 18th to September 27th)

Watermelon seeds (cv. Sugar Baby) were sown the 11th of March on trays. Plants emerged on March 18th and were transplanted to 20 cm diameter the 18th of April. The substrate composition was as follows: 25% peat, 25% vermiculite, 25% perlite and 25% sand, applying 3 kg/m^3 of Plantosan as a fertilizer (final pH mix 6-6.5). Plants were irrigated every two days from transplantation to flowering. Then, irrigation was carried out on a daily

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bases using a droplet sytem.

After plant emergence, two sets of plants were established at random. 12 plants were selected to be treated with EDU, while another set of 12 plants would remained untreated, acting as an experimental control. EDU doses were applied as described in the first bean experiment. A detailed description of the schedule of EDU application is provided in Table 4.
Plants were fertilized on May 24th, 15th and 30th (Hakaphos naranja) and every 2-3 days from June to mid July (Hakaphos naranja, 1gr/l + NH_3PO_4, 1gr/l).

The intensity of O_3-induced visible injury was assessed 55, 62, 69, 79, 87, and 94 DAE, following the procedure described in the previous section. In the last two sampling dates visible injury was reported at the leaf level.

A final harvest was performed at 102 DAE as can be derived from Table 4. Fruit number, yield/plant, g/fruit and above-ground biomass were determined for both treatments.

2.5.- Statistical analyses.

ANOVA was used to assess the response of the different parameters to EDU, the least significant differences test (LSD) was used to determine the differences between means. Before ANOVA analysis was carried out, the normality of the data was confirmed using the Kosmogorov-Smirnov test. The homocedasticity of the variances was performed using the Kochran test.

3.- RESULTS

The results of the different experiments will be presented separatedly in this section.

3.1.- FIRST BEAN EXPERIMENT

3.1.1.- Plant performance and visible injury

Plant performance was very similar in both treatment. No differences were observed in the final amount of trifoliates per plant. Nevertheless, Non-EDU treated plants showed a retardation in both flowering (21% at 29 DAE) and pod formation (27% at 35 DAE) (Fig 1a,b & c).

O_3-induced visible injury appeared in both treatments and began to be observed at 20 DAE, when plants were flowering and presented nine trifoliate leaves. However, EDU significantly protected the plant from O_3, since visible injury expressed as either percentage of affected plants or injury intensity was decreased by EDU application, as presented in Figure 1d. This effect was most apparent at the flowering and fruiting stages, when EDU-treated plants showed the following reductions in ozone symptoms: 50% (36 DAE), 56% (43 DAE), 61% (57 DAE) and 58% (65 DAE). EDU-treated plants did not show Class III or Class IV injury, while 17,6% of control ones showed injury classes III and IV at 57 DAE, subsequently 17,7% and 29,4% of controls showed injury classess respectively at 65 DAE.

Table 1. Schedule for the first bean experiment. July 24th to October 27th

DATE	DAE[1]	EDU DOSE	ASSESSMENT OF VISIBLE INJURY	HARVEST	AOT40 (ppb.h)	PHENOLOGY
Aug. 2nd	9	100 ppm				1st trifoliate
Aug. 13th	20	150 ppm	Plant level		2533	4.2+0.2 tfs.
Aug. 21st	28	200 ppm				Flowering started
Aug. 22nd	29		Plant level		3469	9.1+0.5 tfs.
Aug. 29th	36	250 ppm	Plant level		4183	11.6+0.5 tfs. Pod formation
Sept. 5th	43		Plant level		4907	15.7+0.9 tfs.
Sept. 9th	47	250 ppm				
Sept. 19th	57		Plant & leaf level		5605	16.9+0.6 tfs.
Sept. 27th	65		Plant & leaf level		6446	
Oct. 6th	74			Yes		
Oct. 11th	79			Yes	6956	

Table 2. Schedule for the second bean experiment. August 7th to October 20th

DATE	DAE[1]	EDU DOSE	ASSESSMENT OF VISIBLE INJURY	HARVEST	AOT40 (ppb.h)	PHENOLOGY
Aug. 20th	13	100 ppm				1st trifoliate
Aug. 28th	21	150 ppm				5th trifoliate
Sept. 5th	29	200 ppm				Initial maturation of pods
Sept. 13th	37	250 ppm				8-10 pods > 4cm
Sept. 20th	44	250 ppm	Plant & leaf level		4082	
Sept. 28th	52		Plant & leaf level		4900	8-10 pods>4cm
Oct. 5th	59		Plant & leaf level		5160	
Oct. 20th	74			Yes	5892	

1.- DAE, Days After Emergence

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Table 3. Schedule for the tomato experiment. April 13th to August 18th

DATE	DAE	EDU DOSE	HARVEST	AOT40 (ppb.h)	PHENOLOGY
May 10th	27	100 ppm			6-7 leaves
May 15th					flowering started
May 24th	41	150 ppm			100% flowering (1-3 ramos)
June 7th	55	200 ppm			First fruits in formation.
June 21st	69	250 ppm			abundant fruits next to red coloration
July 5th	83	250 ppm	Yes	15.278	mature fruits
July 12th	90		Yes	16.564	
July 18th	96		Yes	17.229	
July 25th	103		Yes	17.582	
August 5th	114		Yes	19.008	
August 18th	127		Yes	20.487	

Table 4. Schedule for the watermelon experiment. March 18th to September 27th

DATE	DAE	EDU DOSE	ASSESSMENT OF VISIBLE INJURY	HARVEST	AOT40 (ppb.h)	PHENOLOGY
						6-7 leaves. 15 cm stem length
May 12th	55		yes			8.2+0.4 leaves. 25.4+2.5cm stem length
May 19th	62		yes			15.5+0.7 leaves. 47.1+3.4 cm stem length. Flowering started
May 24th	67	150 ppm				
May 26th	69		yes		17.887	
June 6th	79		yes		21.187	First fecunded leaves.
June 7th	80	200 ppm				Steady-state in growth
June 14th	87		yes		23.587	
June 21st	94	250 ppm	yes		25.687	Fruit abortion
July 5th	83	250 ppm				
July 24th	102			yes	35.587	35.587

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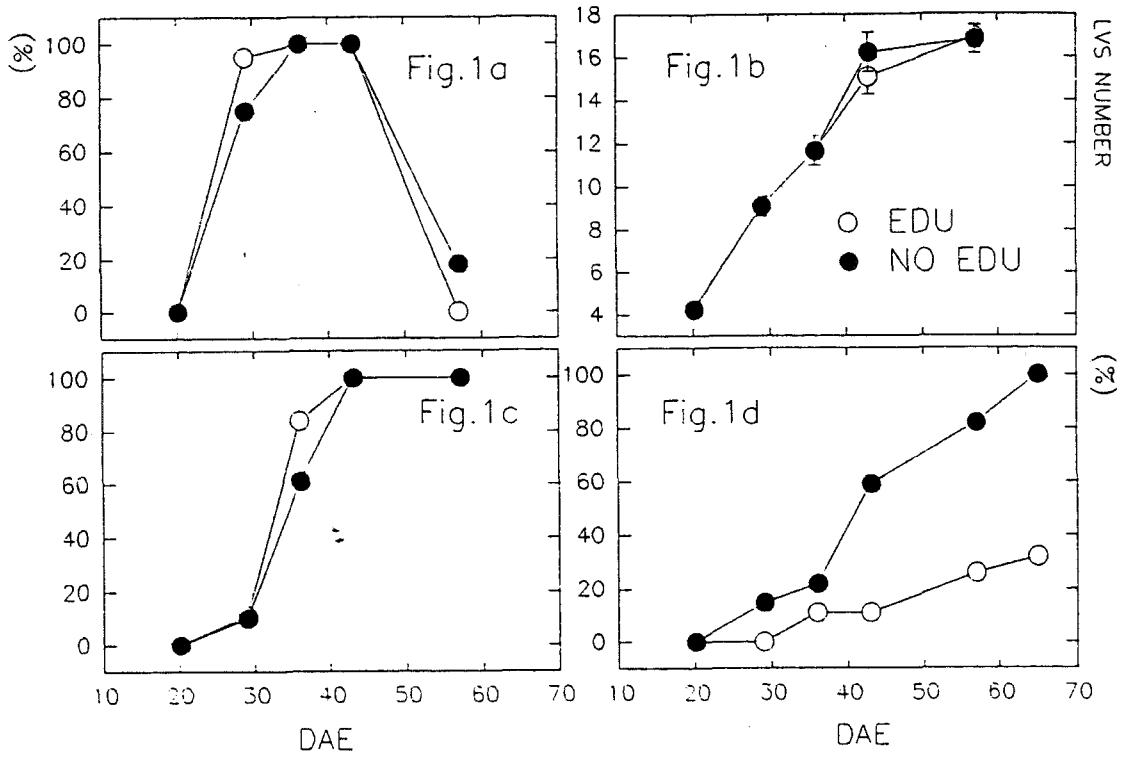


Figure 1.- Evolution of ozone-induced visible injury and plant performance of both EDU and Non-EDU treated bean plants from the first experiment. 1a, % of plant presenting flowers. 1b, number of trifoliated leaves developed. 1c, % of plants showing pod formation. 1d, % of plants showing visible injury.

3.1.2.- Plant yield

Benefitial effects of EDU on plant yield were also recorded (Table 5). Significant ($p < 0.05$) increases were recorded in both dry weight/pod (15,2%) and the amount of seeds per pod (14,9%).

Table 5.- Effects of EDU application on bean plant yield. First experiment (July 24th to October 27th). Means ± standard error. sl=significant limit, n=sample number.

	pods/plant	pods dw/plant	seeds/plant	seeds/pod	dw/pod
EDU	41.75±1.41	46.89±1.61	188.93±6.81	4.56±0.1	1.12±0.03
NO EDU	45.25±1.14	43.21±1.23	174.25±6.23	3.88±0.13	0.95±0.03
sl	0.064	0.08	0.12	0.0003	0.0007
n	16	16	16	20	20

3.1.3.- Pollution climate

Ozone exposure, expressed as AOT40 values, increased sharply from plant emergence to maximum trifoliate development and pod formation, at an almost steady rate of approximately 2.500 ppb.h/20 days. However, this sharp tendency decreased from pod maturation to final harvest, when only 2.049 ppb.h were added to the total exposure in 36 days, as can be derived from Table 1.

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The temperature daily means showed the typical pattern of mediterranean coastal sites, with temperatures ranging between 21 and 24 °C. Daily means of RH levels remained in the range of 80 to 100% during most of the plant cycle, but for five days where RH levels varied between 62 and 75%.

3.2.- Second bean experiment

3.2.1.- Visible injury

O_3-induced visible injury appeared in both treatments and began to be observed after flowering. However, EDU significantly protected the plant from O_3 exposure, since visible injury expressed as either percentage of affected plants or injury intensity was decreased by EDU application, as presented in Figure 2 This effect was most apparent at 44 DAE (50% protection) and 52 DAE (44% protection). Nevertheless, EDU protection was reduced to 20% at 59 DAE.

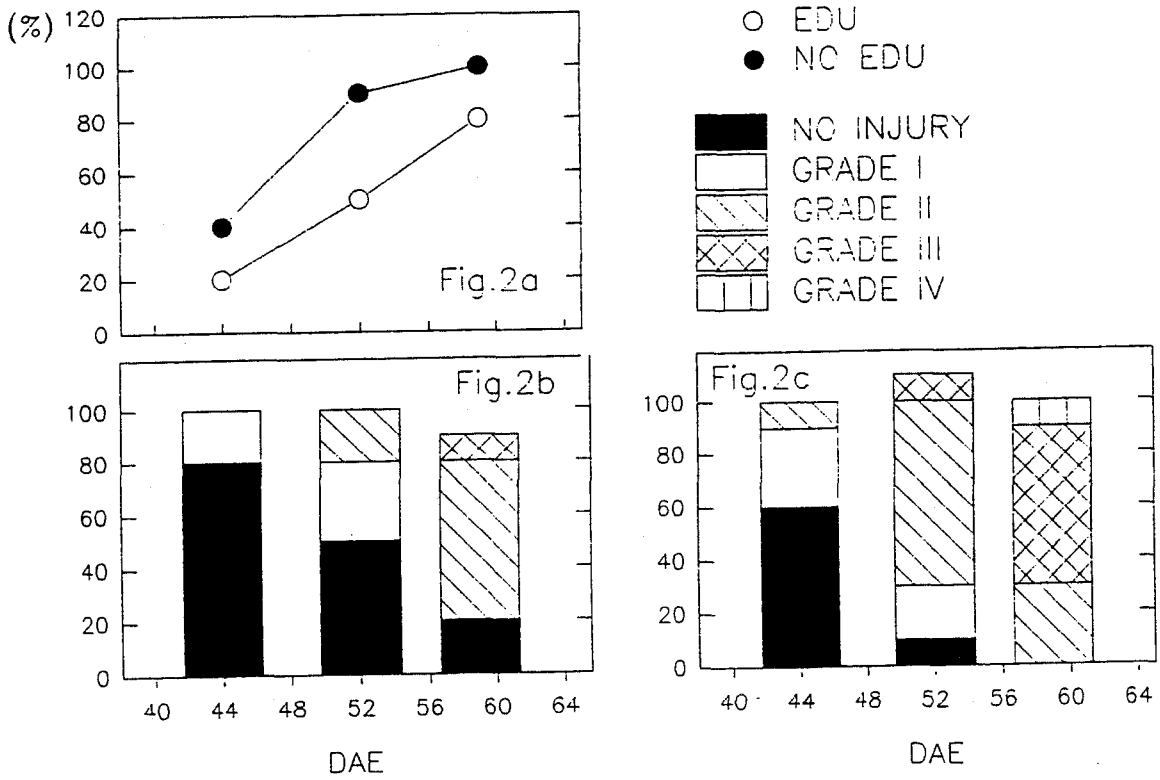


Figure 2.- Evolution of ozone-induced visible injury on the leaves of both EDU and Non-EDU treated bean plants from the second experiment (August 7th to October 20th). 2a, percentage of plants showing visible injury, 2b, foliar injury intensity on EDU treated plants. 2c, foliar injury intensity on Non-EDU treated plants.

3.2.2.- Plant yield

No benefitial effects of EDU on plant yield were recorded, expressed as either number of pods/plant, dry weight of pods/plant or number of seeds per plant (see Table 6).

3.2.3.- Pollution climate

Ozone AOT40 value from emergence till the final harvest were 5.892 ppb.h (see Table 2).

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The variation in temperature and RH levels was very similar to what was observed in the previous experiment.

Table 6.- Effects of EDU application on bean plant yield. Second experiment (August 7th to October 20th). Means ± standard error. sl=significant limit, n=sample number. No significant effects were recorded.

	pods/plant	pods dw/plant	seeds/plant	seeds/pod	dw/pod
EDU	47.87±1.55	44.33±2.80	229.10±10.2	4.70±10.2	0.93±0.03
NO EDU	48.50±0.73	45.58±1.17	228.10±5.10	4.73±0.11	0.89±0.04
sl	0.72	0.58	0.93	0.82	0.41
n	8	8	10	10	10

3.3.- TOMATO EXPERIMENT

3.3.1.- Visible injury

Visible injury was detected on tomato plants. Since EDU was applied after O_3-induced visible injury appeared, no benefitial effects of EDU were recorded.

3.3.2.- Plant Yield

No significant differences between EDU and Non-EDU treatments were recorded in the number of fruits, although an almost significant (sl=0.055) trend to present more fruits was was observed in EDU plants at 113 DAE. However an inverse trend (sl=0.055) was observed previously at 82 DAE (Fig.3a & 3b).

EDU-treated plants significantly ($p<0.05$) showed more fruit weight (g/plant) starting from 95 DAE, when plant yield was a 16% higher. The highest difference ($p<0.05$) was observed in the last harvest date (126 DAE), when EDU-treated plants showed a 34% greater fruit weight. Benefitial effects of EDU were also appreciated at 95 and 102 DAE, when EDU-treated plants showed 23 and 22% greater yield respectiveley. As was observed for fruit number, EDU induced a 36% reduction in fruit weight/plant ($p<0.05$) at the first harvest (82 DAE). This initial adverse effect somewhat balanced the benefitial effects of EDU recorded during the following harvests, determining a final 17% increase ($p<0.05$) in the toatl yield of EDU treated plants, although greater differences were observed in the intermediate harvests (Fig.3c & 3d).

3.3.3.- Pollution climate

Ozone accumulated exposure, expressed as AOT40, was 20.487 ppb.h when the last harvest was performed as can be observed in Table 3.

Daily means of temperature showed ranged between 15 and 26 °C. A steady increase in the temperature profile was appreciated from 61 DAE. RH levels remained in the range of 80-100%.

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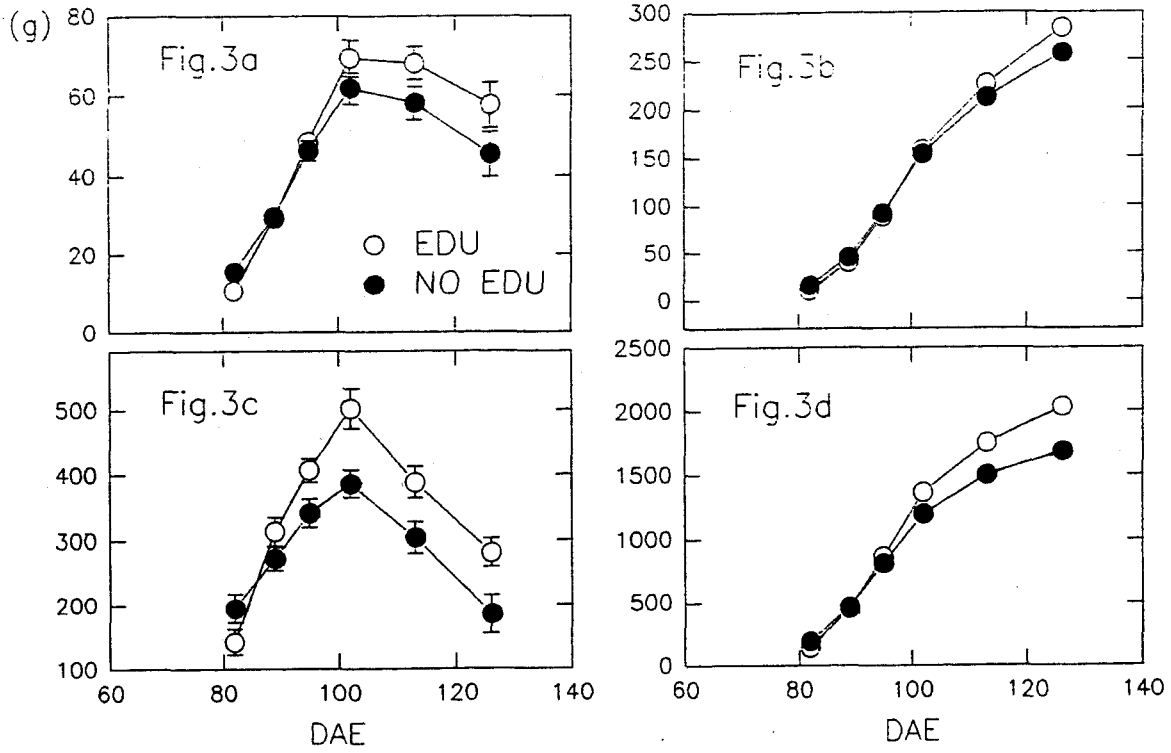


Figure 3.- Accumulated yield per tomato plant for the different harvests. 3a, number of fruits. 3b, accumulated fruit number per plant. 3c, fruit weight per plant. 3d, accumulated fruit weight per plant.

3.4.- WATERMELON EXPERIMENT

3.4.1.- Visible injury

Ozone visible injury first appeared when flowers were already formed (62 DAE). Visible symptoms were also assessed on 79, 87 and 94 DAE but no benefitial effects of EDU were appreciated, expressed either as number of affected leaves or as intensity of injury.

3.4.2.- Plant Yield

Watermelon final harvest was performed on 94 DAE. No EDU effects on yield expressed as either number of fruits, yield per plant, grams per plant or above-ground biomass were recorded (Table 6).

3.4.3.- Pollution climate

Ozone AOT40 levels were above 35.000 ppb.h when the final harvest was performed. Daily mean RH levels remained between 70 and 100%, while the daily mean profile for temperature varied between 12 and 26 °C; a steady increase in temperature levels was observed along the watermelon cycle.

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Table 6.- Effects of EDU application on watermelon yield (March 18th to September 27th). Means ± standard error. sl=significant limit, n=sample number. No significant effects was recorded.

	Fruit number	Total weight (g)	Weight/fruit (g)	Biomass (g)
EDU	2.33±0.23	1503±106	700.4±58	319.4±35.9
NO EDU	2.33±0.23	1438±161	646.5±78	294.7±34.5
sl	1	0.74	0.58	0.58
n	9	9	19	9

4.- DISCUSSION

The pollution climate recorded at the Ebro Delta corresponds to the mediterranean sites located in coastal areas. Moderate temperatures and high relative humidities prevailed during the 1995 growing season for the three horticultural crops studied. Ozone levels were the second highest ever recorded at this site since 1988. Those levels were specially high during spring time. Thus the two crops that were sown in spring time and had the longest vegetative cycle, watermelon and tomato, also showed the highest AOT40 values. The importance of spring time exposure was specially dramatic for watermelon plants, which showed increases of 2.000 ppb.h and even 3000 ppb.h in periods of 7 to 10 days.

In general, EDU protected the plants from ozone exposure at different levels depending on the species. Beans were protected at both visible injury and yield levels, while EDU-protection on tomato plants was observed at the yield level. No benefitial effects of EDU on watermelon plants were recorded.

Although EDU concentrations did not reach to toxic levels in any of the bean experiments, EDU induced a slight delay in flower and pod formation. This would be in agreement with what was observed by Ommen *et al.* (1994), which reported a retardation of plant senescence followed by EDU appliacation, while an accelaration was induced by O_3 exposure.

In the two experiments that were carried out with beans, EDU protected the plants from O_3-induced visible injury expressed as either percentage of affected plants or injury intensity., as reported by several authors (Weidensaul, 1980; Saettler, 1981; Toivonen *et al.*,1982, Adomait *et al.*,1987, Whitaker *et al.*, 1990). EDU application was performed on a weekly bases instead of every fifteen days as recomended by the 1995 ICP-Crops protocol, due to the much shorter vegetative cycle (60 -70 days) that Lit cv presents in the mediterranean region compared to northern areas (Gimeno *et al*, 1993). However, EDU did not completely protect the plant from O_3 visible injury as opposed to what has been observed by Brunschön-Harti *et al.* (1994) using a similar protocol for EDU application.

No benefitial effects of EDU on plant yield were recorded in any of the bean experiments. However, in the first experiment (from the 24th of July till the 27th of September), EDU treated plants showed higher dry weight per pod and more number of seeds per pod. Smith (1987) obtained a similar result; the Non-EDU treated plants formed more pods per plant and compensated the effect observed at the pod level. In fact, those plants that showed more visible injury tended to present more pods, although with a poorer appearance. The disparity

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of the results form both experiments coul be attributed to the difference in O_3 exposure recorded in both experiments. Interestingly the long-term critical level for crop yield protection has been established at 5300 ppb.h (Fuhrer & Acherman, 1994); the AOT40 values recorded in the second experiment were very close to the critical level (5892 ppb.h) and no effects on yield were recorded. On the contrary, the accumulated exposure for the first experiment was much higher (6956 ppb.h) and it could explain the higher pod weight and the higher seed number/pod recorded for the EDU-treated plants. Moreover, this difference in ozone exposure could not be explained using a 10 hour mean index from emergence to harvest since the same value applied for both experiments (45 ppb).

Although ozone-induced visible injury appeared on tomato leaves before EDU was applied, Non-EDU treated plants showed less yield (expressed as g/plant) than EDU treated ones from 95 Days After Emergency (DAE). The total ozone exposure was 20.480 ppb.h. Differences in yield response were apparent from an accumulated exposure of 17.229 ppb.h. EDU application at early stages of the development of tomato plants induced yield losses in the first harvest. Probably, it would be necessary to perform more experiments on EDU application at different stages of development to be able to avoid this kind of effects.

EDU did not significantly protected against the expresssion of O_3 injury in watermelon plants. No EDU effects on yield, expressed as either number of fruits, yield per plant, grams/fruit or above-plant biomass were recorded, which could be attributable to poor growing conditions or unadequate EDU dose. A new protocol needs to be defined for this crop.

The results showed that ambient O_3 levels during 1995 did alter a wide range of horticultural crops, thus confirming the relevance of this pollutant in the mediterranean region.

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Response of wheat to CO_2 enrichment, tropospheric ozone and different nutrient supply

A. Fangmeier, U. Grüters, B. Vermehren and H.-J. Jäger

Institut für Pflanzenökologie, Heinrich-Buff- Ring 38, D-35392 Gießen

Abstract

Spring wheat *Triticum aestivum* cv. Minaret was exposed to three CO_2 and ozone levels, in combination with differend nitrogen supply, in open-top chambers in 1994 and 1995. CO_2 enrichment increased grain yield by 31 to 42 % dependig on season, and shoot biomass by 39 to 52 %. Stem biomass was most responsive to CO_2 enrichment which was due to accumulation of water soluble carbohydrates, in particular of fructans. CO_2 and nitrogen supply interacted positively on grain yield and shoot biomass. Grain quality, however, was reduced by CO_2 enrichment which was related to reduced leaf nitrogen demand. The cultivar under investigation was rather insensitive to ozone. Only during intermediate harvests, enhanced leaf senescence was observed, causing LAI depressions without effects on final yield. However, when peak ozone concentration occurred during early development, yield depressions were found. Since these data did not fit into the AOT40 concept, some critical remarks and suggestions for improvement are given.

Introduction

From the beginning of the Industrial Revolution 200 years ago, global carbon dioxide concentrations have been continuously rising from approximately 280 µl l^{-1} (IPCC, 1990) to more than 360 µl l^{-1} in 1994 (Keeling *et al.*, 1995). This is the highest concentration ever found on earth for the past 220.000 years (Jouzel *et al.*, 1993). At the same time, at least in industrialized regions of central Europe, increasing levels of tropospheric ozone have been observed (Sandroni and Anfossi, 1994; Staehelin et al., 1994; Grünhage & Jäger, 1995).

Vegetation responds directly to CO_2 enrichment as was shown in numerous experiments. Crop response to elevated CO_2 has been studied intensively (see reviews e.g. by Kimball, 1983; Cure & Acock, 1986; Rogers & Dahlman, 1993). C_3 crops show an average increase in (vegetative) biomass of 58 %, C_4 species (both crops and wild species) of 22 %, and CAM species of 15 %, when CO_2 concentrations are approximately doubled (Poorter, 1993). Ozone is known as major air pollutant both for natural vegetation and crops (Krupa and Manning, 1988; Runeckles and Chevone, 1992). In European studies, ozone has been shown to reduce yield of cereals such as wheat, and several attempts have been made to evaluate critical levels for ozone preventing crop yield loss (Skärby *et al.*, 1993; Fuhrer, 1994).

In the field, one environmental factor never acts alone, rather, numerous interactions occur. CO_2 is expected to ameliorate ozone effects (Allen, 1990) via its influence on plant water relations (Kimball *et al.*, 1993; Tyree and Alexander, 1993). On the other hand, effects of CO_2 enrichment are modified by nutritional state of the plants (Van Kraalingen, 1990; Conroy and Hocking, 1993), in particular by nitrogen and phosphorus supply (Conroy, 1992).

In this study, a three-way factorial design was applied to a spring wheat cultivar in open-top chambers to test the response to CO_2 enrichment, nitrogen nutrition, and tropospheric ozone.

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Data from the 1994 and 1995 growing seasons will be presented, and some critical remarks concerning the AOT40-concept of critical levels for ozone will be made with regard to inconsistencies between our data and predicted data from this concept.

Materials and Methods

Plant culture

Spring wheat *Triticum aestivum* cv. 'Minaret' was sown into pots 40 cm in height and 10 cm in diamter filled with loamy sand on April 28 in 1994 and on April 27 in 1995. Five seeds were sown into one pot, and plant density was reduced to two per pot two weeks after sowing, yielding a canopy densitiy of 140 plants m^{-2}. Two applications of pesticides to control aphid infestation had to be carried out, using Pirimor (Spiess-Urania) in 1994 and Neudosan (Neudorf) in 1995. The plants were regularly watered using a drip irrigation system to minimize drought stress. Loss of nutrients by leaching was prevented by the construction of the pots.

CO_2 and ozone fumigations and nitrogen application

Exposures took place in open-top chambers of the Raleigh type (Heagle *et al.*, 1973) 3.15 m in diameter, 2.40 m in height) driven at two air changes per minute. No filters except dust filters were used. For a detailed description of the system see Fangmeier *et al.* (1992). CO_2 was desired to be maintained at three concentrations: ambient (NF treatment), ambient + 160 µl l^{-1} (NF+ treatment), and ambient + 320 µl l^{-1} (NF++ treatment), in combination with two different ozone levels: ambient air concentrations (NF treatments) and tracking ambient air by 1.5 (NF1.5 treatments), in a full factorial design, using two chamber replicates, respectively. Two levels of nitrogen nutrition were applied in a split-plot design: 270 and 150 kg N ha^{-1} in 1994, and 300 and 150 kg ha^{-1} in 1995. In 1994, additional fertilizer treatments (120, 210, and 330 kg ha^{-1}) were applied to gain more information about CO_2-N-interactions. All other nutrient amounts except nitrogen applied via fertilizer were identical in all treatments.

Harvests and growth, biomass and yield estimations

In both years, three main harvests were taken at growth stages 31 (beginning of stem elongation), 65 (anthesis half-way), and 93 (ripening) according to the key by Tottman and Broad (1987). Five replicate pots with two plants each from each chamber/treatment replicate were taken at these harvests, respectively. In 1995, additional harvests were taken at growth stages 24, 39, 55, 69, 77, 83, 85, and 92 to provide reasonable time resolution of biomass accumulation and redistribution. Because of limited space in the chambers, these additional harvests were carried out with one (early harvests) or three pot replicates (harvests during grain filling) per replicate chamber/treatment only.

At any of the harvests, vegetative organs of harvested plants were separated into green and senescent leaves (leaf blades, respectively), flag leaves and older leaves, flag leaf sheaths, stems including leaf sheaths of leaves other than flag leaves, peduncles, and roots. Ears were separated into those from main shoots and tillers, and glume weight, grain weight, grain number and 1000 grain weight were estimated for these samples. Prior to weighing, vegetative samples were oven-dried at 70 °C for 48 hours, and grains were dried at 35 °C until no further weight loss was observed. Green leaf area was estimated using a LiCOR LI-3000A Portable Area Meter eqipped with a LI-3050A Belt Conveyer. Leaf area index was calculated from these readings based on a canopy densitiy of 140 plants per m^2.

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Biochemical analyses

Nitrogen analyses were carried out with an autoanalyser, and carbohydrate contents were estimated according to a method described by Lever (1977) after extraction with ethanol and H_2O/Triethanolamine.

Statistics

Whenever more than one pot replicate had been harvested, chamber/treatment means were calculated from the replicate pots. These means or the pot data from single pot harvests were subjected to Multiple Regression Analyses, using the mean CO_2 and ozone concentrations from sowing until harvest from the particular chambers, and the cumulative amount of applied nitrogen, as independent variables.

Results and Discussion

Climatic conditions and trace gas concentrations

Daily averages of temperature and relative humidity inside and outside the chambers are shown in Figure 1 for the 1994 and the 1995 growing season. The chambers increased daily temperatures by up to 2°C. The maximum hourly temperature differences were 4°C at hot, sunny days which occurred during the grain filling period in both seasons. Chamber effects on relative humidity were within the accuracy of the sensors.

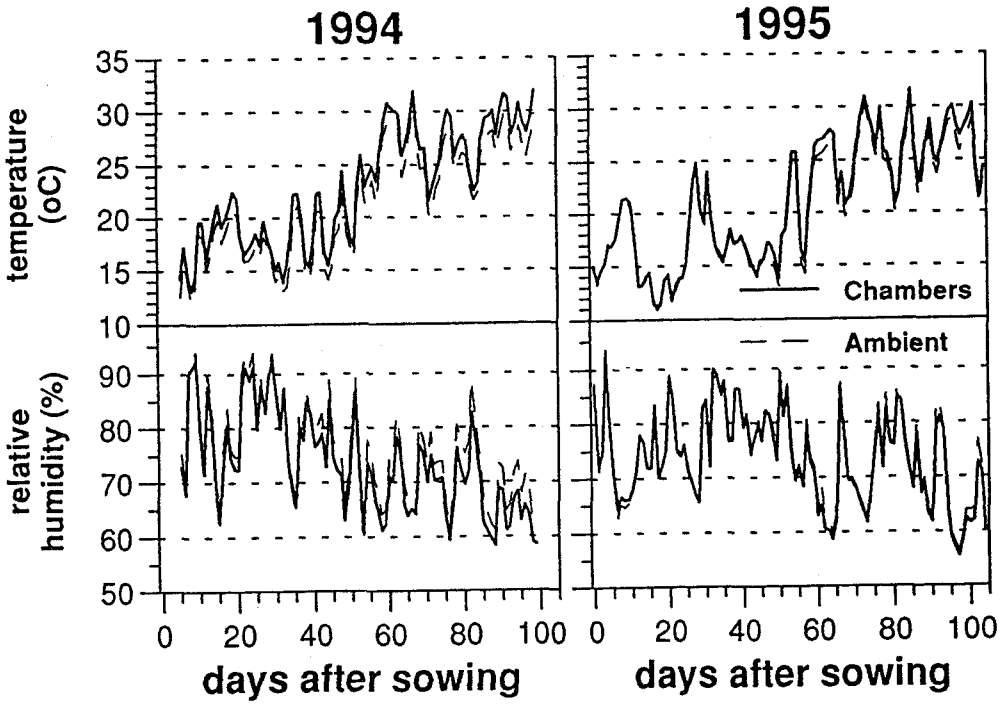


Figure 1: Temperature and relative humidity inside and outside of open-top chambers used to expose spring wheat to CO_2 and ozone (daily averages during the growing seasons 1994 and 1995). Day 0 after sowing was April 28 in 1994 and April 27 in 1995.

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To illustrate the performance of the gas dispensing system, average hourly seasonal concentrations of CO_2 and ozone achieved in any of the chambers of the 1994 growing season are shown in Figure 2.

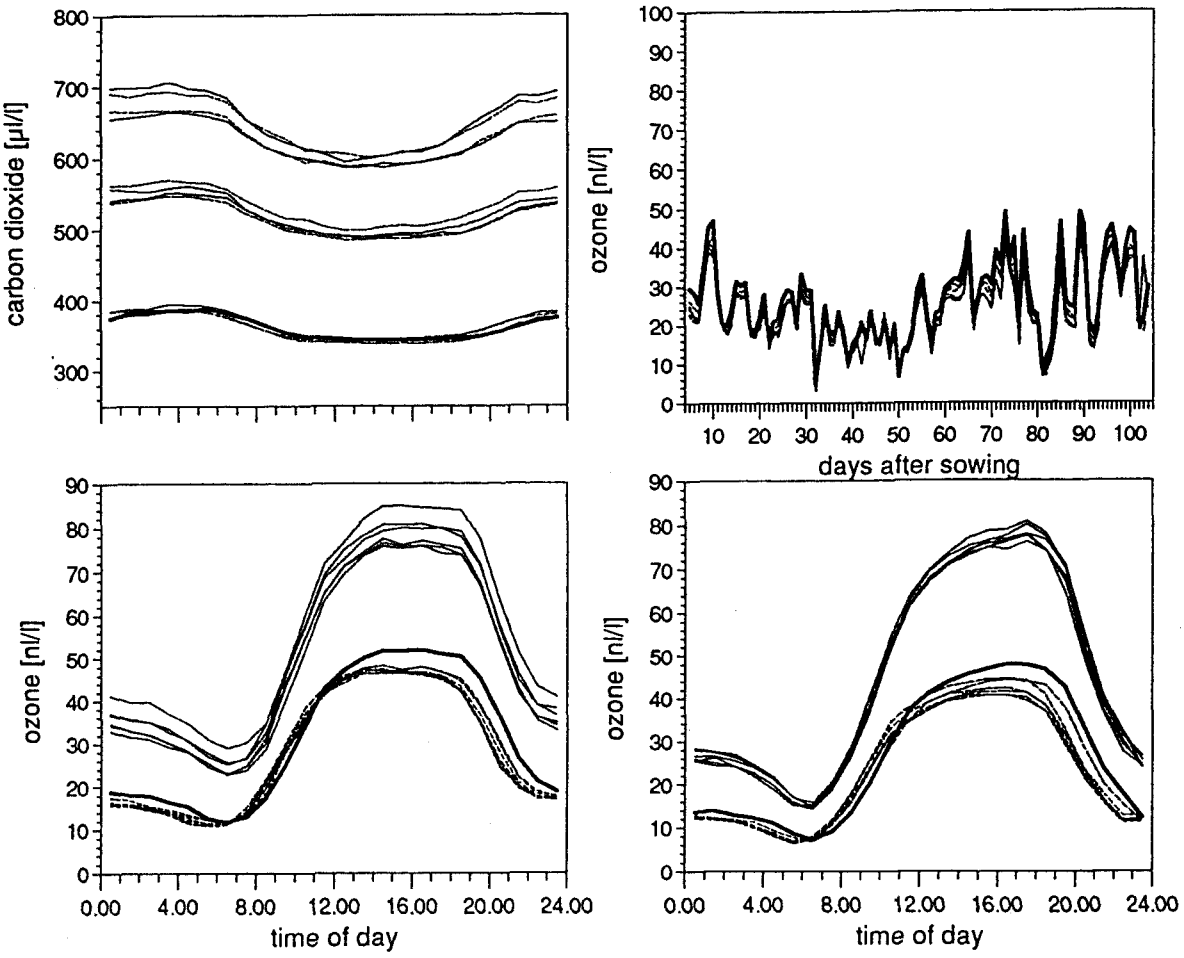


Figure 2: top left: saisonal hourly CO_2 means (from beginnign of exposure until final harvest) in ambient air and in open-top chambers in the 1994 growing season, bottom left: saisonal hourly ozone means in 1994, bottom right: saisonal hourly ozone means in 1995, top right: daily 24 hr ozone means in 1995. All data are based on hourly means continuously recorded in any of the chambers throughout the experimental seasons.

The performance of the gas dispensing system was excellent in both seasons with one exception: in the 1995 growing season, far too high ozone concentrations were achieved on day 9 and day 10 after sowing in one of the NF1.5 chambers driven at ambient CO_2 levels (see top right graph in Fig. 2). Emergence took place on day 7 after sowing, thus, seedlings of approximately 6 to 10 cm in height were exposed to extraordinary high ozone concentrations. The effects of these erroneous treatment will be shown and discussed in the „Ozone effects" section. For any other description and statistical evaluation of treatment effects, data from this particular chamber were excluded from the analyses.

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CO_2 effects

Grain yield was significantly increased by CO_2 enrichment. At high nitrogen supply and ambient ozone levels, 31.6 % more grain mass was yielded in 1994 and 41.8 in 1995 when CO_2 was raised by 278 or 240 µl l^{-1} above ambient (Table 1). Grain yield increase resulted solely from increased grain numbers, which in turn were due to increased tillering at elevated CO_2. However, also the number of grains per ear was somewhat increased by CO_2 enrichment (data not shown). Thousand grain weight was reduced at elevated CO_2 (data not shown) by approximately 5.8 % (average from both seasons).

Table 1: Effects of CO_2 enrichment on yield, yield components, and biomass partitioning at ripening (growth stage 93 according to Tottman and Broad, 1987) of spring wheat *Triticum aestivum* cv. Minaret, expressed as % change in high CO_2 treatments compared with ambient CO_2 levels (averaged data from two chamber replicates, respectively).
All data relate to well fertilized plants exposed at ambient ozone concentrations.

Experimental season	1994	1995
CO_2 enrichment actually achieved in open-top chambers in the high CO_2 treatment (µl l^{-1} above ambient)	278 µl l^{-1}	240 µl l^{-1}
Grain yield	+ 31.6 %	+ 41.8 %
Grain number per m^2	+ 38.9 %	+ 52.2 %
Total shoot biomass	+ 41.2 %	+ 41.3 %
Total leaf biomass	+ 18.6 %	- 9.9 %
Stem biomass (without peduncles)	+ 62.2 %	+ 52.1 %
Peduncle biomass	+ 82.3 %	+ 91.5 %

The yield increases found in this study are rather high compared with those reported from experiments with other cultivars. Mitchell *et al.* (1993) detected yield increases of 15 % in winter wheat, Weigel *et al.* (1994) exposed two spring wheat cultivars which yielded 24 and 33 % more grain biomass, and Cure and Acock (1986) found an average response of 35 % yield increase at doubled CO_2 concentrations (average of several observations). The reason for the relatively high response in our study could be related to the comparably low plant density (140 plants m^{-2}) which might have led to higher light penetration to lower canopy layers, thus reducing radiation limitations in the CO_2 response (Körner, 1995).

Total shoot biomass was approximately as responsive to CO_2 enrichment at final harvest as was grain yield. However, biomass partitioning was significantly altered by CO_2 enrichment, stems and pecuncles showing the highest accumulation of additional carbon. These organs were even more responsive at intermediate harvests: shortly after anthesis, biomass of stems from tillers was increased by 107 % in the high CO_2 treatment (data not shown).

This pattern of biomass partitioning at elevated CO_2 in wheat is due to the carbohydrate metabolism of cereals (or of *Poaceae* in general). Water soluble carbohydrates (WSC), in particular frutan, serve as long-term pools in vegetative tissues. Storage takes place mainly in vacuoles of parenchyma in stems (including peduncles) and leaf sheaths (Schnyder, 1993). WSC were analysed in samples from the 1994 growing season, and the corresponding results for stems of main shoots are shown in Figure 3. The biomass increase of the stems at elevated CO_2 could nearly completely be related to the accumulation of WSC, in particular of fructan. It is also obvious that the WSC pools were depleted again from day 71 after sowing, i.e. during grain filling (anthesis took place on day 63 after sowing), suggesting that WSC pools in the stems contributed to grain yield increase under CO_2 enrichment.

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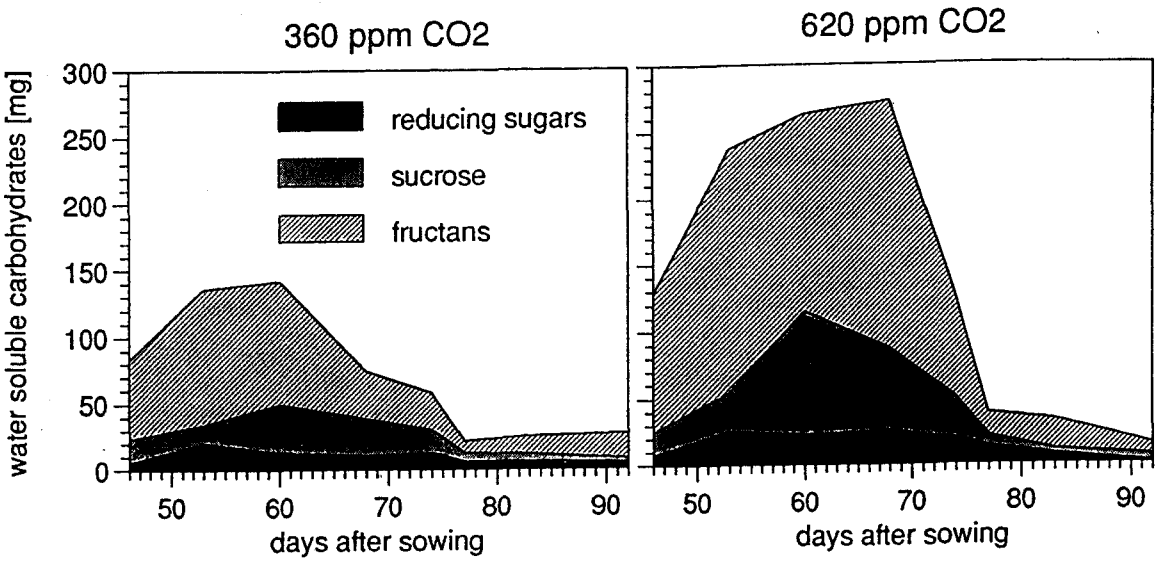


Figure 3: Amount of water soluble carbohydrates (mg per plant) in main tillers of spring wheat *Triticum aestivum* cv. Minaret exposed to CO_2 enrichment in open-top chambers. All plants were well fertilized and exposed to ambient ozone levels.

CO_2 by nitrogen fertilizer interactions

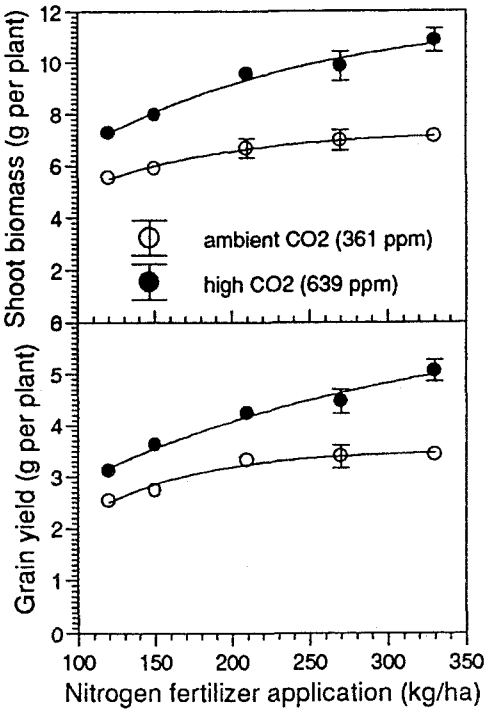


Figure 4: Interactive effects of CO_2 enrichment and nitrogen fertilization on shoot biomass and grain yield of spring wheat *Triticum aestivum* cv. Minaret exposed in open-top chambers. Data represent means ± standard deviation from two chamber replicates.

CO_2 enrichment and additional nitrogen fertilization interacted positively, as shown in Figure 4. Increase in shoot biomass due to CO_2 enrichment ranged from 22.9 % at 120 kg N ha^{-1} to 47.4 % at 330 kg N ha^{-1}, and that in shoot biomass from 31.4 % (120 kg N) to 52.8 % (330 kg N).

Concerning grain quality, significantly reduced nitrogen and protein concentrations were found in grains from plants exposed to elevated CO_2. For example, grain nitrogen content was reduced from 2.65 % of dry weight at ambient CO_2 to 1.92 % at high CO_2 in plants fertilized with 330 kg N ha^{-1}, and from 1.84 % to 1.48 % in plants fertilized with 120 kg N ha^{-1}, thus suggesting severe impacts of CO_2 enrichment on bread-making quality of wheat flour.

Interactive effects of CO_2 enrichment and nutrient supply have been found in many other studies (Conroy, 1992; Linder and McDonald, 1993; Overdieck, 1993). Several reasons are discussed to explain reduced nitrogen content in plant tissues, a response which is commonly found at elevated CO_2, such as a „dilution" by the accumulation of additional carbon

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skeletons, a reduced uptake and xylem transport of nitrogen via reduced transpiration rates, a reduced demand for Rubisco or reduced nitrogen fluxes through the photorespiratory cycle (Hocking and Meyer, 1991, Conroy, 1992; Conroy and Hocking, 1993). Our results (Vermehren *et al.*, in press) suggest that the latter effects of CO_2 on metabolism of the leaves are responsible for the observed reductions of grain nitrogen content. Taking into account that redistribution of nitrogen from vegetative organs rather than nitrogen uptake from the soil is responsible for nitrogen nutrition of the grains (Van Kraalingen, 1990), reduced nitrogen demand of the leaves may easily explain reduced grain nitrogen contents. In Figure 5, the time course of the nitrogen amounts in stems, leaves and ears of spring wheat in the ambient and high CO_2 treatments are shown, clearly indicating that the nitrogen demand of the leaves was significantly reduced at elevated CO_2.

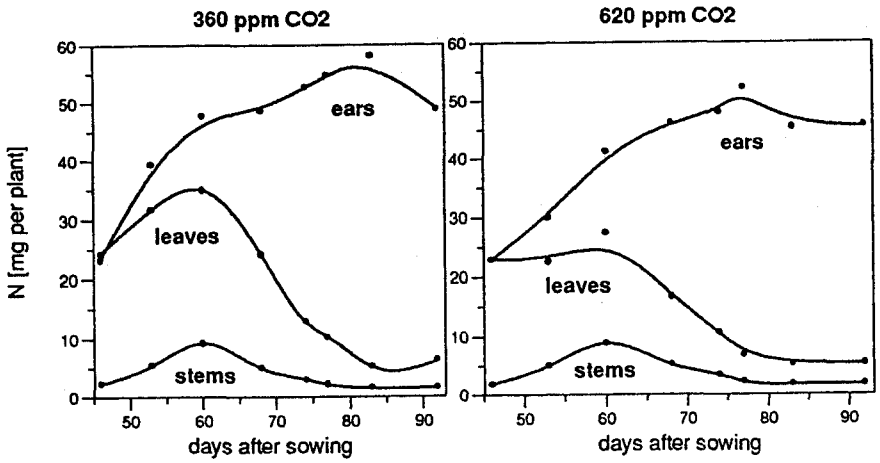


Figure 5 (left): Time course of nitrogen amounts in stems, leaves and ears of the main shoots of spring wheat *Triticum aestivum* cv. Minaret exposed in open-top chambers.

Ozone effects

Ozone had no significant effect on yield or biomass at final harvest in either season. However, a slight (but significant) reduction in 1000 grain weight was observed in 1994, which is in

Table 2: Results from Multiple Regression Analyses of yield and biomass data at final harvest. Based on chamber / treatment means.

Parameter		Significance of CO_2	Significance of O_3	Significance of N	Adj. R^2
Grain yield	'94	<.001	n.s.	<.001	.899
	'95	<.001	n.s.	.0285	.584
Grain no.	'94	<.001	n.s.	<.001	.799
	'95	<.001	n.s.	<.001	.814
Shoot mass	'94	<.001	n.s.	<.001	.879
	'95	<.001	n.s.	.0040	.746
Leaf mass	'94	<.001	n.s.	<.001	.667
	'95	.0118	n.s.	.0097	.447
Stem mass	'94	<.001	n.s.	<.001	.860
	'95	<.001	n.s.	.0316	.772
Pedun-cle	'94	<.001	n.s.	.0163	.820
	'95	<.001	n.s.	n.s.	.664

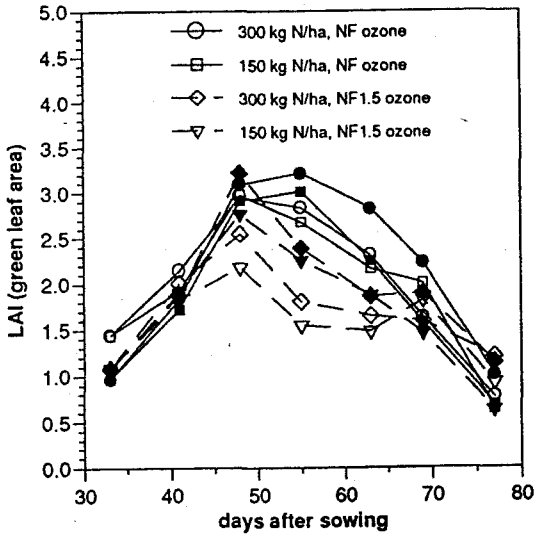


Figure 6: Time course of LAI in spring wheat at different CO_2, ozone, and nitrogen fertilizer treatments. Open symbols: ambient CO_2, closed symbols: high CO_2.

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agreement with several other findings on ozone effects on wheat yield (Fuhrer *et al*, 1989, Fangmeier *et al.*, 1994). Results from the statistical evaluation of yield and biomass data at final harvest are give in Table 2. The most pronounced effect of ozone was on leaf senescence which was enhanced in the NF1.5 treatments. Thus, during intermediate harvests, leaf area index (LAI) was significantly reduced by ozone (Figure 6). Shortened life time of the leaves, on the other hand, did not lead to depressions of grain yield.

These statements relate to data from those chambers where ambient and 1.5 times ambient ozone concentrations were excellently maintained. Data from 1995 from the one particular chamber with a failure of the gas dispensing system (see section 'Climatic conditions and trace gas concentrations' were not taken into account. In this chamber, however, the plants suffered from high ozone exposure (Figure 2) on days 9 and 10 after sowing, i.e. on days 2 and 3 after emergence, for their whole life cycle. Compared with the replicate chamber driven with the same treatment, grain yield at final harvest was reduced by 27.8 % and shoot dry weight by 22.0 %. Grain yield reduction was mainly based on decrease of grain number per plant (-19.6 %), but also on 1000 grain weight (-10.7 %).

The question remains whether common exposure indices such as the AOT40 concept are able to predict effects like those described above. AOT40 values in any of the open-top chambers used in this study and in ambient air were calculated for the exposure period 1995 (Table 3). Obviously, no relation at all can be found between AOT40 values and the effects of ozone found in this study. According to Fuhrer (1994), AOT40 of approximately 40,000 ppb hr should lead to 70% yield loss compared with clean air. Although no clean air treatment was included in thus study, the data obviously do not fit into the regression line provided by Fuhrer (1994). It may be argued that the potted plants used in this study will react less susceptible to ozone than field-grown plants in the open-top chamber experiments used for establishing the AOT40 regression line. On the other hand, differences of orders of magnitude in the responses to ozone may unlikely solely be explained by soil conditions. Rather, the cultivar exposed in this study appears to very insensitive to ozone which is confirmed by other groups using the same cultivar under field soil conditions.

Table 3: AOT40 values achieved in open-top chambers and in ambient air during the 1995 growing season for spring wheat. AA = ambient air, A + xxx = ppm CO_2-addition to ambient air, NF = non-filtered, NF1.5 = tracking ambient air by factor 1.5. Data from the particular chamber where ozone levels were exceeded on days 9 and 10 after sowing are printed in bold letters.

Ozone treatment	CO_2 treatment	Chamber replicate	AOT40 [ppb hr]
AA	AA	-	10172
NF	ambient	1	7593
NF	ambient	2	6756
NF	A + 160	1	5719
NF	A + 160	2	5571
NF	A + 320	1	5285
NF	A + 320	2	7984
NF1.5	ambient	1	38908
NF1.5	**ambient**	**2**	**37162**
NF1.5	A + 160	1	40188
NF1.5	A + 160	2	37452
NF1.5	A + 320	1	40957
NF1.5	A + 320	2	37196

A more important criticism of the AOT40 concept is that it does not relate ozone concentrations measured in air ('exposure') and the amount of ozone actually absorbed by leaf tissues ('dose'). In addition, no information at all is included about the ability of tissues to detoxify ozone (and/or the radicals derived from it) before damage occurs. Conceptual models of ozone exposure, ozone dose and plant injury have been developed long ago (see e.g. Guderian *et al.*, 1985, Grünhage and Jäger, 1994). A model designed to create critical levels for ozone should take into account the resistances to ozone uptake (atmospheric resistance, canopy resistance and stomatal resistance) as well as detoxification and repair processes within

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the plant tissues. All these processes (flux, uptake, metabolisation) should be expressed in reasonable units, i.e. as mass per area and time (mol m^{-2} s^{-1}). Knowledge of absorbed doses and of the processes after absorption would delete almost all of the scatter in current data sets.

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Development of visible injury on the leaves of *Prunus serotina* in Ticino, southern Switzerland, as a result of ozone exposure. Preliminary results.

J.L. Innes*, J.M. Skelly+, W. Landolt*, C. Hug*, K.R. Snyder+ and J.E. Savage+

*Swiss Federal Institute for Forest, Snow and Landscape Research, Birmensdorf, Switzerland

+Department of Plant Pathology, Pennsylvania State University, USA

Abstract

In 1993, *Prunus serotina* trees growing in southern Switzerland were found with foliar symptoms typical of ozone injury. The distribution of injury coincided with an area known to experience relatively high exposures of ozone. Consequently, seedlings and young (1 yr old) trees derived from symptomatic and asymptomatic trees were exposed to different concentrations (ambient, 90% of ambient, 50% of ambient) of ozone under controlled conditions using open-top chambers in order to test whether symptoms could be prevented by reducing ozone exposures.

The experiment indicated that the visible injury to leaves observed in the field could be reproduced under experimental conditions. On sensitive individuals, symptoms rapidly developed in ambient air, increasing in severity in line with increasing ozone dose. Seedlings and young trees grown in air containing approximately 50% of the ambient ozone concentrations did not develop symptoms until towards the end of the experiment. The development of symptoms in the filtered-air chambers corresponded with an accumulated exposure of AOT40 2.5 ppm*h, approximately one quarter of the value previously set by the UNECE to prevent damage to European trees by ozone.

Seedlings varied markedly in their symptom development. In general, a consistent response was found between sensitivity as determined by the incidence of symptoms on the parent trees in the field and sensitivity of the derived seedlings and young trees in the chambers. This, together with the results of a field survey conducted in 1994, suggests that a small proportion of the *Prunus serotina* trees in Switzerland develop visible foliar injury at the ozone concentrations encountered in 1994 and 1995. However, this sensitivity occurs at exposures considerably lower than previously realised.

Introduction

Visible injury to trees caused by air pollution has been very difficult to identify in Europe, apart from in the neighbourhoods of point sources. Visible injury is here defined as the development of symptoms on the foliage of trees. These symptoms are measurable but do not necessarily reduce yield. Damage is defined as an effect resulting in an economic loss. Injury does not necessarily imply damage, and the extent of foliar injury may be a poor predictor of growth losses (Temple and Miller 1994). An impact may consist of either injury or damage.

Although smoke injury and damage (caused by particulates, sulphur dioxide, heavy metals and other relatively localised pollutants) is not considered to be a problem in most areas today, many studies have suggested that some pollutants, particularly ozone, may reach phytotoxic

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concentrations. Maps of the cumulative exposure index AOT40 for Switzerland indicate that exceedances occur over very large areas, particularly in the important (for forests) zone at 1000 - 1500 m asl. The threshold value of AOT40 10 (ppm*h) for the April-September growing season is exceeded every year at many Swiss monitoring stations, with the highest exceedances being reported from Rigi (NABEL 1995). This has led to the expectation that widespread damage should be occurring in forests as a result of ozone.

Despite the high concentrations of ozone, visible symptoms of ozone injury have not been widely reported on forest tree species in Europe. Such symptoms are mainly believed to consist of stippling and other forms of discoloration of the adaxial surfaces of foliage and the development of flecking and necrotic tips on the needles of some conifers (Skelly et al. 1988). Although needle chlorosis of conifers has been relatively widespread in Europe, it is not generally considered that this is attributable to ozone injury, since it is reversible and clearly attributable to magnesium deficiency (Hüttl and Fink 1988). Defoliation has also been widely reported, but a link with ozone has not been established and recent evidence suggests that in some areas of the Alps, the problem is associated with nutritional deficiencies (Flückiger and Braun 1995) and water stress (Webster et al. 1996).

This does not rule out the possibility that damage, in the form of reductions in the potential growth rates, may be occurring. Such growth reductions are not inconsistent with the increases in growth that have been reported from a number of European countries, including Switzerland (Kauppi et al. 1992, Becker et al. 1995). Instead, it has been hypothesised that these growth increases might be even greater in the absence of ozone.

Laboratory studies (e.g. Matyssek et al. 1991, 1992, 1993) have indicated that visible injury may occur on some poplar and birch species in Switzerland, but the studies have generally been laboratory-based, using growth or open-top chambers. Fieldwork is required to check whether such symptoms are also present in the field.

Evidence of ozone injury to trees in Switzerland

Detailed studies undertaken in the Swiss NFP14+ programme suggested that gaseous pollutants were not adversely affecting the physiological processes studied on the two *Picea abies* investigated in the study Häsler et al. 1991, Häsler and Herzog 1991). The main tree species present in Switzerland, *Picea abies*, *Abies alba* and *Fagus sylvatica*, are believed to be relatively insensitive to ozone, except at concentrations well above those experienced in the field. Of the three, *Fagus sylvatica* is probably the most sensitive, with seedling studies using Swiss material suggesting that ozone induces reductions in biomass gains and reductions in cold tolerance

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(Braun and Flückiger 1995). Other species, such as certain *Betula* sp. and a number of introduced American species, are more sensitive. Consequently, when setting out to investigate whether ozone is affecting trees in Switzerland, a basic starting point must be the examination of those species most sensitive to ozone. If no indications of effects are found on these species, then there is little value in looking for similar effects on species which are known to be less sensitive to ozone.

The presence of observable effects is not solely dependent on the sensitivity of the species to ozone. The dose received by the trees, which is not necessarily directly related to the exposure, is also critical. In a country with major regional variations in ozone exposures, such as Switzerland, the possibilities for identifying any effects will be greatest where the most sensitive species coincide with highest exposures. In Switzerland, these criteria are met in southern Ticino, where high exposures of ozone may be affecting a number of sensitive species, including *Prunus serotina, Robinia pseudoacacia* and *Liriodendron tulipifera.* Of these, *Prunus serotina* is known as being particularly sensitive to ozone (Davis et al. 1977, Davis and Skelly 1992, Simini et al. 1992, Samuelson 1994, Neufeld et al. 1995). Within any given species, there is a range of sensitivity (c.f. Patterson and Rundel 1995), and the more sensitive individuals of an "insensitive" species may be more liable to injury by ozone than a less sensitive individual of a "sensitive" species. In the case of *Prunus serotina,* is not clear why some individuals are more sensitive than others. Bennett et al. (1992) found that the total leaf thickness and the palisade mesophyll thickness were smaller and the percentage of spongy mesophyll layer was higher for ozone-sensitive individuals, and similar observations have been made for *Betula pendula* (Pääkkönen et al. 1995). However, precisely how such differences control the sensitivity of individual plants to pollutants remains uncertain (Tingey and Andersen 1991).

In 1993, symptoms typical of ozone injury were identified on *Prunus serotina.* The symptoms were identified in southern Switzerland (Canton Ticino), an area known to experience relatively high concentrations of ozone. The symptoms consisted of red-brown stippling of the adaxial surface of sun-exposed leaves, and was extremely well-developed on symptomatic trees. At a site near Magliaso, TI, there was a range of different responses, with some trees showing no symptoms, and others showing very strong symptom development.

In 1994, a survey of the extent of these symptoms was undertaken. All documented (Ceschi 1992) sites for *Prunus serotina* in Ticino were visited, together with a number of supplementary sites. The symptoms were widespread in the southern part of Ticino. At the same time, seedlings grown from the seed of the symptomatic and asymptomatic trees identified in 1993 near Magliaso, TI, were exposed to known concentrations of ozone in the continuously stirred tank reactor facility at Pennsylvania State University, University Park, Pennsylvania, USA.

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Seedlings from the symptomatic trees developed the same symptoms when exposed to ozone, but showed no such symptoms when grown in clean air. The extent and severity of symptoms increased with increasing exposure time and increasing ozone concentration. The sensitivity of seedlings obtained from one of the most sensitive trees in Ticino was similar to that of the highly sensitive R12 type used in experiments at Pennsylvania State University and originating from central Pennsylvania. These results all suggested the need for a more detailed examination of the potential role of ozone in causing the stippling of foliage of *Prunus serotina* in southern Switzerland. In particular, it was felt that it would be useful if it could be tested whether the symptoms could be reproduced under near ambient conditions in Ticino, and whether the symptoms could be reduced or prevented by removing a proportion of ozone from the ambient air.

This work was extended in 1995 under the auspices of the Swiss Forest Invstigation Programme, in a joint project financed by the Swiss Forest Damage Inventory at Federal Institute for Forest, Snow and Landscape Research, the FLAM sub-programme of the Federal Forest Administration and the Forest Administration of the Canton of Ticino.

Experimental design

Four chambers were installed at the Lattecaldo Forest Nursery (600m a.s.l., north-west aspect) in southern Ticino.Two chambers were equipped with activated charcoal filters and two received unfiltered ambient air. In addition, two open plots were established within the randomized design. Each of the six plots were planted with 24 seedlings and 8 young trees. Each ("Heagle" design - Heagle et al. 1973) chamber contained 24 *Prunus serotina* (16 seedlings and eight 1-yr old trees), 4 *Fagus sylvatica* and 4 *Fraxinus excelsior* young trees. The *Prunus serotina* consisted of provenances already tested at Pennsylvania State University in 1994 and new collections obtained during the 1994 field survey throughout Ticino and included material obtained from both symptomatic and asymptomatic trees.

Assessment period and methods

Assessments of symptom development began on July 31, 1995 and finished on 18 September 1995. Visual assessments of each plant were made by the same observer at weekly intervals, with the last assessment taking place on September 18, 1995. Foliar injury symptoms were expressed as the percentage of adaxial stipple on all leaves of individual trees. The scoring was based on the Horsfall-Barratt system developed for visible injury (Horsfall and Barratt 1945) and given in the ForestHealth Expert Advisory System (Nash et al. 1992). Prior to the start of

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each scoring session, a calibration exercise was undertaken by the observer using the ForestHealth programme.

Ozone exposure characteristics

Ozone was monitored continuously throughout the experiment. Sampling points were positioned ca. 1 m above the ground in the centres of each of the chambers and in the centre of one of the open plots. Cumulative values (AOTs) were used to characterize ozone concentrations, together with the frequency of hours above specific thresholds. In all cases, all ozone values, regardless of time of day, were used in the calculation of the AOT values. The AOT values refer to the cumulative exposures experienced by the plants after being set out in the experiment. Each chamber showed a slightly different set of ozone exposures. Ozone exposures in the two filtered plots were approximately 50% of those in the unfiltered plots. In turn, the values in the unfiltered plots were only about 90% of those in the open-air plots.

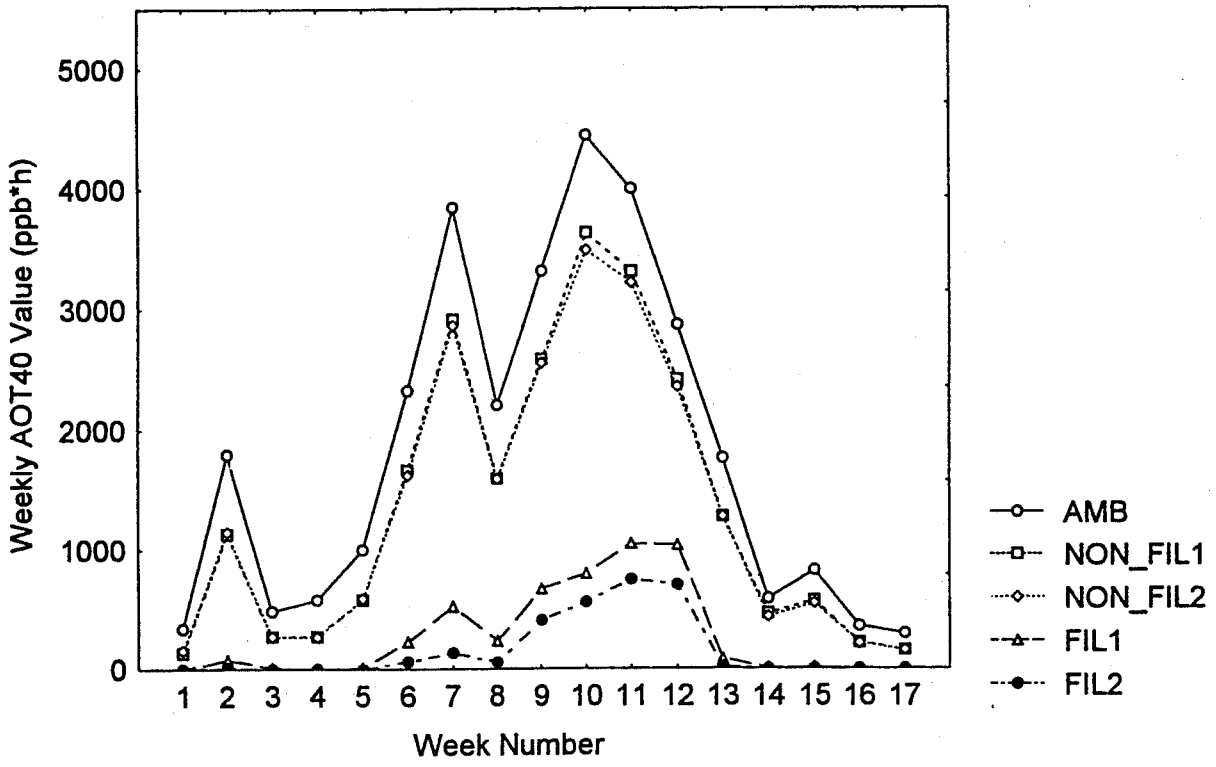


Figure 1. Weekly AOT40 exposures in the ambient, unfiltered and filtered plots.Week numbers correspond to the weeks after the start of filtration, with week 1 being the exposure for the week ending on 21 May 1995.

The differences in exposure between the filtered and unfiltered chambers were marked, particularly for higher AOT values (Figure 1). In the filtered chamber, the cumulative AOT 40 value reached ca. 2.7 ppm*h, whereas in the open plot, the AOT40 value reached ca. 32

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ppm*h. Maximum hourly concentrations during the summer of 1995 were not exceptional. In the open plot, and no hourly mean concentration in excess of 0.16 ppm was recorded within the open plot (ambient exposures).

Plant responses

The typical reddish-brown stipple on the adaxial surface of sun-exposed leaves that has been widely associated with ozone injury developed on the seedlings and young trees. The symptoms first developed in ambient air, and increased as the cumulative exposure increased. Seedlings grown in air containing approximately 50% of the ambient ozone concentrations did not develop symptoms until towards the end of the experiment. The development of symptoms in the filtered air chambers corresponded with an accumulated dose of AOT40 2.5 ppm*h, approximately one quarter of the value previously set to protect trees in Europe from damage by ozone (UNECE 1994). However, it is important to remember that seedlings of *Prunus serotina* have higher stomatal uptake rates than mature trees (Frederiksen et al. 1995) and injury may occur sooner on the leaves of seedlings than those of mature trees.

The development of symptoms on the oldest leaves first is interesting as it suggests that the seedlings were responding to long-term rather than short-term exposures. Indeed, the highest concentrations experienced during the summer did not induce an increase in the occurrence of symptoms the following week, as might be expected if the plants were responding to ozone episodes. However, this remains an area of uncertainty, and more detailed observations (daily assessment of each leaf on each plant) would be required if the problem were to be fully resolved. Other studies have also reported greater effects in older foliage (e.g. Pell and Dann 1991) and the phenomenon does not appear to be restricted to *Prunus serotina*. In addition, morphological assessments are required to check whether there are differences between leaves produced early in the season and those produced later, as such differences could affect the senstivity of the leaves to ozone (e.g., Bennett et al. 1992).

Conclusions

Until now, there have only been tentative identifications of ozone injury on free-grown forest trees in Europe. These are based on the observation of symptoms similar to those attributed to ozone on other species or in other countries. Most experience has come from experimental studies using open-top chambers which, by their nature, modify the environmental conditions. In this study, a number of steps were taken to demonstrate cause-effect mechanisms between ozone and the injury observed on *Prunus serotina*.

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1. The foliar injury was first identified on species known to be sensitive to ozone injury. The investigation was concentrated in an area known to experience relatively high ozone exposures. Injury similar to that observed in experimental studies was identified on trees growing in the forest, on forest margins or in open situations.
2. The type of symptoms that were observed, specifically the stippling of the adaxial surfaces of leaves, are known to be induced under conditions of ambient ozone exposures.
3. The symptoms seen in the field were reproduced under controlled conditions. The extent of the symptoms increased with increasing exposure to ozone. These findings were obtained first under strictly controlled conditions in continuously-stirred tank reactors located at Pennsylvania State University and secondly in the open-top chambers in Ticino.
4. The responses of sensitive and insensitive trees were consistent in the field and in the experiment.

Taken together, these findings provide convincing evidence that the stippling observed on *Prunus serotina* in various parts of southern Ticino is attributable to ozone exposures. The steps taken here are similar to those taken for the attribution of ozone as the cause of "weather fleck" in tobacco (Macdowall et al. 1964) and chlorotic flecking in *Pinus ponderosa* (Miller et al. 1963), both of which studies are now widely accepted. The ozone exposure at which the injury was observed suggests that some individuals of *Prunus serotina* are extremely sensitive to ozone, with symptoms developing at an exposure of AOT40 2.5 ppm*h. However, this introduced species is fairly restricted in its distribution, and the results cannot be extrapolated to other species or other areas within Switzerland.

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GROWTH STAGE AS A MODIFIER OF OZONE RESPONSE IN WINTER WHEAT

Gerhard Soja

Austrian Research Centre Seibersdorf, Division Life Sciences, A-2444 Seibersdorf, Austria

Abstract. This paper reports the results of two experiments quantifying the effect of growth stage on the ozone sensitivity of winter wheat.

Vernalized winter wheat was cultivated in plant containers and exposed to two ozone levels in greenhouse compartments adapted as fumigation chambers. Ozone levels were kept constant at 80±6 $nl.l^{-1}$ for 8 $h.d^{-1}$. A set of eight different ozone exposure treatments was achieved by alternatingly exposing the plants to elevated ozone or to control conditions at certain stages of development: tillering, shooting till ear emergence and ear emergence till three weeks after anthesis. Fumigation started one month after emergence and lasted for three months. The maximum ozone treatment was exposed to an AOT40 of 29.9 ppm-h, the other treatments to correspondingly less.

Grain yield, straw yield and grain yield components (thousand grain weight and number of grains per ear) were analysed for yield reductions relative to non-fumigated control plants. For grain yield, the weights of the three monthly ozone AOT40 (second month before ear emergence, one month before ear emergence and one month after ear emergence) were related like 0.27 : 0.72 : 1. This result indicates the importance of giving a higher weight to ozone exposure after ear emergence for adequately describing the actual ozone-induced yield reductions. Additionally, non-linear functions with two variables (ozone dose before and after ear emergence) were established for each yield parameter.

From these results it is suggested to re-assess the results from other fumigation experiments, validate or modify the proposed model and to use it for fine-tuning the current critical level for ozone.

1. Introduction

Wheat is known to be one of the most ozone-sensitive agricultural crops. It has been shown that ambient ozone concentrations during the vegetation period are high enough to cause significant yield reductions (Heagle *et al.*, 1979; Kress *et al.*, 1985; Kohut *et al.*, 1987; Fuhrer *et al.*, 1989, 1992; Pleijel *et al.*, 1991; De Temmermann *et al.*, 1992; Fangmeier *et al.*, 1994). These and other results of the European Open-Top Chamber Programme have resulted in the establishment of a critical level for ozone, based on AOT40 (accumulated exposure over threshold of 40 $nl.l^{-1}$). This ozone dose (5.3 ppm-h for a yield reduction of 10 %) is calculated for a growth period of three months, giving equal weight to the AOT40 for all days of the observation period (Fuhrer and Achermann, 1994).

On the other hand, there is some evidence that ozone exposure has not the same effectiveness during the different stages of plant development (Lee *et al.*, 1988; Younglove *et al.*, 1994). Therefore growth stage is considered, among others, as a modifying factor for ozone injury. Consequently research is needed to quantify these modifying factors. These results shall contribute to a more detailled definition of Critical Levels, differentiated according to environmental parameters. This study wants to contribute to these efforts by quantifying the effect of growth stage on ozone-induced yield reduction of wheat.

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2. Material and Methods

Seeds of winter wheat (*Triticum aestivum* cv. Perlo) were germinated at +20 °C for 24 h and immediately afterwards vernalized for 60 days at +1 °C. These vernalized seedlings were planted in pots with 12 l of a standard soil mix (Frux ED 73). Two weeks after emergence, the seedlings were thinned to 15 plants per pot. For calculation of the statistics, one pot was treated as one replicate.

The pots were kept in environmentally controlled greenhouse compartments adapted as fumigation chambers. The temperature was held at 15/10 (±1) °C during the first three weeks after emergence, afterwards at 20/12 °C (day/night). Plants were irrigated as necessary, drought stress was avoided. For protection against powdery mildew two sprays with Vigil (Diclobutrazol, 0.1 %) and against aphids two sprays with Decis (Deltamethrin, 0.05%) were necessary. All plants were treated uniformly.

The experiments were performed in 1994 and 1995, with the experimental periods preceding the natural growth periods by two months. This had the advantage of low ozone concentrations for the control without the need of air filtration, resulting in low ozone levels comparable to carbon-filtered open-top chambers.

Fumigation started one month after emergence (stage 21) and lasted for three months (stage 71). Ozone was generated from pure oxygen by silent discharge (Fischer 502, Meckenheim Germany). Ozone concentrations in the fumigation compartments were held constant at 80 $nl.l^{-1}$ (± 6 $nl.l^{-1}$).daily from 9 a.m. to 5 p.m. with a continous ozone analyzer (Horiba APOA 350E; Horiba Europe, Langenfeld, Germany) controlling the output of the ozone generator. The Horiba instrument also measured the ozone levels in control chambers and ambient air at hourly intervals using an electronically controlled sample manifold. Control plants accumulated an AOT40 of 0.4 ppm-h till the end of the fumigation period. Ozone concentrations in the control chamber increased from 15 $nl.l^{-1}$ before to 30 $nl.l^{-1}$ (± 10 $nl.l^{-1}$) after anthesis.

For exposing the plants to different ozone doses during defined stages of development, the pots of the individual treatments were alternatingly exposed in the fumigated or in the control chamber. Total fumigation lasted for 12 weeks. Moving from the control to the fumigation chamber or vice versa occurred every four weeks (only in 1994 the third exposure period lasted for five weeks), resulting in different treatments with different ozone doses as shown in table 1. The treatment with 50 % of the maximum ozone dose was moved every two weeks. The three exposure periods coincided with defined stages of development of the wheat plants: tillering, shooting till ear emergence and ear emergence till three weeks after anthesis.

Statistical analyses was based on 8 replicates per treatment (= 8 containers). ANOVA, Duncan-test and multiple linear regressions were calculated with WinSTAT software (G. Greulich Software, Staufen, Germany), surface-fitting was performed with TableCurve 3D (Jandel Scientific, Erkrath, Germany).

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Table 1: Effect of ozone fumigation during different stages of plant development on wheat dry matter production and yield parameters. Numbers in columns (separately for each year) followed by different letters are significantly different at α=0.05 (Duncan's Multiple Range-Test).

ozone AOT40 (in ppm-h)			yield and yield parameters			
second month before ear emergence	one month before ear emergence	one month after ear emergence	relative grain yield (100 = control)	relative straw yield (100 = control)	relative thousand grain weight (100 = control)	relative number of grains per ear (100 = control)
experiment 1994:		ozone data	experiment 1994:		yield data	
9.545	0	0	99.0 a	100.0 a	100.6 a	95.0 ab
0	8.887	0	89.8 ab	101.8 a	87.5 ab	90.3 abc
0	0	11.427	67.2 bc	80.1 b	70.3 b	74.7 bcd
9.545	8.887	0	78.0 b	89.6 ab	89.9 ab	87.5 abcd
9.545	0	11.427	66.6 bc	80.1 b	82.3 ab	60.7 cd
4.773	4.444	5.714	73.4 bc	80.4 b	79.2 ab	60.8 cd
9.545	8.887	11.427	44.6 c	67.7 c	66 b	57.9 d
0	0	0.400	100.0 a	100.0 a	100.0 a	100.0 a
experiment 1995:		ozone data	experiment 1995:		yield data	
9.800	0	0	103.9 a	105.4 a	93.3 b	105.5 a
0	7.777	0	90.5 bc	99.8 a	92.3 bc	96.7 ab
0	0	9.566	70.6 de	83.5 b	88.1 bc	75.6 c
9.800	7.777	0	61.2 e	72.8 c	89.7 bc	76.3 c
9.800	0	9.566	59.9 e	73.4 c	86.3 c	79.1 c
4.900	3.889	4.783	79.8 cd	87.2 b	91.5 bc	88.3 b
0	0	0.400	100.0 ab	100.0 a	100.0 a	100.0 ab
			(g per pot)	(g per pot)	(g)	($^{-1}$)
absolute data of control in 1994: (mean ± s.d.)			41.7 ± 6.3	89.4 ± 7.0	37.2 ± 3.3	32.2 ± 3.7
absolute data of control in 1995: (mean ± s.d.)			57.7 ± 5.7	152.6 ± 11.4	41.2 ± 1.1	40.2 ± 2.1

3. Results and Discussion

The experiments in 1994 and 1995 were designed to expose the plants to similar ozone concentrations in both years, yielding comparable results for both experimental years. Maximum ozone concentrations did not exceed ambient ozone levels during periods of severe photooxidative stress. Total ozone dose for the treatment with the highest accumulated dose (table 1) was about two times higher than the average AOT40 during the

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months May to July (14.6 ppm-h for the period 1992-1995 at Seibersdorf/Austria; data not shown).

The results in table 1 show that there existed large differences between the treatments. The same ozone dose could reduce wheat productivity to an extremely different extent, depending on timing of ozone exposure. An ozone AOT40 of 7.8-11.4 ppm-h was able to increase yield slightly (not significantly) by 0-5 % or to reduce yield by up to 33 %. According to the critical level of this dose should reduce wheat grain yield by 14-21 %. Our results show that an ozone dose was less effective if the plants were exposed to it during early developmental stages. It was even completely ineffective to reduce grain yield if the plants were exposed to the elevated ozone concentrations only before shooting stage (during the second month before ear emergence). A similar ozone dose only during shooting stage (one month before ear emergence) caused a yield reduction of about 10 %, whereas after ear emergence the yield reduction increased to about 30 %.

Combinations of ozone exposures during different developmental stages usually increased the severeness of yield reduction. Consequently, the most pronounced effects were caused by continously high ozone levels from two months before till one month after ear emergence. In the average of both years, the effect of the ozone dose of the two months before ear emergence roughly equalled the effect of the ozone dose one month after ear emergence; but in this respect the two experimental years showed controversial trends. Plants receiving 50 % of the maximum ozone dose for all three months (14.9 ppm-h) exhibited about the half yield reduction of plants exposed to the full ozone dose. This indicates that on the basis of the accumulated AOT40 during a whole growth period a linear reduction of grain yield will appear when the plants experience the same distribution of ozone doses during their development. This confirms the results of the European Open Top Chamber Programme which found a linear exposure-response relationship for wheat when exposure was expressed as AOT40 (Fuhrer and Achermann 1994). But in the case of a non-uniform distribution of ozone dose during plant development, a correction for developmental stage would be necessary.

Both yield components were significantly affected by ozone exposure, but the reduction of the number of grains per ear was slightly higher than the reduction of thousand grain weight (table 1). Fuhrer *et al.* (1992), however, found higher reductions in thousand grain weight; this difference might be due to the use of a spring wheat cultivar instead of winter wheat and due to different cultivation and exposure techniques.

Straw yield was considerably less reduced than grain yield. Only AOT40 of >17 ppm-h before ear emergence or ≥10 ppm-h after ear emergence produced straw yield reductions of >10%. The higher sensitivity of grain compared to straw productivity has also been observed by Fuhrer *et al.* (1989, 1992), Pleijel *et al.* (1991) and Soja & Soja (1996). These differences reflect the importance of a high green leaf area, of unimpaired photosynthetic capacity and of abundant carbohydrate reserves during the period of grain filling. If the plants are injured by ozone exposure, these conditions are only partly fulfilled and consequently grain development is more inhibited than the previous growth of the vegetative plant parts. A decreased accumulation and export of assimilates under ozone stress has also been observed by Balaguer *et al.* (1995), although Mortensen & Engvild (1995) did not find reduced translocation velocity.

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The different importances of the ozone doses during certain growth stages is even more evident when the relative yield is expressed as a multiple linear function of the ozone doses during these stages. In table 2 the equations are given for calculating the relative grain yield, straw yield and the grain yield components. Both experimental years were combined for this analysis. The regression coefficients show that the relative yield was mainly determined by the ozone dose after ear emergence. Comparing the three growth stages tillering, shooting and post-shooting, the ozone dose during the last month of these stages determined 50-60 % of the yield reduction. Ozone dose during shooting stage determined 30-40 % of the yield reduction, whereas ozone exposure before shooting contributed only 10-20 % to the final yield reduction. Thousand grain weight was an exception: this yield parameter was not influenced by the ozone dose during the earliest growth stages; the weights of the ozone doses before and after ear emergence were related like 2:3. This analysis shows that the number of grains per ear was slightly (though not significantly) influenced by early ozone exposure, whereas the thousand grain weight did not show any after-effect of early ozone exposure. This means that the translocation of assimilates to the developing grain was mainly determined by the extent of disposable carbohydrates after flowering (Slaughter *et al.* 1993). However, the pollination of a single earlet flower could also be disturbed by earlier ozone exposures.

Figures 1-4 demonstrate the dependence of ozone-induced yield reduction on the timing of ozone exposure graphically. The relative yield is shown as a function of two variables: ozone dose before (x) and ozone dose after (y) ear emergence. Variable x is composed of the ozone dose during one month before ear emergence (M-1) and a fraction of the ozone dose in the second month before ear emergence (M-2). According to the relative importance of early ozone exposure, this fraction is smallest for thousand grain weight (fig. 3) and highest for straw yield (fig. 2). The graphs also show that grain yield is the most ozone-sensitive productivity parameter tested in these experiments (fig. 1). The form of the graph indicates that the effectiveness of ozone exposure before ear emergence was small at low ozone doses but increased exponentially with higher doses. Ozone exposure after ear emergence, however, influenced grain yield in a linear way. For straw yield (fig. 2), ozone exposure after ear emergence affected productivity even less than linear. This can be explained by the growth pattern of wheat: in the generative stage of development the allocation of assimilates to the grain becomes more important than leaf and stem growth. Similar differences could be found in the grain yield components: thousand grain weight that is mainly determined by assimilate translocation after flowering was exponentially influenced by ozone dose after ear emergence (fig. 3). The number of grains per ear, however, were linearly co-influenced by ozone dose before ear emergence (fig. 4).

The results largely agree with the findings of Pleijel et al. (this volume) who observed increased ozone sensitivity after flowering. Our model somewhat underestimates the yield reductions observed in their experiment but the difference is still within the standard error of the model. A possible cause for the underestimations is the different experimental approach: pot experiments (as in our case) may yield dose-response-relationships with decreased ozone sensitivities compared to experiments with soil-grown plants (Skärby et al., 1993).

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Table 2: Dependence of wheat productivity and yield parameters on ozone AOT40 during two months before and one month after ear emergence. Multiple linear regression equations are given; y = relative yield, M_{-2} = AOT40 in ppm-h in the second month before ear emergence, M_{-1} = AOT40 in ppm-h one month before ear emergence, M_{+1} = AOT40 in ppm-h one month after ear emergence.

parameter	relative yield =	r^2	adj. r^2	std.err.
grain yield	$104.1 - 0.833\ M_{-2} - 2.22\ M_{-1} - 3.08\ M_{+1}$	0.874	0.840	7.14
straw yield	$104.1 - 0.745\ M_{-2} - 1.07\ M_{-1} - 1.98\ M_{+1}$	0.762	0.697	6.78
thousand grain weight	$99.9 + 0.0089\ M_{-2} - 1.28\ M_{-1} - 1.81\ M_{+1}$	0.831	0.785	4.67
number of grains per ear	$102.5 - 0.526\ M_{-2} - 1.50\ M_{-1} - 2.68\ M_{+1}$	0.796	0.740	7.85

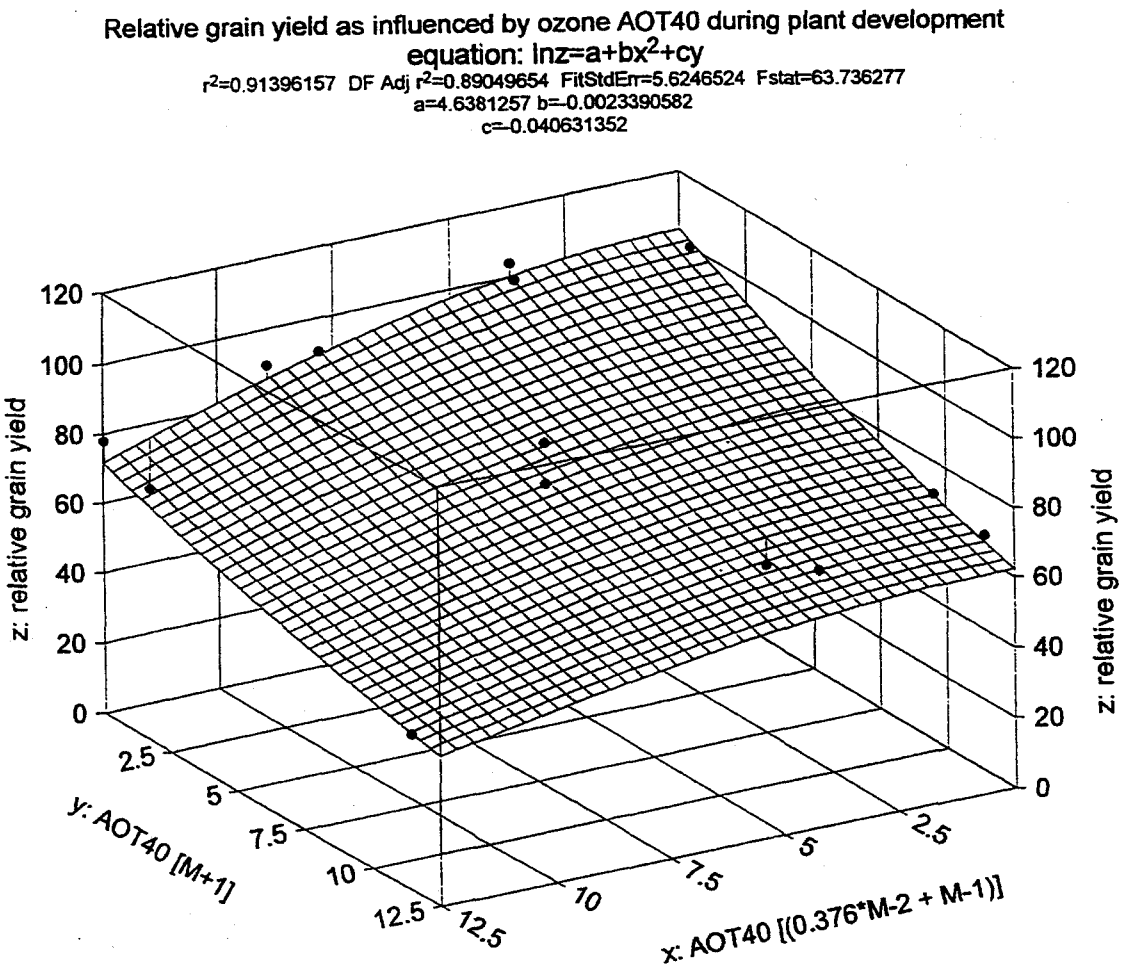


Figure 1: Relative wheat grain yield as influenced by different ozone doses during plant development. X-axis: sum of AOT40 (in ppm-h) of the first (M-1) and the second month (M-2) before ear emergence. For M-1 the multiplication factor 1, for M-2 the factor 0.376 has to be used; y-axis: AOT40 during one month after ear emergence (M+1); z-axis: grain yield of winter wheat relative to non-fumigated control

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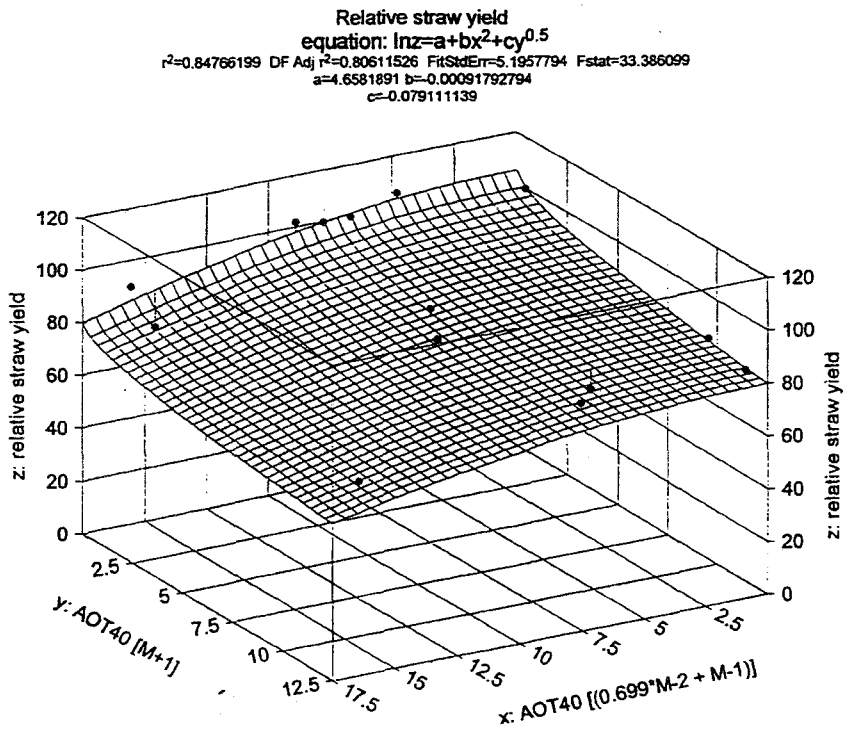


Figure 2: Relative wheat straw yield as influenced by different ozone doses during plant development. X-axis: sum of AOT40 (in ppm-h) of the first (M-1) and the second month (M-2) before ear emergence. For M-1 the multiplication factor 1, for M-2 the factor 0.699 has to be used; y-axis: AOT40 during one month after ear emergence (M+1); z-axis: straw yield of winter wheat relative to non-fumigated control

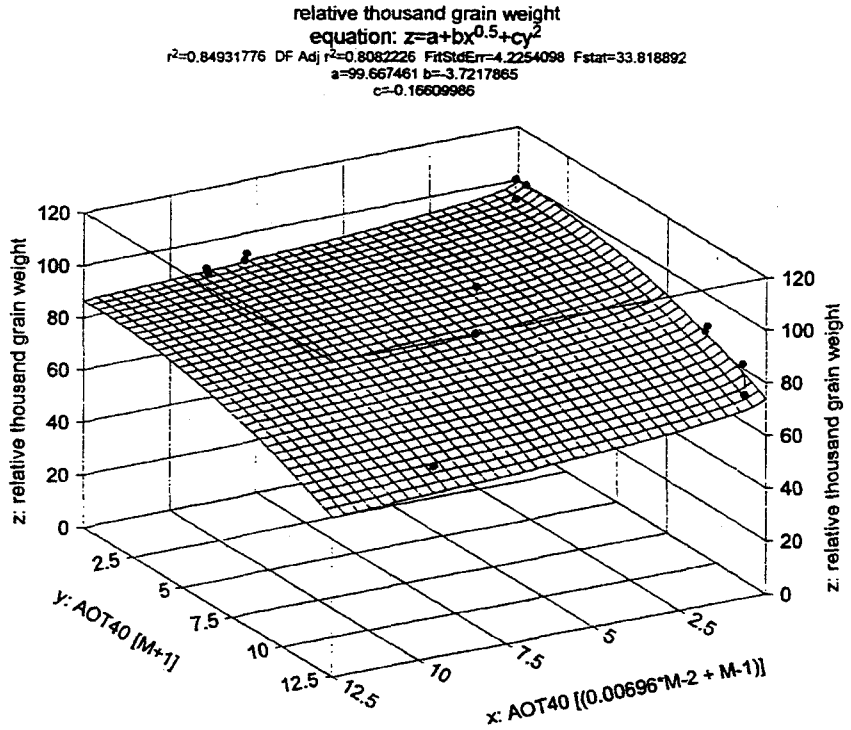


Figure 3: Relative thousand grain weight of wheat as influenced by different ozone doses during plant development. X-axis: sum of AOT40 (in ppm-h) of the first (M-1) and the second month (M-2) before ear emergence. For M-1 the multiplication factor 1, for M-2 the factor 0.00696 has to be used; y-axis: AOT40 during one month after ear emergence (M+1); z-axis: thousand grain weight of winter wheat relative to non-fumigated control.

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4. Conclusions

- Ozone exposure after ear emergence is more effective in reducing wheat productivity than ozon exposure before ear emergence.
- The resulting model will require further validation with soil-grown wheat under ambient conditions.
- One of these validation exercises could be the re-assessment of results of the European Open Top Chamber Programme that was the basis for the current critical level of ozone.

5. Acknowledgements

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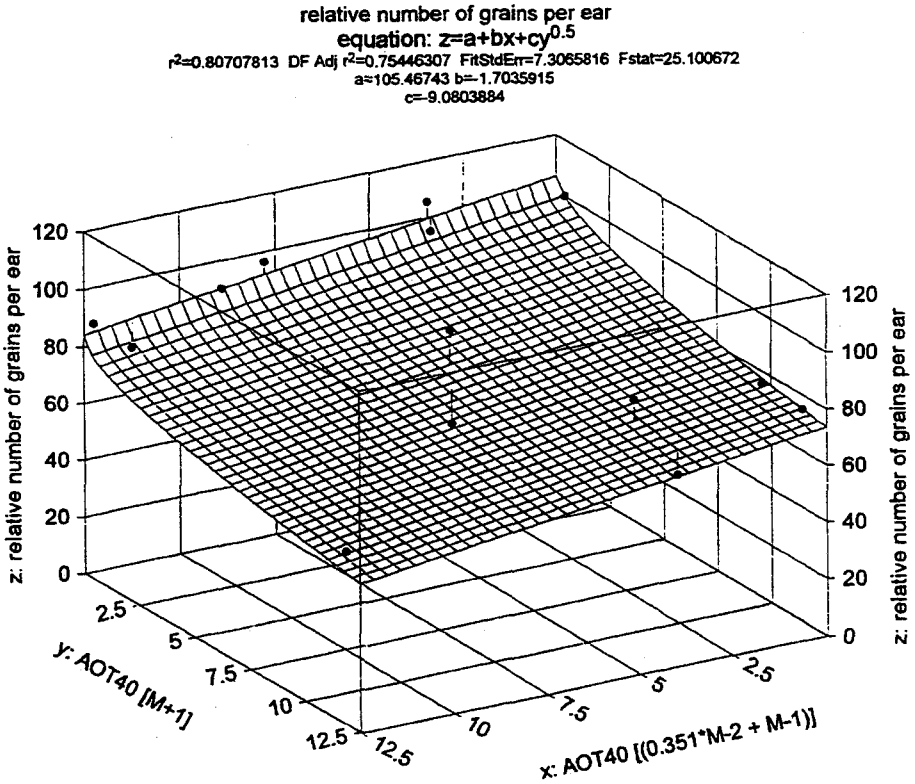


Figure 4: Relative number of grains per ear as influenced by different ozone doses during plant development. X-axis: sum of AOT40 (in ppm-h) of the first (M-1) and the second month (M-2) before ear emergence. For M-1 the multiplication factor 1, for M-2 the factor 0.351 has to be used; y-axis: AOT40 during one month after ear emergence (M+1); z-axis: number of grains per ear of winter wheat relative to non-fumigated control.

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GROWTH STAGE DEPENDENCE OF THE GRAIN YIELD RESPONSE TO OZONE IN SPRING WHEAT

H. Pleijel [1], H. Danielsson [1], J. Gelang [2], E. Sild [2] and G. Selldén [2]

[1] Swedish Environmental Research Institute (IVL), P. O. Box 47086,
S-402 58 Göteborg, Sweden
[2] Botanical Institute, Carl Skottsbergs Gata 22,
S-413 19 Göteborg, Sweden

Abstract: Field grown spring wheat was exposed to three different ozone treatments in open-top chambers: *NF* (non-filtered air), *NF+pre* (non-filtered air with 2 500 ppb-hours above 40 ppb (AOT40) during 38 days before anthesis) and *NF+post* (non-filtered air with 2 500 ppb-hours ozone above 40 ppb 26 days during and after anthesis). The pre-anthesis treatment period had an AOT40 in *NF+post* of 195 ppb-hours and the post-anthesis period an AOT40 in *NF+pre* of 86 ppb-hours. During the establishment phase of the plants, before the *NF+pre* started, the plants received an AOT40 of approximately 500 ppb-hours and after the end of the *NF+post* treatment the AOT40 was around 250 ppb-hours. The respective treatment periods in *NF+pre* and *NF+post* had long-term, 12-hour daytime (08.00-20.00), averages of ozone concentration between 40 and 45 ppb, while it was 20 to 25 ppb in the NF treatment.
The grain yield in *NF+post* was 11% lower than in *NF* and was statistically significant. The difference in grain yield between *NF+pre* and *NF* was small, a 2% positive treatment effect, and not statistically significant. The protein content of the grain was significantly higher in the *NF+post* treatment, 15.0%, compared to 13.7% in the *NF* treatment and 13.9% in the *NF+pre* treatment.
It is concluded that ozone exposure is much more effective in decreasing the grain yield of wheat between the onset of anthesis and end of grain filling than before anthesis. The increase in the protein content of the grain, which has been associated with ozone effects on the grain yield of wheat in a number of studies, was found in the present study only in the *NF+post* treatment. This confirms the conclusion that the post-anthesis period is more sensitive to ozone exposure in wheat than the pre-anthesis period.

1. Introduction

There is today abundant evidence that present ambient ozone concentrations are high enough to cause significant yield loss in wheat in the USA (Kohut *et al.*, 1987) and in Europe (De Temmermann, 1992; Fuhrer *et al.*, 1989, 1992; Pleijel *et al.*, 1991, Skärby *et al.*, 1993). Although pooled data from a number of studies on spring wheat show a fairly consistent response pattern and can be related to the exposure index AOT40 (Accumulated Exposure over Threshold 40 nl l^{-1} ozone) with high correlation (Fuhrer & Achermann, 1994), a variation between different experiments in the relative yield loss caused by a certain AOT40, remains. One possible reason for that could be that ozone exposure was performed only after anthesis in some experiments (Fuhrer & Ashermann, 1984), and both before and after anthesis in others.

It has been suggested by for example Lee *et al.* (1988), that the sensitivity to ozone by seed crops is maximal during a period between flowering and seed maturity, while the sensitivity is lower before and after that period. It would then be feasible to hypothesise that a certain AOT40 would produce a larger effect given mainly during the period of maximum sensitivity than if given over the whole growing period of the crop.

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The aim of the present study was to determine the effect of ozone, as a certain number of ppb hours above 40 ppb (AOT40), given either before or during and after anthesis (flowering) of a field grown spring wheat crop.

2. Experimental

The experiment was conducted in a wheat field at Östad, 50 km north-east of Göteborg, Sweden (N 57°54', E12°24'). No major air pollutant sources are located in the vicinity of the experimental site. A spring wheat cultivar, *Triticum aestivum* L., cv. Dragon, was sown with 12.5 cm row spacing (240 kg seeds ha^{-1}). Further details on agricultural practices are presented in Table 1.

Table 1. Timetable of events in 1995 ESPACE experiment for the cultivar Dragon

Event	Time
Sowing	950503
NPK fertilisation	950503
Emergence	950513
Herbicide, Pirimor	950728
Installation of chambers	950524
Start of irrigation	950524
Frequency and amount of irrigation	every other day, 10 mm
Start of ozone exposure, *NF+ pre*	950531
End of ozone exposure, *NF+ pre*	950709
Start of ozone exposure, *NF+ post*	950709
End of ozone exposure, *NF+ post*	950803
Removal of chambers and harvest	950830

The plants were exposed to three different ozone regimes in open-top chambers (OTC) (n=5): *NF* (non-filtered air), *NF+pre* (non-filtered air with 2 500 ppb hours above 40 ppb (AOT40) during 38 days before anthesis) and *NF+post* (non-filtered air with 2 500 ppb hours ozone above 40 ppb 26 days during and after anthesis). The OTCs were 1.24 m in diameter and 1.60 m tall, including frusta. Air was continuously blown and distributed via a manifold placed 0.1 m above the canopy at a rate of about 10 m^3 min^{-1}. The chambers were equipped with roofs. For controlling the chamber effect the experimental design also included ambient air plots (n=5). Because of problems with weeds, three plots were excluded. This reduced the replicate numbers to 4 in both the *NF+pre*- and the *NF+post* - treatment and also in the AA-treatment. The AOT40 was chosen to enable a direct test of the critical level concept suggested in Fuhrer & Achermann (1994). The AOT40s of the different periods are presented in Figure 1. During the pre-anthesis treatment period the AOT40 in *NF+post* was 195 ppb hours and during the post-anthesis period the AOT40 in *NF+pre* was 86 ppb hours. During the establishment phase of the crop, before the *NF+pre* started, the plants received an AOT40 of approximately 500 ppb hours and after the end of the *NF+post* treatment the AOT40 was around 250 ppb hours. During the respective treatment periods in *NF+pre* and *NF+post* the 12 hour daytime (08.00-20.00, local time) long-term average of the ozone concentration was between 40 and 45 ppb, while it was 20 to 25 ppb in the *NF* treatment. Starting the 24:th of may and throughout the experiment all plots were irrigated every other day with a water volume corresponding to 10 mm precipitation each time. The air temperature and the relative humidity inside one chamber

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and also in one ambient plot, was measured with a Rotronic YA-100 hygrometer/thermometer at 1.1 m above the ground.

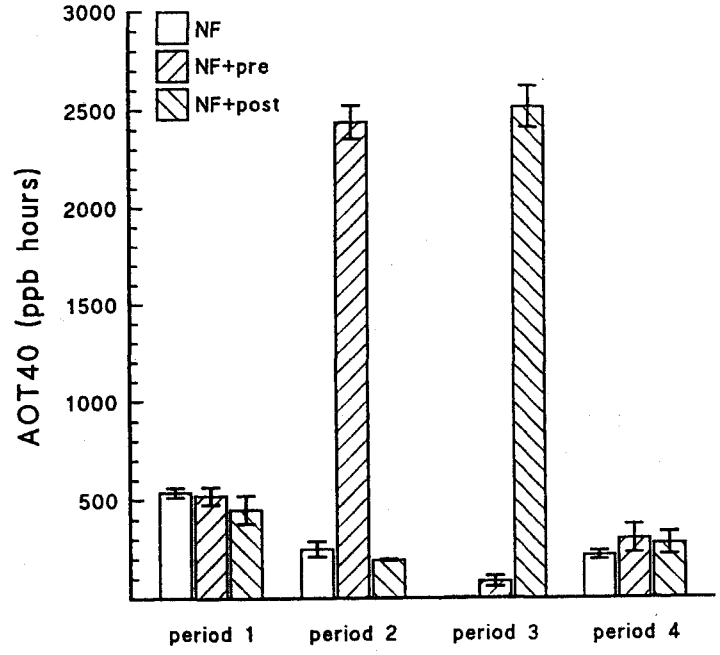


Figure 1. Ozone exposure as AOT40 for the four periods: 1. Establishment, 2. Pre-anthesis exposure period, 3. Post-anthesis exposure period, 4. The period from end of grain filling to maturation. The error bars represent ± standard error.

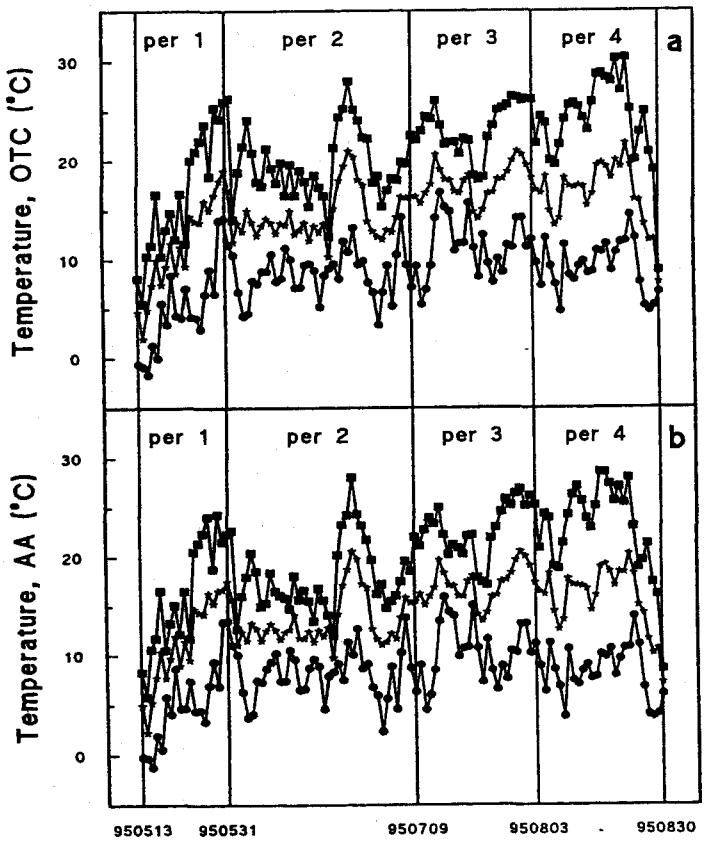


Figure 2a and 2b. The min (•), max (▪) and mean (✶) temperature during all four periods inside open-top chamber (2a) and in the ambient air (2b).

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3. Results and discussion

On average the daily maximum, mean and minimum temperature was slightly higher inside the chamber than in the ambient air, especially in June (Figure 2 a,b). The seasonal mean temperature was 0.7°C higher inside the chamber than in the ambient air (Table 2). During most days the relative humidity was slightly lower inside the chamber than in the ambient air. The situation was the opposite during the nights. The differences between the chamber and the ambient air were larger during the nights than during the days.

Table 2. Seasonal mean temperature inside one chamber and in the ambient air

Temperature °C	Chamber	AA
Minimum	8.7	8.2
Maximum	21.0	19.9
Mean	15.2	14.5

The grain yield (Figure 3) in *NF+post* was 11% lower than in *NF* and this difference was statistically significant. The difference in grain yield between *NF+pre* and *NF* was small, a 2% positive treatment effect, and not statistically significant. The protein content (Figure 3) of the grain was significantly higher in the *NF+post* treatment, 15.0% compared to 13.7% in the *NF* treatment and 13.9% in the *NF+pre* treatment.

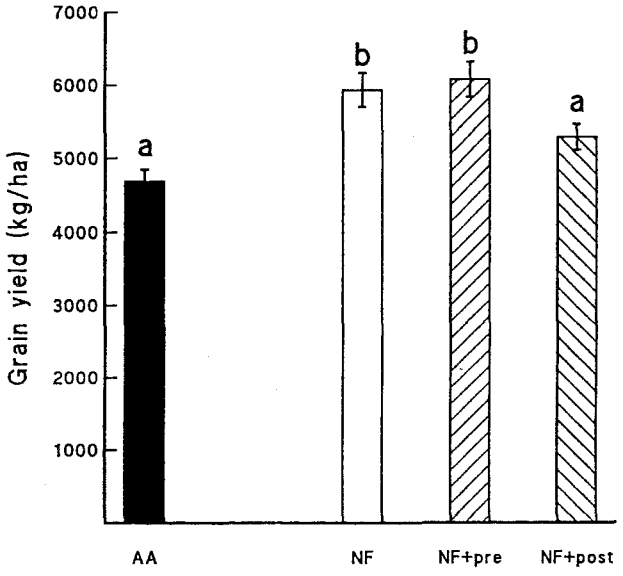


Figure 3. Grain yield (dry weight) of spring wheat exposed to ambient air (*AA*), non-filtered air (*NF*), non-filtered air plus extra ozone added either before (*NF+pre*) or after (*NF+post*) anthesis. Error bars show standard error. Significances refer to ANOVA and LSD-test at the 95% level.

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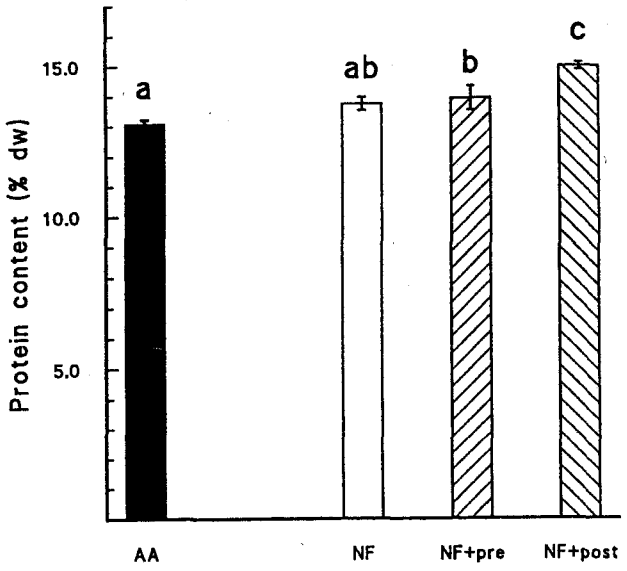


Figure 4. Protein content (Kjeldahl) of the grain of spring wheat exposed to ambient air (*AA*), to non-filtered air (*NF*) or non-filtered air plus extra ozone added either before (*NF+pre*) or after (*NF+post*) anthesis. Error bars show standard error. Significances refer to ANOVA and LSD-test at the 95% level.

Towards the end of the grain filling period, the green colour of the flag leaves decreased much earlier in the *NF+post* treatment compared to the *NF* and *NF+pre* treatments. This is reflected in Figure 5, where the Green Leaf Area Index (LAI) from anthesis until short before end of grain filling is shown. Thus, the premature senescence symptoms frequently associated with ozone injury in wheat (Grandjean & Fuhrer, 1989; Ojanperä *et al.*, 1992) was observed only for the *NF+post* treatment. As discussed in Pleijel *et al.* (1995) the shortening of the green leaf area duration is likely to account for the loss of above-ground biomass by ozone.

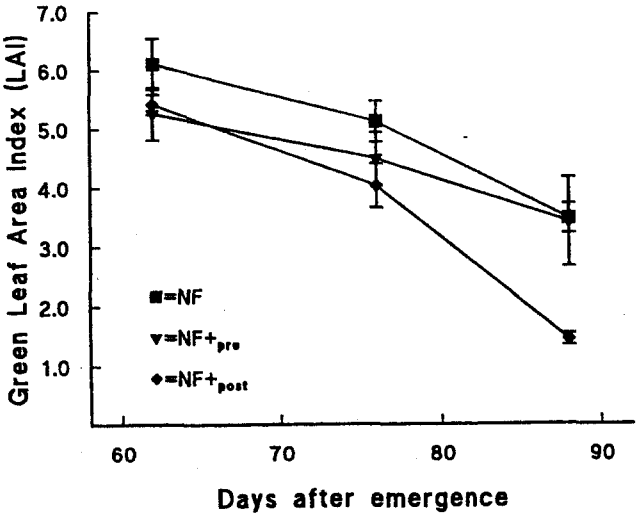


Figure 5. Green Leaf Area Index (LAI) of the wheat crop in the three chamber treatments from mid anthesis through most of the grain filling period.

It is concluded that ozone exposure is much more effective in decreasing the grain yield of wheat between anthesis and end of grain filling than before anthesis. This is well in

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accordance with what has been observed in beans, where ozone exposure after flowering had a much larger effect on pod yield than exposure before flowering (Younglove *et al.*, 1994) and in wheat (G. Soja, this volume). The increase in the protein content of the grain, which has been associated with ozone effects on the grain yield of wheat in a number of studies (Pleijel *et al.*, 1991; Fuhrer *et al.*, 1992), was found in the present study only in the *NF+post* treatment. This confirms the conclusion that the post-anthesis period is more sensitive to ozone exposure in wheat than the pre-anthesis period. This result should be considered in future work with critical ozone levels for crops.

4. Acknowledgements

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ASSESSMENT OF AMBIENT OZONE ON BEAN: RESULTS OF 1994 EXPERIMENTAL SEASON IN NORTHERN ITALY

I. Fumagalli [1], L. Mignanego [2]

[1] ENEL spa - Centro Ricerca Ambiente e Materiali, Milano, Italy

[2] Phytos snc - Biassono (MI), Italy

ABSTRACT

Within the International Co-operative Programme on the effects of air pollutants on crops (ICP-Crops) promoted by the UN/ECE Convention on Long-Range Transboundary Air Pollution, a series of experiments were performed in Italy on different crops. Results on bean of the 1994 experimental season carried out at a field site near Milan are presented. The aim of the trial was to assess the effects of tropospheric ozone on bean plants exposed to ambient air and treated with the antioxidant ethylene diurea (EDU). Additionally bean plants were exposed to filtered (F) and unfiltered (NF) air in open-top chambers. The plants were sowed in June and harvested in September. Main parameters investigated have been ozone visible injury on leaves, photosynthesis, number and weight of pods at final harvest.

Air quality data and the diagnostic features of plants about this trial allow to identify ozone as an important air pollutant with levels often above threshold for phytotoxic effects. In particular the number of injured leaves have been significant higher in -EDU and unfiltered air than +EDU and filtered air. Physiological measurements show a decrease in the photosynthetic rate in relation of ozone level. Effects of ozone dose is also confirmed by biomass data: the dry weight of pods in unfiltered air treatment was lower in comparison to filtered air treatment.

INTRODUCTION

Air pollution may be inducing visible injury and adverse crop response in vegetation. The effects of gaseous air pollutants have been recognised for many years on plants (Heagle et al. (1973), Roberts et al. (1983)) and within of gas investigated, tropospheric ozone is an important phytotoxic air pollutant (Heck et al. (1984), Krupa et al. (1995)).

On mediterranean crops, deleterious effects of ozone have been described through different experimental approaches (Badiani (1994), Gimeno et al. (1994), Schenone (1992)).

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Within the International Co-operative Programme on the effects of air pollutants on crops (ICP-Crops) promoted by the UN/ECE Convention on Long-Range Transboundary Air Pollution, a series of experiments have been performed in Europe on different crops.

These experiments involve standard protocol and specific sensitive plant with the aim to biomonitor the effects of air pollution, ozone in particular, in different climate condition.

This papers presents the experimental results concerning a trial carried out during the 1994 season on bean plants using both OTCs facilities and EDU plants administered. The results are compared to relative plants of control.

MATERIALS AND METHODS

The experiment was performed at Redecesio di Segrate (Milan) in summer 1994. Plants of bean (*Phaseolus vulgaris* cv Lit) were grown in 4 litres pots in a mixture of soil and peat (2:1). The plants were sowed on 16 June and harvested on 12 September. When the first trifoliate leaf had expanded (Day 0) the plants were placed inside the open-top chambers and in an external plots. Every two weeks, starting from Day 0, bean exposed to open-air were treated with ethylene diurea (+EDU) and water (-EDU) according to UNECE protocol. Twenty plants for each treatments (+EDU, -EDU, F, NF) were used. Visible injury on leaves and photosynthesis, using a portable analyser (ADC), were tested periodically during plant growing. At final harvest the dry weight of pods was determined. Air quality was monitored by automatic analysers from May to October. AOT40 (Accumulated exposure Over a Threshold of 40 ppb) during the diurnal hours with solar radiation above 50 W/m^2 and 7 hours mean have been determined for ozone pollutant.

RESULTS

Air quality data of pollutants monitored (SO_2, NO, NO_2, O_3) have showed as only ozone levels recording during the bean experiment were high. The 7-hours ozone mean from emergence (June, 23) to final harvest (September, 12) was 67 ppb. In the same period the accumulated dose above the threshold of 40 ppb (AOT40) for daylight hours was 20115 ppb.hours (Table 1). These ozone levels were sufficiently high to induce visible injury on leaves of -EDU and NF plants. The first appearance of O3 visible injury on trifoliate leaves have been recorded at 33 days after emergence with an AOT40 accumulated over 3 consecutive days of 931 ppb.h. The number of injured leaves was about 10% of total leaves at final harvest. Ozone symptoms were practically absent in +EDU and F plants. Physiological measurements show a decrease in the photosynthetic rate in -EDU and NF plants respect to the control plants (+EDU and F) both in

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August and September with significative differences (Figure 1). Effects of ozone dose is also confirmed by biomass data: the dry weight of pods in NF treatment was 28.5% lower in comparison to F treatment. A smaller reduction (-18%) was found in -EDU treated plants versus EDU treated plants (Figure 2).

CONCLUSION

The two different experimental approaches (air filtering and the antioxidant use) gave the same estimate of crop loss, so EDU seems to be a useful tool for studying yield effects of ambient ozone. In this experiment an evaluation of air quality data and of the diagnostic features of plants allows to identify ozone as an important air pollutant with levels often above threshold for phytotoxic effects. These results confirm previous tests with different plant species but same experimental design carried out in northern Italy by ENEL-CRAM.

From the above consideration, it is possible to conclude that ozone seems to be an important phytotoxic gas for crops in northern Italy, able to produce visible leaf injury and adverse crop response.

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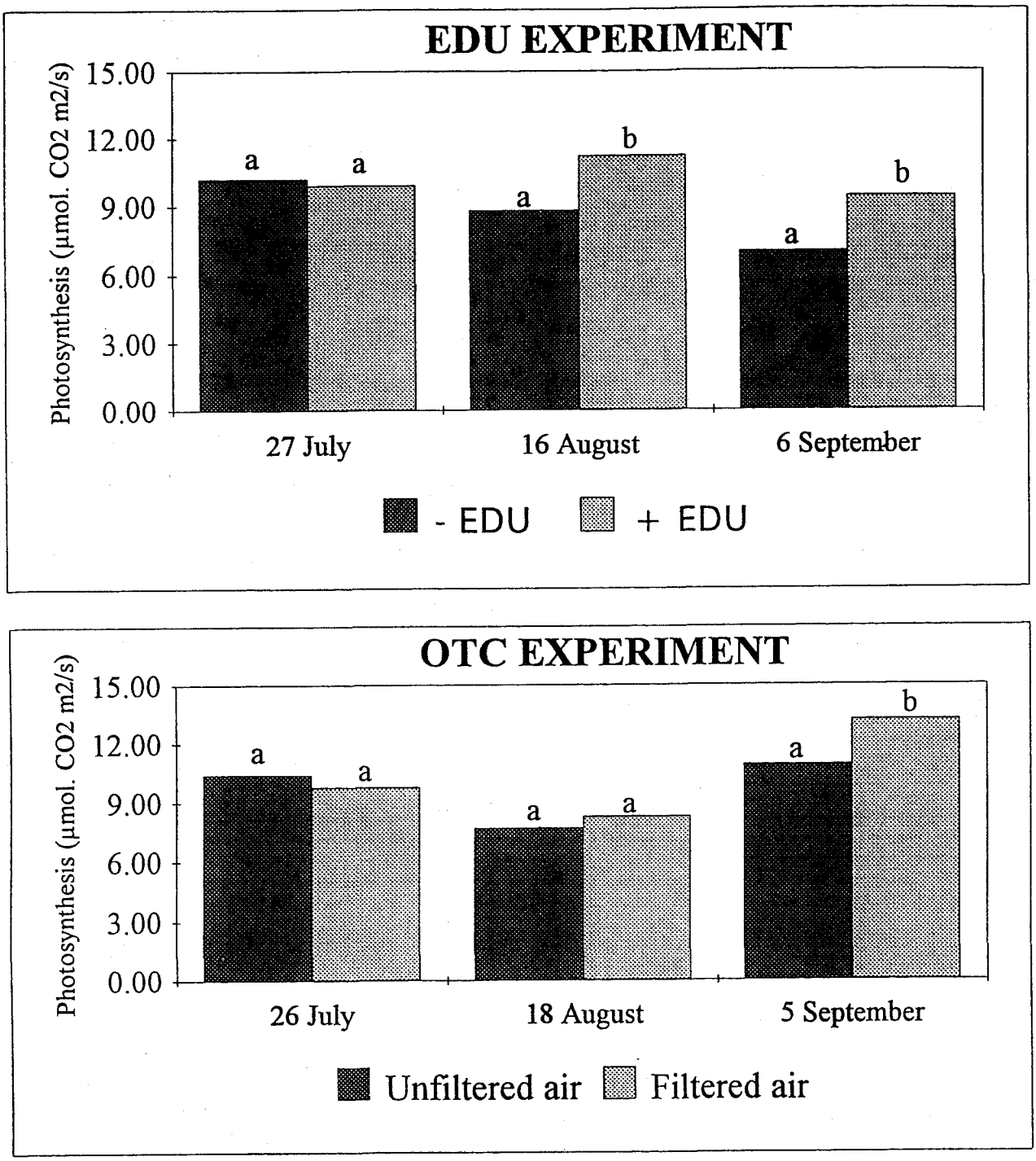


Fig.1 : Photosynthesis assessment on bean plants grown in open air (EDU experiment) and in open top chambers (OTC experiment) at Redecesio di Segrate (Milan). Different letters indicate significant differences at $p<0.05$ between treatments.

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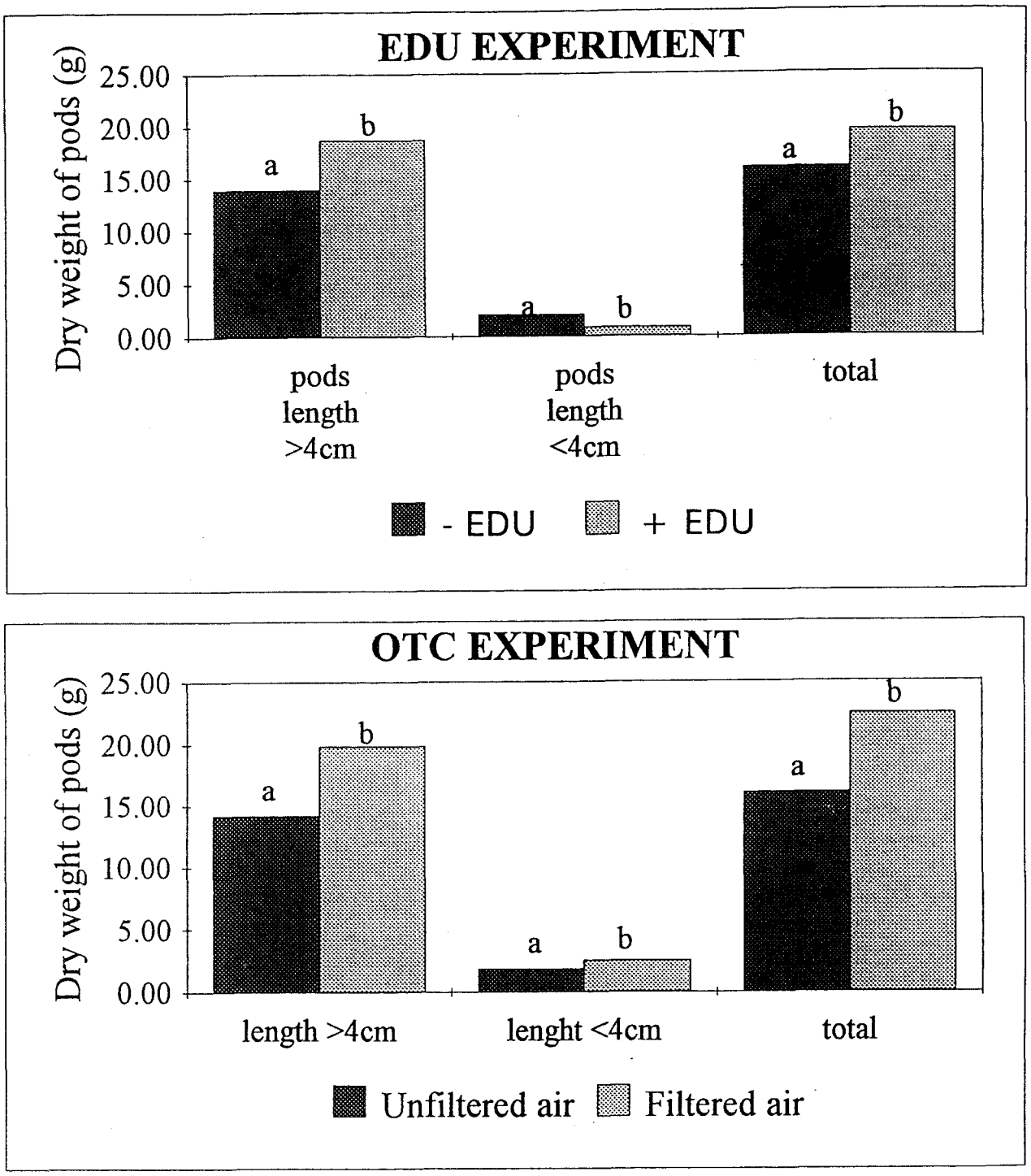


Fig. 2 : Dry weight of pods assessed at final harvest of bean plants grown in open air (EDU experiment) and in open top chamber (OTC experiment) at Redecesio di Segrate (Milan). Different letters indicate significant differences at $p < 0.05$ between treatments.

TABLE 1: Ambient ozone levels during different period of 1994 bean experiment performed at Redecesio di Segrate (Milan).

	7h mean (ppb) 10:00 - 17:00	12h mean (ppb) 7:00 - 19:00	24h mean (ppb)	**AOT40 (ppb*h)[1]**
Emergence to flowering 23/06 → 22/07 (29 days)	**71**	60	46	**8290**
Flowering to final harvest 22/07 → 12/09 (52 days)	**65**	54	37	**11825**
Emergence to final harvest 23/06 → 12/09 (81 days)	**67**	56	40	**20115**

[1] AOT40 is calculated for daylight hours (> 50 Wm^{-1}) as the sum of the differences between the hourly concentration and 40 ppb for each hour when the concentration exceeds 40 ppb.

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Effects of Chronic Ozone Stress on Growth and Reproduction Capacity of Native Herbaceous Plants

E. Bergmann, J. Bender and H.J. Weigel

Institut für Produktions- und Ökotoxikologie, Bundesforschungsanstalt für Landwirtschaft, Bundesallee 50, D-38116 Braunschweig

Abstract: Ozone (O_3) sensitivity of 17 native herbaceous species was investigated in a two year open-top chamber study. Plants forming homotypical collectives were exposed to charcoal-filtered air (CF) plus near ambient levels of O_3 during their entire life cycle (annual species) or during one growing season (perennial species). Half of the plants were harvested at the end of vegetative growth for measuring the above-ground biomass. From the remaining plants seeds were collected over several weeks to determine total seed yield or 1000 seed weight. On 8 species the leaf biomass production was reduced significantly due to an exposure of about 26 ppb O_3 (24 h mean). For some species this depression in vegetative growth corresponded to a loss in seed mass or number of flowers. However, *Malva sylvestris* was found to be the most sensitive species in terms of 1000 seed weight reduction.

1. Introduction

The UN Economic Commission for Europe (UNECE) defined a critical level (CL) for O_3 for agricultural crops, which is based mainly on experiments with spring wheat (Fuhrer & Achermann, 1994). In order to prove the application of this CL to other crops, the UNECE Working Group on Effects (ICP-Crops) is currently trying to validate this CL using other crop species and particularly O_3-sensitive clover cultivars. With respect to semi-natural and natural vegetation this CL remains to be tested and especially knowledge of the sensitivity range of members of natural plant communities is required. There are only a few screening experiments with native plant species showing different sensitivities in terms of visible injuries in fumigation experiments with O_3 concentrations of 200 ppb for 4 h (Ashmore et al., 1987 and Cornelius et al., 1985). In contrast, Reiling & Davison (1992) showed that the relative growth rate of a number of native species was already affected by a moderately enhanced O_3 concentration of 70 ppb for two weeks.

Basing on the ecological strategies proposed by Grime (1979) different predictions can be made to describe the reaction of three primary strategy types to stresses, including environmental pollution. Generally, it is expected that exposure to low concentrations of air pollutants like O_3 for a long period may impact ruderals by causing a switch from vegetative to generative growth. Although seed production may be reduced the effect of stress on the following generation will be low (Wolfenden et al., 1992). In contrast, the response of competitors to O_3 is of particular interest because they have the capacity to produce morphogenetic changes.

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Here, we describe effects of near ambient O_3 concentrations on growth and reproduction of native herbaceous species exposed in open-top chambers. Species tested were selected to represent the most frequently occurring species within the main plant communities typical for different habitats as well as different life-forms and regenerative and ecological strategies (sensu Grime).

2. Materials and Methods

2.1 Plant material and cultivation

Within the growing seasons of 1994 and 1995 17 species were exposed to O_3 either for one life cycle (annuals) or one growing period (perennials) starting at a young seedling stage. Plants were grown from seeds (Bornträger GmbH, Offstein) and young seedlings were transplanted into pots at a density of 2 to 5 individuals per dm^2, depending on the plants' size. Two soil types, a loamy soil and a loamy sand, were used and both soils were pre-fertilized with N-P-K-fertilizer corresponding to a nitrogen content of 120 kg per ha. Plants were exposed to O_3 in open-top chambers for individual exposure periods related to species' life cycles. At the time of flowering half of the plants were harvested for biomass measurements and injury assessments. Leaves were separated into healthy, senescent, dead or injured ones and dry matter mass of leaves, stems, flowers or buds were estimated separately. The remaining plants were further exposed until seed maturity. For quantitatively seed collection a paper bag was pulled on the ripening inflorescences (*Chenopodium album*, *Daucus carota*, *Sonchus asper* and *Urtica dioica*) or mature seeds or fruits were collected over several weeks (*Capsella bursa-pastoris*, *Malva sylvestris*, *Papaver dubium* and *Solanum nigrum*). From four species of the Compositae (*Chamomilla recutita*, *Galinsoga parviflora*, *Matricaria discoidea* and *Senecio vulgaris*) the total number of capitula was determined and an aliquot of seeds was collected to determine the 1000 seed weight.

2.2 O_3 exposure

O_3 was added to charcoal filtered air (CF) at a constant rate of 30 ppb for 8 hours per day (reference treatment) or additionally to about 50 % of the ambient air concentration simulating elevated O_3 (O_3 treatment). The mean daily O_3 profiles are shown in FIGURE 1. On average, the O_3 treatment resulted in an AOT40 of about 750 $ppb*h*week^{-1}$ and in a 24 h mean of 26 ppb. Compared to the O_3 concentrations in ambient air the AOT40 of the O_3 treatment was elevated by a factor of 1.5, whereas the 24 h mean was only 86 % of that in ambient air.

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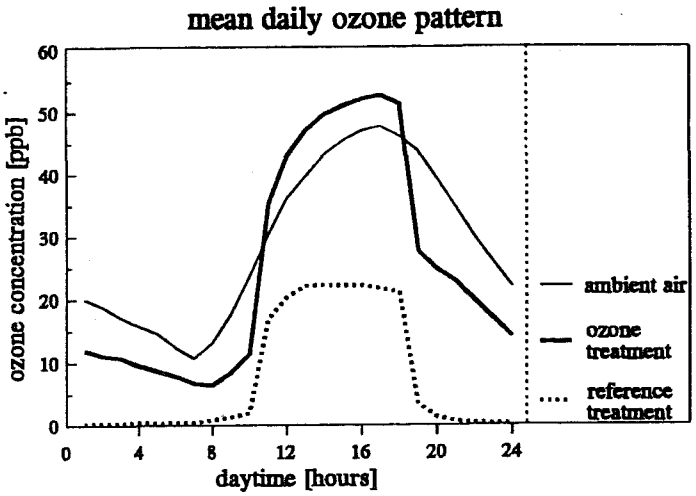


FIGURE 1: Mean daily O_3 pattern in open-top chamber treatments compared to ambient air. The constant addition of 30 ppb O_3 took place, when sunlight (PAR) exceeded 500 $\mu E*m^{-2}*s^{-1}$, O_3 dosage related to ambient O_3 was performed during the whole season for 24 hours per day. Each treatment was replicated in two chambers.

3. Results

3.1 O_3 exposure

TABLE 1: O_3 exposure indices for individual exposure periods of native herbaceous plant species in open-top chamber treatments and ambient air during vegetative growth. AOT40 = accumulated O_3 exposure above a threshold of 40 ppb within the exposure period.

	days of	*AOT40 [ppb*h]*			*24 h mean [ppb]*	
species	*exposure*	*O_3 treatm.*	*ambient*	*ref.*	*O_3 treatm.*	*ambient*
Capsella bursa-pastoris	36	2814	1951	9.0	21.0	29.0
Chamomilla recutita	47	4604	2607	9.5	27.0	28.5
Chenopodium album	29	3016	2233	9.5	26.5	30.0
Cirsium arvense	42	3950	2142	9.5	27.0	28.5
Daucus carota	88	7432	5938	9.0	21.8	29.0
Galinsoga parviflora	38	3530	2325	9.0	26.0	28.5
Malva sylvestris	42	3950	2233	9.5	27.0	28.5
Matricaria discoidea	55	4901	2652	9.5	25.5	28.0
Papaver dubium	73	8400	7136	9.5	26.0	28.0
Rumex obtusifolius	34	4169	2790	9.5	27.5	30.5
Senecio vulgare	36	3332	2209	9.0	25.5	28.5
Solanum nigrum	50	4248	2457	9.5	26.0	28.0
Sonchus asper	28	6477	5901	9.0	33.5	37.0
Stellaria media	41	2658	1321	9.5	25.0	27.0
Tanacetum vulgare	69	5518	4122	9.0	21.3	28.6
Trifolium arvense	51	7059	6658	10.0	28.0	34.3
Urtica dioica	49	3256	2264	8.5	20.0	28.0

TABLE 1 shows differences in the O_3 exposure levels between single exposure periods. Especially the AOT40, which reflects the exposure duration as well varied considerably between the species tested. This is primarily due to the fluctuating O_3 concentrations in ambient air, to which the chamber treatment was related. However, all exposure periods

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corresponded to ambient pollution in the same way and the O_3 concentrations applied were adapted to the ambient O_3 patterns that actually occurred when a certain species was growing.

3.2 Vegetative Growth

At the time of main flowering or after 5 to 6 weeks of O_3 exposure (*Rumex obtusifolius* and *Cirsium arvense*) leaf and total shoot biomass production were recorded (TABLE 2). In three species out of 17 (*Papaver dubium*, *Solanum nigrum* and *Malva sylvestris*) the O_3 treatment caused a significant reduction in leaf biomass by more than 30 %, which was also reflected in a reduction in total shoot biomass. Five more species (*Senecio vulgaris, Chamomilla recutita, Galinsoga parviflora, Trifolium arvense* and *Sonchus asper*) showed a decrease in leaf biomass up to 20 %. In all these plants, single leaf mass and leaf number per plant were reduced, however, forming smaller leaves seemed to be the predominant effect. Only *Trifolium arvense* formed significantly fewer leaves, when treated with elevated O_3 concentrations. In two species (*Chenopodium album* and *Rumex obtusifolius*) leaf biomass production was stimulated by O_3 by more than 20 %, however, these effects were not significant. This increase in leaf biomass corresponded to an increase in leaf number in case of *R. obtusifolius*, but not for *Ch. album*.

TABLE 2: Effect of O_3 treatment on shoot biomass production of native herbaceous plant species. Values are expressed as the ratio between O_3 treated plants and reference plants. * indicate significant differences between both means ($LSD_{95\%}$), (n.d. = not determined).

species	*no. of leaves*	*leaf d.m.*	*single leaf d.m.*	*shoot d.m.*	*n*
Capsella bursa-pastoris	0.885	0.932	1.074	0.945	16
Chamomilla recutita	0.962	0.729	0.819	0.836	18
Chenopodium album	1.154	1.448	1.218*	1.095	18
Cirsium arvense	1.201	1.010	0.834	0.955	18
Daucus carota	0.980	1.046	1.064	0.818	18
Galinsoga parviflora	1.006	0.774	0.805*	0.797	10
Malva sylvestris	0.953	0.623*	0.631*	0.781*	18
Matricaria discoidea	1.165	1.174	1.006	0.855	14
Papaver dubium	0.892	0.578*	0.623*	0.618*	18
Rumex obtusifolius	1.350	1.285	1.039	1.567	18
Senecio vulgaris	0.864	0.724*	0.827*	0.884	18
Solanum nigrum	0.826	0.586*	0.691*	0.767*	18
Sonchus asper	0.915	0.798	0.874	0.839	18
Stellaria media	n.d.	1.035	n.d.	0.999	18
Tanacetum vulgare	1.166	0.914	0.855	0.971	18
Trifolium arvense	0.828*	0.765*	0.938	0.686	14
Urtica dioica	0.908	0.873	0.953	0.900	18

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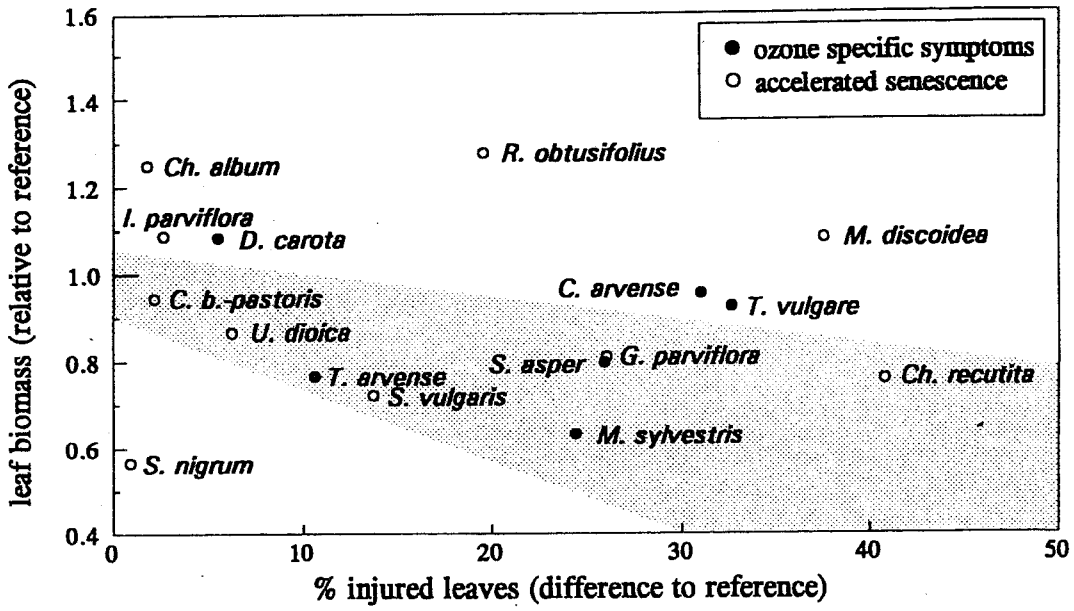


FIGURE 2: The relationship between leaf biomass response to O_3 and the extent of visible injuries. Number of injured leaves is expressed as percent of total leaf number, leaf biomass is given as the ratio of dry matter between O_3 treated plants and reference plants. For *Papaver dubium* leaf injuries were not recorded.

The remaining seven species showed only minor or no O_3 effects on leaf biomass. For *Cirsium arvense* and *Tanacetum vulgare*, the formation of smaller leaves due to O_3 was compensated by an increase in the number of leaves, leading to a net effect on leaf biomass close to zero.

Within these response types a correlation of leaf biomass response to the occurrence of visible leaf injuries could be detected for only about half of the species tested (FIGURE 2). Effects on leaf biomass ranged from + 20 % (*Ch. album*) to - 40 % (*S. nigrum*) relative to the reference treatment although the number of senescent leaves was not affected by O_3 in both species. Leaf biomass of species exhibiting O_3 symptoms was also affected in different directions ranging from about + 29 % (*R. obtusifolius*) to - 38 % (*M. sylvestris*). The two camomile species were highly affected by O_3 in terms of leaf injuries (37 % and 40 % of all leaves more were senescent on O_3 treated plants than on reference plants) but leaf biomass was either decreased by 27 % in *Ch. recutita* or increased by 17 % in *M. discoidea*. During plant development *M. discoidea* formed more leaves, when older leaves were injured by O_3 resulting in an increase in total leaf biomass, while *Ch. recutita* did not change leaf number even when a high number of leaves was senescent. Thus, it may be concluded that forming additional leaves, when existing leaves are injured, seems to be one mechanism to cope with an O_3 impact. This was found for *R. obtusifolius* and *M. discoidea* for example, whereas the growth of *Ch. album* was stimulated without the appearance of any visible symptoms.

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3.3 Reproduction Organs

In 12 species all plants tested set flowers during the exposure period, while *C. arvense, D. carota* and *R. obtusifolius* usually flower in the second year. *U. dioica* is complete dioecious and the number of female plants differed between O_3 treatments. Time of flowering or of seed set was not affected by O_3 in any species (data not shown).

Most species tested revealed less reproduction organs, when exposed to elevated O_3, whereas the 1000 seed weight remained unaffected (TABLE 3). This loss in reproduction organs was partly related to the extent of leaf injuries. *S. nigrum* and *C. bursa-pastoris* were impaired by O_3 in terms of fruit or seed production, but showed no impact in terms of leaf injuries (FIGURE 3).

TABLE 3: Effect of O_3 treatment on reproduction parameters (seed mass or number of inflorescences revealed totally or until an intermediate harvest). Values are expressed as the ratio between O_3 treated plants and reference plants. * indicate significant differences between both means ($LSD_{95\%}$). Number of plants that produced flowers are indicated as flow., (th.s.wt. = 1000 seed weight. # = fruits fresh weight, n.d. = not determined)

	seed mass / plant [g]		*no. of inflorescences*		*th.s.wt.*	*plants*
species	*total*	*at harvest*	*total*	*at harvest*		*flow. / total*
Capsella bursa-pastoris		0.718			0.944	12 / 12
Chamomilla recutita			0.694*		n.d.	18 / 18
Chenopodium album	0.875				0.975	18 / 18
Cirsium arvense	n.d.				n.d.	2 / 14
Daucus carota	0.494*				0.992	5-9 / 18
Galinsoga parviflora			0.720		1.045	10 / 10
Malva sylvestris	0.460*				0.839*	18 / 18
Matricaria discoidea			0.716*		0.960	28 / 28
Papaver dubium		1.243			1.046	18 / 18
Rumex obtusifolius	n.d.				1.012	1-2 / 18
Senecio vulgaris			0.795		0.965	18 / 18
Solanum nigrum	0.710#				n.d.	18 / 18
Sonchus asper	0.877				0.973	18 / 18
Stellaria media		0.974			0.975	18 / 18
Tanacetum vulgare			0.908		n.d.	18 / 18
Trifolium arvense				1.311	n.d.	3-4 / 12
Urtica dioica	0.813				0.915	6-10 / 18

If the response of reproduction is related to the response of leaf biomass three different response types could be identified (FIGURE 4). For most species a loss in reproduction organs was likely to correspond more or less to a loss in leaf biomass. In three species seed production was reduced, while leaf biomass was stimulated to some extent (*D. carota, M. discoidea* and *Ch. album*). If in these species seed production was not a function of leaf biomass production, the way in which O_3 has affected the reproduction capacity may be of special interest for further investigations.

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On the opposite, *P. dubium* and *T. arvense* revealed more inflorescences or seeds in response to the O_3 treatment while leaf biomass was decreased. Generally, it should be noticed that biomass and injury determinations and seed production were studied on different individuals.

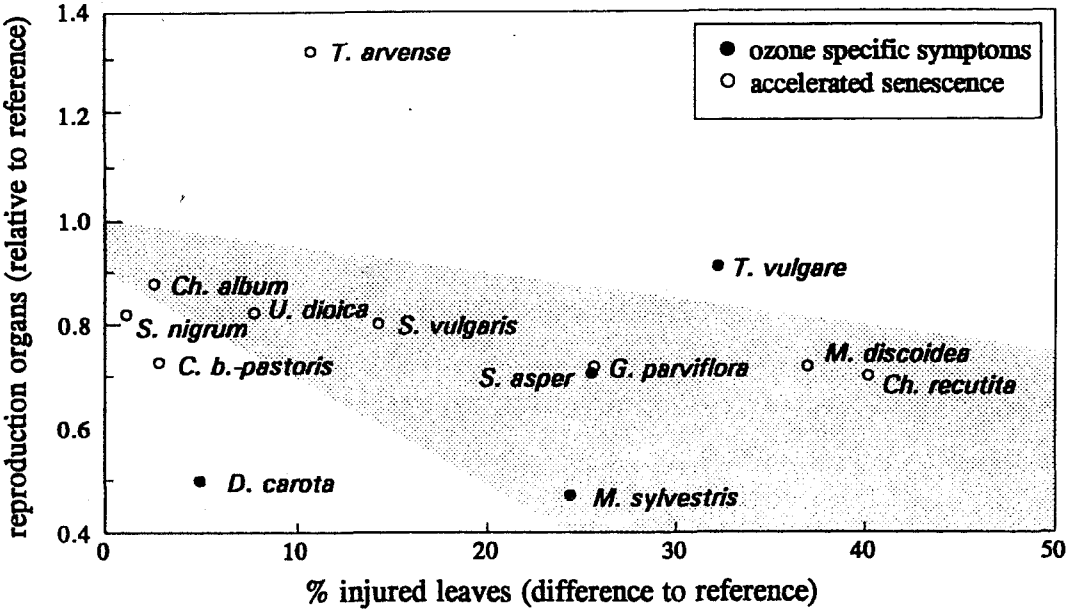


FIGURE 3: The relationship between the effect of O_3 on the formation of reproduction organs and the extent of leaf injuries (see FIGURE 2). Reproduction response is given as the ratio between O_3 treated plants and reference plants (see TABLE 3).

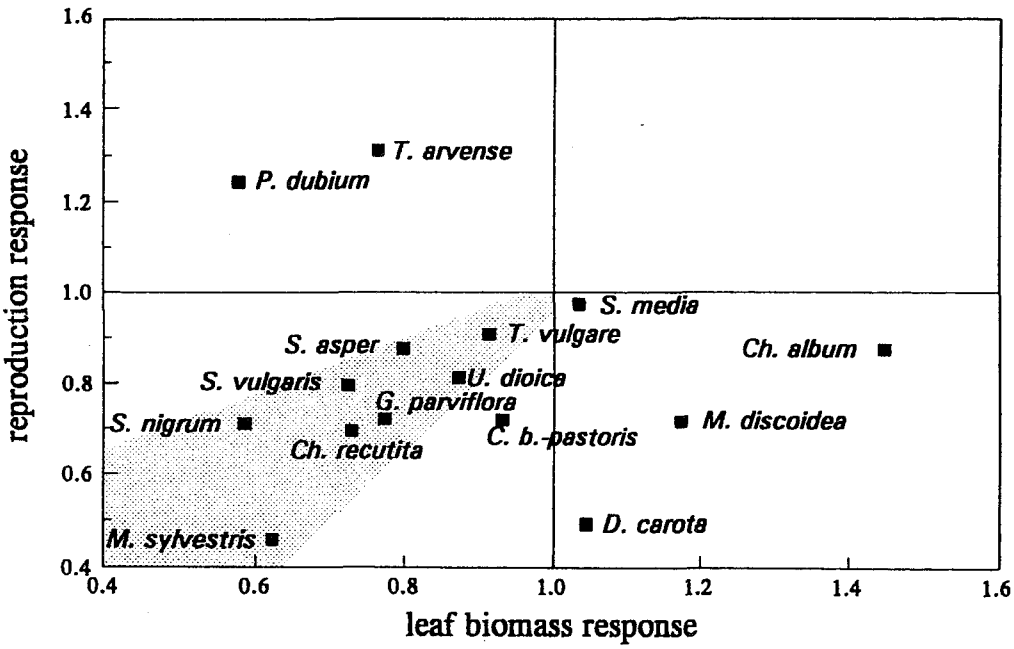


FIGURE 4: The relationship between leaf biomass and reproduction response to O_3. Both parameters are expressed as the ratio between O_3 treated plants and reference plants.

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4. Conclusion

In general the present results show that production of leaf biomass and reproduction organs was not necessarily related to the degree of visible leaf injuries, whereas for most species formation of reproduction organs correlated with leaf biomass production.

The most responsive species to O_3 in terms of effects on reproduction was *M. sylvestris*, in which both number of seeds and 1000 seed weight were reduced. Moreover, this decrease in reproduction capacity corresponded with a high rate of injured leaves and a loss in leaf biomass due to O_3.

Ashmore et al. (1987) observed higher O_3 sensitivities of plants from more calcareous habitats and a higher proportion of sensitive species within phylogenetically more "primitive" families. These authors reported that only very few members of the family of Compositae exhibited O_3-symptoms which would classify these plant species as rather O_3 insensitive. In our experiments this family was represented by seven species and all of them were affected by O_3, although the pattern of response differed between species. Reiling & Davison (1992) suggested a relationship between O_3 sensitivity of native plant species and their seed bank characteristics.

Considering all 25 species, that have been tested in our experiments with respect to visible leaf injuries, leaf vitality of 17 species was significantly impaired by O_3. Out of these, six species showed O_3-specific symptoms. Within this group of species a relationship between O_3 sensitivity and their habitats or life-forms could not be proved. It has to be considered that beside the different O_3 response patterns of the species tested, the O_3 exposure regimes may also contribute to the differences in O_3 sensitivity found between the species. As also shown in our studies, non-specific leaf colorations and accelerated senescence was highest with the highest chronic O_3 dose, whereas the extent of O_3-specific visible injuries was best related to exposure characteristics that emphasize more peak concentrations (Bergmann et al. 1995).

Acknowledgements

We thank Andrea Bürger and Carina Trenkler for plant cultivation and excellent technical assistance.
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Growth response of clonal Norway spruce (Picea abies (L.) Karst.) on ozone and elevated CO_2 under different nitrogen and water regimes

Häberle KH*, Lippert M, Payer HD

GSF-Forschungszentrum für Umwelt und Gesundheit, Expositionskammern, Neuherberg, Postfach 1129, D-85764 Oberschleißheim.

*present address: Lehrstuhl für Forstbotanik, LMU München, Hohenbachernstr. 22, D-85354 Freising.

Abstract

Model systems of 5-year-old Norway spruce seedlings were subjected to different concentrations of ozone (20 vs. 80 nl l^{-1}) and CO_2 (350 vs. 550 nl l^{-1}) as well as to different water supply (watered vs. droughted) and nitrogen treatments (normal vs. surplus) for one growing season in a complete factorial experiment in the GSF phytotron. According to the workshop topic single and combined effects of ozone on biomass production, development of the stem diameter, and changes in the nutritional status are reported with respect to their potential physiological causes. Ozone prolonged the period of radial stem growth and reduced the concentrations of P, K and Mg, whereas most of biomass fractions and the root/shoot ratio remained unchanged. Interactions of ozone especially with the CO_2- and water supply show that it is necessary to take the environmental conditions into consideration when discussing ozone effects on plants.

Introduction

European forest species respond to experimental ozone treatments with both biochemical changes and visible injury (SENSER et al. 1987; LANGEBARTELS et al. 1989) not only at excessive concentrations but also at levels not much higher than in reality (LEONARDI et al. 1990; WALLIN 1990; LANGEBARTELS et al. 1992; MATYSSEK et al. 1992, 1993). Norway spruce is known as a relatively ozone resistant species (CHAPPELKA & CHEVONE 1992), but knowledge about interactive effects of ozone with other environmental factors are scarce (MOONEY et al. 1991). In nature a stress does not occur as an isolated event: Enhanced ozone levels coincide with high radiation loads and are often accompanied by elevated temperatures and drought events. Consecutively not only water and nutrient availability from the soil are modified but also the stomatal aperture which determines ozone uptake. Furthermore tree growth is supposed to become enhanced with increasing atmospheric CO_2 concentrations and nitrogen immissions (CEULEMANS & MOUSSEAU 1994,

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VITOUSEK 1994). Air pollutants and nutrient deficiencies potentially counterbalance this development.

Material and methods

In a multifactorial investigation on Norway spruce the CO_2 and ozone concentrations in the air, the nitrogen and water supply were crosswise varied on two levels each (16 treatments) under controlled conditions in closed environmental chambers (GSF-phytotron, see PAYER et al. 1993).

After almost 2 years of precultivation, during which the differing nitrogen supply had already begun, 5-year-old clonal Norway spruce trees were exposed to the following experimental conditions in the GSF phytotron from 16/6/92 to 12/1/93:

CO_2:		ambient air	(referred to as **350-CO_2**)
	vs.	ambient air + 200 $\mu l*l^{-1}$	(referred to as **550-CO_2**)
O_3:		20 $nl*l^{-1}$ 24 $h*d^{-1}$	(referred to as **20-O_3**)
	vs.	80 $nl*l^{-1}$, 11 $h*d^{-1}$ with shortterm peaks up to 140 $nl*l^{-1}$; during nighttime 20 $nl*l^{-1}$.	(referred to as **80-O_3**)
N:		100 mg N kg^{-1} dry soil	(sufficient supply, referred to as +N)
	vs.	250 mg N kg^{-1} dry soil	(N-surplus, referred to as ++N)
H_2O:		non limiting water supply according to the demand of the trees	(referred to as **+H_2O**)
	vs.	periodically dry, not watered between July, 17th and 31st and again between September, 7th and October, 5th	(referred to as **-H_2O**)

256 trees were planted in 1990 in groups of four into 64 pots of 40 L volume each. The simulation of the climate in the chambers followed closely the climatic conditions of higher altitudes of the Bayerischer Wald. The soil applied in the experiment also originated from this region. Light intensity, air humidity, air- and soil temperature followed an idealized basic pattern in their diurnal course which was regularly repeated. At the end of October day length, temperature and ozone fumigation were gradually reduced. The whole experiment which comprised four different factors on two levels each in a completely crossed design resulted in 16 different treatments. A detailed description is given in HÄBERLE (1995).

Biomass determination: The shoots and needles which had developed during the exposure as well as the previous year`s shoots and the remaining above-ground biomass were separated and dried to constant weight (65°C, 3 days). Roots were washed, oven-dried, and weighed. The root/shoot ratio was calculated per pot from the total below-ground biomass divided by the total above-ground biomass.

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Nutrient analysis: Spruce needles were ground after drying and digested by nitric acid under pressure. The liquid samples were analyzed by several spectrometrical methods (ICP, DCP, and Z-AAS). Nitrogen was determined by elementary analysis of the dried and ground raw material. The analyses were performed by the Central Analytical Department of the Institute of Ecological Chemistry at GSF.

Dendrometric survey: The stem diameter increment was recorded continuously with one tree per pot. This tree was equipped with a micromorphometric instrument ("dendrometer") fixed to the stem two centimeters above ground. The resistance of a potentiometer (1 kOhm, Megatron, Munich) which was pressed against the stem by a spring, provided data with an accuracy of ± 0.5 µm every half an hour (HÄBERLE et al. 1995). The length of the growth period was determined from the duration of increase in stem diameter.

The results were evaluated by means of four-factorial analyses of variance (SAS statistical system). The results are shown in graphs followed by a table containing the levels of significance of treatment effects and interactions. The symbols have the following meaning:

Effects: + = increase, - = decrease, * = interaction

Levels of significance:

+++,---,***:	p<= 0.001,
++, --, **:	0.001<p<= 0.01
+, -, *:	0.01 <p<= 0.05

Results and discussion

The accumulated O_3 doses (AOT0) achieved at the end of the experiment were 96 ± 1 for 20-O_3 and 214 ± 5 µl l^{-1} h for 80-O_3 respectively, corresponding to comparable doses at these ozone monitoring stations of the german Umweltbundesamt with the lowest and highest values. According to the workshop topic, the discussion is stressed on ozone effects.

After four month of ozone fumigation (cumulative ozone dose AOT40 about 60 µl l^{-1} h) five per cent of the trees (80-O_3) developed visible symptoms ("mottling") in previous years' needles. There was no correlation found to a distinct treatment combination within the elevated ozone level.

The biomass production of shoot and root fractions was not affected by 80-O_3 with the exception of the previous years' shoot. Its dry weight was reduced by nine per cent (fig. 1, tab. 1).

Ozone reduced the seasonal growth period of well watered young Norway spruce by 13 days. In trees which had experienced drought events this ozone effect was 3 days only indicating compensatory effects of water deficiency which extended the growth period in general by 12 days. CO_2 reduced the growth period by 8 days but was counterbalanced by O_3. Additive and interactive responses resulted in a maximal difference of approximately 3 weeks (fig. 2, tab. 1 and 2).

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The N-fertilization which was started on two levels in 1991 led to well-distinguished nutritional statuses in the needles, when further treatments were applied in 1992. The nutrient contents were considered not to be limiting at this stage, although they were low (not shown). With the development of the new flush a considerable proportion of nutrients was transferred thereto from the older needles. This resulted in a decrease of the nutrient content in one-year-old needles on both N-levels. Inspite of the resulting shortage of N,P, and K in the previous year's needles, the nutrient ratios were well balanced in the +N treatment. In the ++N treatment, however, N/P and N/K ratios exceeded the limit for a balanced nutrition in previous year's needles (tab. 3, HÜTTL 1991). 80-O_3 reduced the concentrations of phosphorus, potassium and magnesium concentrations of current years` needles (tab. 4). In contrary to the other factors (CO_2, N and periodic drought) the ozone induced changes in nutrient concentrations cannot be explained by a changed biomass production (HÄBERLE 1995).

Along with the decreased dry weight of the previous year`s shoot (1991) it can be suggested that ozone disturbed the allocation of nutrients and carbohydrates (WILLENBRINK & SCHATTEN 1993). The shortened growth period at elevated O_3 concentrations points into the same direction. Whereas the drought and CO_2 effects on the growth period can be explained by reaching the sink capacity slower or faster, respectively, the reaction on ozone is unexpected. May be again disturbed transport processes, which are leading to a misfunction in sink-source regulation, can be made responsible for this observation. Probably all these findings indicate problems in phloem loading in spruce caused by ozone, as it was found in birch (MATYSSEK et al. 1992), although the disturbances were not severe enough to change biomass accumulation and the root/shoot ratio significantly.

Further indications on interactive ozone effects result from additional measurements in the described experiment. Net photosynthesis, carboxylation efficiency, and maximum photosynthetic capacity were reduced by ozone, when the trees were well watered in a balanced nutritional state under ambient CO_2 (LIPPERT et al. 1993, 1996). The photosynthetic parameters were enhanced, however, by 50 - 60 % by preceding drought events. Results of former studies indicate such effects to depend on the genetic disposition as well (clone effects). SCHWANZ et al. (1996) found the activities of antioxidative enzymes (catalase, peroxidase) to be influenced by different treatments. Three weeks after the end of a drought episode in summer, needles from spruce trees grown at elevated CO_2 showed increased enzyme activities as compared to needles from trees grown at ambient CO_2. This response was increased, if elevated ozone was present as an additional stress factor.

Interactive effects of ozone are also known from other tree species. PEARSON and MANSFIELD (1993) reported on an experiment with beech, where stomatal conductance decreased due to ozone fumigation in well watered trees, but increased in droughted trees.

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Conclusions

i) The ozone treatment caused visible chlorosis („mottling") in needles of Norway spruce, known as an ozone tolerante species. Also the length of the growth period was influenced by ozone. These findings proofed the chosen cumulated ozone dose between June and October (AOT40 ca. 60 $\mu l\ l^{-1}\ h$) to be sufficient to cause first reactions on a whole-plant level in spruce.

ii) Although the direct effects of ozone on growth and biomass production of the young spruce trees remained relatively small, it is obvious that in combination with other stress factors unpredictible reactions can occur. This should not be neglected in the discussion about anthropogenic climate change.

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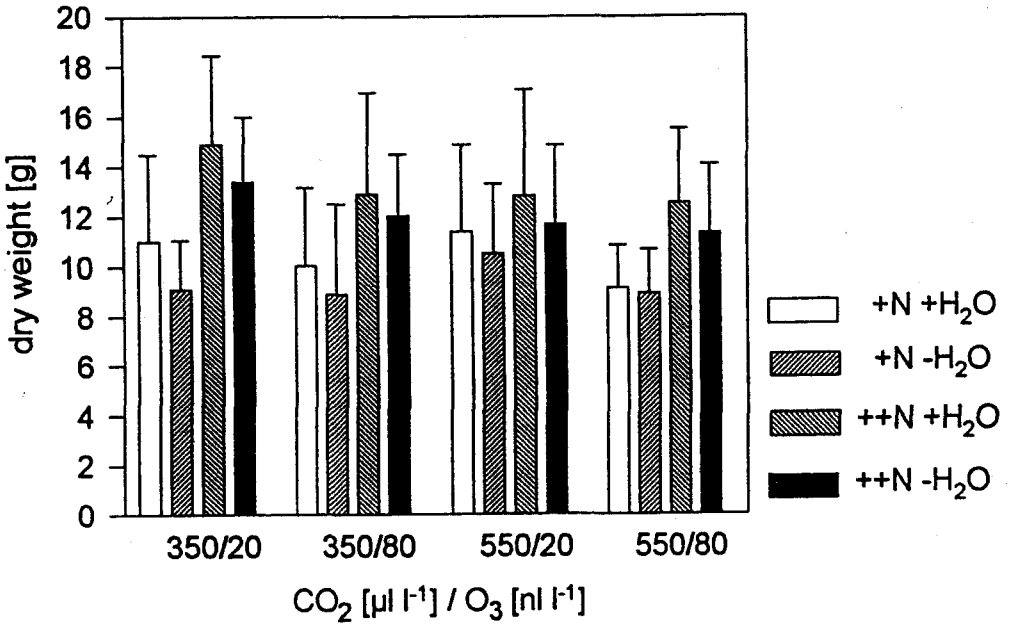


Fig. 1: Dry weight of the shoots developed in 1991, determined at the end of the exposure period in January 1993.

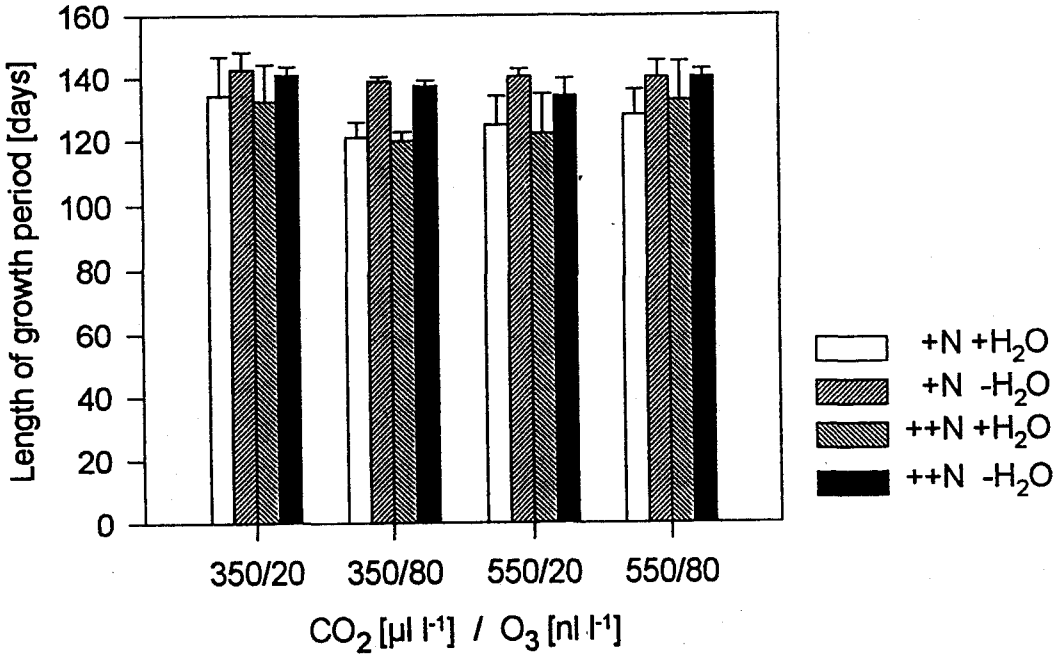


Fig. 2: Period of radial stem growth measured by dendrometers.

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Tab. 1: Level of significance for main and combined effects of treatments (four-factorial ANOVA) on the dry weight (DW) of the plant fractions, root/shoot ratio and the length of the growth period.

Main effects and interactions	DW shoot `92	DW shoot `91	DW total shoot	DW root	DW S+R	R/S ratio	Growth period 1992
n	256	256	64	64	64	64	64
CO_2	++			+		+	
O_3		-					
H_2O(-)	---	-	---	---	---		+++
N	+++	+++	+++	+++	+++	---	
CO_2 O_3							**

Tab. 2: Interactive effects between CO_2 und ozone on the length of the growth period.

treatment		mean	Level of significance (p-value)				
CO_2	O_3	[days]	No.	1	2	3	4
350	20	138	1	-	0,0036	0,0102	0,4108
350	80	129	2		-	0,6969	0.0300
550	20	130	3			-	0,0714
550	80	135	4				-

Tab. 3: Nutrient concentrations in current year`s needles at the end of the experiment in the two nitrogen groups. (m=mean, s=standard deviation, *= interaction CO_2*N), # limit of good supply (HÜTTL 1991).

Element		N	P	K	Ca	Mg	S	Fe	Mn	Zn	N/P	N/K	N/Mg
		[mg g^{-1}]											
+N	m	10,2*	1,0	4,7	6,3	1,1	0,6	0,04	0,56	0,05	10,0	2,3	9,8
	s	2,2	0,2	1,0	1,2	0,3	0,1	0,03	0,23	0,01	2,6	0,7	3,9
++N	m	20,6*	0,8	3,1	4,6	0,9	0,7	0,03	0,48	0,03	25,2	7,0	25,9
	s	3,5	0,1	0,8	1,6	0,3	0,1	0,02	0,28	0,01	5,3	1,9	9,8
limit#		12	1,1	4	2	0,6	0,8	0,003	0,02	0,013	6-12	1-3	8-30

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Tab. 4: Level of significance for main and combined effects of treatments (four-factorial ANOVA) on concentrations of macronutrients of current year's needles (1992) at the end of the experiment.

Main effects and interactions	N	P	K	Mg	Ca
n	256	256	256	256	64
CO_2	-	--			+
O_3		---	-	-	
H_2O(-)	+++		+++		
N	+++	---	---	---	--
CO_2 N	***				

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The classification of chromosomal aberrations - a method for the assessment of various kinds of environmental stress (1989-1995)

Maria Müller, Michael Tausz, Dieter Grill

Institute of Plant Physiology, University of Graz, Schubertstraße 51, A-8010 Graz

Abstract

The classification of chromosomal defects as a plant test system with spruce trees (*Picea abies* (L.) Karst.) has been used for an early detection of environmental influences on forest trees. The results obtained from different natural sites and from fumigation experiments recommended this test system to be an easy and sensitive screening method for damages caused by environmental stresses on spruce trees. Despite the widespread occurrence of injuries caused by different air pollutants, the mechanisms of the damaging process and the plant defence systems against pollution attacks are often poorly understood. Considering the importance of our forest ecosystems it is essential to characterize their short- and long-term responses to a variety of environmental and pollution impacts.

Introduction

Since the beginning of the eighties an accumulating evidence of dieback of forest ecosystems has been observed, known as 'novel forest decline'. The damage is characterized by its wide geographic dissemination, its appearance on all major tree species, as well as its long duration (Wild and Schmitt 1995). The phenomenon is commonly thought to be caused by multiple factors. Among others there is strong evidence for a participation of air pollutants in inducing forest damages (Cowling 1988, Schulze 1989). The concentration of air pollutants can vary strongly, locally as well as temporally (Moosmayer 1992). The SO_2 emissions in many European countries have decreased in the past 10-15 years. Consequently, there has been a concern about the role of ozone as a contributing factor to forest decline (Guderian 1985). In contrast to accumulating agents as SO_2, fluoride, or chloride, the indication of these non-accumulating substances is difficult.

Therefore it is of interest to find new methods or to modify old test systems for an early detection of environmental influences on forest tree species. One of these test sytems is the classification of chromosomal abnormalities, that comprises a sensitive method for the assessment of genotoxic effects caused by chemical treatments. The cytological studies are relatively simple to perform and give good information about effects of cell division and chromosomes (Levan 1938, Fiskesjö 1985). In our search for an easy and reliable test assay for the detection of genotoxic substances on forest tree species we have carried out a test system by the classification of chromosomal defects with spruce trees as bioindicator plants. Some other systems with forest trees exist today: Fiskesjö (1989) examined the aluminium toxicity in root tips of spruce trees, beech and oak and found disturbances in dividing cells, as mitotic effects as well as growth restriction and browning of the roots and so-called 'Al-structures'. Other investigations with spruce trees showed that this forest tree species is mitotically highly active throughout the whole year and therefore the trees may be affected by

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pollutants also during their morphological state of dormancy (Matschke et al. 1994). Another genotoxicity test, basing on the evaluation of sister chromatid exchange frequencies has been worked out by Schubert and Rieger (1994). The authors suggested their test a suitable and reliable tool to observe genotoxic effects in spruce plants. Some of these test systems were carried out under laboratory conditions with high concentrations of genotoxic substances only for short exposure times. In 1989 we performed a plant test system by the classification of chromosomal aberrations according to the basic work with spruce trees of a Slovenian working group (Druskovic 1988). Our test system has been used under natural conditions (Grill et al. 1993, Müller and Grill 1995, Müller et al. 1991, 1992, 1994b, 1995, Tausz et al. 1994) as well as under defined conditions of single air pollutants as ozone alone (Müller and Grill 1994, Müller et al. in press) and in combination with elevated levels of CO_2 (Müller et al. 1994a). These studies showed that the classification of chromosomal aberrations in spruce is a sensitive measure to monitor various environmental effects on trees.

In the present paper the results obtained with our test system from natural sites as well as from ozone-fumigation experiments under defined conditions from 1989 to 1995 are reviewed and discussed.

Chromosomal analysis

After the preparation of the root tips the meristem cells are separated and interphases and all mitotic stages can be observed (Müller and Grill 1992). Due to the effect of the metaphase blocker, metaphases are the most frequent mitosis stages. The normal metaphase of Norway spruce consists of $2n = 24$ chromosomes (e. g. Miyake 1903, Sax and Sax 1933, Price 1989). The most common type of chromosomal aberrations in all variants including the field studies was chromosome stickiness, resulting in connections, clumped metaphases and amorphous chromatin masses. This type of aberrations may be the result of improper folding of the chromosome fiber into single chromatides. Thus, chromosomes become attached to each other by subchromatid bridges (McGill et al. 1974, Klasterska et al. 1976). Stephen (1979) also suggested that stickiness is a type of physical adhesion that involves mainly the proteinaceaous matrix of the chromatin material. Chromosome stickiness reflects highly toxic effects, usually of an irreversible type, probably leading to cell death (Liu et al. 1993). Further defects found less frequently in this series of experiments are breaks, fragments and rings, which are genotoxic effects and they may transfer the damage to the following generations of cells (Fiskesjö 1994). Under conditions of constant ozone-treatment a significantly increased number of sticky chromosomes was observed. Under field conditions also an increased number of chromosome stickiness was noticed, but considerably more breaks and fragments occurred (Müller et al. 1994, Müller and Bermadinger-Stabentheiner in press). It is difficult to decide whether certain influences or combinations lead to specific chromosomal abnormalities. Druskovic (1988) suggested acute influences creating abnormalities on single chromosomes, as breaks and fragments, and constant long-lasting influences causing stickiness.

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Field studies

From 1989 to 1995 we have applied our plant test system to natural spruce stands in Austria. The data show that the classification of chromosomal aberrations is a valuable tool in environmental monitoring under natural conditions. Our method works with young spruce plants (up to five years old) and the results of the young trees are well correlated to results of the older trees of the stand (Müller et al. 1991, 1992). The data suggest an intensive site effect, that is more pronounced than possible effects of the soil or of the provenance of the individual tree (Müller et al. 1992, 1994). The results of our method provide valuable information about the condition of the trees (Müller et al. 1991, 1992), including that of spruce trees at higher elevation sites (Müller and Bermadinger in press). Plants at higher elevated sites have to cope with lower temperatures, higher irradiation and higher concentrations of naturally occurring photooxidants, with the leader substance ozone, compared to plants growing at lower elevated sites (Tranquillini 1979, Bolhar-Nordenkampf and Lechner 1989). However, at natural sites it is difficult to decide which factors are the most decisive in affecting plants. In experiments each different factor may be varied while other factors are kept constant, thus enabling the researcher to test the plant´s specific reaction (Fangmeier et al. 1992).

Ozone fumigation experiments

Ozone fumigation experiments were undertaken in climate chambers (GSF-Munich), in open-top chambers (Institute of Plant Ecology Giessen), and in greenhouses (Research Centre Seibersdorf).

In all the experiments obervations of visible damage were made on a qualitative basis during the fumigation, at the end of the fumigation period, and during the time when the plants were kept under natural air and weather conditions for the observation of long-term after-fumigation effects. No visible evidence of injuries or of stunted growth due to the effect of increased ozone was observed in any of the plants (Müller and Grill 1994, 1995, Müller et al. 1995, in press). These data suggested that the spruce clones used in our experiments were not sensitive to ozone treatments. The well known symptoms of ozone injury including bleaching, chlorotic mottling, changes in pigmentation, and necrosis (Kress et al. 1982, Williams 1986, Polle et al. 1993) could not be observed. Visible damage had not been found in other experiments in which concentrations of about 50-70 nl ozone l^{-1} were used (Alscher et al. 1989, Wallin et al. 1990). It has been suggested that acute short-term exposure to high ozone concentrations (> 200 nl l^{-1}) generally results in visible damage, whereas long-term, chronic exposure to lower ozone concentrations generally leads to reduced growth without foliar damage (Heagle 1989).

The results of the investigations under defined conditions of enhanced levels of ozone (Table 1) showed an intensive influence of ozone on the genetic material in the root tips of spruce. The amount of chromosomal defects increased with increasing ozone dose (Müller and Grill 1994, Müller et al. 1995, Müller et al. in press). This suggests the genetic material of the root tips of spruce trees as a sensitive indicator for oxidative stress, although the roots did not come in contact with ozone directly. In other studies with different test systems by the classification of genetic defects ozone has been identified as an agent that is genotoxic to plants (e. g. Ma et al. 1982). Furthermore, ozone was found genotoxic to microorganisms (e.

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g. Dubeau and Chung 1982), insects (e. g. Erdman and Hernandez 1982) as well as to cell cultures in vitro (e. g. Thomassen et al. 1991). Results from in vivo cytogenetic studies with laboratory animals after inhalation exposure are contradictory (e. g. Rithidech et al. 1990). The investigations concerning genotoxicity of ozone to higher plants were carried out with high ozone concentrations at first. In an early study (Fetner 1958) *Vicia faba* was exposed to 4000 µl l^{-1} ozone in the air for 15, 30, and 60 minutes and this treatment induced chromosome-type aberrations in the root meristem cells. The effects of ozone on *Vicia* were also studied by Janakiraman and Harney (1976) by investigations of the meiotic chromosomes of the buds. These exposures to concentrations of 2 µl l^{-1} for 4 or 8 hours caused chromosome-type aberrations, too. Further test systems for studying the genotoxic effects of volatile air pollutants in the laboratory and for field monitoring were carried out with *Tradescantia*. Either somatic mutations in stamen hairs (Schairer et al. 1979) or micronucleus formation in the meiotic pollen mother cells as test systems were used (Ma et al. 1982). Ozone exposure for 6 hours at 5 µl l^{-1} induced positive response in the stamen hair system, whereas the test for micronucleus formation (5.5 hours at 5 µl l^{-1}) was negative. In many of these investigations high ozone concentrations were added to the respective test system and the direct effects produced due to the ozone treatment were then determined after a certain time.

Table 1: Average hourly ozone concentration (nl l^{-1}) calculated using all 24 hours per day for the entire exposure. 24-hour dose (ppm * h)
Effects on Norway spruce root cells treated with ozone for 42 days (climate chambers), for 50 days (greenhouses) and for 101 days (open-top chambers), respectively. The chromosomal aberrations were scored in metaphase. For the variants 6 to 10 trees were used, and if possible 200 metaphases per tree were examined for chromosomal defects. Numbers in brackets are the standard deviation.
The symbols indicate significant differences between the two variants of each experiment for immediate effect (*** $p < 0.05$, Mann-Whitney's U-test)

Climate chamber studies	control	ozone
24-hour average	20	100
24-hour dose (42 days)	20.16	100.80
Chromosomal aberrations	2.1 (0.2)	3.9 (0.7)***
Greenhouse studies		
24-hour average	23.30	56.17
24-hour dose	27.76	67.40
Chromosomal aberrations	3.3 (0.3)	7.5 (0.4)***
Open-top chamber studies		
24-hour average	6.86	24.42
24-hour dose	16.63	59.20
Chromosomal aberrations	2.2 (0.6)	6.3 (0.3)***

In our experiments we worked with concentrations found also in our environment under so called natural conditions. In this connection the distance from the site of ozone influence (the top of the tree) to the root meristems must be taken into consideration. Via stomata ozone enters the mesophyll, where it can be dissolved in water and converted into reactive oxygen species such as superoxide anions, hydroxyl radicals, and hydrogen peroxide (Grimes et al. 1983, Mehlhorn et al. 1990, Kanofsky and Sima 1991). The active oxygen species are most probably responsible for the plasma membrane lipid peroxidation that has been demonstrated to occur after ozone treatment (Pauls and Thompson 1980, Heath 1987,

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Chevrier et al. 1990). There is also increasing evidence that aldehydes generated endogenously during the process of lipid peroxidation are causally involved in some of the physiological effects associated with oxidative stress in cells and tissues (Esterbauer et al. 1991). The role of aldehydes with chromosomal damage in plants is still unknown. In trees the rather long lived aldehydes must be translocated in the phloem to the root tips and the toxic peroxidation products may damage the phloem. Phloem damage interrupts translocation of assimilates, which may correlate with reductions in root growth. Nevertheless, biomass measurements of pine trees exposed to ozone showed no differences in root biomass compared to control trees. However, in this study the fumigation period seemed to be a rather short time compared with the total life span of a tree (Spence et al. 1990). Even Alscher et al. (1989) observed no effects of ozone on root biomass or carbohydrate content in red spruce seedlings in the first field season. However, in the second year starch content of the roots decreased and sugar content and its rate of accumulation increased with increasing ozone dose, but root-biomass was not affected (Amundson et al. 1991). Various studies have noted accumulations of soluble sugars and starch in shoots, that were explained by reduced or delayed phloem translocation (Tingey et al. 1976, Vogels et al. 1986, Gorrissen and Van Veen 1988, Spence et al. 1990, Willenbrink and Schatten 1993). If ozone reduces only the phloem loading, the long-term after effect may not be evident for years. For explaining our observations we suggest, that the effects in the root tips are the results of secundary reactions, e. g. caused by the translocation of toxic peroxidation products as aldehydes to the root tips, or caused by hormones.

The results of the investigation of spruce trees under normal air and weather conditions for a long-term after-fumigation effect revealed an increased number of chromosomal abnormalities in formerly ozone treated trees (Müller and Grill 1994, Müller et al. in press). The increased number of chromosomal aberrations proved a long-term effect of ozone on the genetic material of spruce plants. Our experiences from studies at natural sites suggest that the genetic material of spruce trees responds to changing environment with changes (either increase or decrease) in the number of chromosomal aberrations (Müller et al. 1992). This is corroborated by the observation of an increase of the aberration rate of experimental control variant plants as a consequence of their exposure to field conditions. Approximately 4 to 5 % of chromosomal defects can be found in meristems of spruce trees grown in natural areas (Müller et al 1991, 1992). If no long-term after-fumigation effects were manifest in the genetic material of the former fumigated variants all series of our experiments should show the same frequency of chromosomal defects under field conditions. But in our experiments the formerly ozone-fumigated variant responded under field conditions between 5.6 and 8.1% of chromosomal abnormalities, while the former control variants responded with 4.2 to 4.3%. Post-fumigation long-term effects of ozone have been found for other parameters previously: Among others they were observed for plant growth and vitality in loblolly pine (Spence et al. 1990), for secondary metabolites and antioxidants as catechin (Langebartels et al. 1990) and for pigments in spruce needles (Lütz 1992).

Conclusion and Outlook

Our investigations showed that the rates of chromosomal aberrations in the root tip meristems of Norway spruce trees are strongly dependent on environmental conditions, in particular on air pollutant impacts. Based on this knowledge the measurement of

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chromosomal aberrations provides a sensitive tool for bioindication of (otherwise hardly detectable) stress impacts. The question of mechanisms and pathways of the induction of chromosomal damage by the action of air pollutants, such as ozone, remains still unclear and needs to be clarified in further studies.

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Critical Load Exceedances and Ecosystem Protection Percentages

Maximilian Posch

Coordination Center for Effects, RIVM
P.O.Box 1, NL-3720 BA Bilthoven, The Netherlands

The concept of a critical load function for sulfur (S) and nitrogen (N) is introduced. This poses some difficulties in defining the exceedance of the critical loads as the amount by which deposition has to be reduced. This paper summarizes the methods for generalizing the concept of critical load and exceedance percentiles by so-called introducing protection isolines, which can be used for assessing simultaneous S and N emission reduction strategies.

Introduction

Critical loads of acidity and – derived from it – critical depositions of sulfur have been used in the negotiations on emission reductions leading to the Second Sulphur Protocol, signed in Oslo in June 1994 (UN/ECE 1994). A single critical load value considered representative for an entire EMEP-grid cell has been obtained by taking a percentile of all critical load values within that grid cell. Sulfur deposition patterns resulting from various emission scenarios have then been compared to these percentile critical loads by calculating the so-called exceedance, a single number per grid cell, obtained by subtracting the critical load from the sulfur deposition. And these exceedance values were then used in integrated assessment models to (cost-)optimize sulfur emission reductions over Europe.

Critical loads of nitrogen are required for the negotiations of a second step to the nitrogen protocol. In addition to sulfur, the deposition of nitrogen also contributes to the acidification of ecosystems, and this combined acidification effect is described by establishing an acidity balance. Such a balance equates the sources of S and N (i.e. their deposition) with their sinks (such as N immobilization, N uptake and denitrification), the counteracting net input of base cations and the leaching of alkalinity (ANC). Fixing the alkalinity leaching at a critical value leads to an equation which has to be fulfilled by a pair of deposition (N_{dep},S_{dep}) in order to avoid "harmful effects" on ecosystems. In general, there will be no unique such pair, rather all such pairs lie on a function which has been termed the critical load function of the ecosystem. In contrast to sulfur, which is considered a tracer in the soil, i.e. the S leaching equals the S deposition, there are sinks of N in the soil and (some of) these sinks are dependent on the amount of nitrogen deposited. Therefore, reducing N deposition by an given amount does not imply that the same amount can be deposited as sulfur without changing the acidity balance, and thus a unique exceedance cannot be defined.

As a consequence of the above, also simple percentiles cannot be computed. In this paper we summarize how the concept of a percentile can be generalized to critical load functions, leading to so-called percentile functions or ecosystem protection isolines. Although unique exceedances cannot be computed, it is still possible to calculate the percentage of ecosystems protected in a grid cell and thus to evaluate deposition scenarios. Furthermore, for a given pair of N and S deposition it is possible to classify – for a given protection percentage – the options available for reducing the deposition of one or both of these pollutants, in this way indicating the options available for reduction policies.

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The critical load function for N and S

Future negotiations on emission reductions in Europe will focus on nitrogen. Since both sulfur and nitrogen contribute to acidification they have to be considered together. In the following we shortly summarize the methodology for computing critical loads for S and N using the so-called simple mass balance (SMB) approach. A detailed derivation can be found in Posch et al. (1995) and the literature quoted therein. From the simplified charge balance in the leachate of a homogeneous soil compartment one obtains:

$$S_{dep} + N_{dep} - \left(BC_{dep} - Cl_{dep} + BC_w - BC_u + N_i + N_u + N_{de} - ANC_{le}\right) = 0 \qquad (1)$$

where $BC = Ca + Mg + K + Na$ and ANC stands for Acid Neutralization Capacity. The subscripts *dep*, *w*, *u*, *i*, *de* and *le* stand for deposition, weathering, net growth uptake, immobilization, denitrification and leaching, resp. Defining a critical ANC-leaching, $ANC_{le(crit)}$, which links a soil-chemical criterium (e.g. a critical (Ca+Mg+K)/Al ratio) with a potentially harmful effect, we get:

$$S_{dep} + N_{dep} - \left(CL_{max}(S) + N_i + N_u + N_{de}\right) = Ex \qquad (2)$$

with the abbreviation (essentially the critical load of acidity):

$$CL_{max}(S) = BC_{dep} - Cl_{dep} + BC_w - BC_u - ANC_{le(crit)} \qquad (3)$$

If $Ex = 0$, we call S_{dep} and N_{dep} critical loads of S and N; if $Ex > 0$, we say critical loads are exceeded and if $Ex < 0$, critical loads are not exceeded. Note, that critical loads of S and N are not unique: any pair of S_{dep} and N_{dep} leading to $Ex = 0$ is a (pair of) critical load(s).

Furthermore, N processes can be deposition-dependent, e.g., denitrification:

$$N_{de} = \begin{cases} f_{de}(N_{dep} - N_i - N_u) & \text{if } N_{dep} > N_i + N_u \\ 0 & \text{otherwise} \end{cases} \qquad (4)$$

where f_{de} ($0 \le f_{de} \le 1$) is the so-called denitrification fraction. Inserting Eq.4 into Eq.2 yields

$$Ex = S_{dep} + (1 - f_{de})N_{dep} - CL_{max}(S) - (1 - f_{de})CL_{min}(N) \qquad (5)$$

with the abbreviation

$$CL_{min}(N) = N_i + N_u \qquad (6)$$

For $Ex = 0$ we obtain a function in the (N_{dep}, S_{dep})-plane, the so-called **critical load function** (see Fig.1). For $N_{dep} < CL_{min}(N)$, S_{dep} can be considered alone, and the maximum critical load for S is given by $CL_{max}(S)$. The maximum critical load for N is obtained by setting $S_{dep} = 0$ in Eq.5 and solving $Ex = 0$:

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$$CL_{max}(N) = CL_{min}(N) + \frac{CL_{max}(S)}{1-f_{de}} \quad (7)$$

In addition to the acidifying aspect of nitrogen, the effects of N deposition on the nutrient status (eutrophication) of an ecosystem should be considered when determining critical loads. A critical load for nutrient nitrogen, $CL_{nut}(N)$, is derived from the mass balance for N by defining an acceptable N leaching, $N_{le(acc)}$:

$$CL_{nut}(N) = N_i + N_u + \frac{N_{le(acc)}}{1-f_{de}} \quad (8)$$

As mentioned above, single critical loads of S and N cannot be defined, and consequently there is no single exceedance of a critical load. This is best illustrated by an example (see Fig.1). Let the point E1 denote the current deposition of S and N; reducing N_{dep} substantially, one reaches the point Z1 and therefore non-exceedance without reducing S; on the other hand, one can reach non-exceedance by only reducing S_{dep} till reaching Z3; finally, with a smaller reduction of both S_{dep} and N_{dep} one can reach non-exceedance as well (e.g. point Z2). In practice, external factors, such as the costs of emission reductions, will determine which path will be followed to reach zero exceedance.

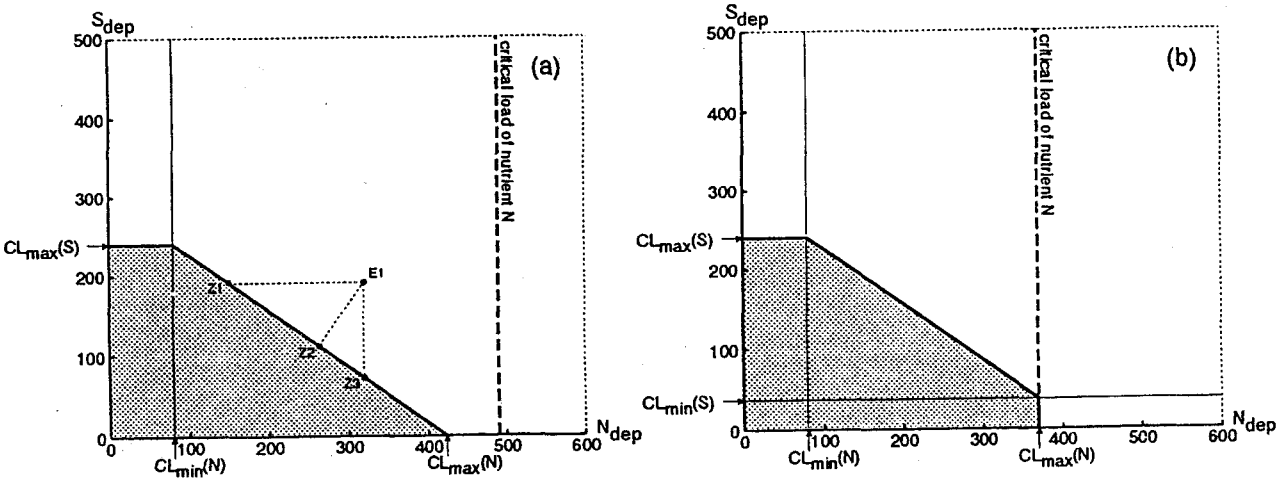


Figure 1: Relationship between nitrogen and sulfur deposition and the exceedance of critical loads for a hypothetical ecosystem. Also shown is the critical load of nutrient nitrogen (dashed vertical line) for two cases: (a) $CL_{nut}(N) \geq CL_{max}(N)$ and (b) $CL_{nut}(N) < CL_{max}(N)$.

The critical load of nutrient N can easily be incorporated into the critical load function. Since this critical load is independent of the sulfur deposition, it can be represented by a line parallel to the S deposition axis. In relation to the critical loads function for acidity, two cases for the critical load for nutrient N are possible: (a) $CL_{nut}(N) \geq CL_{max}(N)$, in this case the nutrient critical load is not binding (Fig.1a), or (b) $CL_{nut}(N) < CL_{max}(N)$, in which case $CL_{nut}(N)$ determines the maximum allowable N deposition (Fig.1b); in the latter case the sulfur critical load is given by

$$CL_{min}(S) = CL_{max}(S) - N_{le(acc)} \quad (9)$$

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Conditional critical loads of N and S

If critical loads are to be used in an analogous fashion as for the Oslo Protocol negotiations, a procedure is required for deriving (a) single number(s) for every ecosystem.

Future negotiations on nitrogen abatement may not involve further S deposition reductions in view of the recent completion of the new Sulphur Protocol. In this event the critical load functions can serve as a valuable tool for determining critical loads of nitrogen for a *fixed* S deposition. By intersecting the critical load function for each ecosystem with a chosen sulfur deposition S_{dep} the *conditional critical load* for nitrogen, $CL(N|S_{dep})$, is obtained and computed as follows:

$$CL(N|S_{dep}) = \begin{cases} CL_{min}(N) & \text{if } S_{dep} \geq CL_{max}(S) \\ CL_{min}(N) + \dfrac{CL_{max}(S) - S_{dep}}{1 - f_{de}} & \text{if } CL_{min}(S) \leq S_{dep} < CL_{max}(S) \\ CL_{nut}(N) & \text{if } S_{dep} < CL_{min}(S) \end{cases} \quad (10)$$

Note, that if $CL_{nut}(N) > CL_{max}(N)$, the third line in Eq.10 becomes superfluous. Similarly, a conditional critical load for sulfur, $CL(S|N_{dep})$, is computed as

$$CL(S|N_{dep}) = \begin{cases} 0 & \text{if } N_{dep} \geq CL_{max}(N) \\ CL_{max}(S) - (1 - f_{de})\left(N_{dep} - CL_{min}(N)\right) & \text{if } CL_{min}(N) \leq N_{dep} < CL_{max}(N) \\ CL_{max}(S) & \text{if } N_{dep} < CL_{min}(N) \end{cases} \quad (11)$$

Note, that $CL(S|N_{dep})$ does not depend on the value of $CL_{nut}(N)$, which is obvious since S is acidifying only. In Fig.2 the calculation of conditional critical loads is depicted for the various deposition values.

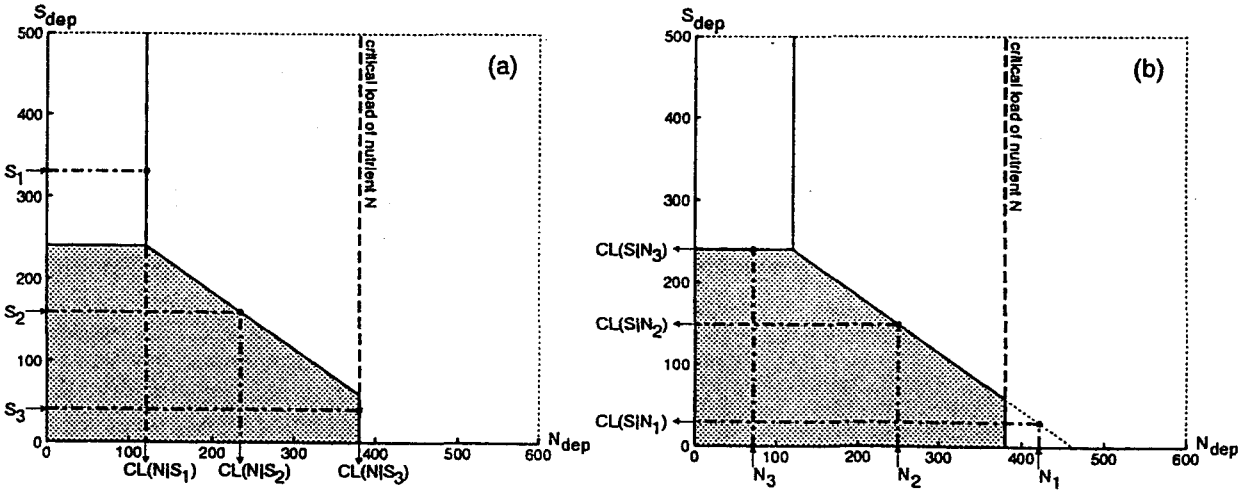


Figure 2: (a) Computing conditional critical loads of nitrogen $CL(N|S_1)$, $CL(N|S_2)$ and $CL(N|S_3)$ for different sulfur deposition values S_1, S_2 and S_3 (see Eq.10); (b) computing conditional critical loads of sulfur $CL(S|N_1)$, $CL(S|N_2)$ and $CL(S|N_3)$ for different nitrogen deposition values N_1, N_2 and N_3 (see Eq.11).

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Percentiles and protection isolines

For presenting the spatial variation of critical load values, such as $CL(N|S_{dep})$ or $CL(S|N_{dep})$, the cumulative distribution function of all values within a chosen grid cell is constructed and then a percentile, i.e. a protection percentage, is chosen as the representative critical load for that grid cell. Presentation of results and communication with non-technical users (policy makers) is easy in such cases, but the inherent flexibility of the critical load function, allowing to determine (cost-)optimal reductions considering both N and S emissions, is lost.

In order to be useful for the analysis of emission reduction strategies involving both N and S, an aggregation procedure is required for deriving a single function for every grid cell from all the critical load functions. This is done in the following way: for every pair of N and S deposition one checks, whether it lies below or above the given critical load functions, thus determining the percentage of protected ecosystems. Connecting those points in the (N_{dep},S_{dep})-plane, which protect a prescribed percentage p of ecosystems, yields the $(100-p)$-percentile function (or *protection isoline*). The computational procedure for obtaining a protection isoline is described in Posch et al. (1995), and an example is shown in Fig.3.

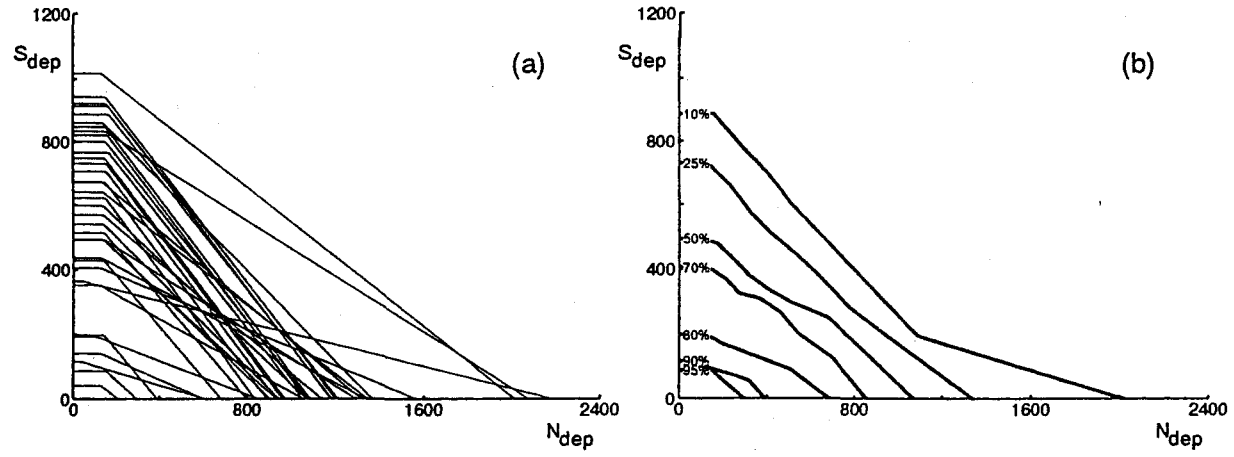


Figure 3: (a) Critical load functions (n=36) within a grid cell, and (b) non-intersecting protection isolines derived from them with the procedure outlined above (see also Posch et al. 1995).

A single function, e.g. the 5-th percentile function (=95% protection isoline), replaces the individual critical load functions in the same way as a percentile is used as a surrogate for a set of critical load values within a grid cell. The selected percentile function for each grid cell can then be linked to models which (cost-)optimize emission reductions for S and N. Both the costs of the individual abatement strategies and the shape of the percentile functions determine the optimal emission reduction strategy and thus the allocation of S and N deposition.

When comparing the nitrogen and sulfur deposition in a grid cell with a protection isoline, five cases can arise:

(0) The point (N_{dep},S_{dep}) lies below the percentile function, i.e. no exceedance;
(1) Reductions in N_{dep} or S_{dep} are interchangeable, i.e. non-exceedance can be achieved by either N reductions or S reductions alone;
(2) Some reductions in S deposition are mandatory;
(3) Some reductions in N_{dep} are mandatory;
(4) Both reductions in N and S are required to achieve non-exceedance.

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In Fig.4 it is shown which case occurs in each EMEP grid cell for the 5-th percentile protection isoline, comparing it with the 1990 sulfur and nitrogen deposition. In producing Fig.4 the protection isoline for S and acidifying N has been intersected with the 5-th percentile of $CL_{nut}(N)$ in every grid cell, in this way considering both effects of nitrogen.

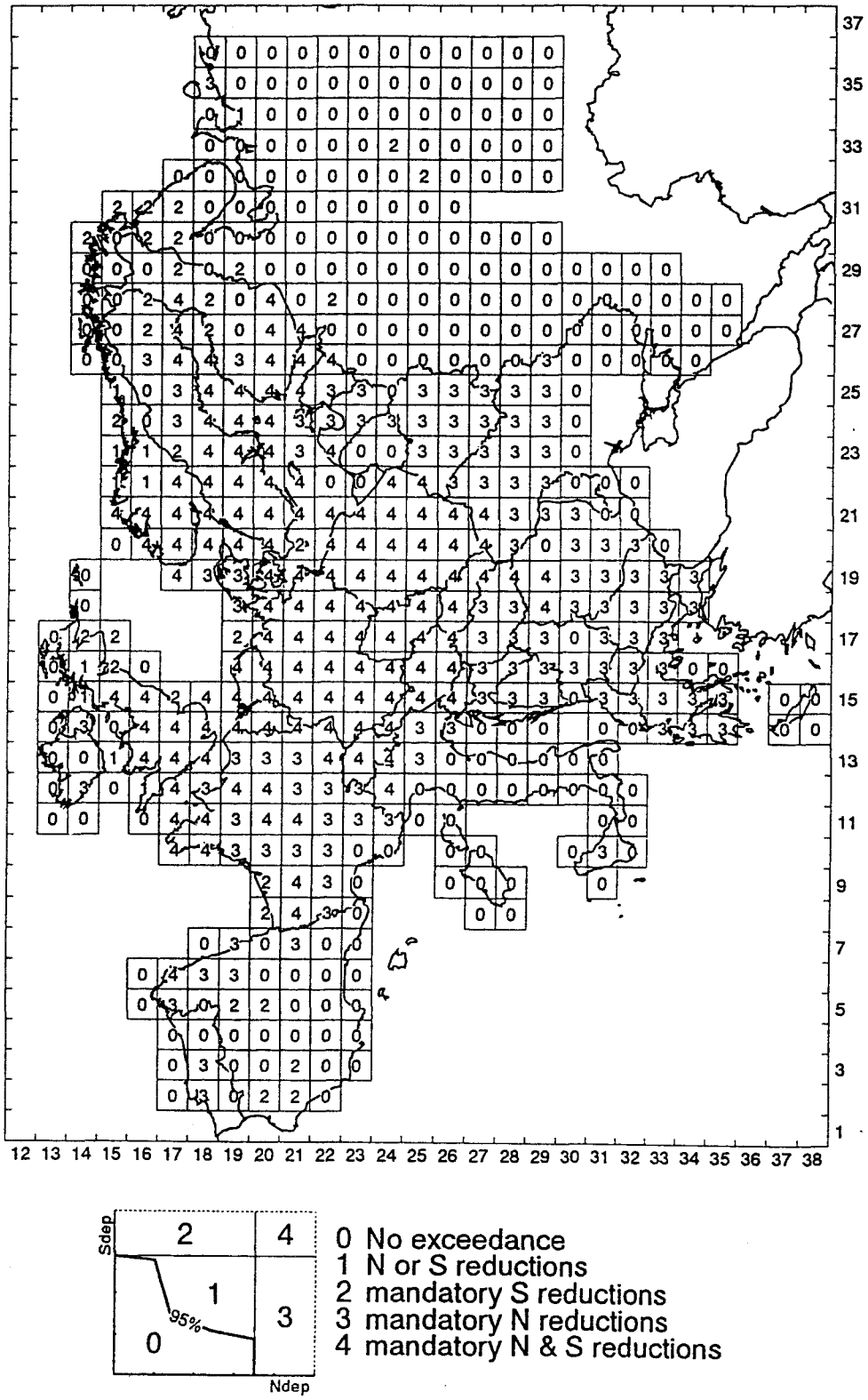


Figure 4: Type of nitrogen and/or sulfur reductions required in each EMEP grid cell when comparing the 5th percentile function (95% protection isoline) with the 1990 sulfur and nitrogen deposition (preliminary results).

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Concluding Remarks

Generalizing the notion of a percentile to critical load functions, ecosystem protection isolines can be computed for each grid cell in Europe reflecting combinations of N and S depositions at which no exceedance occurs of both the critical load of acidity and eutrophication. This methodology allows a flexible allocation of emission reductions taking into account abatement costs. Work remains to be done on procedures for including other effects of N, such as ozone, or other pollutants.

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GENERALISATION OF DEPOSITION IN EUROPE

Jan Willem Erisman, Erik van Leeuwen, Addo van Pul and Geert Draaijers

National Institute of Public Health and the Environment (RIVM)
P.O. Box 1
3720 BA Bilthoven
The Netherlands

Abstract In describing the effects of acidification on the level of ecosystems, acid loads should be available at least on the size of ecosystems. Until now no deposition maps on this resolution are available, hampering accurate estimation of exceedances of critical loads in Europe. Current methods and models for estimating deposition of acidifying components differ in the used horizontal resolution i.e. varying from 5x5 km up to 150x150 km. These include inferential models applied in different countries of Europe, where enough measurements of concentrations and meteorological parameters are available. For Europe as a whole long-range transport model results based on emission estimates (EMEP, TREND) have been available. Too few measurements are available in Europe to apply the inferential method for deposition mapping. A so called merging method was developed, the EDACS model, combining long-range transport modelling and inferential methods to map deposition of acidifying components on a small scale of Europe. Maps over small scale fluxes of Europe with this method are presented in this review. The maps are produced in close co-operation with EMEP. Base cation deposition has not been considered up until now, although several regions can be found in Europe where base cation deposition is important in relation to acidification in a possitive sense. In this background report a method to determine wet and dry base cation deposition and preliminary results are given.

1. Introduction

1.1 Historical Background on long-range transport

The first hypotheses of long-range transport of atmospheric components (see Erisman and Draaijers, 1995 for an overview) were related to the sea as a source of chloride in rain and at the surface. Maggraf already hypothesised in 1750 that the chloride he found in a precipitation sample must have come from the ocean. Meyrac (1852) in France showed that NaCl could be transported over long distances, with the ocean as the source. Smith (1872) stated: *'One of the uses of storms seems to be to supply the world with salt'*. That components could travel over long distances was also known because Sahara dust and components emitted by volcanoes, which must have been transported over at least 2500 km through the air were identified in several places in Europe. Ehrenberg (1850) demonstrated that material which he investigated by his microscope must have been originating from the eruption of the volcano St. Vincent in 1812. Ehrenberg hypothesised that material not originating from volcanoes, originates from dust transported from south America by south-westerly winds to Europe (cf. Ludwig, 1862). Brogger (1881) suggested dirt on snow observed in Norway was the result of dust emissions from Great Britain (Last, 1991). In the Belgium Commission report on damage to vegetation (Rapport à M. le Ministre de l'intérieur par la commission d'enquête, Bruxelles, 1856), so-called 'safe distances' were given for the relation between damage and factories, suggesting transport of pollutants. Smith (1872) discussed these distances and hypothesised that they should be a function of wind direction and wind speed, and should not only be related to hydrochloric acid production, but also to smoke stacks producing sulphuric acid.

The first maps of sulphur deposition over large regions, the UK in this case, were made by Meetham (1950). He estimated sulphur and chloride deposition in the UK as the result of

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smoke emission. The first maps of deposition in Europe were made by Enger and Eriksson (1955). Acidification as a result of deposition of sulphur became of interest in the early sixties (Odén, 1960). In the eighties the concern about the excess nitrogen deposition and its contribution to acidification was recognised as an environmental threat (e.g. Van Breemen *et al.*, 1982). It was only 15 to 20 years ago that air pollution was assumed capable of causing effects not only close to sources. The most feared effects were those to humans because they were exposed to the highest concentrations of pollutants near to source areas (industry, cities). It was thought that the atmosphere was large enough to deal with the relatively low concentrations of pollutants emitted by anthropogenic activity. Furthermore, it was believed that the cleaning capacity of the atmosphere was thus that pollutants would be degraded fast and effectively. The only concern was therefore to keep the concentrations near sources as low as possible. Therefore, high chimneys were built to overcome local pollution. Unfortunately it was not foreseen that long-range transport of pollutants could lead, for instance, to acidification of Scandinavian lakes and destruction of the ozone layer.

In Scandinavia scientists hypothesised that the acidification of their lakes was due to acid rain mainly caused by emissions other than their own. To determine whether this was possible, a long-range transport model was developed. This has led to the development of the EMEP (European Monitoring and Evaluation Programme) langrangian long-range transport model which is used under the UN ECE Convention on Long Range Transboundary Air Pollution to determine the concentration and deposition distribution over Europe, to determine the budgets for countries and to provide the so-called blame matrices (Eliassen and Saltbones, 1983; Iversen *et al.*, 1991; Tuovinen *et al.*, 1994). Deposition was treated in this model only as a loss-term. It was thought that the atmospheric deposition had to be known only for large regions as long-term averages. It became clear recently that deposition variation in time and space is very large, and that for determining the relation between effects and deposition, receptor specific estimates are needed (Erisman and Draaijers, 1995).

The long-range transport modelling is well developed during the past twenty years. The emphasis of the studies was on determining the large scale transport and deposition of pollutants. The local scale estimation of deposition was not addressed.

1.2 Introduction to the issue of generalisation of deposition

Despite the large efforts which have been put in deposition research and measurements during the last decades, it is not possible to derive deposition maps entirely based on deposition monitoring results. This is caused by the stochastic nature of the dry deposition. On an annual basis, wet deposition can be treated as smoothed fields over regions. By installing a monitoring network with enough samplers to cover the spatial gradients, wet deposition can be monitored and wet deposition maps can be derived from interpolated annual averaged data. For several years now this is done on national levels (e.g. RIVM, 1994, UK RGAR, 1990, Lövblad *et al.*, 1993; see Van Leeuwen *et al.*, 1995 for an overview). For Europe as a whole it is currently possible to obtain a reasonable coverage with all available national and European monitoring networks. Van Leeuwen *et al.* (1995) compiled European wet deposition maps of different components of Europe using monitoring results of about 800 sites.

Contrary to wet deposition, dry deposition maps cannot be obtained from interpolating long-term dry deposition measurements. There is a serious lack of monitoring data. Currently dry deposition of SO_2 and O_3 is monitored at 2 sites, in Portugal and Spain (EC SREMP project),

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SO_2 and NH_3 at the Oostvaardersplassen in the Netherlands (Mennen *et al.*, in prep.; Wyers *et al.*, in prep.) and SO_2, NH_3 and NOx at three sites, in Scotland, Germany and the Netherlands (EC LIFE project). This makes only six monitoring sites in Europe, with only one forested area. There are three ways of deriving dry deposition maps, i.e. using measurements of concentrations, meteorological parameters and surface condition parameters from which the dry deposition flux can be inferred (inferential technique, Hicks *et al.*, 1987); using emission estimates and long-range transport model (LTRAP) calculations; or using a combination of the two (Erisman and Draaijers, 1995). This background document will provide some information on these three methods, but will mostly focus on the combination method, the merging technique (Van Pul *et al.*, 1992; 1995). The main results will be highlighted here.

With regard to acidification, usually little emphasis is placed on the role of base cations such as Na^+, Mg^{2+}, Ca^{2+} and K^+ in relation to acid deposition. Base cations play an integral role in the chemical processes of acid deposition since the acidity of any material is a function of both its acidic and basic compounds. Besides their ability to neutralize acid input, base cations are important nutrient elements for ecosystems. The depletion of exchangeable base cations in sensitive soils is currently thought to represent a major detrimental effect of acid deposition on terrestrial ecosystems (De Vries, 1994). Atmospheric input may be a quantitatively important source of base cations to vegetation in nutrient-poor conditions. Up to now, no reliable information is available on the spatial pattern of base cation deposition over Europe (Draaijers *et al.*, 1995). There is lack of knowledge on the spatial and temporal variation of emissions and concentrations, hampering accurate deposition mapping. Here, a method is proposed to estimate local-scale deposition fluxes based on the inferential modeling technique, as presented in Draaijers *et al.* (1995). In this background document the method is explained and preliminary results of local scale deposition fluxes are presented.

2. Long-range transport modelling

The way deposition is modelled depends on its purpose. It may be considered as a loss term in long-range transport modelling, or as an input term in determining the deposition to ecosystems. For the latter it is important to consider the scale on which the deposition is desired. If the scale is comparable to the scale considered in the long-range transport (LTRAP) model, the deposition estimates of this model can be used directly. But if the desirable scale is much smaller than that considered in the LTRAP model, sub-grid processes should be taken into account.

Modelling the dispersion of trace gases in the atmosphere, including their physical and chemical transformations, is an essential element of the general study of trace gas behaviour and in the determination of the functional relationships between emissions and concentration or deposition levels. Measurements and models are strongly intertwined. On the one hand, measurements are necessary for parametrisation and validation of models, while on the other model results may render support in the evaluation, generalisation or extrapolation of measurements. Dispersion models exist in all sizes and complexities. The choice for a model or model concept is determined by the spatial and temporal scale on which air quality and deposition estimates have to be made (cf. Van Dop, 1986). In general, the models can be divided into different categories according to their mathematical framework: Eulerian models with a fixed co-ordinate system, Lagrangian models, where the co-ordinate system moves with the representative parcel of air or hybrid models merging both concepts. Here we will describe

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two Lagrangian-type LTRAP models that are used to determine the annual spatial distribution of pollution in Europe. These are the EMEP (Eliassen and Salbones 1983, Iversen *et al.*, 1991) and OPS (Operationeel Prioritaire Stoffen model, Van Jaarsveld, 1990; 1995) models. Several Eulerian-type of models were developed during recent years, but most of them have not been used to map annual deposition in Europe. In the USA the RADM (Regional Acid Deposition Model) model is applied for deposition mapping (Chang *et al.*, 1987). The EURAD (European Acid Deposition Model) model, was designed to simulate the transport, chemical transformation and deposition of atmospheric pollutants over Europe (Jakobs, 1994). Until now this model has not been applied for mapping annual deposition fluxes. Several studies have been done simulating episodes. EMEP is currently involved in developing a Eulerian type of model for application of mapping annual deposition of Europe (Jacobson *et al.*, 1995; Johnson and Berge, 1995). This model is under development and has not yet been tested enough for application purposes and will therefore not be described here. In the future, however, it is expected that this model will replace the current Lagrangian EMEP model.

The deterministic Lagrangian EMEP model is a receptor-oriented one-layer trajectory model originally developed at the Norwegian Meteorological Institute (Eliassen and Saltbones, 1983) and operationally applied in the framework of the 'Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe'. The European Monitoring and Evaluation Programme (EMEP) under the UN ECE Convention on Long Range Transboundary Air Pollution, consists of a monitoring programme and modelling of atmospheric transport, conversion and deposition. The aim of EMEP is to provide basic information about emissions, levels of concentrations and deposition. The model calculates 96-h trajectories in reverse time on the basis of the 850 hPa wind field from the arrival of an air parcel at a receptor. Receptor points are the grid points of a 150 x 150 km grid system covering Europe. The trajectories arrive at the receptor points four times a day. During the air parcel's travel along the trajectory, the gridded sulphur and nitrogen emissions are immediately mixed in the vertical up to the top of the constant mixed layer. The model is used every year to estimate sulphur and nitrogen pollution in Europe and to determine the country-to-country receptor matrices and country budgets (e.g. Tuovinen *et al*, 1994). The model has been revised during the last year in order to include the resistance analogy to determine land use specific dry deposition velocities (cf. Erisman *et al.*, 1994; Sandnes, 1995). Furthermore, the model resolution has been increased from 150x150 km to 50x50 km. Land use and resistance parametrisation have been incorporated in close cooperation with RIVM (van der Velde *et al.*, 1995 and Erisman *et al.*, 1994, respectively).

The statistical transport model OPS is able to describe both short- and long-distance transport, and average concentrations and depositions can be computed for time scales ranging from 1 day to more than 10 years (Van Jaarsveld, 1990; 1995). It can account for both point sources of various heights and area sources of various shapes and heights. The sources need not be distributed on a regular grid system. It yields realistic results both within area sources and near point sources, as well as at long distances from sources. The receptor system is therefore determined by the resolution of the emissions. The concentrations and depositions in Europe are described on a 50 x 50 km grid system, whereas those in the Netherlands are estimated on a 5 x 5 km grid system. Computations are made for a limited number of meteorological situations (classes) with a representative meteorology for each class. Among the discretisations, a total of 12 wind-direction sectors and 6 atmospheric stability classes are distinguished. The basis for the model is formed by the Gaussian plume formulation for a point source. It is assumed that the plume is reflected only once at the surface and at the top of

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the boundary layer. Moreover, it is assumed that at larger distances from the source the plume is vertically distributed homogeneously over the whole boundary layer, apart from an attenuation near the surface due to dry deposition (Van Jaarsveld, 1994; Asman and Van Jaarsveld, 1992).

3. Deposition modelling

There is a difference between deposition modelling and long-range transport modelling, i.e. the focus of the two is different: deposition modelling is aimed at estimating the input to ecosystems as accurately as possible; long-range transport modelling, on the other hand, is aimed at determining the distribution of pollutants (concentration and deposition) based on emission estimates and dispersion calculations. In long-range transport modelling, deposition is a loss term in which the accuracy needed depends on the modelled scale. When both the pollution distribution and the deposition needs to be known as accurately as possible, a compromise is required on the detail the main processes determining transport and deposition are modelled. Methods merely based on measurements and on a merge between measurements and LTRAP model results can be distinguished in deposition modelling.

3.1 Inferential technique

As an alternative to the direct measuring methods, knowledge on dry deposition processes can be used to infer dry deposition fluxes from basic information on routinely measured air concentrations and meteorological parameters. The flux is inferred as the product of ambient concentration of the chemical of interest and its dry deposition velocity. The dry deposition velocity is derived using a multiple-resistance transfer model (Hicks *et al.*, 1987). The resistance model provides a framework for coupling individual processes, some being surface dependent and/or pollutant dependent (Wesely, 1989. Erisman *et al.*, 1994; Erisman and Draaijers, 1995). The three major resistances are the aerodynamic resistance R_a, a near surface boundary resistance R_b, and the surface resistance R_c.

Flux measurements on a routine basis using the inferential technique are made in two networks in the USA (Hicks *et al.*, 1991; Clarke *et al.*, 1992). Weekly mean measurements of HNO_3, SO_2, NO_3^-, SO_4^{2-} and NH_4^+ concentrations in these networks are combined with meteorological parameters to obtain weekly average fluxes (Hicks *et al.*, 1985). This technique has also been applied in studies done in the UK (UK Review Group on Acid Rain, 1990), in Sweden (Lövblad *et al.*, 1991) and in the Netherlands (Erisman *et al.*, 1989; Erisman, 1992; 1993).

The quality of results obtained by this method depends on the availability and quality of data, and on the description of the resistances. R_a and R_b can be estimated from measurements of wind speed and radiation and information on surface roughness, together with parametrisations given by Garland (1977) and Wesely and Hicks (1977). R_c values can be obtained from flux measurements and from chamber methods. The most important parameters determining R_c should be included on a relevant time and spatial scale (see also section 4).

Eder and Dennis (1990) developed a technique to allow estimation of the annual and monthly dry deposition of Ca^{2+}, Mg^{2+}, Na^- and K^-. The technique is based on the strong correlation between concentrations within precipitation and the surface-level air at 23 stations in Ontario,

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Canada. This method has been applied in the inference modelling of base cation deposition over the Netherlands by Erisman (1995), and over Europe by Erisman and Draaijers (1995) and Draaijers *et al.* (1995).

3.2 Merging technique

In applying abatement strategies on a national or European scale, it is desirable to know both the deposition and the distribution of pollution as accurately as possible. When emission abatement strategies are based on the critical load concept, the deposition has to be determined on the ecosystem level. Desired emission reductions based on the target levels should be known per country and/or per individual source or activity. Therefore, long-range transport modelling and ecosystem-scale deposition modelling should be linked. The deposition model which was developed within the framework of BIATEX is the EDACS model (Estimation of Deposition of Acidifying Components in Europe, Van Pul *et al.*, 1995). Wet deposition in this model is estimated using interpolation of wet deposition measurements (Van Leeuwen *et al.*, 1995). Dry deposition is determined using the inferential technique (Hicks *et al*, 1987). The dry deposition flux is inferred from LTRAP concentrations and land-use-specific dry deposition velocities.

3.3 Framework for the description of atmosphere - surface exchange

A common framework has been provided to describe the exchange of a range of gases and particles with very different chemical and physical properties. The most widely used is that of the resistance analogy (Chamberlain, 1966). In this framework, three stages in dry deposition to the surface are distinguished. The flux of a trace gas is given as:

$$F = V_d(z)\, c(z) \qquad [1]$$

where $c(z)$ is the concentration at height z and V_d is the dry deposition velocity; z is the reference height above the surface. If the surface is covered with vegetation, a zero-plane displacement is included: $z=z-d$. d is usually taken as 0.6-0.8 times the vegetation height (Thom, 1973). The absorbing surface is often assumed to have zero surface concentration and the flux is therefore viewed as being linearly dependent on atmospheric concentration. This holds only for depositing gases and not for gases that might also be emitted, such as NH_3 and NO. For these gases a non-zero surface concentration, a compensation point c_p, might exist, which can be higher than the ambient concentration, in which case the gas is emitted. For these gases the flux is estimated as:

$$F = V_d(z)\, [c(z) - c_p] \qquad [2]$$

V_d is widely used to parametrise gas uptake at the ground surface. V_d is the inverse of three resistances:

$$V_d(z) = \frac{1}{R_a(z-d) + R_b + R_c} \qquad [3]$$

The three resistances represent the three stages of transport, the aerodynamic resistance R_a for the turbulent layer, the laminar layer resistance R_b for the quasi-laminar layer enveloping receptor surfaces and the surface or canopy resistance R_c for the receptor itself. The atmospheric resistance to transport of gases across the constant flux layer is assumed to be similar to that of heat (e.g. Hicks *et al.*, 1989). R_a is approximated following the procedures used by Garland (1978), while the quasi-laminar layer resistance R_b can be approximated by the procedure presented by Hicks *et al.* (1987).

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3.3.1 Surface resistances

The surface or canopy resistance, R_C, is the most difficult of the three resistances to describe. R_c values can be obtained from theoretical considerations based, for instance, on solubility and equilibrium calculations in combination with simulations of vegetation specific processes, such as accumulation, transfer processes through stomata, mesophyll and cuticles, absorption, etc. (Baldocchi *et al.*, 1987; Wesely, 1989). R_c values presented in the literature are primarily based on field measurements of V_d and on chamber studies. By determining R_a and R_b from the meteorological measurements, R_c can be calculated from V_d measurements as the residual resistance using Equation [3]. Values of R_c can then be related to surface conditions, time of day, etc., yielding paramtrisations. For use in a generalisation framework R_c parametrisations must be based on routinely available data, i.e. ambient temperature, relative humidity, global radiation, wind speed, land use class, type of soil and vegetation. In the next section the derivation of R_c values from experimental and modelling work, a.o. conducted within EUROTRAC, is explained. The parametrisation is extensively described in Erisman *et al.* (1994). The basic assumptions are only briefly discussed here, and some example values resulting from application of the parametrisation under different conditions are given.

The surface resistance of gases consists of a series of resistances reflecting different pathways of uptake. They are either determined by the actual state of the receptor, or by a memory effect. R_c is a function of the canopy stomatal resistance R_{stom} and mesophyll resistance R_m; the canopy cuticle or external leaf resistance R_{ext}; the soil resistance R_{soil} and in-canopy resistance R_{inc}, and the resistance to surface waters or moorland pools R_{wat}. In turn, these resistances are affected by leaf area, stomatal physiology, soil and external leaf surface pH, and presence and chemistry of liquid drops and films. The stomatal resistance, external leaf surface resistance and soil resistance act in parallel:

$$\textit{vegetative surface: } R_c = \left[\frac{1}{R_{stom} + R_m} + \frac{1}{R_{inc} + R_{soil}} + \frac{1}{R_{ext}}\right]^{-1}$$

$$\textit{water surfaces: } R_c = R_{wat} \qquad [4]$$

$$\textit{bare soil: } R_c = R_{soil}$$

$$\textit{snow cover: } R_c = R_{snow}$$

In Table 1 values of surface resistances obtained with the parametrisations presented in Erisman *et al.* (1994) are given for three different pollution climates as examples.

SO_2

The deposition of SO_2 on vegetation has been shown to be regulated mainly by stomatal resistance and the presence of surface water on the foliage. The stomatal uptake can be modelled satisfactorily. However, an understanding of the variations of surface resistance due to surface wetness chemistry is limited. It is assumed here that when the surface is wet, deposition of SO_2 is regulated by external leaf uptake. At low SO_2 concentrations, the buffering capacities of vegetation leaves are probably sufficient to maintain low R_c values. At high SO_2 concentrations other neutralising components, such as NH_3, might become important. The surface wetness may be caused by increased relative humidity, dew, guttation, rain or fog, and cloud water deposition. Deposition of SO_2 to soil is dependent on the soil conditions (snow cover, frozen), relative humidity and on the soil pH.

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NH_3

NH_3 is emitted from fertilised soils and from pasture during grazing. In addition, emission takes place from crops, ungrased pasture and semi-natural vegetation under dry, warm conditions during daytime at low ambient concentrations. In other cases NH_3 is mainly deposited. Deposition in these cases is regulated by stomatal uptake and by deposition to external leaf surfaces. R_c values for semi-natural vegetation and forests are low (in the range of 0-50 s m^{-1}), and depend on surface wetness.

TABLE 1 Examples of averaged surface resistances (s m^{-1}) for different gases during daytime under different pollution climates, with the stomatal resistance (R_{stom}) also given

	Moist (rain or *rh*>90%), moderate temperature		Winter, snow covered surface		Dry, warm summer (*rh*<60%), well watered soil	
	Coniferous forest	Grassland	Coniferous forest	Grassland	Coniferous forest	Grassland
R_{stom}	200	60	400	-	150	50
SO_2	0#	0#	500	500	300	100
NH_3	0#	0#	500	500	500*	500*
NO_2	320	100	640	1000	240	80
HNO_2	0# (?)	0# (?)	500	500	300	100
HNO_3	0#	0#	10	50	0#	0#
HCl	0#	0#	10	50	0#	0#
PAN	500	500	1000	1000	500	500
O_3	320	100	640	1000	240	80

* Under these conditions NH_3 is expected to be emitted from the vegetation. The upward flux is estimated with this R_c value in Equation [3].

\# R_c values will be set to 1 s m^{-1} to avoid unrealistically high V_d at low values of R_a and R_b.

NO_X

NO is not taken into account here as a depositing pollutant. Deposition of NO_2 is mainly regulated by stomata. The deposition to external leaf surfaces, water surfaces and soils is one order of magnitude less than stomatal uptake.

PAN, HNO_2, HNO_3, HCl

PAN deposition is slow with an overall surface resistance of 500-1000 s m^{-1}. HNO_2 is treated as SO_2 until more information becomes available. Thus when the surface becomes moist, uptake of HNO_2 is regulated by stomata and external leaf surfaces. HNO_3 and HCl deposit efficiently to all surfaces with negligible R_c values. Only at low temperatures with a surface covered with snow does R_c increase up to 50-100 s m^{-1}

O_3

The uptake of ozone by vegetation is mainly via the stomata. The resistance of the remaining plant parts (the cuticle) to ozone is found to be larger than 1000 s m^{-1}. Ozone is destroyed slowly at a wetted surface or free water due to its low water solubility, and its destruction may depend on the chemical content of the water. The surface resistance to ozone deduced from measurements above water and snow amounts to about 1000-2000 s m^{-1}. The soil resistance to ozone is largely dependent on the type of soil and the soil water content. The soil resistance is typically 100 s m^{-1} under dry soil conditions. This value increases typically up to 500 s m^{-1} at a waterlogged stage of the soil (20-30% water content).

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3.3.2 Particles

The resistance analogy is not used for particles. Transport from the free atmosphere to the receptor surface is more-or-less similar to gas transport. However, transport processes through the quasi-laminar layer differ considerably between gases and particles. Whereas gases are transported primarily through molecular diffusion, particle transport and deposition basically take place through sedimentation, interception, impaction and/or Brownian diffusion. The currently available particle deposition models cannot be used for generalisation because most of the input data are not available for different kinds of surfaces. Ruigrok *et al.* (1994) gives an overview of available particle deposition models and ranges in V_d resulting from these models when applied to a specific situation. This range is very large. They simplified the Slinn (1982) model to include all relevant routinely available parameters. Slinn (1982) proposed use of a simplification of his model by distinguishing a turbulent and a surface deposition velocity for forests, or roughness lengths above 0.5 m. The surface deposition velocity depends on the collection efficiency of the surface and thus surface properties:

$$V_d = [V_{dt}^{-1} + V_{ds}^{-1}]^{-1}, \qquad [5]$$

where the surface deposition velocity is given as $V_{ds} = E\ u_*^2/u_h$, with u_h the wind speed at canopy height, E is the collection efficiency and V_dt is the turbulent contribution, represented by the inverse of the aerodynamic resistance. E is different for different size classes as different processes become important. E is determined by parametrisation of modelled values based on data such as u_*, z_0, d, u_{50}, rh, surface wetness, etc. (Ruigrok *et al.*, 1994).

Current models for generalisation, such as DEADM (Erisman, 1993a) and EDACS (van Pul *et al.*, 1995), use a parametrisation of V_d for *low vegetation ($z0 < 0.5$ m)* based on relations of Wesely *et al.* (1985):

$$V_d = \frac{u_*}{300} \quad \textit{for stable conditions and}$$

$$V_d = \frac{u*}{300}\left[1 + \left(\frac{-300}{L}\right)^{2/3}\right] \textit{ for unstable and neutral conditions} \qquad [6]$$

Routinely available data for generalisation include meteorological parameters such as wind speed, radiation, temperature, relative humidity, u_* and L, and land use statistics and $z0$ maps.

4. EDACS

The outline of the EDACS model to estimate local and regional scale deposition fluxes is presented in Figure 1. The basis for the two estimates is formed by calculations with the EMEP long-range transport model. Ambient concentrations of the acidifying components (daily averages) and concentrations in precipitation (monthly averages) calculated with the EMEP model are used along with a detailed land-use map and meteorological observations to estimate small-scale dry deposition fluxes (Figure 1). By using calculated concentration maps, the relationship between emissions and deposition is maintained and scenario studies, budget studies and assessments can be carried out on different scales. Wet deposition is added to the dry deposition to estimate total local scale deposition in Europe. Wet deposition can either be obtained directly from the EMEP model, or from measurements made in Europe. The latter method is used here and will be described in the next section. The method for estimating dry deposition will be explained in section 4.2.

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4.1 Wet deposition

Up to now, wet deposition maps on a European scale are based on long-range transport model results (e.g. the EMEP model; Iversen *et al.*, 1991), whereas for most components wet deposition maps based on measurements are only available on national scales. Van Leeuwen *et al.* (1995) recently used measurements in Europe to estimate the wet deposition in Europe. Concentrations of acidifying components and basic cations were mapped on a 50x50 km scale over Europe for 1989 using results of field measurements made at approximately 800 locations (the number of locations differs per component). Information on concentrations of ions in precipitation in 1989 was obtained from the EMEP database and from organisations responsible for wet deposition monitoring in their countries. Concentrations measured with bulk samplers were corrected for the contribution of dry deposition onto the funnels of these samplers. Sulphate concentrations were corrected for the contribution of sea salt, because only non-marine sulphate contributes to the acidification process. By assuming that the ratio of sodium to sulphate in sea-spray is the same as in bulk sea water, and that all the sodium in a sample is of marine origin, it is possible to calculate non-marine sulphate. At about 200 sites only information about the sulphate concentration was available and the sodium concentration was not measured. At those sites interpolated values from the sodium map were used for correction. Point observations were interpolated to a field covering the whole of Europe using the Kriging interpolation technique. An extensive uncertainty analysis was performed to assess the quality of the maps (see section 4.5).

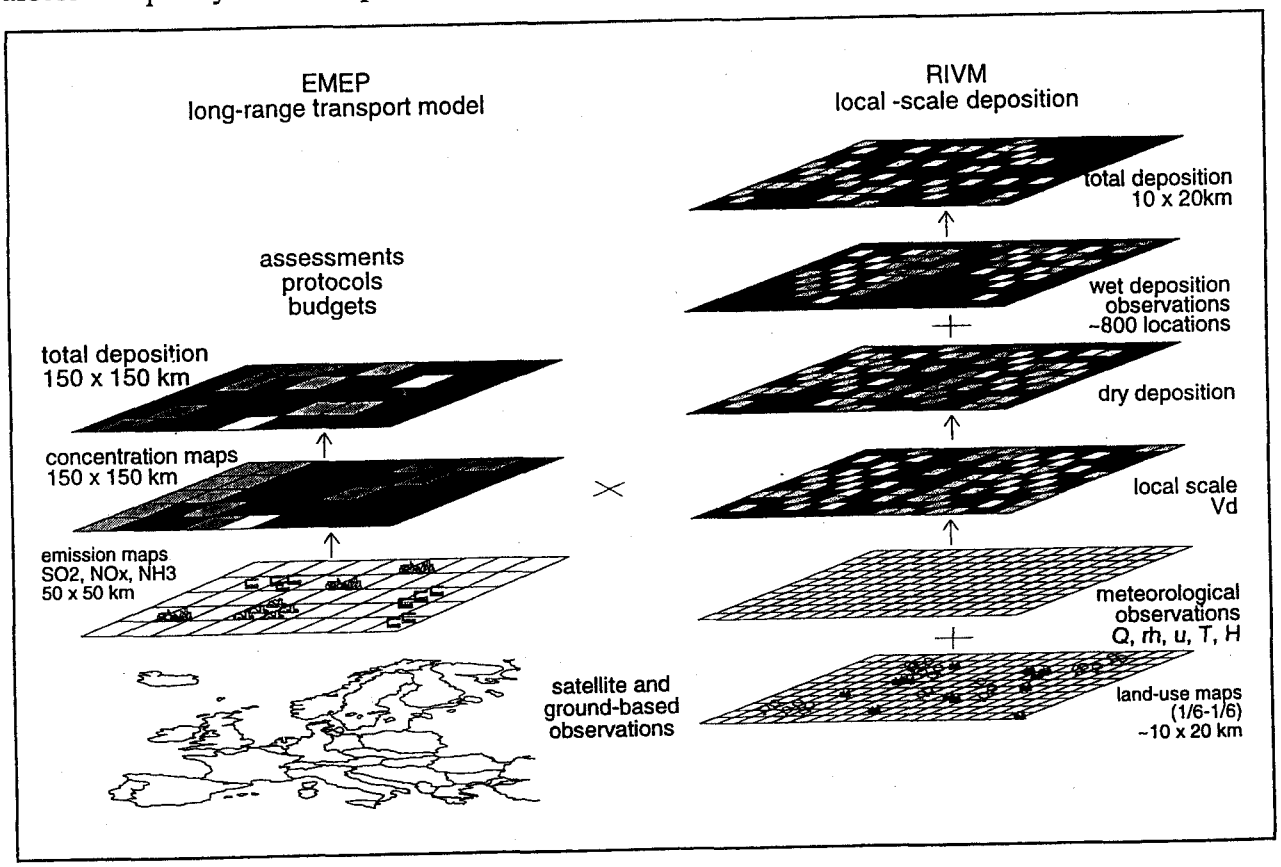


Figure 1. Outline of method to estimate local scale deposition fluxes.

As an example, the wet deposition flux map of non-marine sulphate (Figure 2) is presented. The map clearly resemble European emission and climate patterns. Large emission sources can be recognised in the concentration maps (not shown here), whereas in the flux maps

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climate patterns and orographic effects are also observed. In mountainous areas large fluxes due to orographic rains can be seen. Especially in the former Yugoslavia the large long-term mean precipitation cause large fluxes in all deposition maps. Besides originating from large amounts of rainfall, large fluxes can also arise from high concentrations in precipitation.

Large fluxes (400-1000 mol ha^{-1} a^{-1}) can be observed in Eastern and Central Europe (Germany, Poland, Czech and Slovak Republics, Austria, Hungary, northern Italy, The Ukraine and former Yugoslavia). Large sulphate concentrations are mainly caused by SO_2 emissions from industry and power stations. Concentrations and fluxes obtained in Turkey should be interpreted with care, as they are not based on measurements in Turkey itself but are solely the result of interpolation (in this case extrapolation) from surrounding countries.

Fgure 3 shows the total base cation wet deposition in Europe (van Leeuwen *et al.*, 1995). Total deposition of base cations is calculated as K^++Ca^{2+}+Mg^{2+} (Draaijers *et al.* 1995). Na^+ is not taken into account, because sodium inputs almost entirely originates from sea spray and does not contribute to the neutralisation of acid input. Furthermore, Na^+ does not play a role as a nutrient for ecosystems. Deposition fields are found to resemble the geographic variability of sources, climate and land use (Draaijers *et al.* 1995).

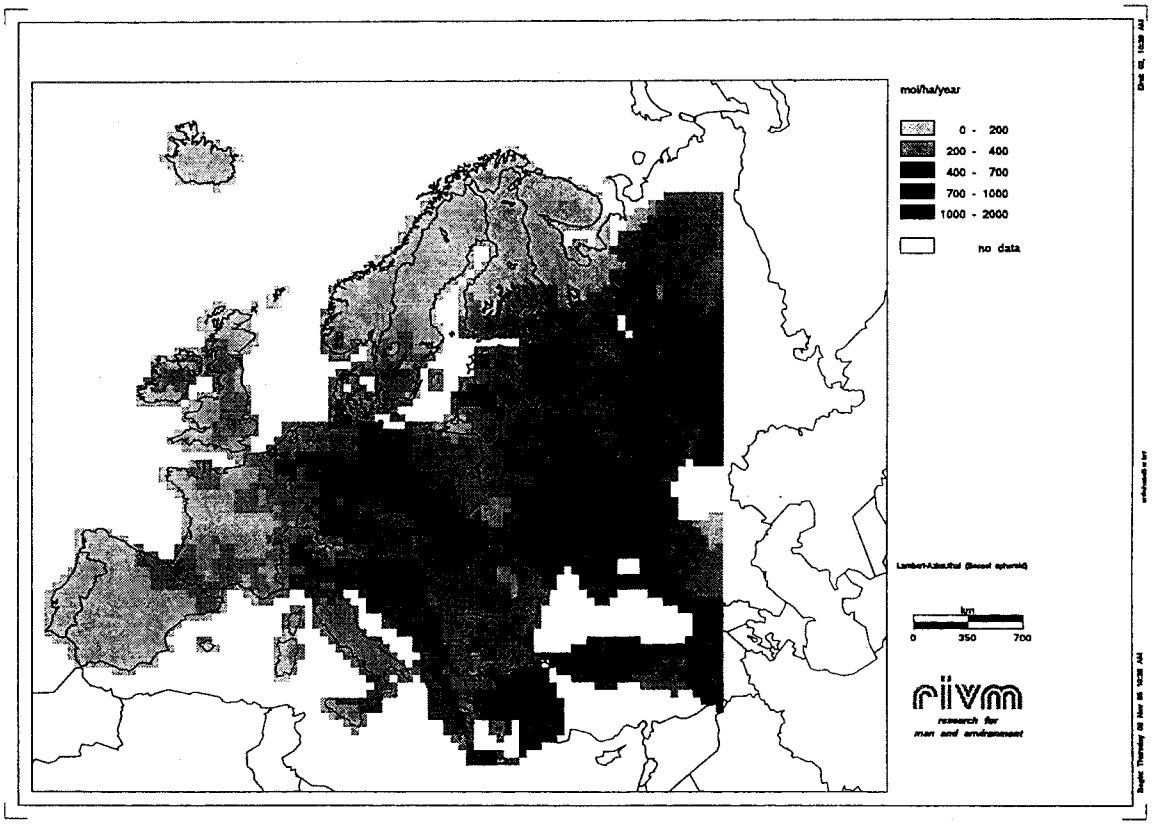


Figure 2. Wet deposition of SO_4^{2-} in Europe on a 50x50 km basis in 1989 in mol ha^{-1} a^{-1} (Van Leeuwen *et al.*, 1995).

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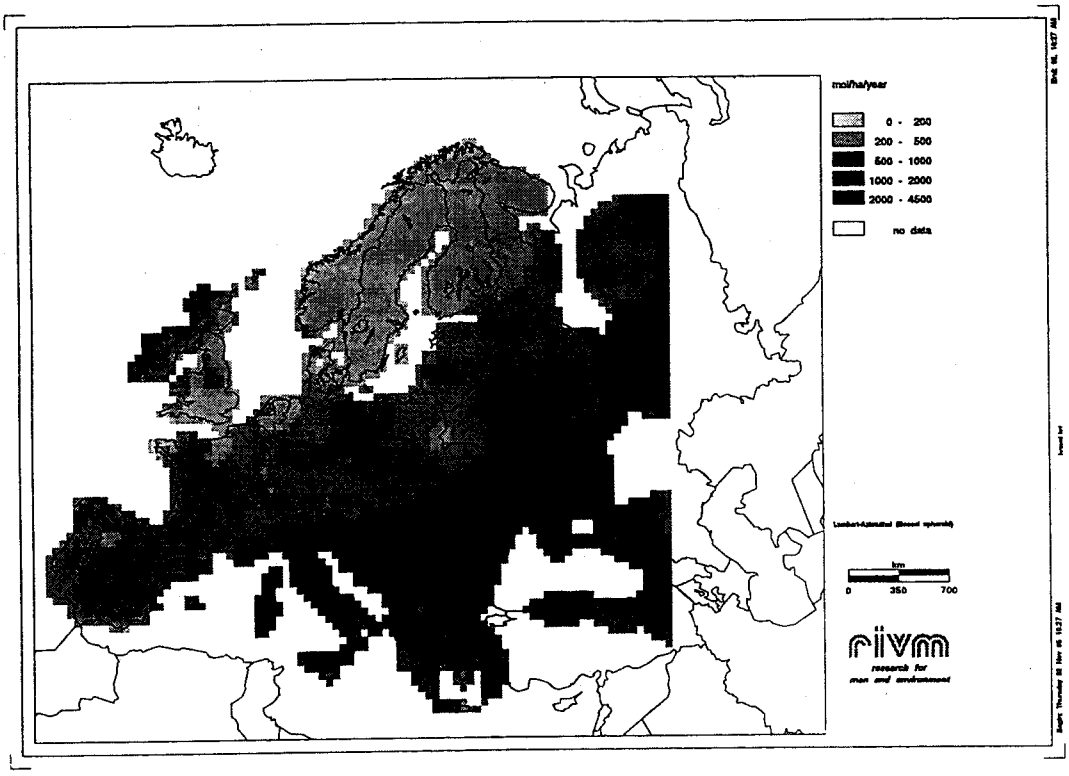


Figure 3. Wet deposition of $K^+ + Ca^{2+} + Mg^{2+}$ in Europe on a 50x50 km basis in 1989 in mol ha^{-1} a^{-1} (Van Leeuwen *et al.*, 1995).

4.2 Dry deposition

The method for dry deposition has been developed by Van Pul *et al.* (1992; 1994; 1995). Details can be found in these references. It is based on that used for the Netherlands (Erisman, 1992; 1993a). Dry deposition in EDACS is inferred from the combination of long-range transport model concentrations provided by EMEP and parametrised dry deposition velocities (Hicks *et al.*, 1987; Van Pul *et al.*, 1995). Concentrations at 50 m above the surface (blending height) are used. At this height it is assumed that concentrations and meteorological parameters are not influenced by surface properties to a large extent. Dry deposition velocities of gases and particles at this height are calculated on a small scale using a land-use map, routinely available meteorological data and the inferential technique (Erisman *et al.*, 1994; Van Pul *et al.*, 1995) Resistances are modelled using observations of meteorological parameters in Europe and parametrisation of surface exchange processes. Parametrisations of the surface resistance used for different receptor surfaces and pollution climates in Europe were described in Chapter 3. Meteorological parameters are obtained from the ODS (Observational Data Set: Potma, 1994) every 6 h and interpolated over Europe on a $1/6^0$ x $1/6^0$ grid. Meteorological parameters are wind speed, friction velocity, radiation, temperature, humidity and precipitation.

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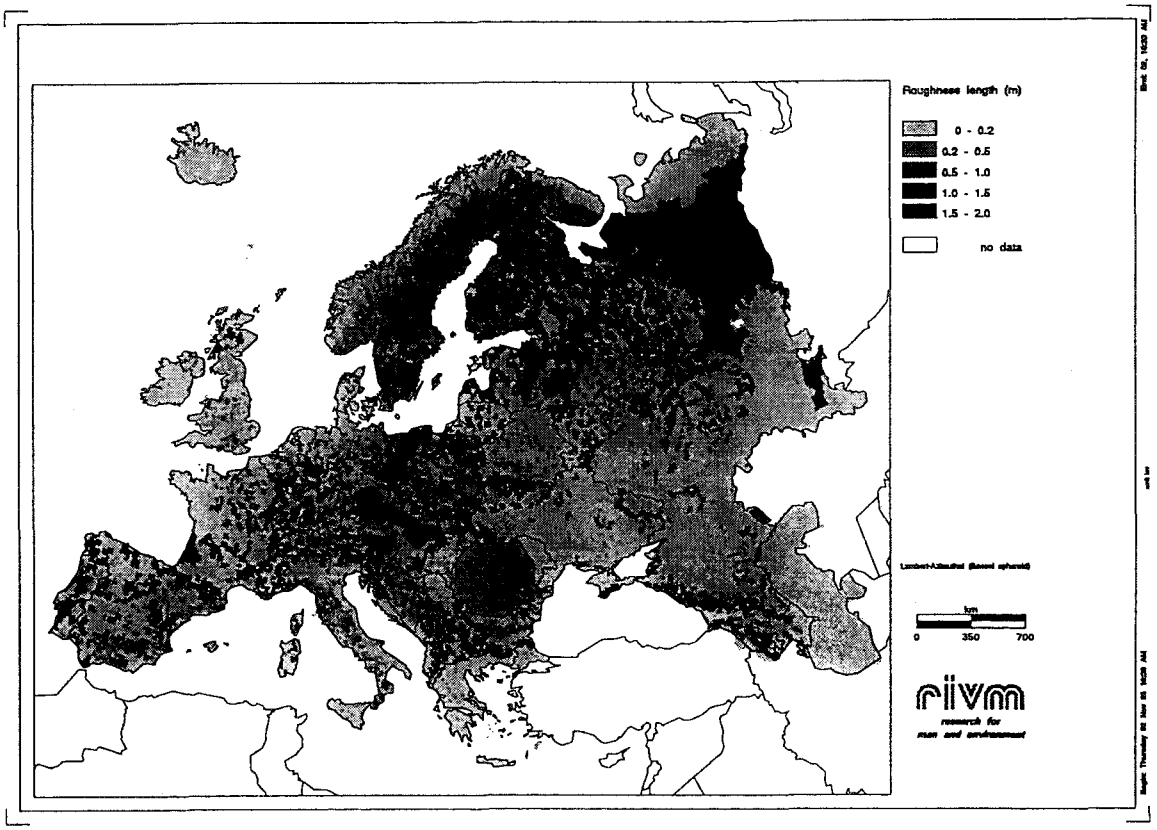


Figure 4. Summer roughness length map of Europe on a $1/6^0$ x $1/6^0$ grid (in cm) (after Van Pul *et al.*, 1995).

The land-use map for Europe was constructed by RIVM on a $1/6^0$ x $1/6^0$ (ca. 10 x 20 km) using ground-based and satellite observations (Van Velde *et al.*, 1994). Figure 4 shows roughness length maps for the summer season as derived from the land-use map and z_0 classifications according to Erisman (1992). The roughness length is used for estimating atmospheric transport to the surface. For each land-use class, the coverage within a grid cell and the deposition velocity are calculated. The average deposition velocity for a grid cell is then calculated by weighting the land-use specific deposition velocities with the surface area within that specific grid cell. Site-specific dry deposition might be calculated likewise, provided that detailed land-use and surface-roughness data of the site is available. Here aggregated estimates are presented.

Figure 5 to 7 show maps of the annual average dry deposition on a $1/6^0$ x $1/6^0$ scale of NOy, NHx and SOx in Europe. The effect of land use (roughness) and the difference in V_d is clearly shown. In areas with forested terrain the dry deposition is increased relative to the EMEP values, and, for example, in dry areas, the dry deposition is decreased as a result of a difference in V_d estimates. There is thus a clear sub-grid effect. The variation in dry deposition within EMEP grids is determined by variation in land use. This is the result of a variation in the aerodynamic resistance due to differences in roughness (see Figure 4), or of a variation of the surface resistance due to differences in vegetation and surface conditions.

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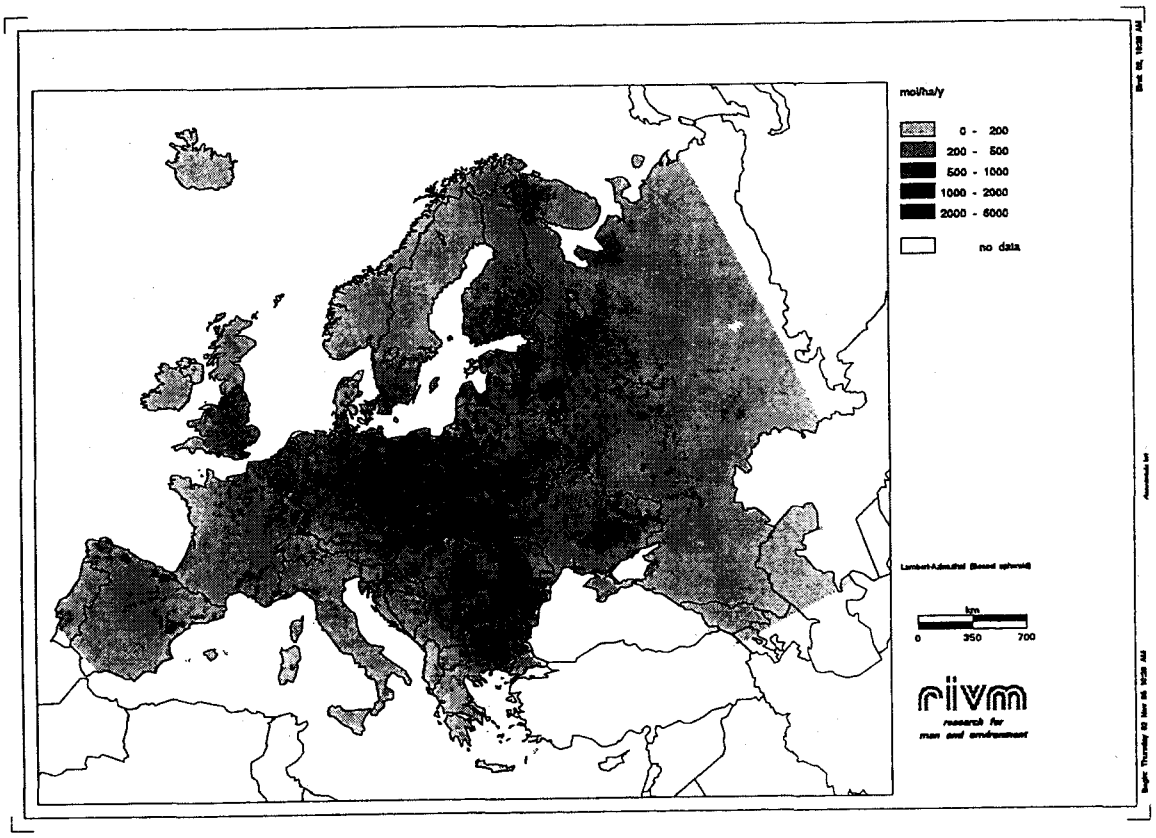


Figure 5. Dry deposition of SOx in Europe on a $1/6^0$ x $1/6^0$ scale in mol ha^{-1} a^{-1} (Van Pul *et al.*, 1995)

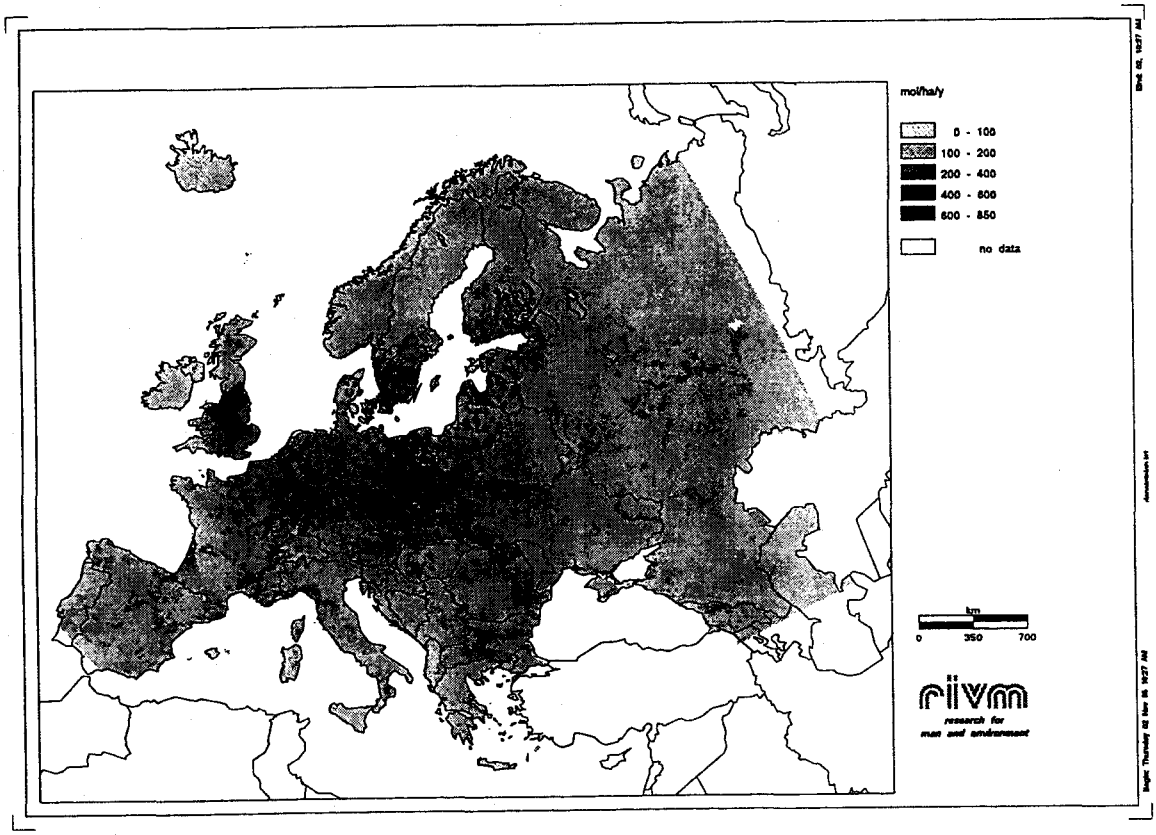


Figure 6. Dry deposition of NOy in Europe on a $1/6^0$ x $1/6^0$ scale in mol ha^{-1} a^{-1} (Van Pul *et al.*, 1995)

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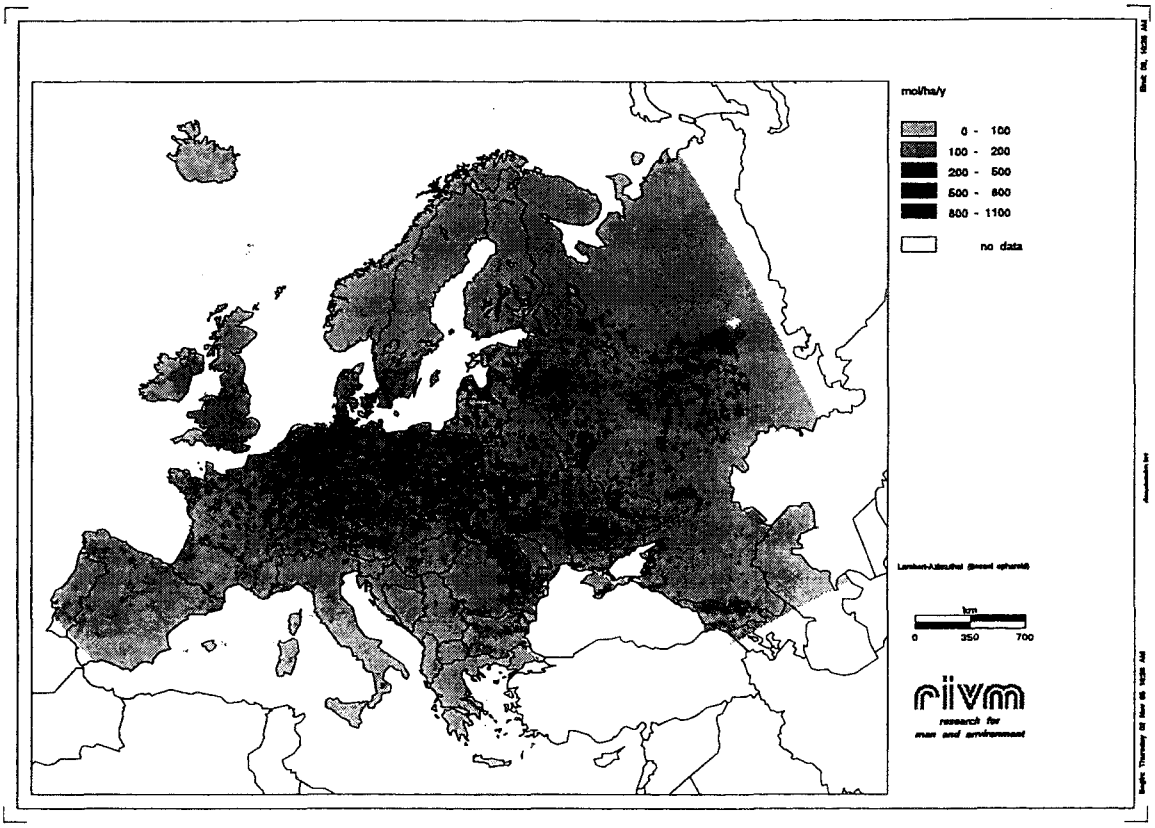


Figure 7. Dry deposition of NHx in Europe on a $1/6^0$ x $1/6^0$ scale in mol ha^{-1} a^{-1} (Van Pul et al., 1995)

The dry deposition flux of base cations is calculated as the product of the dry deposition velocity and air concentration at a reference height above the surface. The parametrisation of the dry deposition velocity for particles was explained in section 3.3.2. Six-hourly based dry deposition velocity fields were aggregated to annual means before they were combined with annual mean air concentration fields, yielding dry deposition estimates on a small scale over Europe (Draaijers *et al.*, 1995). This in contrast to the acidifying components where 6 hour average concentrations and deposition velocities are used.

Surface-level air concentrations were estimated from precipitation concentrations using scavenging ratios (Draaijers et al., 1995). These were derived from simultaneous measurements of base cation concentrations in precipitation and surface-level air performed by Eder and Dennis (1990) in Canada and Römer and Te Winkel (1994) in The Netherlands. This approach to estimate air concentrations is based on the premise that cloud droplets and precipitation efficiently scavenge aerosols resulting in a strong correlation between concentrations within precipitation and the surface-level air (Eder and Dennis, 1990). This assumption will only be valid for well-mixed conditions at sufficient distance from sources. Factors that will influence the magnitude and variability of scavenging ratios include particle size distribution and solubility, precipitation amount and rate, droplet accretion process and storm type (Galloway et al., 1993). Event scavenging ratios can range several orders of magnitude even for single species at a single location but scavenging ratios have been found reasonably consistent when averaged over one year or longer (Galloway et al., 1993). Therefore, annual mean precipitation concentrations were used to infer annual mean air concentrations of Na^+, Mg^{2+}, Ca^{2+} and K^+. Precipitation concentration data were taken from Van Leeuwen et al. (1995) as described in section 4.1.

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Deposition of Na^+ and part of the deposition of Mg^{2+}, Ca^{2+} and K^+ will be the result of sea spray. In sea spray these ions are mainly associated with Cl^-, and thus do not contribute to neutralization of atmospheric acid. To estimate what fraction of Mg^{2+}, Ca^{2+} and K^+ is of non-sea salt origin, correction factors have been derived based on the composition of sea water (Asman et al., 1981). It is assumed that Na^+ originates exclusively from sea spray and that the ratio between the concentration in sea spray for the component to be corrected and Na^+ is the same as in bulk sea water. Using such a correction will only yield reliable estimates on non-sea salt Mg^{2+}, Ca^{2+} and K^+ in areas where sea salt is the only source of Na^+ in ambient air.

Figure 8 shows a map of the annual average dry deposition of base cations in Europe. A clear pattern of increasing fluxes with decreasing distance to seas, in particular the Atlantic Ocean, can be observed. Large fluxes are also found e.g. northwest of the Black Sea, where they probably originate from wind erosion of salt containing soils. Large deposition of non-sea salt $Mg^{2+}+Ca^{2+}+K^+$ in south and southeast Europe is mainly the result of wind erosion of calcareous soils, agricultural tillage practices and traffic on unpaved roads. In this part of Europe, the prevalence of warm and dry conditions in the summer period will enhance the suspension of alkaline particles in the atmosphere. Sahara dust might be an important additional source of alkaline particles. Antilla (1990) found 80% of the annual total deposition of particulate matter in Corsica caused by Sahara dust. The relatively high fluxes in the border area between Germany, Poland and the Czech Republic, as well as in e.g. Estonia, can be attributed to intensive industrial activity.

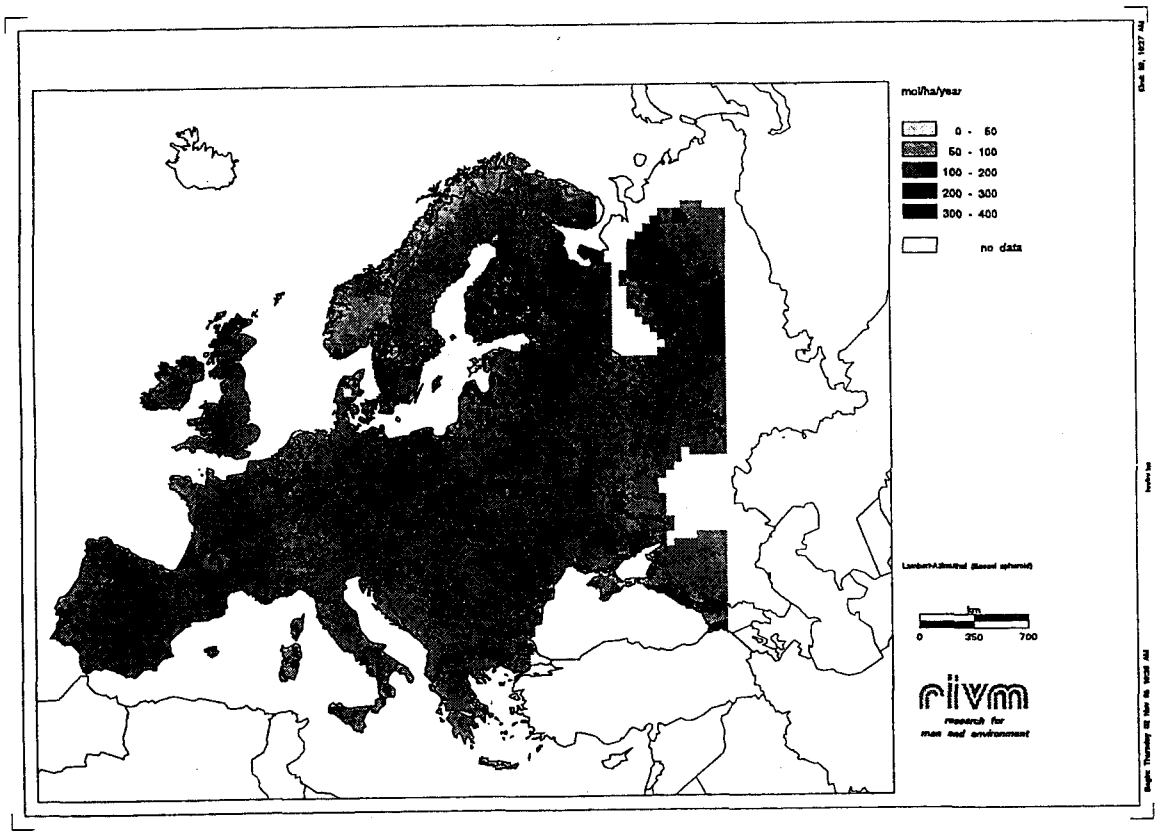


Figure 8 Dry deposition of base cations in Europe on a $1/6^0$ x $1/6^0$ scale in mol ha^{-1} a^{-1} (Draaijers *et al.*, 1995)

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4.3 Total deposition

Total deposition is the sum of dry and wet deposition. Total deposition of SOx is given as an example in Figure 9. Highest deposition of total sulphur occurs in the 'Black Triangle', i.e. the border area between Germany, Poland and the Czech Republic. Relatively high sulphur fluxes are also found in parts of the Ukraine and former Yugoslavia. The highest deposition of total nitrogen is observed for the area including northern France, Belgium, The Netherlands, Germany, the Czech and Slovak Republics and Poland. Lower depositions are found over southwestern Europe, Ireland and northern Scandinavia. High deposition of oxidised nitrogen (>0.7 g N m^{-2} a^{-1}) is found in an area mainly covering the Netherlands, Germany and large parts of Poland, whereas high deposition of reduced nitrogen is also found over large parts of Ireland, Great Britain, France and Eastern Europe.

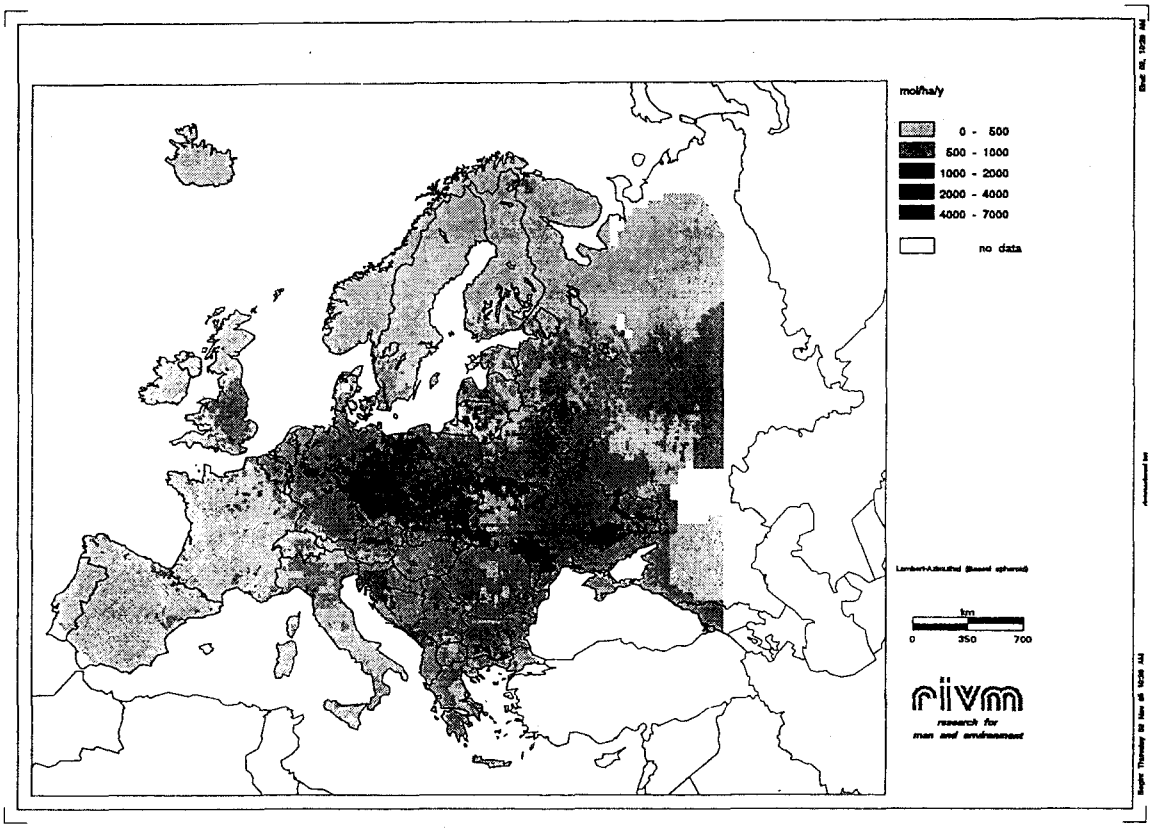


Figure 9 Total deposition of SOx in Europe on a 1/6 x 1/6° scale in mol ha^{-1} a^{-1} (Van Pul *et al.*, 1995)

Figure 10 shows the total base cation deposition which to some extent is available for neutralising acid deposition (Ca^{2+}+Mg^{2+}+K^{+}). Note that the lowest input is found in regions where the buffering capacity of the soil is already relatively low, e.g. in Scandinavia. Highest fluxes are found near the coast of Ireland and Scotland, in the 'Black Triangle', Ukraine, former Yugoslavia and some parts of Italy. With the exception of forested areas, the contribution of wet deposition to the total deposition of base cations is higher than dry deposition. Highest sodium fluxes are found in coastal areas, reflecting sea salt being the main source. For Ca^{2+}, Mg^{2+} and K^{+} relatively high fluxes are estimated e.g. in the Ukraine, in the former Yugoslavia and the southern and eastern part of Europe. Main sources of high input can be combustion processes, calcareous soils and high rainfall.

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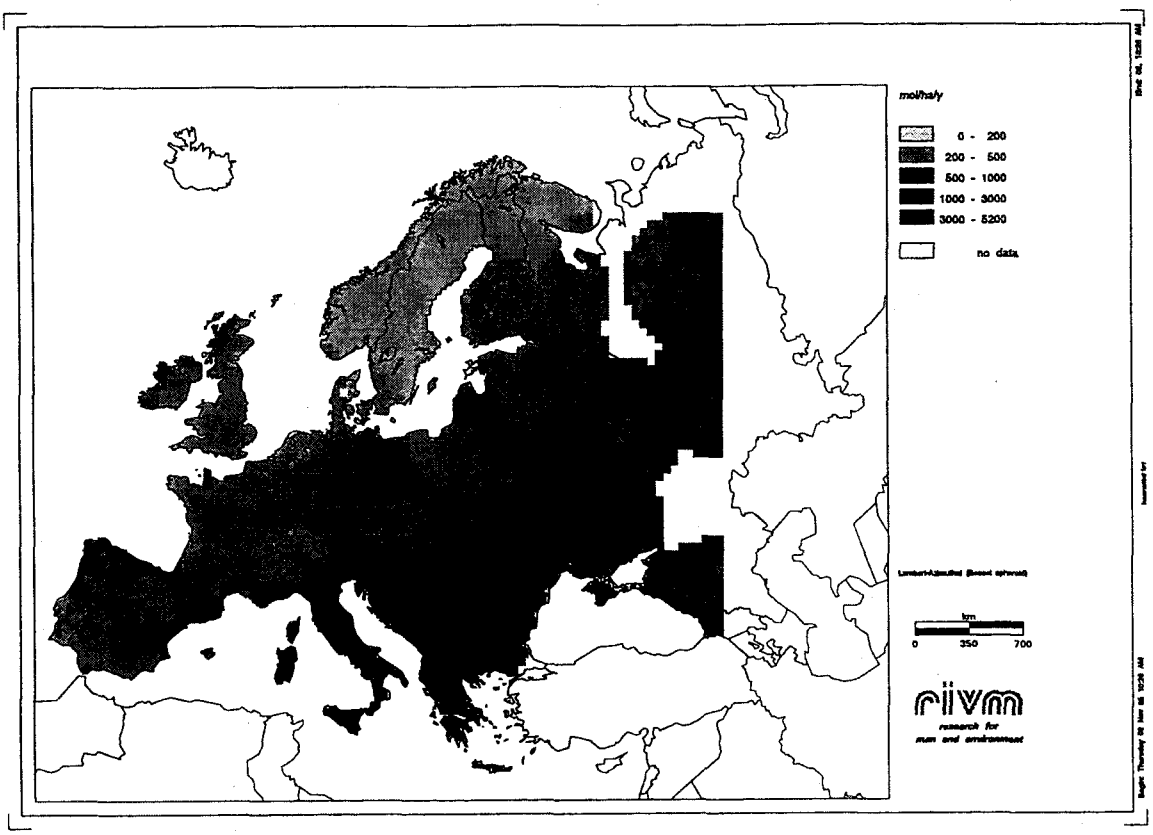


Figure 10 Total deposition of base cations ($Ca^{2+}+Mg^{2+}+K^{+}$) in Europe on a 1/6 x 1/6° scale in mol ha^{-1} a^{-1} (Draaijers *et al.*, 1995)

4.4 Uncertainty in the EDACS results

4.4.1 Wet deposition

The wet deposition maps are subject to several sources of uncertainty. These are divided into three main categories: *(1)* uncertainty associated with the measurements, *(2)* uncertainty associated with assumptions and simplifications in the methods used, and *(3)* uncertainty caused by the interpolation procedure. Van Leeuwen *et al.* (1995) give an extensive description of these uncertainties. Furthermore, they tried to quantify the contribution of all individual uncertainty sources to the total uncertainty. The numbers presented should be regarded as a best guess. The main source of uncertainty appears to be different for different regions of Europe. Therefore, a division into three categories is made, i.e. *(a)* West, northwest and Central Europe as 'good quality areas' where data quality is assumed to be good and sufficient (representative) data is present, *(b)* areas on edges of the maps, i.e. East, southeast and south-west Europe as 'poor quality areas', where less data is available of which the representativeness and quality can be questionable and *(c)* mountainous areas (e.g. the Alps) and upland areas (e.g. United Kingdom and Scandinavia), even though located in areas with many data, as 'complex terrain areas'. Total uncertainty in concentrations and in rainfall amounts have been estimated separately. For both variables the most dominating uncertainty (which could be quantified either in this study or by literature) was taken to represent total uncertainty, as all uncertainties are distributed around the same mean. In this approach uncertainties are assumed to reflect random, i.e. non-systematic errors. Total uncertainty was estimated using error propagation methods. Two cases are considered: first a propagation of

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errors without correlation (conservative), and second, a propagation with full positive correlation (worst case). Numbers are presented as percentages reflecting the relative deviation from the estimated value for an average 50x50 km grid cell in terms of one standard deviation, implying that the probability that the real value is the estimated value $\pm x\%$, is 67%. No distinction between different components was made. In the conservative case *(1)*, the overall uncertainty for an average 50x50 km grid cell is estimated to be 50% in West, north-west and Central Europe, whereas in the worst case *(2)* the overall uncertainty is estimated to be 70% here. In areas on the edges of the maps, i.e. East, southeast and southwest Europe, *(1)* is estimated to be 60% and *(2)* 80%, whereas in complex terrain areas *(1)* is estimated to be 70% and *(2)* 100%.

4.4.2 Dry deposition

ACIDIFYING COMPONENTS In the dry deposition estimates probably the highest uncertainty is introduced by using the simple resistance formulation for such a complex process as dry deposition, and, more specifically, the surface resistance parametrisations used to determine the surface exchange of gases and particles. The resistance model represents a simple approach to a highly variable process. It assumes a constant flux layer, i.e. there are no surface inhomogeneities, edge effects or chemical reactions. How much these simplifications contribute to the total uncertainty in the annual average deposition fluxes has not been quantified. The uncertainty in the surface resistance parametrisation is the largest uncertainty in this simplified scheme. Therefore more and more accurate parametrisations are needed for various vegetation species and surface types. Moreover, there is a lack of measurements which can be used to test these parametrisations, especially for southern and eastern European climates and surfaces. The vegetation and land use classes used in EDACS should be expanded when more parametrisations become available. Surface wetness is found to be one of the major factors influencing the deposition process of soluble gases. In the present version of EDACS only rain and indication of dew are used. Because of the great influence of surface wetness, processes leading to surface wetness should be taken into account in greater detail. The overall uncertainty in the surface resistance due to these factors is different for each component and surface type. This uncertainty varies on an annual basis, between 20 percent and more than 100%.

In the current version of EDACS, the EMEP-LTRAP modelled concentrations on a 150 x 150 km grid are used. The uncertainty in the concentrations are estimated to amount 40 - 70% using a statistical analysis with the EMEP measurements (Krüger, 1993). These concentrations represent the background situation in Europe. In source areas the uncertainty can be even higher. It is assumed that the concentration distribution within a grid is homogeneous. This is not the case in a grid which contains industrialised areas or many scattered sources such as with NH_3 and NOx. For such conditions, sub-grid concentration variations are present and will lead to underestimates of the deposition in that grid. To obtain an indication of the errors, a small-scale, short range model can be useful here to resolve sub-grid concentration gradients for dense source areas. The uncertainty in the deposition in an EDACS grid cell due to these gradients is estimated at 25% (Berg and Schaug, 1994).

The deposition in EDACS is based on the EMEP-LTRAP concentrations which in turn are dependent on EMEP deposition parametrisations. The deposition in the EMEP model and in EDACS are parametrised in different ways. By using other dry deposition velocities in EDACS, a mass inconsistency between EMEP-calculated deposition and the small-scale maps

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of EDACS might be introduced. As the result of a close cooperation between EMEP and RIVM, EMEP is currently using the same V_d parametrisations and land use data as those used in EDACS. Therefore, in the near future the problem of mass inconsistencies between the two models will be completely solved.

The accuracy of the presented results depends on the availability and quality of the input data, such as the land-use map and the meteorological observations. In the gridded version of the RIVM land-use database, forest is not subdivided into deciduous and coniferous forest. All forest is classified as coniferous forest (in the newest version of the land use maps this is improved). This will probably lead to overestimates of the deposition velocity to deciduous forests for all components during winter. However, the stomatal resistance in winter will generally be large due to the low temperatures. This overestimate will therefore not be very large.

The dry deposition is calculated on a daily basis. However, due to the daily averaging of the concentration and deposition velocity, a loss in temporal interpolation is introduced. This error is component-specific and estimated to be smaller than 20% (van Pul *et al*, 1993).

BASE CATIONS For dry deposition of base cations uncertainty is introduced by *i)* the parametrisation of the deposition velocity, and *ii)* the scavenging ratios and precipitation concentration maps used to estimate air concentrations. Ruijgrok et al. (1994) assessed the uncertainty of the model on which the parametrisation of the deposition velocity was based. The overall uncertainty in modelled deposition velocities integrated over the size distribution representative for alkaline particles at the Speulder forest was found to equal 60%. For other sites additional uncertainty will arise due to limited availability and accuracy of relevant land-use information and meteorological parameters. The uncertainty in deposition velocity caused by variation in size distribution of alkaline particles amounts 30-50%, assuming an average mass median diameter (MMD) of 5 μm and taking a geometric standard deviation (σ_g) of 2-3 to represent the variation (Ruijgrok *et al.*, 1994). The MMD of particles at a particular site will depend on the distance to sources and on e.g. ambient relative humidity (Fitzgerald, 1975). Theoretical models (Slinn, 1983) and field measurements (Kane *et al.*, 1994) suggest that the scavenging efficiency increases with particle diameter. Using the relationship between particle mass median diameter and scavenging efficiency presented by Kane *et al.* (1994), the uncertainty in estimated ambient air concentrations caused by variation in size distribution can be calculated to amount 50-100%, assuming a mean MMD of 5 μm and taking a σ_g of 2-3. Large errors in air concentrations will arise in areas very close or far from major sources and/or in areas with an exceptional precipitation climatology.

Using error propagation methods and assuming presented uncertainties in deposition velocities and air concentrations represent random errors, the total uncertainty in dry deposition of base cations for an average grid can be calculated to amount 80-120% (Draaijers *et al.*, 1995). Systematic errors in dry deposition may arise from e.g. *i)* using scavenging ratios which are based on only a limited set of simultaneous ambient air and precipitation concentration measurements, *ii)* neglecting complex terrain effects in the parametrisation of the deposition velocity, *iii)* using annual mean air concentrations and deposition velocities for flux calculation, thereby neglecting temporal correlations.

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4.4.3 Total deposition

The results of two studies comparing different models showed that the country budgets calculated with different models were reasonably in agreement (Derwent *et al.*, 1989; Erisman *et al.*, 1993). However, on a regional scale large differences between estimated total deposition fluxes were identified. The uncertainty in regional scale deposition estimates strongly depends on the pollution climate and on landscape complexity of the area under study. The uncertainty is determined by the uncertainty in wet, dry or cloud and fog deposition. Furthermore, deposition estimates yield higher uncertainty in areas built up by complex terrain and with strong horizontal concentration gradients. The influence of roughness transition zones and forest edges to the deposition in the Netherlands is quantified by e.g. Draaijers (1993). He showed that there is a systematic underestimation of the dry deposition to forests as the result of these factors in the order of 5 - 10% for acidifying components and 20 - 25% for base cations. Estimates for other types of complex terrain, such as mountainous regions, are not available.

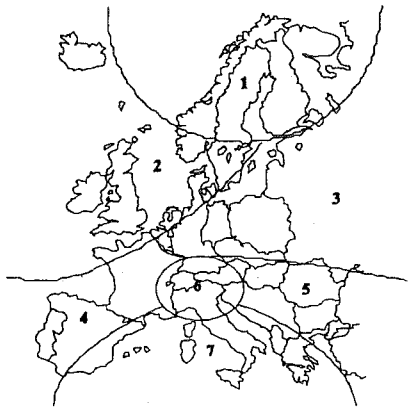


Figure 11 Distribution of the pollution regions in Europe in Table 2 (see text for explanation).

Table 2 shows the uncertainties associated with major key factors in deposition for seven (pollution) regions in Europe. The seven regions are shown on a European map in Figure 11. These regions are chosen because *i)* there is a marked difference in dominating deposition process (in region 1 the main input is wet deposition, in region 6 the main input is fog or cloud deposition), *ii)* there is a marked difference in climate (region 2 exhibits a sea climate, whereas region 7 exhibits a Mediterranean sea climate and region 4 mainly an inland climate), or *iii)* there is a marked difference in industrial activity (region 1 is remote, region 3 is mainly dominated by large old industrial complexes and region 2 exhibits high reduced nitrogen emission densities). This is only a crude classification. Furthermore, Table 2 shows only a crude estimate of uncertainty. Because of the strong local variations in dry deposition and the associated uncertainty local scales should be considered.

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Table 2 Uncertainty of key factors influencing deposition estimates of S and oxidised and reduced N in different pollution regions in Europe

Key Factors	NOy							NHx							SOx						
Regions[a]:	1	2	3	4	5	6	7	1	2	3	4	5	6	7	1	2	3	4	5	6	7
emission/type of	-	++	++	+	++	+	+	-	++	+	+	+	+	-	-	+	++	+	+	+	-
source	2	2	2	2	2	2	2	2	3	3	3	3	3	2	1	1	3	1	3	1	1
concentration	++	++	++	++	++	++	++	++	++	++	++	++	++	++	++	++	++	++	++	++	++
	2	2	3	3	3	3	2	1	3	3	3	3	3	2	1	2	3	3	3	2	2
wind speed	-	+	+	-	-	+	-	+	++	++	+	++	++	-	+	++	++	+	++	++	-
	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
roughness length	-	+	+	+	+	+	-	+	+	+	+	+	+	+	+	+	+	+	+	+	+
	1	1	2	2	2	2	2	1	1	2	2	2	2	2	1	1	2	2	2	2	2
surface wetness	-	-	-	-	-	-	-	+	++	++	+	++	+	+	+	++	++	+	++	++	+
	1	1	1	1	1	1	1	3	2	3	2	3	3	3	3	2	3	2	3	3	3
orography	-	-	-	-	-	++	-	++	-	-	+	+	++	-	+	-	+	+	+	++	-
	1	1	1	1	1	1	1	2	1	1	2	2	2	1	2	2	2	2	2	2	1
co-deposition	-	-	-	-	-	-	-	-	++	++	+	+	+	-	-	++	++	+	+	+	+
	1	1	1	1	1	1	1	3	2	3	3	3	3	3	3	2	3	3	3	3	3
surface	++	++	++	++	++	++	++	++	++	++	+	++	+	+	++	++	++	+	++	+	-
resistance	2	2	2	2	2	2	2	3	3	3	3	3	3	3	3	3	3	3	3	3	3
dry deposition	-	+	+	+	+	+	-	-	++	++	+	++	+	-	-	++	++	+	++	+	-
	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
wet deposition	++	+	+	+	+	++	+	++	+	+	+	+	++	+	++	+	+	+	+	++	+
	1	1	1	1	2	2	2	1	1	1	1	2	2	2	1	1	1	1	2	2	2
cloud and fog	+	++	-	-	+	++	-	+	++	-	-	+	++	-	+	++	-	-	+	++	-
	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3

legend:

importance of key factors:
++ dominating
+ important
- neutral
-- unimportant

uncertainty:
1 low <30%
2 median 30-70%
3 high > 70%
[a]the distribution of these regions is given in Figure 11

SYNTHESIS

This background document describes the current state of knowledge on modelling atmospheric deposition of acidifying components to ecosystems, most information is devrived from Erisman and Draaijers (1995). Methods and their limitations are described to come to a generalised description of deposition in Europe. The need for receptor or ecosystem specific deposition estimates is fairly new. Up to now deposition was treated as a loss term from the atmosphere and not as a highly variable spatial input parameter to sensitive systems. The emphasis in this document is put on the methodology development and on the application and evaluation of resulting current loads. The methodology which is used to assess the loads is based on the coupling of measurements and models. Measurements are used to explore and understand processes and to derive parameters which are used in models for generalisation. Measurements are also used to evaluate model results and to determine long-term trends. Models are used for generalising experimental results and to extrapolate them in space and time. For this, parameters derived from experiments are used. The results can be used to estimate critical load exceedances and to determine where ecosystems are at risk. Abatement strategies can be optimised in order to protect the most sensitive ecosystems by implementing the most effective measures on emission reduction.

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The inferential method is a good alternative for the combination of large-scale deposition estimates and filtering factors and percentile values of critical loads which are currently used for policy development. The method applied here for acidifying components playing a role in the issue of acidification can easily be used for other components related to other issues, such as heavy metals, persistent organic components, etc.

At the end of this document the uncertainty in the estimates in Europe is assessed. Especially the uncertainty in small scale estimates are large. This has to do with the concentration estimates, which are calculated on a coarse grid (150 x 150), and with the surface resistance estimates. Although surface resistance parametrisations are based on measurements, the number and accuracy of most of these measurements is not enough to be representative for all receptor types and pollution climates in Europe. Measurements of dry deposition fluxes of different components are therefore needed for different areas of Europe. An uncertainty analysis is an essential part of assessments. Especially in this kind of research where the resulting numbers can have large economic and political impact. The uncertainty analysis is in first instance used to check the results: how do they compare with other, independent estimates: either modelled or measured. Moreover, it can help to trace errors in procedures. Furthermore, it is useful to determine what uncertainty of sub-parts contribute most to the total uncertainty, in order to focus future research on.

At this moment, model development is hampered by lack of data which serve as input (detailed emissions) and as material for evaluation of modelled results. With the evolution of computers and their capacities, it has become possible to develop more detailed models, gradually moving from statistical approaches to real-time modelling. It must be kept in mind, however, that models will always be simplifications of reality and therefore simplifications are always needed. Conclusions might be that an optimum must be found between desired and possible detail of the descriptions, data availability and computer resources and process knowledge.

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IMPORTANCE OF SPATIAL DEPOSITION VARIATIONS FOR CRITICAL LOADS EXCEEDANCES

G. Lövblad

Swedish Environmental Research Institute (IVL), P.O. Box 47086, S-402 58 Göteborg, Sweden

Abstract

In order to study uncertainties in critical loads exceedances on the large -European- scale, exceedance has been calculated on a small scale and compared with the European scale for two areas in southern Sweden. In the small scale calculation, local critical loads for acidity were compared to local deposition. In this way, deposition variations due to pollution gradients and to ecosystem structure were considered. The results were similar for the two areas studied. Calculations based on local exceedances indicate, that greater emission reductions are necessary, 25% or more, than is predicted by the large-scale European calculations in order to protect 95% of the ecosystems. This difference is of importance for the further need for sulphur emission abatement. Consideration to scale will be even more important when studying nitrogen critical loads exceedances. The local deposition variations for nitrogen - oxidized as well as reduced - in Sweden are much greater than for sulphur, since local air pollution influence is of minor importance for sulphur in most parts of the country.

1 Introduction

Critical loads exceedances have formed the basis of optimal European abatement strategies for sulphur, as agreed to in the new sulphur protocol. A new protocol for NO_x is presently discussed and negotiations will start next year. This protocol will most likely consider the multipollutant, multieffect aspects related to emissions of nitrogen and be based on effect-related concepts such as critical loads and levels.

The exceedance of critical loads in a specific ecosystem is determined by comparing its critical load with the local deposition. The critical loads exceedances over larger areas must, however, be determined on a relevant scale, short enough to make it possible to consider the spatial variability in deposition and critical loads and large enough to enable the calculations to be carried out in a practical way.

For the sulphur protocol, deposition, critical loads and critical loads exceedance over Europe were calculated on a 150 x 150 km scale. Critical loads variability were dealt with by calculating the exceedance of the 5-percentile of the critical loads frequency distribution. In this way, 95% of the sensitive ecosystems is assumed to be protected. However, the deposition variability was not considered. Only one deposition value, the average deposition over the 150 x 150 km grid area, was used.

2 Description of the study

This study was made in order to investigate the effect of spatial variability in deposition on the critical loads exceedances in Sweden. The calculations were made for sulphur, since deposition of sulphur is known relatively well and since data on deposition as well as critical loads and critical loads exceedance are available on both the 150 x 150 km and the 50 x 50 km scale. The area studied was two EMEP grid squares in southern Sweden (Figure 1). For these grid squares (Figure 1a),

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EMEP model calculated deposition is available and large scale exceedance calculations have been made. Data on deposition, landuse and critical loads are available on a 50 x 50 subgrid net (Figure 1b). The squares were chosen for three main reasons:

1. the deposition is considerable as well as the local dry and wet deposition variations,
2. the grid squares contain mainly non-sea areas,
3. the grid squares contain Swedish land areas, for which deposition as well as landuse and critical loads data, produced in a uniform way, are available,
4. a large part of the area is acidified and considerable deposition reductions are necessary.

In order to compare large scale and small scale exceedance estimates, the same deposition (EMEP model calculated deposition for the period 1985 - 1993, Tuovinen et al., 1994) was used. To consider small scale variations within the square, the deposition was distributed to the ecosystems over the grid squares in relation to its location, within 50 x 50 km scale and in relation to type of ecosystem in accordance with monitoring results. The total integrated deposition over the ecosystems within the grid area was the same as the deposition calculated by EMEP. The pollution gradients for sulphur are relatively weak, so the variations considered on the 50 x 50 km scale are to a large extent due to variations in landuse. Since the variations in deposition are considered, the scale of variation considered is in reality smaller than the 50 x 50 km.

Deposition data and their variations are derived from throughfall and wet deposition measurements. To compare monitoring results with model calculated and the resulting critical loads exceedances, monitoring data from 1989 was used in parallel with the 1985 - 1993 results. Deposition derived from throughfall and wet deposition data for 1992 was used to study the exceedance of critical loads at present with a deposition which is approximately 40% lower than 1989 (Kindbom et al., 1995). The expected deposition in 2010 - assumed to be approximately 45% lower than 1990 (Erik Berge, pers comm.) was used for estimating the exceedance after the emission reductions made in accordance to the new sulphur protocol.

The influence of a number of aspects on the exceedance such as the enhanced forest edge deposition, monitoring uncertainties on the results of exceedance calculations, was also studied.

3 Variability in deposition over Sweden

Deposition varies on both a spatial and a temporal scale. This study deals mainly with the spatial variations, but to some extent longterm temporal changes between years are considered. Temporal variation in air pollution and deposition are mainly influenced by climatic variations. To some extent also temporal - seasonal, weekly and diurnal - emission variations are of importance. For the annual variations in deposition, which can be considerable, ± ca 20% (Kindbom et al., 1994), the

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Figure 1a EMEP gridsquare over Europe 150 x 150 km

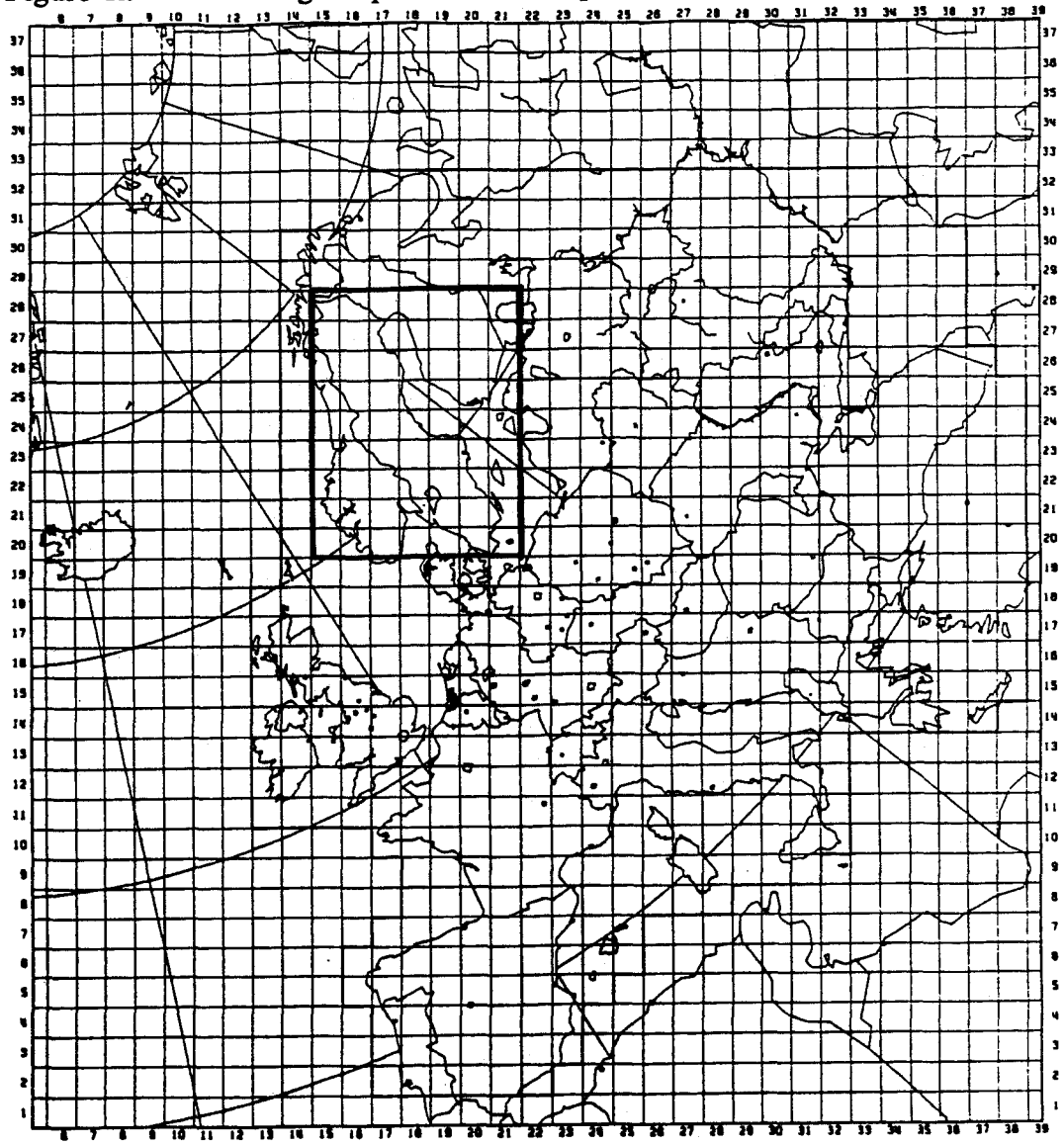
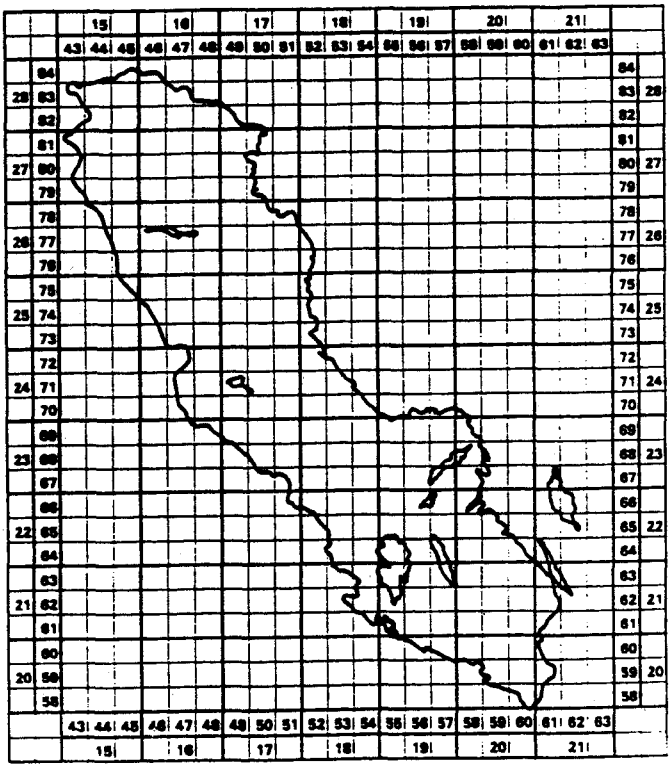


Figure 1b
EMEP subgrid net
50 x 50 km



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interannual climatic variations are usually most important. In the critical loads concept, it is assumed that effects should be avoided on a long-term scale. For this reason, use of the average deposition over a number of years is most relevant.

3.1 Different deposition processes

Three main processes contribute to the total deposition of sulphur; wet deposition, dry deposition and fog and cloud droplet deposition. The contributions are of varying magnitude in different parts of the country. More detailed descriptions of the deposition processes, how to estimate deposition, and the influence on different processes on total deposition amount are found in Lövblad et al. eds., 1993. The variations are due to pollution gradients and to variations in factors such as precipitation amount. There is a large-scale south to north decreasing gradient due to long-range transport of air pollution. Local scale pollution variations are small for sulphur in most Swedish areas except for the area around the largest cities.

3.2 Wet deposition

Wet deposition is the deposition from the atmosphere via precipitation. It is measured using bulk samplers, which sample not only precipitation, but also dry deposition of particles and soluble gases. For sulphate, however, bulk and wet-only measurements in parallel at some Swedish stations, have shown that dry deposition will, in most cases, give only minor contributions (<5%) to the measured results (Granat, 1989). Wet deposition is estimated and mapped in Sweden (Lövblad, 1992 a and b), using monitoring data.

The variation in wet deposition is controlled by the amount of precipitation and the pollution concentration. The large-scale pollution gradient influence the wet deposition over Sweden (Kindbom et al., 1994). In addition, local pollution from high-emission areas contribute to the regional variations. This has been shown e. g. in the Stockholm region (e.g. Kindbom et al., 1995) and is also indicated for the Göteborg area by the data used for this study. It is most probably seen also in a few other source areas in southern Sweden. Contributions from single sulphur emission sources are generally small (Persson et al., 1994) and relatively unimportant in Sweden.

The largest variations between near-by sites are due to differences in precipitation amount (Granat 1991). There is a decreasing gradient in precipitation amounts from coast to inland and also an increasing gradient with altitude, see e.g. SMHI, (1994). The western parts of the southern Highlands receive large amounts of precipitation. In relatively short distances from the west coast towards inland, there is a sharp precipitation gradient from around 500 at the coast to more than 1000 mm per year in the inland, which influence the wet deposition variation within the studied grid squares.

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3.3 *Dry deposition*

Dry deposition includes a number of processes for the transfer of gaseous SO_2 and particulate sulphate from the atmosphere to soil, vegetation, lake and sea surfaces. Uptake of carbon dioxide by plants for photosynthesis, will result in uptake also of other gases in the atmosphere, which is one important dry deposition process. The dry deposition is controlled by concentrations of pollutants in the air, atmospheric conditions and characteristics of the receptor in relation to both the atmosphere and the pollutant. The rougher, the larger and the more reactive a surface is, the greater the deposition. For example, forests induce atmospheric turbulence, which enhances atmospheric movements and chances of contact between pollutants and receptor surfaces. An alkaline receptor surface will favour acid deposition, a lipophilic surface will favour deposition of certain organic compounds, a wet surface will favour deposition of soluble gases, etc. More details about the influence on deposition by the structure and physical-chemical characteristics of the receptor as well as by the climate, is found in van Ek & Draaijers (1991) and Draaijers (1993).

No routine methods are available for dry deposition monitoring, and no measurement data are available for Sweden. Estimations of dry deposition to open fields with natural and semi-natural vegetation are made from bulk deposition measurements. This procedure may underestimate deposition to some types of vegetation in the most polluted areas in Sweden. However, it will not considerably alter the magnitude of deposition to larger areas and it is not considered to change the conclusions from this study. For agricultural and urban areas no detailed estimates are made since critical loads exceedances are not expected to occur in these areas.

Deposition to various forest categories and its variation is estimated using throughfall measurements results from the regional network and different research projects. Throughfall is considered to measure the total deposition of ions, which are not taken up by or leached from the canopy, i.e. sulphate, sodium and chloride (e.g. Ivens 1990; Ivens et al., 1990; Erisman 1992). Estimates of deposition to forests is presently based on models derived from monitoring results (Lövblad, 1992 a and b). Deposition is estimated to different types of forests; spruce, pine, deciduous and mixed forests, young to old forests, and forest edges. For forests growing on exposed slopes, no deposition data are available today. Enhanced deposition in such areas is not possible to estimate in this study.

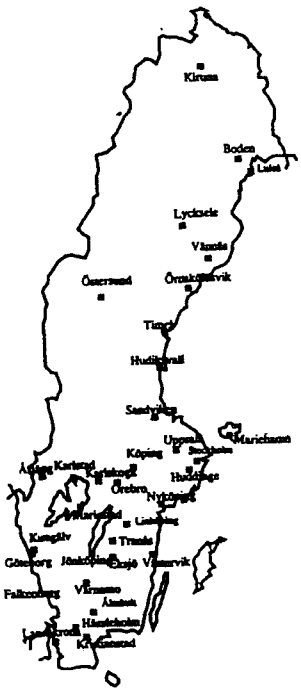
Also for the air concentration of sulphur compounds, there is a large scale decreasing gradient from south to north, which will influence the magnitude of dry deposition. The rural SO_2 concentrations are around 4 $\mu g \cdot m^{-3}$ in the south and below 1 $\mu g \cdot m^{-3}$ in the north. Air concentrations of SO_2, measured during the winter half year 1994/95 in urban and rural areas over Sweden (Svanberg et al., 1995), are summarized in Table 1. The table shows the concentration in town centres (SO_2-U) and at two rural sites approximately 10 km outside the city (SO_2-A and SO_2-B). In addition, the concentration measured at the nearest EMEP station (Sjöberg et al., 1995) is presented (SO_2-E). The table shows that differences between the two rural sites (SO_2-A and SO_2-B) are usually small, ≤ 0.3 $\mu g \cdot m^{-3}$. Exceptions are found around the four most southern/south-western cities, and around one city in the inner

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southern part of Sweden. The differences between urban and rural concentrations are also small. For 17 out of 22 cities, the differences between urban and rural concentrations, i. e. the local contributions, are ≤1 μg $SO_2 \cdot m^{-3}$. Only in 5 cities is the local SO_2 contribution larger. In 5 areas, the urban concentration is somewhat lower than the rural. The conclusion is that the local variations in SO_2 concentrations in Sweden is relatively small.

Table 1 Sulphur dioxide concentrations in Swedish urban and rural air during six winter months 1994/95, μg $SO_2 \cdot m^{-3}$ (Svanberg et al. 1995).

City	SO2-U Urban	SO2-A Rural	SO2-B Rural	SO2-E EMEP Rural	Differ. Rural A - Rural B	SO2-R Estim. Rural	Differ. Urban - Rural
Uppsala	1.8	2.2	1.9	1.6	0.3	1.9	-0.1
Nyköping	2.7	2.1	2.1	1.6	0.0	1.6	1.1
Linköping	2.0	1.7	2.0	1.6	0.3	2.0	0.0
Jönköping	3.0	2.5	2.6	n.s.	0.1	2.6	0.4
Värnamo	2.2	2.7	2.8	n.s.	0.1	2.7	-0.5
Eksjö	2.8	2.5	2.4	n.s.	0.1	2.4	0.4
Tranås	2.6	2.3	2.8	n.s.	0.5	2.3	0.3
Västervik	2.5	2.6	2.7	2.6	0.1	2.6	-0.1
Kristianstad	5.4	4.0	4.6	3.9	0.6	3.9	1.5
Hässleholm	5.0	4.9	3.8	3.9	1.1	3.9	1.1
Landskrona	4.4	4.9	6.4	3.9	1.5	4.9	-0.5
Falkenberg	3.3	2.8	4.7	2.9	1.9	2.9	0.4
Kungälv	3.3	3.5	2.9	2.9	0.6	2.9	0.4
Årjäng	3.9	-	1.6	n.s.	-	-	-
Karlskoga	2.9	1.6	1.7	n.s.	0.1	1.6	1.3
Köping	1.0	1.6	1.6	n.s.	0.0	1.6	-0.6
Sandviken	1.7	1.4	1.4	n.s.	0.0	1.4	0.3
Hudiksvall	1.5	1.2	1.1	n.s.	0.1	1.1	0.4
Timrå	1.3	1.0	1.3	n.s.	0.3	1.0	0.3
Östersund	2.0	1.4	1.1	0.5	0.3	0.8	1.2
Vännäs	1.1	0.8	1.0	n.s.	0.2	0.8	0.3
Luleå	1.6	1.1	1.1	n.s.	0.0	1.1	0.5
Boden	1.4	1.1	1.0	n.s.	0.1	1.1	0.3



n.s. means that no EMEP stations are representative for the area.

As a comparison, an air concentration of 1 μg $SO_2 \cdot m^{-3}$ is equal to a deposition of approximately 0.1 g $S \cdot m^{-2} \cdot year^{-1}$, i.e. 1 kg $S \cdot ha^{-1} \cdot year^{-1}$, with a deposition velocity of 0.8 cm s^{-1}. The results indicate, that dry deposition is of little importance in northern Sweden, where rural SO_2 concentrations are below 1 $\mu g \cdot m^{-3}$. In addition to the gaseous SO_2, sulphate particles in the air are deposited, but deposition velocity as well as concentration in the air is lower. Consequently, deposition contributions from sulphate particles is estimated to be much lower, as far as sea salt deposition is excluded.

Throughfall measurements can be used to determine the total deposition to forests and its variability. Dry deposition is estimated as the difference between throughfall

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and wet deposition. Measurements in spruce forests (Lövblad et al., 1994; Hallgren-Larsson et al., 1993; 1994 a, b) over Sweden also indicate that dry deposition is relatively unimportant in the north. Dry deposition, including fog and cloud droplet deposition to forests is usually 1 - 2 kg·ha^{-1}·year^{-1} in Norrland, the northern part of Sweden.

As mentioned, local sulphur emissions contribute to the local sulphur deposition variability only in some areas. This is also indicated by the results from throughfall measurements in spruce forests, (i. e. Hallgren-Larsson et al. 1993, 1994 a, b; Lövblad et al., 1994). Clear gradients are seen in the forest deposition pattern. Throughfall monitoring results have formed the basis for mapping sulphur deposition to Sweden (Lövblad et al., 1992a; Lövblad, 1993). On a local scale, the variations in landuse are large, and in most areas considered, it is much more important for the total and the dry deposition than pollution gradients. For this reason, deposition was mapped for different types of ecosystems. The dry part of the deposition to spruce forest was related to the presence of air concentrations of SO_2, sulphate particles and ammonia.

Also results from other forest types than Norway spruce forests are available, even though less measurements are made (see Table 2 below). However, the magnitude of the deposition to Scots pine forests and to deciduous forests can be estimated, but with a considerably lower accuracy than for spruce forests. The results indicate, that the deposition is relatively similar for pine and deciduous forests, that it is considerably lower than for spruce and that the variations are larger between different ecosystems.

3.4 Fog and cloud droplet deposition

Today very little is known about fog and cloud droplet deposition over Sweden. On-going work (Ferm & Westling, pers. comm.) in a mountainous region in middle Sweden indicate that this process may contribute considerably during wintertime at high altitude. Fog and cloud water deposition contribution is not possible to separate from the dry deposition in throughfall, but the contribution is included in the total. Due to lack of data this deposition process is not considered further in this study.

3.5 Total deposition

Variations in total deposition is studied using throughfall monitoring results (Hallgren-Larsson et al., 1993, 1994 a, b). The main variations in total deposition locally, are due to variations in landuse. The average deposition to different types of forests and different parts of southern Sweden and the variation, calculated as standard deviation, is presented in Table 2. The same relative variations between different ecosystems are included in the deposition estimates for the period 1985 - 1993, and the years 1989 and 1992.

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Table 2 Results from sulphur deposition measurements in southern Sweden; Average deposition, kg S·ha^{-1}·year^{-1}, standard deviation and number of sites (n) within each area/county.

Area /county	Wet deposition	n	Total deposition to Norway spruce forests	n	Total deposition to Scots pine forests	n	Total deposition to deciduous forests	n
Skåne	5.7±0.9	9	15.6±7.6	6	no sites	0	9.1	1
Blekinge	5.4±0.3	6	12.7±0.6	4	7.0	1	8.7	1
Kronoberg	5.4±0.8	9	12.2±2.8	6	7.6	2	5.1	1
Halland	6.7±1.3	6	14.2±2.6	4	8.5	2	no sites	0
Jönköping	6.0±2.0	9	10.6±2.1	5	6.1	3	10.3	1
Älvsborg	6.3±1.3	12	10.7±2.7	8	no sites	0	no sites	0
Göteborgs and Bohus län	6.5±1.4	12	12.5±4.6	10	10.4	2	no sites	0

The deposition to forest edges is enhanced in comparison with the forest deposition. From monitoring data (Hasselrot & Grennfelt, 1987; Beier & Gundersen, 1989; Ivens et al. 1988) the forest edge deposition, as the mean deposition over the whole edge area, from the edge and 50 m into the forest, is in this study assumed to be 20% higher than the local forest deposition. Further details are given in Lövblad et al., 1995 a, b).

Throughfall is normally measured in grown-up forests in Sweden. If forests are clear-cut, the deposition will decrease to approximately the level of wet deposition (Hultberg, 1985). New forest trees will grow up and the deposition will gradually increase. After 20 - 30 years, the trees in southern Sweden, will reach the size and the deposition of the adult forest. If the forest cutting is made in the traditional selective way, this temporal deposition variation will not be of importance. For this study, no data are available for estimating the deposition to young forests. The effect of clear-cuts is, however, considered, since the clear cut areas are included in the open field landuse class, with a very small dry deposition.

4 Variability in landuse

Landuse data are of importance for two reasons, for estimating deposition and for estimating ecosystems at risk. For deposition estimates, landuse is important for the dry deposition process. As mentioned above, dry deposition is of importance only in southern to middle Sweden.

Landuse data were obtained from the Swedish University of Agriculture (SLU). The data were divided into five main different categories of importance for this study: forests, open fields (with natural/semi-natural ecosystems), agricultural land, urban areas (including roads and similar types of anthropogenically influenced areas) and lakes. The forests are further subdivided into classes, different species (Table 3) and into forest - forest edges.

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Table 3 Forest classes, considered in the deposition estimates.

Forest class	Mixture of species	Deposition estimate based on data for
1	Scots Pine (incl. Contorta) ≥70%	Pine/deciduous forest
2	Norway Spruce ≥70%	Spruce forest
3	Coniferous ≥70%	0.5*Pine/dec. + 0.5*spruce forest
4 - 5	Deciduous ≥70%	Pine/deciduous forests
6 - 7	50 - 60% coniferous, 40 - 50% deciduous	0.3*spruce + 0.7*pine/dec. forests
8 - 9	30 - 40% coniferous, 60 - 70% deciduous	0.2*spruce + 0.8*pine/dec. forests
10 - 11	≤ 20% coniferous, 80% deciduous	Pine/deciduous forests

The variability in landuse within the studied area (18 subgrids) is described in Table 4 below. As can be seen, there is a difference in landuse between the two EMEP squares. Square 20;21 is dominated by forests. Square 19;21 contains forests to a large part, but considerable more agricultural land than 20;21. The mixture of forests within different subgrid squares is also presented in Table 4. Spruce dominates the forests in both EMEP squares, however, deciduous forests are more common in square 19;21.

In addition, the edge part of the total forest area was estimated from map studies (Lövblad 1995a). The length of borders between open land, lakes, roads etc. and forests was measured. From measurements at forest edges (Hasselrot and Grennfelt, 1987; Ivens, 1988; Beier & Gundersen, 1989), the width of the edge, with enhanced deposition was considered to be 50 meters. In the most heterogeneous landscape studied, the forest edges were estimated to cover 10 - 15% of the forest area, and in less patchy landscapes, 5 - 10% of the total forest area (Table 5). The general assumption in the calculations for this study was that 10% of the forest area, receives enhanced deposition due to the forest edge effect.

5 Variations in critical loads

Critical loads data are available for Sweden based on soil analysis and model calculations from a large number of sites all over the country (Data from Sverdrup et al., 1995). The data, however, cover only forest soils. For other types of soil, there are no systematically derived data. Assumptions have been made, in lack of other information, that there is a similar distribution of critical loads between forest soils and soils in other types of natural or semi-natural open field ecosystems. For urban and agricultural land, critical loads are not defined. These areas are heavily influenced by human activities. Agricultural land is treated with fertilisers and lime and deposition from the atmosphere is of minor importance. Urban areas consist to a large extent of impervious surfaces or rubble stone which makes the critical loads concept inapplicable. In the calculations made for this study, critical loads exceedance within urban and agricultural areas were set as the lowest exceedance in the square.

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Table 4 Landuse classes within the two studied EMEP grid squares 19;21 (subgrid squares 61;55 to 63;55) and 20;21 (subgrid squares 61;58 to 63;60) in % of the non-sea surface within the 50x50 km subgrid squares. Data from SLU. Forest classes in % of total forest area.

EMEP subgrid square		EMEP square	open land*	lakes	agri-cult. land	urban areas **	for-est areas	spruce class 1	pine class 2	coni-fer. class 3	decid class 4 -5 +10-11	Mix. class 6 -9
61	55	19;21	31	4	22	6	38	39	18	20	14	9
61	56	19;21	15	13	8	6	58	44	23	20	6	7
61	57	19;21	18	7	5	4	67	61	12	17	5	5
62	55	19;21	13	19	28	5	34	33	27	18	9	14
62	56	19;21	15	3	39	3	39	36	22	14	18	10
62	57	19;21	21	5	12	2	60	49	17	20	6	8
63	55	19;21	2	85	8	1	4	40	30	15	10	5
63	56	19;21	12	4	43	7	34	37	27	14	15	8
63	57	19;21	8	31	18	4	38	29	37	18	12	4
61	58	20;21	19	11	7	6	58	48	22	18	5	7
61	59	20;21	18	12	7	3	61	52	12	21	9	6
61	60	20;21	17	12	7	2	62	57	13	14	11	6
62	58	20;21	24	7	7	4	58	44	28	17	6	6
62	59	20;21	15	10	7	2	66	52	19	17	7	5
62	60	20;21	16	4	4	4	71	38	35	18	3	6
63	58	20;21	16	8	12	3	62	51	18	16	7	8
63	59	20;21	17	7	9	4	63	36	32	21	5	6
63	60	20;21	14	5	7	3	72	34	38	16	7	6

* includes grazing fields, meadows, bogs, moor land etc.

Table 5 Estimated part of forest which can be assumed to be forest edges. Results from studies of topographical maps (Lövblad, 1995a)

Maps studied to estimate the edge part of the forest		Forested part of mapped area (%)	Part of edges in % of total forest area	
Map	mapped area km^2		facing towards south	facing towards north
Uppsala SE	275	62.5	8.1	8.3
Åmål SW	288	71.1	7.3	7.5
Örebro NW	287	63.9	6.4	6.3
Norrköping SE	288	66.6	8.0	8.3
Göteborg NE	138	66.6	7.6	7.5
Kungsbacka SE	550	56.7	4.8	4.8
Tingsryd SW	120	61.5	12.2	11.6
Lessebo SW	576	78.2	8.1	7.7
Trelleborg NE + Malmö SE	581	8.5	13.5	11.1

Table 6 summarizes the critical loads data for the area studied. The critical loads were studied in order to find out if there are any systematic differences between forest species grown in forest soils and the critical load value of the soil (Table 7). The table indicate that soils where spruce is found are usually more insensitive than the soils where deciduous forests are growing. It is unclear whether this is a general

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conclusion. Similar results are seen also for the rest of southern Sweden (Lövblad, 1995a). However, the spruce plots dominate the sampling sites as well as the area of the studied grid squares.

Table 6 Critical loads for forest soils within the EMEP grid squares 19;21 and 20;21. Percentiles are calculated only forest classes where more than 20 sites have been studied (meq·m^{-2}·year^{-1}).

Grid	19;21					20;21				
Forest class*	arithm.mean	n	95%	50%	5%	arithm. mean	n	95%	50%	5%
1	128	16				106	34	139	103	77
2	195	43	280	125	121	137	72	184	131	92
3	120	25	187	125	56	105	30	170	98	60
4	119	7				105	9			
5	149	1								
6	135	6				117	6			
7	87	2				99	1			
8	141	3				106	1			
9	130	2				104	2			
10										
11						112	3			
total	153	105	260	151	56	120	158	174	118	75

* See Table 3 above.

Table 7 Comparison of critical loads for different forest classes (meq·m^{-2}·year^{-1}).

	Comparison between spruce and other types of forests		Comparison between deciduous and other types of forests	
	Mean of CL spruce forest	Mean of CL Other forest	Mean of CL Decid. forest	Mean of CL Other forest
19:21	195	124	119	156
	n=43	n=62	n=8	n=97
20;21	137	106	107	121
	n=72	n=86	n=12	n=146

As mentioned, for natural open field areas, the same distribution of critical loads as for forests have been assumed.

6 Exceedance calculations

Calculations of critical loads exceedances were made for sulphur for two EMEP grid squares (19;21 and 20;21). Only terrestrial ecosystems were considered in the calculations. The calculations were made for sulphur, but the way of calculation is principally the same also for nitrogen. In the calculation of critical loads exceedance, deposition of base cations and nitrogen are necessary input data. Estimates of the deposition to different types of forests and open fields natural ecosystems from the Nordic mapping were used (Lövblad et al., 1994).

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6.1 Small scale exceedance calculation

Critical loads of acidity are available for a number of forest sites within each subgrid square. Each critical load site was considered representative for a part of the forested grid area in relation to the critical loads distribution within different forest classes and in relation to the landuse data. Each critical load site was also considered representative for a proportional part of the forest edge area within the square. 10% of the forest area was considered as forest edges, which were assumed to obtain 20% higher deposition than the forest. The soils of the open field areas within the grid square is assumed to have a similar distribution of critical loads as forest soils.

Deposition of sulphur for the different forest classes for forest edges and for open field areas was estimated within each 50 x 50 km EMEP subgrid square, totally 18 subgrid squares, for the four time periods 1985 - 1993 (from Tuovinen et al., 1994), for 1989 (from Lövblad 1993), for 1992 (from Hallgren-Larsson et al., 1994 a, b) and for the year 2010 (as -45% is expected in relation to 1990). Deposition data for 1985 -1990 was used for nitrogen and base cations (Lövblad et al., 1992 a) for all time periods. These estimates for nitrogen and base cations are less accurate than for sulphur deposition.

A calculation was also made including uncertainties, ±20%, in relation to the average deposition, which is approximately the standard deviation for monitoring results within the subgrid square/county (see Table 2) for deposition to spruce forests and for wet deposition.

The critical loads exceedances were calculated locally for each "critical load site". Data on nitrogen and base cation uptake and nitrogen immobilisation, necessary for the calculations were obtained from the national critical loads mapping (See e. g. Sverdrup et al., 1995). Exceedances were calculated in the following way (see e. g. Downing et al., 1993):

$CLS = S_f * CLA$

$CDS = CLS + S_f * (BC_{dep} - BC_{uptake})$

$EXC = S_{dep} - CDS$

$S_f = S_{dep} / (S_{dep} + N_{dep} - N_{uptake} - N_{immob})$
If $N_{dep} > (N_{uptake} + N_{immob})$

Where CLS = Critical load of sulphur
CLA = Critical load of acidity
S_f = Sulphur fraction
CDS = Critical sulphur deposition
BC_{dep} = Base cation deposition
BC_{uptake} = Base cation uptake
EXC = Critical loads exceedance
S_{dep} = Present sulphur deposition
N_{dep} = Nitrogen deposition
N_{uptake} = Nitrogen uptake
N_{immob} = Nitrogen immobilisation

The result of the calculations is a number of "critical load site areas" with local critical loads exceedances (Figure 2), arranged after ascending exceedances. In addition, agricultural land and urban areas are included. These areas are defined as being the most insensitive, having the lowest exceedance. From these data is calculated the 95 percentile - by area - of exceedance, equal to the deposition

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reduction necessary to protect 95% of the ecosystems or the total terrestrial area of the grid square and the part of the grid area where critical loads are not exceeded.

Figure 2 Principal calculation procedure for the small scale exceedance. Calculated exceedance values are arraged in ascending order and the 95-percentile of exceedance by area is calculated.

Critical load database

Grid sq.	Crit. load	Area
19;21	CL1	A1
	CL2	A2
	CL3	A3
	.....	.....
	CLn	An

Landuse data base

Grid sq.	Landuse categ.		Area
	Forest	class 1	Y1
	Forest	class 2	Y2
	Forest	class 3	Y3
		.....	.....
	Forest	class 11	Y11
	Open fields		Y12
	Agriculture land		Y13
	Urban areas		Y14

Forest area = 90% of total forest area

Grid sq.	Crit. load	For. class	Area	Deposition	Area	Exceed.
19;21	CL1	Fcl1	Afo1	Estimated in rel.	Afo1	EXfo1
	CL2	....	Afo2	to forest class	Afo2	EXfo2
	....	....	.....	and subgrid square	.....	....
	....	....	.....		.....	....
	CLn	Fcl11	Afon		Afon	EXfon

Forest edge area = 10% of total forest area

Grid sq.	Crit. load	For. class	Area	Deposition		
19;21	CL1	Fcl1	Afe1	Estimated as 20%	Afe1	EXfe1
	CL2	....	Afe2	enhanced comp.	Afe2	EXfe2
	....	....	.....	to forest deposition	.....	....
	....	....	.....	in rel to forest class	.....	....
	CLn	Fcl11	Afen	and subgrid square	Afen	EXfen

Open field area, the same critical load distribution as for forest soils

Grid sq	Crit. load	Area	Deposition	Area	Exceed.
19;21	CL1	Aof1	Assumed equal	Aof1	EXof1
	CL2	Aof2	to bulk deposition	Aof2	EXof2
	....	....	in rel. to subgrid	....	...
	CLn	Aofn	square	Aofn	EXofn

Agricultural land and urban areas

Exceedance is defined as the lowest exceedance in the gridsquare	Aur.+Aag.	EXmin

6.2 *European large scale exceedance calculation*

Calculations on the large-scale are made for the two 150 x 150 km EMEP grid squares. Deposition of sulphur (one value per grid square) is obtained by the EMEP model calculations for the period 1985-1993 (Tuovinen et al., 1994). The critical load value for the 5% most sensitive ecosystems in the grid squares are found in Table 6 and is used to calculate the critical sulphur load according to the formulas in

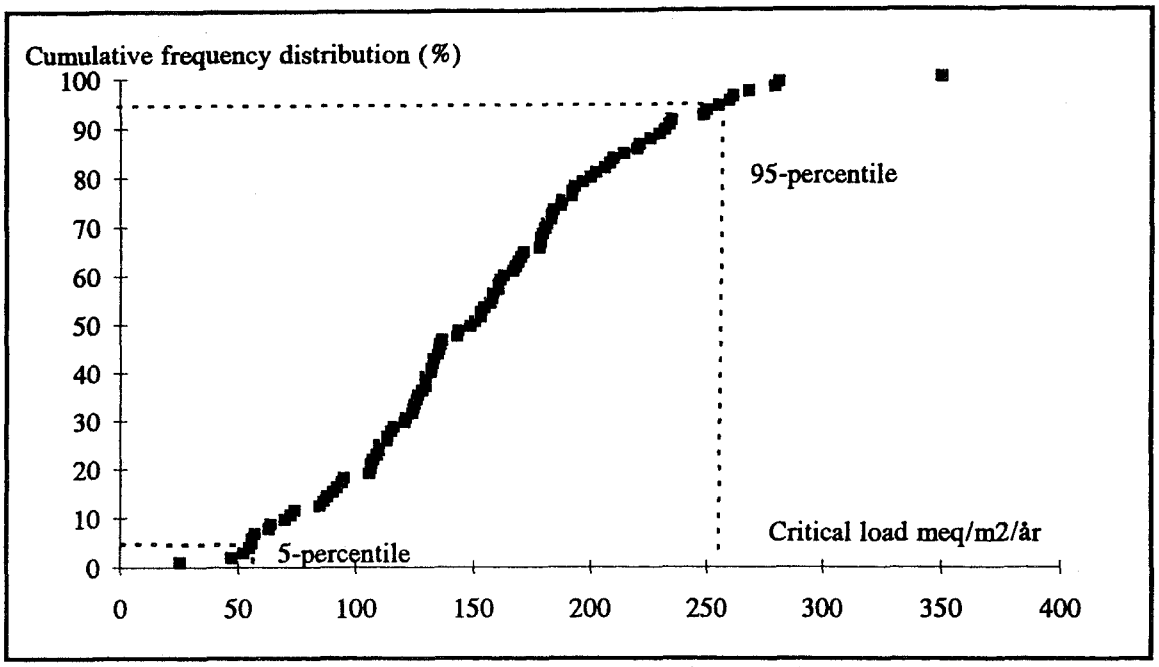
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chapter 6.1. The exceedance is determined as sulphur deposition minus critical sulphur deposition. Data on nitrogen immobilisation, nitrogen uptake and base cation uptake are estimated as mean values for the nine subgrid squares within each EMEP square from the national mapping (see e. g. Sverdrup et al., 1995).

7 Results

Results from the calculations are summarized in Table 8. The large-scale exceedance for grid square 19;21 is shown in Figure 2. The 5-percentile defines the 5% most sensitive ecosystems. This value is compared to the deposition.

Figure 3 Large-scale exceedance calculation. Cumulative distribution of forest soil critical loads (%) in EMEP grid square 19;21 (meq·m^{-2}·year^{-1}).



Exceedance is calculated by comparing the 95-percentile for critical load and deposition calculated using the EMEP model (Tuovinen et al., 1994).

The small scale exceedance calculations for grid square 19;21 and 20;21 are shown in Figure 4 - 5. The grid squares differ from a deposition point of view. The square to the west receives somewhat more pollution. The EMEP model calculated deposition to the squares are 0.97 and 0.91 g S·m^{-2}·year^{-1}, which is equivalent to 61 and 57 meq·m^{-2}·year^{-1}, respectively.

Also critical loads and landuse data differ. Grid square 19;21 is, on the average, less sensitive than 20;21, but the critical loads variations are larger. Average critical loads (Table 6) for forest soils are 153 for square 19;21 and 120 meq·m^{-2}·year^{-1} for 20;21. Square 19;21 contains more agricultural land (26% of total non-sea area) compared to 20;21 (7%). Both grid squares are dominated by forests, 52% in square 19;21 and 67% in square 20;21. The part of open fields is similar in the two squares, 16 and 14%, respectively. Urban areas (incl. roads and similar landuse) are small within the squares, 5 and 3%.

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Figure 4 Cumulative distribution (%) of critical loads exceedances calculated in the small scale, and aggregated to the whole EMEP square 19;21 (meq·m^{-2}·year^{-1}). Deposition data are EMEP weighted deposition 1985 - 93.

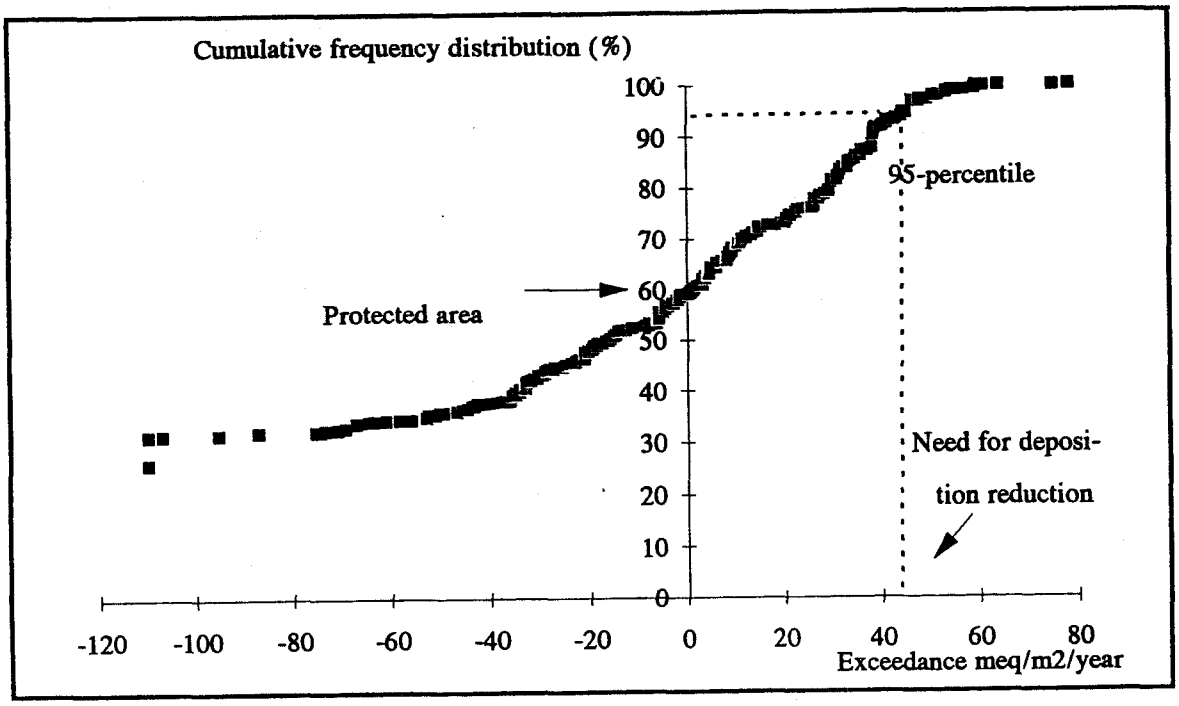
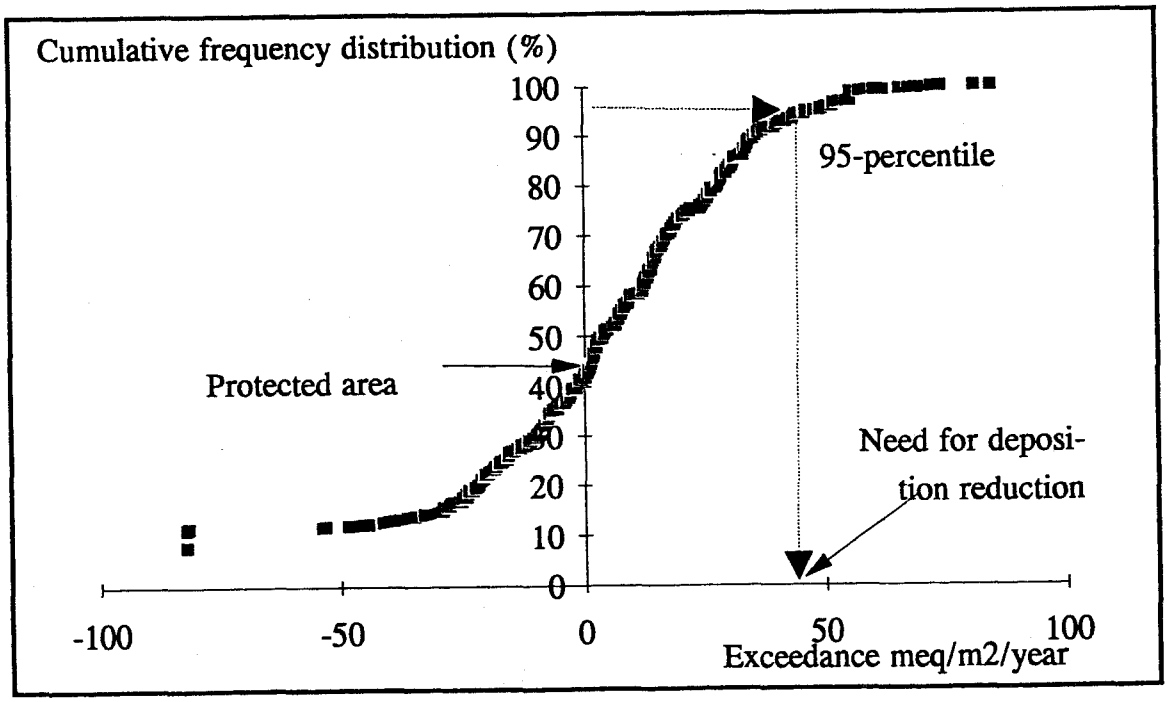


Figure 5 Cumulative distribution (%) of critical loads exceedances calculated in the small scale, and aggregated to the whole EMEP square 20;21 (meq·m^{-2}·year^{-1}). Deposition data are EMEP weighted deposition 1985 - 93.



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Table 8 Summarized results from the exceedance calculations. Part of the area protected against critical loads exceedance (%) and the exceedance (meq·m^{-2}·year) in the 95 % most sensitive ecosystems.

Calculation	Sulphur deposition estimate	Grid square 19;21		Grid square 20;21	
		Area protected	Exceed-ance	Area protected	Exceed-ance
On large scale	EMEP deposition*	no data avail.	36	no data avail.	36
On small scale considering forest edges, incl. agricultural land and urban areas	Weighted EMEP deposition*	59%	45	42%	46
On small scale considering forest edges, incl. only terrestrial ecosystems	Weighted EMEP deposition*	40%	47	34%	50
On small scale considering forest edges, incl. agricultural land and urban areas	Deposition estimate for 1989	61%	44	37%	54
On small scale considering forest edges, incl. only terrestrial ecosystems	Deposition estimate for 1989	44%	45	28%	56
On small scale considering forest edges, incl. agricultural land and urban areas	Deposition estimate for 1992 "Present situation"	68%	32	47%	34
On small scale considering forest edges, incl. only terrestrial ecosystems	Deposition estimate for 1992 "Present situation"	54%	35	39%	34
On small scale considering forest edges, incl. agricultural land and urban areas	Deposition estimate for 2010 "Future situation"	72%	21	75%	14
On small scale incl. agricultural land and urban areas, but without considering forest edges	Weighed EMEP deposition*	60%	45	43%	43
On small scale considering forest edges, incl. agricultural land and urban areas. Uncertainties ±20% are included in the sulphur deposition estimate.	Weighted EMEP deposition*	60%	49	44%	50
On small scale considering forest edges, incl. agricultural land and urban areas. Uncertainties ±20% are included in the sulphur, nitrogen and base cation deposition estimate.	Weighted EMEP deposition*	69%	55	45%	52

* 1985 - 1993 mean deposition, calculated by the EMEP model

The difference between small scale and large-scale calculations are demonstrated for square 19;21 by Figure 2 and 3 and by Table 8. The large-scale exceedance in both squares is 36 meq·m^{-2}·year^{-1}. Small scale calculations result in a greater exceedance, equivalent to a 25% higher need for sulphur deposition reductions, 45 and 46 meq·m^{-2}·year^{-1}.

If variation in deposition between sites in the same region, ±20%, is included in the calculations, the 95 percentile for square 19;21 increases and is about 60% higher, and for square 20;21 more than 50% higher.

If insensitive areas, agricultural and urban areas, are excluded in the exceedance calculations, there is only a minor change in exceedance for 19;21. The 95-

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percentile increases from 45 to 47 meq·m^{-2}·year^{-1}. There is a larger influence in square 20;21 where the exceedance increases from 46 to 50 meq·m^{-2}·year^{-1}, when only ecosystems within the grid square are considered.

There is only a small effect on the exceedance by the enhanced deposition to forest edges. If forest edges are not considered, the exceedance in square 20;21 is 43 meq·m^{-2}·year^{-1} compared to 46 meq·m^{-2}·year^{-1}, if edge deposition is considered. For square 19;21 there is no change in exceedance due to the edge effect. However, the forest edge area is assumed to be approximately 10% of the total forest area, which is 52 and 67% respectively in the two squares. The forest edge area in the two squares is consequently 5 and 7%.

The calculations show almost no difference for square 19;21 if EMEP weighted deposition for 1985 - 1993 is used or the deposition for 1989 estimated from monitoring data is used. Monitoring result based deposition estimates for 1989 integrated over the grid square in relation to landuse (Lövblad, 1993) indicate a deposition of 0.97 g·m^{-2}·year^{-1}, which is equal to the EMEP calculated deposition for 1985 - 1993. However, for square 20;21 there is a difference. The integrated deposition for 1989 is 1.11 g·m^{-2}·year^{-1}. The difference in integrated deposition as well as in exceedance is assumed to be due mainly to the difference in landuse. Square 20;21 is to a large part forested and spruce forests dominates.

Comparison between 1989 and 1992, show that the decrease in deposition, observed by monitoring data on air concentrations, wet and total deposition from 1990 over large parts of Sweden (Kindbom et al., 1994), result in a lower exceedance of critical loads in both grid squares. The exceedance have decreased 27% in square 19;21 and 37% in square 20;21. Also, this difference is assumed to be due to landuse differences between the squares.

8 Conclusions and discussion

The small scale calculations made, result in a 25% higher exceedance and thus in a 25% greater need for deposition reductions than the large (European) scale calculations in both squares. If variation in deposition between sites in the same region, ±20%, is included in the calculations, the 95-percentile for square 19;21 increases and is about 60% higher, and for square 20;21 about 50% higher. The calculations show the relatively small importance by enhanced deposition at forest edges for the critical loads exceedance in the two studied 150 x 150 km grid squares. The calculations indicate furthermore, that it is of importance how the exceedance is expressed, in relation to ecosystem area in the grid square or in relation to the total land area.

Similar studies of the influence of scale on exceedance of critical loads were made in Great Britain (Smith et al., 1994; Smith et al., 1995). The results indicate that deposition estimates and calculations on large scale, 20 x 20 km (UK scale) and 150 x 150 km (European scale) considerably underestimate the exceedance in complex terrain, compared to the 1 x 1 km scale. The variations in deposition are, however, much larger in parts of the UK, than in the areas studied in Sweden.

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The scale of calculation introduces uncertainties in the exceedance calculations. However, there are other sources of uncertainties. The input data for this study are partly uncertain and partly lacking. As regards critical loads, data for other soils than forest soils were lacking and a similar distribution was assumed. Other data necessary for the calculations, are highly uncertain, especially nitrogen immobilisation. Also the deposition data are subject to uncertainties. Additional data on sulphur deposition in the area would have improved the calculations. However, it is unlikely that it would greatly alter the conclusions. There is a strong need for better deposition data on base cations and nitrogen to different types of ecosystems, in order to calculate the exceedance of critical loads more accurately. In this context, base cation deposition data are probably the most uncertain and most important to improve. An evaluation of the uncertainties in the total exceedance calculation chain may be important, to separate the small uncertainties from the larger.

Two of the EMEP squares in southern Sweden are studied. If instead square 20;20 was chosen, even greater effect of deposition variations would be expected. The reason is that this area is subject to a large deposition variability, due to influence from the local densely populated region, influence from the Copenhagen region and from long-range transport. Calculations for this square are, however, in practice complicated by the fact that the square covers, not only Swedish land area, but also the sea and a part of Denmark. In areas north of the squares studied, the deposition variability is expected to be of less importance for the exceedance. The dry deposition decrease in importance in relation to the wet deposition when moving northwards.

The data indicate that there is no important relation between the critical load for forest soil and tree species, and thus no relation between critical load and deposition in the area studied. Variations in critical load within the squares studied, expressed as the deviation between 95- and 5-percentile, is approximately a factor 5 (square 19;21) and a factor 3 (square 20;21). The variations in deposition between wet deposition and total forest deposition in different parts of the 150 x 150 km square are a factor 4 in the two squares. These variations are not considered at all in the large-scale exceedance calculations. If calculations were made for nitrogen instead of sulphur, the greater variability in deposition of oxidized as well as reduced nitrogen, is expected to enhance the effect on exceedance calculations. No calculations were made for nitrogen due to lack of much of the input data necessary. For example, the critical loads for nitrogen were not yet available. The greater variability for nitrogen will, however, be of importance when making exceedance calculations in connection with the new NO_x protocol. It is important to find a practical solution to consider these variations in the exceedance estimate, or at least to evaluate the effect of small scale exceedance in different parts of Europe.

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RELEVANCE OF SPATIAL RECEPTOR DISTRIBUTION FOR THE ASSESSMENT OF CRITICAL LOADS

Knoflacher Helmut Markus

Environemtal Planning Department, Austrian Research Centre
A-2444 Seibersdorf

Abstract

The calculation of critical loads and their exceedances under the convention of the UN ECE is based on the combination of two different scale dependent approaches. The calculation of the impacts (concentrations and depositions) is based on a top-down approach with a disaggregation of large scale processes to spatial impact distributions at smaller scales. The identification of receptors and their sensitivities is based on a bottom-up approach with an extrapolation of local parameters up to large area units. Important constraints, especially economical factors and the peculiarities of investigation methods, are strongly influencing the possibilities to join these two approaches. It is therefore necessary to use methodological approximations to close the gaps between both approaches. The spatial disaggregation of the impact system is depending on the peculiarities of the specific substances and is discussed on other places. In contrast to it, the accuracy of the methods for extrapolation in the receptor system is strongly influenced by structural and functional characteristics of specific landscapes and is also depending on the definition of the target receptor.

1. Introduction

The calculations of critical loads and their exceedances are treated in two different systems

- the impact system and
- the receptor system (fig. 1).

The impact system comprises the generation of air pollutants by human activities, the transport processes (fig. 1) in the atmosphere and the spatial distribution of concentrations and depositions onto the ground based receptors. The parameters for the

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description of this system are based on data from meteorological and air-chemical monitoring methods.

Under the convention of the UN ECE only long range transports out of three different spatial scales of transport processes are considered. This approach entails some methodological problems. Nations are in generally more interested to control their "home made" impacts, monitoring stations are therefore located on sites with potential high impacts from medium range transports. The information density about the spatial and temporal distribution of impacts from long range transports, at the scale of receptor observation, is therefore not very high. These problems and their methodological consequences are treated in other papers and will not be discussed here. Of importance is the fact, that impacts of long range tranports are influencing areas far away from direct human activities. The investigation density and consequently the knowledge about different processes in such areas is roughly lower than in more densely populated areas. It is therefore not so easy to get sufficient data for the calculation of critical loads. A further problem is the fact, that depending on the geomorphological situation, some effects can cumulate over reaction chains in the landscape and can cause critical effects which could not be identified by single spot investigations.

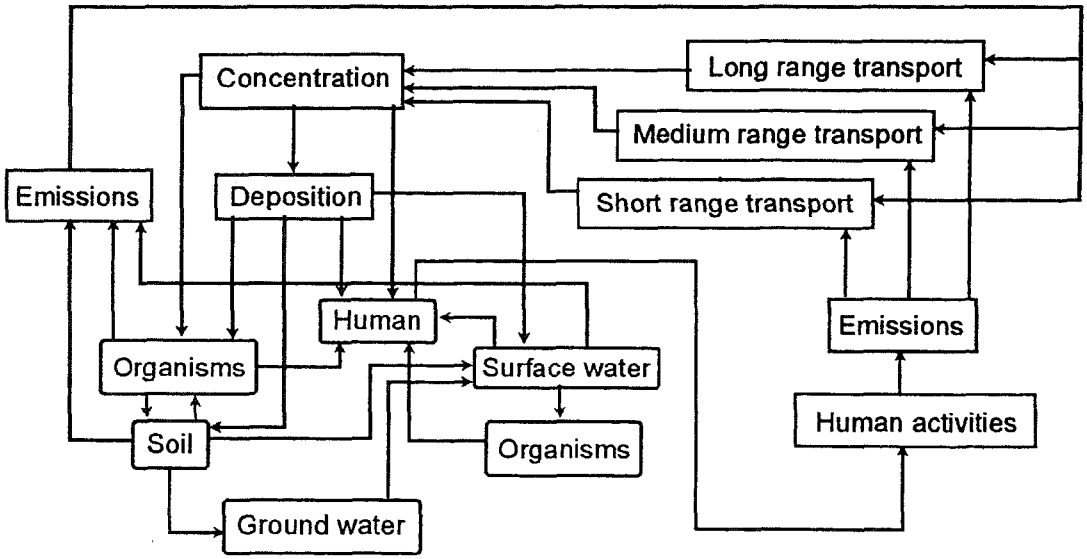


Figure 1: Interactions of impact (human activities to depositions) and receptor (human to groundwater) systems.

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2. *The interactions of the impact and the receptor systems*

The accuracy of the calculation of exceedances is strongly dependent on the accuracy of receptor and impact data. These preconditions can not be reduced only to a problem of data, it is also necessary to include the processes behind the data. It has also to include aspects of temporal and spatial dimensions as of dynamic processes. Although it is principally clear, that the calculation of critical loads has to deal with system processes, the shortcomings of contacts between different groups of specialists reduced the discernment and acceptancy of the approaches of other groups.

For the assessment of impacts under system conditions it is necessary to know

- the principal system interactions,
- the definition of the relevant receptor(s) and
- the definition of critical conditions.

In the system interactions of air pollutants and ground based receptors different types of receptors and different critical conditions can be defined. The primary ideas were influenced by the observations of soil and lake acidification and its consequences for the health of forests and aquatic organism. By extending the spectrum of impacts the potential number of effects can be increased indefinitely. But for practical purposes it is necessary to reduce the number of critical effects to an amount which can be handled by investigations and monitoring methods.

In this context it is necessary to point out the potential influence of long range transports on different receptors. Because of the distribution over large areas and the long time of persistency in the atmosphere (fig. 2) impacts of long range transports are determining the framework conditions for processes in landscapes. Medium and short range transports are important for the spatial variability of impact conditions at larger scales. The importance of these impacts for receptors in large areas is strongly dependent on the spatial densities of relevant human activities and on the spatial distribution of the receptors.

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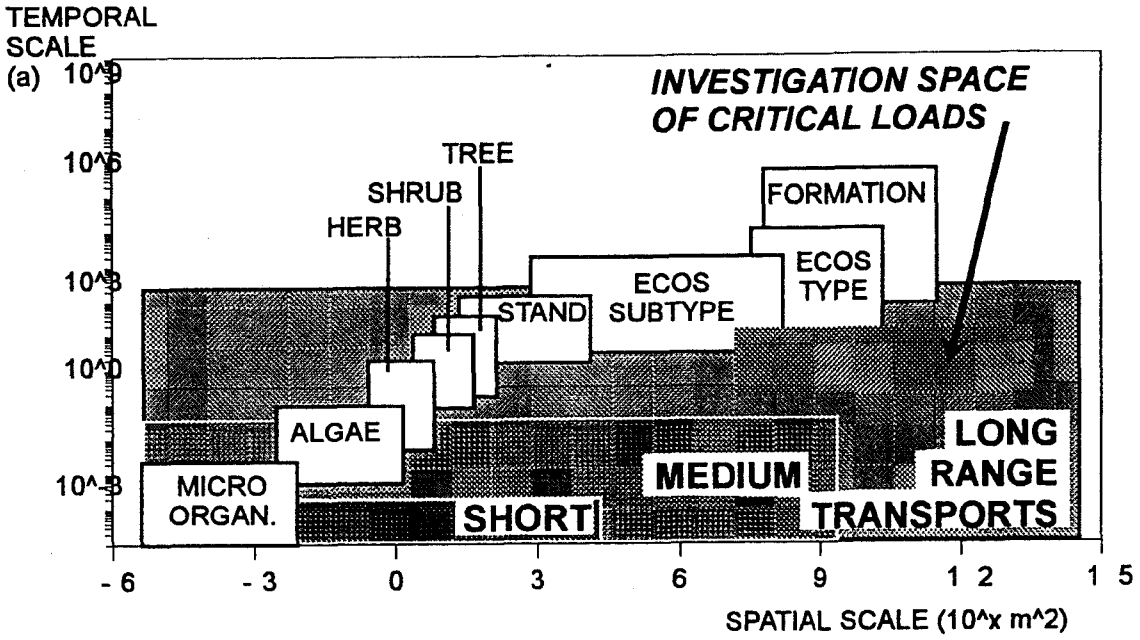


Figure 2: Spatio-temporal relationships of impact and receptor systems.

Primary impacts of air pollutants are only possible on receptors, which are exposed directly to concentrations or depositions (e.g. vegetation, exposed humans, suface water). In all other cases reactions of receptors, which were influenced previously, are influencing the quality and quantity of the remaining impact. Corresponding to the reaction chain in ground based systems the receptors can be characterised by the number of the reaction order as primary-, secondary-, and so on receptors.

This structure of order has to be distinguished from the concept of target receptors, which is influenced by the knowledge and human interests. Examples for target receptors are human, forest trees, fishes, communities of different organism species and so on. The target receptors are in general the objects of experimental investigations about the sensitivities on impacts and are also determining the basis for the definition of critical conditions. These definitions are used for transformations to parameters which can be observed easely in large areas. A typical example for such an approach is the definition of critical loads of acidity. The target receptors for the definition are forest trees or aquatic organisms, but the critical parameters are defined in chemical conditions of soils or surface waters.

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3. *Consequences for the definition of target receptors and critical conditions*

3.1 General aspects

The necessity to use substitutes for the calculation of critical loads should not diplace the insight into the principles of interactions between both systems and the consequences of it. Every exchange between the atmosphere and solids or liquids is strongly influenced by the conditions of the boundary layer. In the context of the human being and the physical foundations of it, surface ecosystems can be interpreted as an autocatalytic layer on the boundary between the atmosphere and the solid earth (KNOFLACHER, 1994). Other conditions are given in surface water systems, where the boundary processes are directly determined by the characteristics of water and the atmosphere.

Depending on structural and functional indicators it is therefore possible to classify the surface systems in basic categories (fig. 3). Every substance, which should be exchanged between the atmosphere and the deeper layers of the earth has to pass one of these categories. Critical conditions can develop in the systems of the boundary layers (ecosystems) or in the subsequent systems, but not independently of the state of the boundary layer. As a consequence of these interdependencies only one target receptor per area unit of the earth surface can be defined.

Because of the limited knowledge about the interdependent effects of several airborne substances on ecosystems it is very often necessary to consider the effects of different impacts seperately. In such cases it is possible, that different target receptors can be important for different substances. The above formulated conclusion has than to be extended, that per impact only one target receptor per area unit of the earth surface can be defined.

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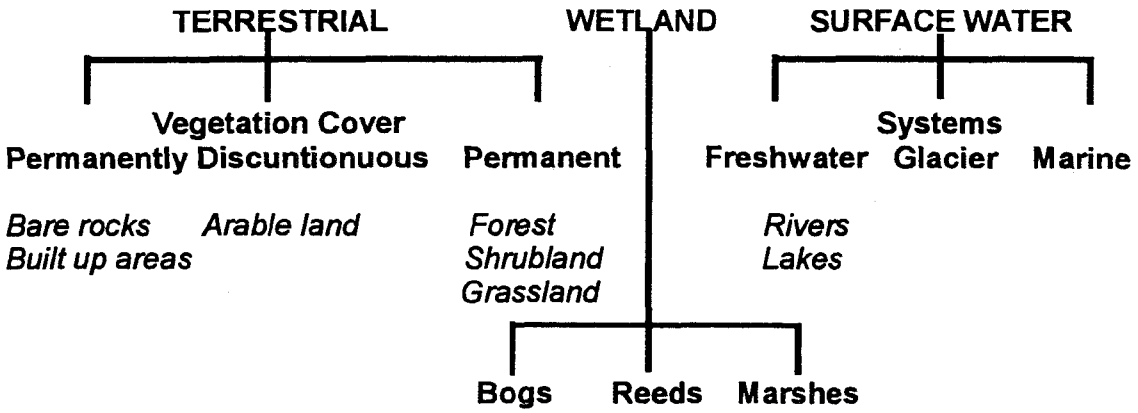


Figure 3: Functio- structural classification of ecosystems.

This definition leads to the question how target receptors can be defined in large areas, or in other words how far it is possible to extrapolate results from laboratory experiments or single spot field studies into larger areas? Different methods are used with different theoretical background and practical applications.

3.2 Specific aspects of receptor definition

The definition of receptors in environmental systems can not be done by simple causal analyses. Entities at specific scales (see also fig. 2) are functional connected with entities at other scale dimensions, so it is possible to define receptors at different scales by scientific methods. Because of the interdependencies between the different entities it is necessary it is at the scientific level also necessary to use a background conception about the purpose of the receptor definition. But the definition of receptors in environmental systems is not only a scientific task. Awareness and interest of the society on environmental problems are strongly influencing the weights of receptor importance. Both, scientific approaches and factors of social acceptance are therefore influencing the concepts of receptor identification and consequently also the receptor definition. Actually following concepts can be distinguished:

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a. the single species concept
b. the compartment concept
c. the landcover concept
d. the ecosystem concept
e. the landscape concept.

ad a: The single species concept is especially used in laboratory investigations and definition of critical levels for several species of organisms (e.g. human, plants). This concept is also supported by databases about the spatial distribution of single species. Under field conditions it has to be considered, that genetic variations within the species and different environmental factors are influencing the sensitivity of organisms at specific sites.

ad b: The compartment concept is predominantly used in the definition and calculation of critical loads (e.g. for soils, surface water, groundwater). Other compartments of ecosystems (e.g. the vegetation compartment) are considered as external values. Because the protection targets are not included explicitly it is necessary to derive critical values from field observations or to define it on the base of expert knowledge.

ad c: The landcover concept (e.g. in relation to the CORINE system) is based on geographical approaches. The definition of elements in this concept is strongly influenced by the survey method (e.g. satellite remote sensing). This concept covers categories of elements with different properties. The element itself can also be inhomogeneous in relation to functional properties as a consequence of the spatial scale.

ad d: The ecosystem concept is ideally oriented on biotic and abiotic factors. In reality it is often reduced to plant communities and their interactions with the abiotic environment (e.g. soil, water, air). It is a prefered concept in nature conservation (sometimes in combination with the single species concept), in general it is focussed on natural ecosystems but it is also possible to use it for other applications. The realization of this concept is dependent on the consideration of interactions between different compartments and of processes within the compartments. In existing examples the hierarchical relationships between different processes in ecosystems are used for the definition of entities at different scale levels (KLIJN et al., 1995).

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ad e: In this context the landscape concept has to consider functional interactions within landscape entities with strong internal relations between heterogeneous subunits (e.g. ecosystems). Receptors can be defined at the level of the subunits, but its weighting has to be defined on the basis of the landscape unit. Theoretically the weighting should consider the dynamic interactions between the subunits. First approaches are using structural criteria for the definition of the importance of the subunits (LENZ and STARY, 1995).

The spatial dimensions of the elements in the different concepts are relatively small in comparison to the spatial units of calculations of long range transports (e.g. 22500 km² or 2500 km²) :

Concept	Sptial dimension of a basic unit
	km²
Single species concept	10^{-8} - 10^{-4}
Compartment concept	10^{-4} - 10^{0}
Landcover concept	10^{-4} - 10^{-2}
Exosystem concept	10^{-3} - 10^{-2}
Landscape concept	10^{0} - 10^{2}

Transformations between elements of the mentioned concepts are in general equivocal, because of different properties of the elements. Additional parameters and specific framework conditions have to be considered in the tranformation procedures. A lot of questionable results can be produced, if transformations are carried out without respect to these constraints. To reduce the risk of potential errors because of these factors it is recommendable to strenghten in future the efforts to harmonize the concepts for receptor identification and definition. Minimum requirements for a sufficient definition of elements are continuity and consistency.

Basic parameters should exist continuous over the defined time period (e.g. decades or centuries). Such a reference matrix is necessary for a long term assessment of human made impacts in the dynamic environmental system. Because of the different dynamic properties of environmental processes (WHITE et al., 1992) a set of parameters with different dynamic characteristics should be used. An important subcriteria in this context is the potential continuity in the monitoring of the parameters. This asks for monitoring methods with a high robustnes against the variability in social interests on environmental problems.

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Consistency is necessary in transformations between different spatial scales. This requires a unequivocal definition of parameter categories which can be aggregated or diaggregated over different scales without a mishmash of relations between the parameters. A operational approach is the classification of the parameters in partial systems (KNOFLACHER, 1992) with an analogue classification of human caused impacts. Based on these classifications it is possible to aggregate complex entities at the desired level of scale and to assess the oberved or predicted effects.

The realization of concepts is also dependent and influenced by the strategy for the realization of a task. In the case of critical loads and levels three kinds of strategies can be used:

- the conservative strategy,
- the opportunistic strategy or
- the evolutionary strategy.

The target of a conservative strategy is the preservation of every defined receptor in an unchangeable status, independent of the consequences for the human system. Because of the ignorance of human society needs in this strategy, it is only realizeable in particular cases (e.g. for protection areas, for particular types of ecosystems or for some species). The application of this strategy in large areas is not very successful because of the increasing number of unsolvabel conflicts with the demands of the human society.

The target of a opportunistic strategy is the accomplishment of a task with a minimum of conflicts under actual circumstances. Results worked out under this strategy are strongly influenced by actual socio-economic conditions and will change from time to time. A complete realization of this strategy over longer time periods can therefore undermine every concept and will produce unpredictable results. If there is little existing knowledge about long term effects the opportunistic strategy is the only way to find broad acceptance for real solutions. In these cases the acceptance of an approach will be increased if receptors are familiar to the public.

The target of the evolutionary strategy is the optimization of dynamic processes in order to avoid critical conditions for all processes considered, but to accept fluctuations within this margin. This strategy can for short periods provide suboptimal conditions, which can provoke temporal conflicts which have to be managed. The realization of the evolutionary strategy needs therefore dynamic approaches for long term optimization of processes with complex interactions.

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For the calculation of critical loads/levels it is necessary to bring parameters of different spatial resolution into the resolution of the desired scale. If the spatial resolution of the original data is too high it is necessary to aggregate it, if the original resolution is too low the information have to be disaggregated. Objective rules, which are in general based on the applied strategy, are necessary for the execution of both procedures. The operationalization of the strategies will therefore produce different results in the definition of critical loads/levels in a single EMEP grid.

In the conservative strategy the rules will be formulated in a way, that the most sensitive value will determine the representative value (V_A) of an entity:

$$V_A = \min\{v_a : a \hat{I} A\}$$

This rule will produce an overrepresentation of sensitive areas, especially if large differences in spatial dimensions have to be treated with the aggregation procedure.

The opportunistic strategy is not strictly correlated with any operational rule. With this strategy it is therefore possible to choose different receptors and to use different rules (e.g. median value, mean, dominant value). Divergent results are consequences of this strategy.

Operational rules in the evolutionary strategy will refer to functions of receptor sensitivities instead of single values for seperated receptors. Additionally to sensitivity function it is possible that also demand functions (as minimum loads) for some substances will be formulated. An important precondition for the realization of this concept is the application of dynamic modelling in the spatial context.

3.3 Technical aspects

The survey and computation of spatio-temporal processes is strongly restricted by the available "space of operation" for the observation of relevant parameters. The "space of operation" can simply be defined by the number of parameters which are synchronously observable within an accepted time period. While the time dimension is primarily determined by the dynamics of the observed processes, the number of observable parameters per time unit is dependent on the available methods and the economic conditions.

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Ecosystems can be interpreted in the landscape context as parallel processors with different combinations and expressions of parameters. A sufficient "space of operation" to cover large areas with observations is only available for processes with low dynamics (e.g. geological processes, some soil processes and long term processes of the atmosphere). In all other cases the "space of operation" can be kept acceptable by a reduction of the number of observed parameters or of the spatial resolution of the observations. The strategy for the monitoring of complex processes under these restrictions is a optimization of the required information density in space and time.

At present, there is no harmonized integrated method available for the monitoring of environmental processes at the landscape scale . Only first approaches for small areas of particular interests are tested in the integrated monitoring programme. However, the most common case is a rather incomplete information based on data of different processes, acquired with different monitoring methods. The application of dynamic models is therefore restricted to some special cases, where sufficient data with harmonized quality is available. For the development of a broad basis for the application of dynamic models in the landscape context it is necessery to strenghten the efforts for an improvement of the existing situation in following main directions:

- The further development of methods for the integration of existing data into complex informations.

- The improvement of environmental theories and as an operational consequence the harmonization of relevant observation methods to close the existing gaps in the information structure.

4. Proposals for optimization

The dynamic modeling of critical loads in large areas is dependent on the availability of extensive data sets and on the possibility of consistent extrapolation of the results over surrounding areas. To meet these requirements with the economical and methodological constraints of field investigations it is necessary to optimize the existing approaches.

One important precondition for the optimization is the focus of the critical loads concept on long range transports. As explained above, impacts from long range transport are determining the framework conditions for regional systems. This can also be interpreted in the landscape context by a differentiation of the responsibilities of different scale

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impacts. Medium and short range impacts and its effects in landscapes has to be considered by national measures.

Assuming that structural or functional macrounits of landscapes are including ecosystems of different sensitivities, it can be derived that all ecosystems of these unit will be protected if the most sensitive system will be protected. Further it is assumed, that the existence of the most sensitive system is necessary for the existence of the macrounit. Based on these assumptions only one receptor per macrounit has to be defined. Depending on the knowledge about the effects of several impacts it is potentially possible to use different receptors for every impact or to use a "hot spot" of impact combinations with one receptor for all considered impacts. As weighting factors the real area of relevant ecosystems in the area or the required area of natural ecosystem (corresponding to the approach of LENZ and STARY, 1995) can be used.

With this approach it will be possible to reduce the number of spots for intensive field studies and monitoring systems. Technically a three step method is necessary for the realisation of the concept. The first step icludes the disaggregation of the investigation area into macrounits. In the second step the relevant systems and the target receptor(s) per macrounit have to be defined on the basis of existing data and knowledge. The final third step should include the calculations of critical loads for the investigation area.

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Critical loads at the landscape scale

M Hornung[1], B Reynolds[2], M Brown[3] and D C Howard[1]

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Institute of Terrestrial Ecology

[1]Merlewood Research Station, Grange-over-Sands, Cumbria LA11 6JU, UK.

[2]Bangor Research Unit, UWB, Deiniol Road, Bangor Gwynedd LL57 2UP, Wales, UK.

[3]Monks Wood Research Station, Abbots Ripton, Huntingdon PE17 2LS, UK.

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Critical loads at the landscape scale

M Hornung[1], B Reynolds[2], M Brown[3] and D C Howard[1]

1. Background

The critical loads approach has been developed as a science-based tool for optimising air pollutant emission reductions at both national and international scales (Bull 1991). As such it works best using regional data bases as input to national and international mapping and evaluation programmes. However there are a range of user communities who wish to apply the approach to much smaller areas. These users wish to know for, for example;

- the implications of national and international emission control strategies at the scale of individual sites or habitats,
- the local and regional impacts of changes in emissions from a particular national source,
- the impacts of national or international controls, or changes in emissions on individual elements within landscapes,
- the impacts of changes in land use in a specific catchment on the exceedance or otherwise of the critical load of the drainage waters.

National conservation agencies are particularly interested in the site or habitat level application of the critical load concept (eg Sketch and Bareham 1993; Woodin and Farmer 1993). Such sites might typically be of the order of 5km^2 down to 1km^2 or less. Furthermore, within a given site, there may be concern as to the effects of changes in atmospheric deposition on particular individual landscape elements, or even species of high conservation value. Environment agencies and rural affairs departments are also interested in impacts at the landscape scale, particularly in regions of heavily fragmented landscapes. They are concerned with the variations in sensitivity of the various landscape elements to a given pollutant load and the impact of given deposition scenarios on selected elements of the landscape (eg Bennett and Banerjee 1995).

National pollution inspectorates or environmental agencies are frequently involved in the assessment of the impacts of emissions from individual sources, whether this be a power plant

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or a large, intensive, animal rearing unit. Critical loads are seen as a way of quantifying the local-scale effects which can then be incorporated into the planning process. As above, the interpretations often need to consider the impacts on the different elements of the landscape affected by the source.

Afforestation of grassland or heathland in some areas can result in large changes in atmospheric inputs of pollutant and non-pollutant ions (Fowler *et al.* 1989). The increase in pollutants inputs can lead to exceedance of the critical load of the catchment stream or lake without an increase of emissions. There is therefore interest in using the critical loads approach in the evaluation of afforestation proposals at the catchment scale. This has been exemplified by a recent study at Green Burn in Scotland where the critical loads approach has been used to test two scenarios for the catchment (Nisbet *et al.* 1995). Excess sulphur inputs were modelled to the catchment for a hypothetical 100% acid moorland vegetation cover and for 60% cover with mature Sitka spruce. Irrespective of land use, deposition greatly exceeded the freshwater acidity critical load throughout the 1980's. However, if nationally planned emission reductions are implemented, deposition will be reduced to the critical load by 2003 for the catchment covered by hypothetical moorland vegetation. A delay of two years beyond this is predicted before deposition to the afforested catchment is reduced to the acidity critical load of the stream indicating that the afforestation is likely to have little impact on freshwater acidification at this site.

The application of the critical load concept at these smaller scales raises both conceptual, methodological and technical issues.

2. Conceptual considerations

In the context of acidic atmospheric pollutants, the critical load concept has been primarily used in connection with the optimisation of controls on transboundary pollution (Grennfelt *et al.* 1994). If site specific critical loads are not incorporated into the development of national maps which become part of the discussion and negotiation of protocols, the link with the control of transboundary pollution is lost. However, application of the concept at this scale is important as a means of identifying those valued sites, habitats or landscape elements which are at risk with current amounts of deposition, or under any given deposition scenario, and

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which sites will be protected by proposed national or international emission control protocols (Holdren *et al.* 1993). Within the UK, for example, the critical loads approach has been used to identify the areal extent of habitats at risk from excess nitrogen deposition. The plant species typical of calcareous species-rich grassland and montane-subalpine grassland were identified using the National Vegetation Classification data base (Rodwell 1991). The number of species from a list of 16 indicator species were then identified and plotted using data from the UK Biological Records Centre. Stock at risk was then defined as those 10 km squares in which more than a specified number of species (12 for the calcareous grassland and 4 for the montane-subalpine grassland) occurred. The choice of the number of indicator species used as a cutoff to define stock at risk was determined by expert opinion. The critical load for nutrient nitrogen determined for the respective vegetation communities at the Lokeberg workshop (Grennfelt and Thornelof 1992) were then assigned to the 10km squares defined by the stock at risk assessment. The exercise shows that under current nitrogen deposition, critical loads are exceeded for the montane-subalpine grassland in north Wales, the northern Pennines, the Lake District, parts of the Southern Uplands of Scotland and the southwest Scottish Highlands. The critical loads for the calcareous species-rich grassland are exceeded in parts of south central England.

The site specific application can be linked back to control when individual emission sources or pollutants with short atmospheric lifetimes are considered. Thus the short distance, downwind impact of a new power plant, changes in the fuel of a power plant or of a large isolated animal husbandry unit could be evaluated and used in making decisions about whether or not the proposed development should be allowed to proceed. This approach has been used in a recent evaluation of the effects of UK power station emissions on terrestrial vegetation. A Gaussian plume atmospheric dispersion model has been used to predict air concentrations of SO_2 and NO_2 under a variety of power station operating conditions. Gas concentrations attributable to the power station were combined with background values from non-power station sources, obtained from monitoring sites, to give average values for each 1 km cell within an area of 79x79 and 21x21 km around the power station. Both annual average concentrations were modelled and effects on vegetation were assessed by mapping the exceedance of critical levels for a variety of vegetation types occurring within the target areas. The distribution of land cover within these target areas were mapped using remotely

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sensed data (Fuller *et al.* 1994) aggregated to give a modal cover for each 1 km square.

The critical loads for different elements within a landscape cannot all be used in the development of emission targets and controls. Emissions cannot be controlled to influence deposition at this scale of resolution. However, the landscape scale approach can indicate the most sensitive element in a given landscape, which elements are at risk under a given emission/deposition scenario and hence possible changes in the landscape. Table 1 provides an example of this application and shows the soil acidity critical load for landscape elements of the partly forested Hafren catchment in Wales calculated using the Simple Mass Balance Equation (SMBE; Sverdrup and de Vries 1994), together with the freshwater steady-state water chemistry (SSWC) (Henriksen 1979) and diatom critical loads (Battarbee 1993) for the catchment stream.

Table 1. Soil acidity and freshwater critical loads for the Hafren catchment, mid-Wales.

Vegetation	Altitude (m)	Area (ha)	Critical load (keq ha^{-1} $year^{-1}$)
		Terrestrial ecosystem	
Conifers	350-400	35.0	1.24
Conifers	400-500	102.4	1.32
Acid grass	400-500	0.2	1.65
Conifers	500-600	41.8	1.47
Acid grass	500-600	79.1	1.74
Acid grass	600-690	98.6	1.83
Catchment	350-690	357.1	1.37
		Freshwaters	
Henriksen	350-690	357.1	0.94
Diatom	350-690	357.1	0.69

From Table 1, it is clear that freshwaters are the most sensitive component of this catchment system and that deposition reductions to meet the SSWC or diatom critical load will protect the terrestrial landscape components from further acidification. The most sensitive elements of the terrestrial landscape are the low altitude forest. This arises because the deposition of

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base cations to these landscape elements is relatively low compared to the predicted net base cation removal by the forest crop.

The interaction and potential conflict between local interest and national emission control policies is also demonstrated by this example. Table 2 shows the modelled current (1987/88) excess sulphate deposition to each landscape element and to the catchment as a whole, together with exceedance values calculated using these different inputs. Not surprisingly, excess sulphate deposition varies with vegetation type (essentially canopy roughness) and altitude, with greatest deposition to the high altitude forest. This is reflected in the pattern of critical loads exceedance within the catchment. The modelled deposition to the catchment as a whole is effectively an area-weighted average figure, and still results in 38% of the catchment area being exceeded.

Table 2. Modelled excess sulphate deposition (SO_4^*) and critical loads exceedance for the Hafren catchment, mid-Wales (ne = not exceeded).

Landscape element	SO_4^* dep	Landscape exceedance	Catchment exceedance
		$keqH^+ ha^{-1} year^{-1}$	
Conifers (350-400)	1.336	0.096	0.166
Conifers (400-500)	1.497	0.177	0.086
Acid grass (400-500)	1.040	ne	ne
Conifers (500-600)	1.734	0.264	ne
Acid grass (500-600)	1.200	ne	ne
Acid grass (600-690)	1.363	ne	ne
Catchment (350-690)	1.406	0.036	0.036

The basis for UK national policy with regard to critical loads exceedance is modelled atmospheric deposition at a 20 km grid square resolution (CLAG 1994). The value assigned to the 20 km grid square containing the catchment is 1.222 $keqH^+ ha^{-1} year^{-1}$ (1986-88 data). Using this input, none of the landscape elements are exceeded, all having a SMBE critical load greater than 1.222 $keqH^+ ha^{-1} year^{-1}$. Thus for national policy, the Hafren catchment area

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would be disregarded as receiving excess sulphate deposition below the critical load. However, using more detailed information, approximately 50% of the catchment area (all the forest) is actually receiving deposited sulphur acidity in excess of the critical load. It should be noted that the landscape deposition estimates are modelled using the same (ITE Edinburgh) deposition model as employed in the UK national critical loads programme, modified to provide data at a much finer scale of resolution.

Although the critical load concept is generally applied at the regional or national scale, the calculation methodologies which have been developed can be readily applied at the smaller scale. Indeed, regional maps can be derived from critical loads calculated for a sample of sites, for example, using a steady state model such as PROFILE (Sverdrup *et al.* 1992).

Most surface water critical load maps are derived from critical loads for individual lakes and this raises the issue of how such individual site data should be aggregated to formulate a national or regional assessment. This has been worked through in a recent example from the northeast United States (Holdren *et al.* 1993). The study compared the distribution of freshwater critical loads determined from individual lake data for the Northeast US region as a whole, with those for the component sub-regions of the Adirondaks, southern New England and the state of Maine. Cumulative frequency distributions of critical loads within the regions showed that whilst the Adirondaks had the largest percentage of low ANC lakes, this region did not contain the most sensitive systems. These were found in the southern New England population. As a population, however, the lakes in the Adirondaks were more sensitive compared to the populations for Maine or the Northeast US as a whole. Thus the choice of regional critical load was strongly determined by the proportions of lakes in each region to be protected. A critical load set on the basis of the Northeast US population statistics would leave a larger proportion of lakes unprotected in the Adirondaks compared to the state of Maine. Perhaps of more concern was the authors' observation that there was a large change in the number of exceeded lake sites within the regions depending on which model was used to calculate the critical load.

Dynamic models are almost always applied to specific sites as the required input data is rarely available regionally. A recent attempt at regional-scale dynamic modelling of the chemical

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response of European soils, using the SMART model, has pointed up some of the uncertainties in this approach (De Vries *et al.* 1994). Errors arising from uncertainties in system inputs and outputs such as precipitation amount, growth uptake and in soil constants such as weathering rates and denitrification factors were relatively small. Larger errors were associated with uncertainties in the initial conditions for soil C/N ratio and base saturation. These uncertainties related to both the poor spatial resolution of the soil mapping (1:5,000,000) and to the spatial variability of the parameters within mapped soil types.

Application of the SMBE for acidity to the calculation of critical loads for individual terrestrial habitats, or specific elements of a landscape elements, requires a critical chemical indicator for that habitat. If the base cation to aluminium ratio for fine roots is used as the critical chemical indicator, then a value is required for that ratio for an indicator species in the habitat. There has been little consideration of this approach to habitats other than forests, although a study in Maryland has considered a variety of non-coniferous tree species (Sverdrup *et al.* 1992).

3. Technical issues

The effects of 'down scaling' raises specific technical issues in relation to i) the availability of data at the appropriate scale and ii) the presentation of the data either in statistical terms or in map form.

3.1 Data availability

The main issue with respect to modelling critical loads at the landscape scale is whether there are suitable input data for the available critical load models to work sensibly at the scale of interest.

Empirical approaches to determining critical loads of acidity or nitrogen for terrestrial systems require data on soil characteristics and/or vegetation cover types. The application of the SMBE for acidity or nitrogen requires data for each input variable to the equation for the site, habitat or landscape element; the equation is then solved for each site or landscape element. The data requirements for the SMBE can involve atmospheric inputs of base cations, various soil attributes, water fluxes, nitrogen losses by denitrification and net nitrogen uptake.

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Dynamic models such as (eg SAFE and MAGIC) or more complex steady state models (eg MACAL and PROFILE) require an even larger array of input data. In landscape scale applications, the relevant data is, ideally required for each individual landscape element.

Thus, for the application of empirical approaches, vegetation/land cover and /or soil maps are required at the landscape scale. Data is rarely available at this scale of resolution for each of the input variables to the simple mass balance equation and information is therefore usually drawn from a mix of data sources with different scales of resolution.

Broadly, two types of data can be identified. Firstly the spatially detailed regional data (eg remotely sensed land cover data) which is available in raw form at high resolution (eg 25m x 25m) but can be aggregated up to $1km^2$ modal information and beyond (Fuller *et al.* 1994). So while the scale may change, the data classification does not. If the boundaries of the site of interest are used to 'cut out' a polygon from the land cover data, critical loads assigned to individual cover types at the national scale will be directly transferrable to the polygon representing the site.

Secondly, there are individual site specific data such as mapped, high resolution soil and vegetation data for example from a national nature reserve or a specific experimental site. In this case there are questions of data compatibility in trying to derive critical loads information from the site specific data and then relating this to the nationally mapped data and vice versa. eg relating detailed species level vegetation data within a well characterised nature reserve to the broader categories used in defining the empirical nutrient N critical loads in the Grange report (Hornung *et al.* 1995).

A third issue arises where the available data exist at a variety of different scales of resolution. There is then the problem of matching the information in such a way as to minimise errors. An example might be the derivation of surface water acidity critical loads for individual catchments where stream chemistry from the site have to be used in conjunction with nationally available atmospheric deposition data at $20km^2$ resolution as the UK. In some cases, in Table 3, quite large errors may be introduced in the critical loads calculated for specific sites if only national, large scale resolution data are used rather than the measured

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catchment values.

Table 3. SSWC critical loads calculated for five Welsh upland catchments using observed stream chemistry with deposition, rainfall and runoff derived from i) measured catchment data and ii) national data base values.

	Cyff	Gwy	Hafren	CI6	LI1
i)	1.747	1.024	0.942	1.517	0.238
ii)	1.261	0.610	0.735	1.386	0.778

In this example, the critical load for any given catchment may be under or over estimated using the nationally available data compared values obtained using the site specific information. As discussed earlier, such catchment-specific assessments may be required in connection with afforestation schemes in upland, acid sensitive catchments.

3.2 Data presentation

Environmental data used for deriving critical loads and the critical loads values themselves, can be presented in one of two ways. Firstly, information can be spatially referenced so that its geographical position on the ground is directly represented on the map. Examples of this information are maps of vegetation and soil distribution. Secondly, information can be presented as a statistical summary for a grid cell, for example the modal soil cover or soil critical loads for a 1km square. The two kinds of data are both equally valid, but the latter has particular limitations with respect to applications at the landscape scale.

i) Spatially referenced data

This information can be readily down scaled for local as opposed to regional modelling, although there will be limits in terms of the accuracy and resolution of the original data collected in the field. The flexibility of high resolution spatial data for landscape scale assessments is demonstrated by the relatively simple trial study examining the effects of scale on setting critical loads for acidity for five sample 1 km squares in the Welsh uplands (Reynolds *et al.* this workshop). Empirical soil acidity critical loads were assigned to the individual mapped soil units within five sample 1km squares in upland Wales and compared to the values assigned to the squares under the national mapping programme under which the

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critical load is set to the value of the most widespread soil. For all the squares the nationally mapped empirical soils acidity critical load was 0.2-0.5 keqH^+ ha^{-1} $year^{-1}$. In two of the sample squares, numbers 321 and 380, the most widespread soil was the most sensitive class of soils occurring within the square (Table 4). Therefore, all the soils in these squares would be protected if the inputs of acidity were reduced to the modal critical load which is the nationally mapped value. However, in three of the sample squares the most widespread soil was not the most sensitive soil occurring within the square. Thus, 43%, 40% and 9% respectively of the area of sample square numbers 260, 348 and 391 would not be protected if the deposition of acidity was reduced to the critical load of the most widespread soil. The soils in these unprotected portions of the squares would be liable to continued acidification. These more vulnerable soils are shallow, acidic rankers or ombrotrophic peats in square 260, peats in square 348 and peats or gleys in square 491. The results demonstrate the inevitable loss of detail and resolution that occurs within national mapping programmes compared to the variability in soil types actually present within 1 km squares.

Table 4. The allocation of soils into different critical loads classes for the sample 1km squares using the empirical classification, values indicate the percentage of the square covered by each soil type.

Critical load	Square number				
keqH^+ ha^{-1} yr^{-1}	260	321	348	380	491
0 - 0.2	43	- -	40	- -	9
0.2 - 0.5	57	93	60	71	91
0.5 - 1.0	- -	3	- -	29	- -
1.0 - 2.0	- -	4	- -	- -	- -
>2.0	- -	- -	- -	- -	- -

Digital data can also be overlaid to produce derived information using, for example a mass balance equation to combine information on soils, vegetation, deposition and runoff, however this may require a number of assumptions and simplifications in order to reduce the complexity of the landscape. For example, digital soil and land cover maps of the 5 sample 1 km squares in Wales were overlaid to produce a single map for each square with individual parcels representing unique soil/land cover combinations. The number of parcels produced

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within each square varied between 128 to a maximum of 556. Land cover and soils data did not match exactly, so that there may have been some erroneous combinations produced by combining information mapped by different groups. However these errors will be orders of magnitude smaller than those produced by combining national datasets which were collected for presentation at a scale of 1:250,000 or greater.

Additional uncertainties arise where information is combined at differing scales of resolution. In relation to the Welsh sample squares, the SMBE has been solved by combining mapped soil and vegetation data with a runoff figure for a $1km^2$ and deposition data for a $20km^2$ (Reynolds *et al.* this workshop). Sensitivity analysis of the model will provide information as to the main sources of uncertainty in the model predictions, which can then be related to the spatial uncertainty in the data.

Steady state models can be used to identify particular landscape elements that might be at risk under a variety of pollutant deposition scenarios. This approach has been used by Sverdrup *et al.* (1994) to assess the effects of acid deposition on the growth of Swedish forests. Provided the information is available, the approach can also provide a focus for more data demanding dynamic models which can be used to assess the detailed responses of individual landscape elements over time.

ii) Statistically summarised data

For the purposes of national and international negotiations, critical loads data may be aggregated in to larger grid cells such as 150km EMEP squares. There is a further loss of detail through summarising data statistically which can leave small, highly sensitive areas unprotected although its effect on the protection of such small landscape elements is not always straightforward. Table 5 shows how the protected proportion of each sample square would vary as a result of reducing acid deposition the critical load of larger grid cells containing the sample square. Reducing deposition to the modal critical load of the EMEP square would provide no protection to three of the sample squares, and limited protection to the other two (Table 5). In contrast, complete protection is provided for the sample squares by reducing deposition to the 5 percentile critical load value of the EMEP squares. This provides a strong argument in favour of the percentile approach in order to protect small areas

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of high conservation value. However, this will not provide information to show which specific areas are at risk within the grid cell nor identify their geographical location. Thus these information are not readily amenable to landscape scale analysis although the underlying data might be spatially referenced. In the UK, the smallest cell size is 1km^2, which generally too large for landscape scale assessments.

Table 5. The effect of 'upscaling' on the percentage of each sample square protected

		Modal			5 Percentile		
Square	1km	10km	20km	EMEP	10km	20km	EMEP
260	57	0	0	0	100	100	100
321	100	100	100	7	100	100	100
348	60	60	60	0	100	100	100
380	99	99	0	29	100	100	100
491	91	91	91	0	91	91	100

iii) Probability data

Within the UK the ITE Land Classification provides a probability of occurrence of 32 land classes for each 1km square in the UK (Bunce *et al.* 1981). Each land class is characterised by land form, topography, landscape, landuse, soils and vegetation and predictions are based on a population of surveyed squares. These data are of limited use at the sub-1km square scale although they can be used to pin-point areas potentially at risk.

4. Conclusions

Increasingly, a number of agencies are seeking to apply the critical loads approach to assess environmental impacts of atmospherically deposited pollutants at relatively local scales. This may arise in connection with specific planning or impact assessments, such as plantation forestry, or in regard to emissions from point sources. In addition there is considerable interest in the effects of nationally applied emission control strategies on landscape scale elements of the environment such as highly valued habitats and designated areas such as national nature reserves. In countries with a highly fragmented landscape, spatial aggregation

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of critical loads data can also leave small but highly sensitive portions of the landscape unprotected. Conceptually there is little problem in using the critical loads approach to assess these local or landscape scale impacts and a relevant literature is beginning to accumulate. The application of the critical loads approach at the landscape scale does not readily link with the requirement to optimise transboundary air pollution emission reductions. However, it can provide a useful basis for setting emission targets for individual point sources.

The main problem with landscape-scale applications of the critical loads concept lies in the availability of data for the various input variables at the desired scale. In particular, uncertainties arise when information at different scales of resolution have to be combined. The magnitude and direction of the resulting errors are not easily predicted and will depend partly on the sensitivity of the individual parameters within the particular critical loads model. Ideally, all the input data bases should be at the same resolution and this should also be appropriate to the scale of the application. Similarly for exceedance calculations, the atmospheric pollutant deposition data needs to be at the appropriate scale for the various vegetation or cover types under consideration. Whilst this seems an obvious requirement, in many cases the appropriate data are not available so that a number of simplifying assumptions have to made. In many cases, the potential errors associated with such assumptions are not adequately appraised.

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UNCERTAINTY ANALYSIS OF SPATIAL VARIABILITY OF NITROGEN CRITICAL LOADS

O.M. Golinets[1], V.N. Bashkin[2] and M.Ya.Kozlov[2]

[1] Pushchino State University
[2] Institute of Soil Science and Photosynthesis RAS, Pushchino, Moscow region, 142292 Russia

ABSTRACT

This study aim of this research is to present the comprehensive estimation of different approaches to analyses of nitrogen critical loads (CL) values under the computer calculations for terrestrial and freshwater ecosystems of Russia: uncertainty analyses and degree of spatial resolution. The CL values are used to set goals for future deposition rates of acidifying and eutrophying compounds so that the environment is protected. In this research CL values for various ecosystems are determined using EM GIS approach. Some software packages (e.g. Statgraphics and UNCSAM) have been used as tools for the analysis. It has been shown that both input source uncertainities and corresponding values of risk assessment were influenced by degree of spatial resolution used for collection of initial information databases. The data for analysis were collected from the large territories of European and Asian part of Russia with various scale. The most influential sources, such as nitrogen uptake by vegetation and nitrogen leaching from terrestrial to freshwater ecosystems are described for the whole territory of Russia.

Key words: Nitrogen, Critical Loads, Uncertainty Analysis, Spatial Resolution.

I. INTRODUCTION

At present there are already many publications devoted to quantitative assessment and mapping of sulphur and nitrogen critical loads at various ecosystems (Hettelingh et al, 1991; Critical loads..., 1992; Downing et al, 1993; Kamari et al, 1994; Bashkin et al, 1994 et al) with carring out of uncertainty analysis for a determination of the most influential input parameters on these values. However, until now the most given uncertainty analysis has been connected with regional values of critical loads. Analyses of regional variability and uncertainty especially in different scale are quite rare (Hettelingh, 1989; De Vries, 1991; Hettelingh and Janssen, 1993, Kozlov et al, 1995) and the majority of similar investigations are devoted to water quality data (Hettelingh et al., 1992; Posch et al., 1993; Kamari et al., 1993). Otherwise, time very often the uncertainty analyses should be done for multiscale conditions that is rather commen when one consider the large spatial variability of terrestrial ecosystems of Russia, China, USA, Ukraine etc countries having no real opportunity for the creation of information DBs with homogenous resolution. Consequently there is very important to estimate an adequacy of technique of uncertainty analyses to data sets created at different scale basis.

Consequently the aim of this research is to present the comprehensive estimation of different approaches to analyses of nitrogen critical loads (CL) values under the computer calculations for terrestrial and freshwater ecosystems of Russia: uncertainty analyses, risk assessment and degree of spatial resolution.

2.MATERIALS AND METHODS

2.1. Subjects of investigation

The main methodological and conceptual ideas in calculation of critical load values of nitrogen and sulphur for terrestrial ecosystems of Russia as well as the uncertainty analysis have been described earlier (Bashkin et al., 1993; Bashkin et al., 1994; Kozlov et

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al, 1995). Three different scale databases have been used for the goals of this study: DB for the calculation of critical loads for the European part of Russia in LoLa grid scale (1grad. longitude and 0.5grad. latitude); DB for the Asian part in LoLa grid scale (1grad. longitude and 1grad. latitude) and DB for the North-Western part of Russia (St-Petersburg, Karelia and Murmansk regions) in LoLa grid scale (15min. longitude and 30min latitude).

Quantitative assessment of CL using EM GIS is related to the subdivision of the whole area under study into "elemental" taxones with relatively homogeneous characteristics and to the collection of all available information for use in steady-state mass balance (SSMB) equations. Accordingly, an area comprising the whole European part of the Russia, characterized by great complexity of natural and anthropogenic conditions was chosen. This area was subdivided into 23 soil types and subtypes. Regarding the Asian part of Russia, this territory was divided into 48 soil types and subtypes and 45 vegetation types but for the aims of the given study the ecosystems of Regosols, Humic Cambisols, Cryic Gleysols, Gelic Podzols, Andosols, Dystric Cambisols, Eutric Cambisols, Luvic Phaerozems, Chernozems and Kastanozems soils were characterized. In the North-Western part 39 soil types and subtypes with corresponding ecosystems of tundra and forest types were divided.

Consequently, the complexity of soil cover, for ex.,in the European part of Russia is associated not only with a great number of soil types and subtypes but also with their different spatial distribution. One can see that the most widespread soils are Podzols (S10), Luvisols (S11), complexes of Podzols and Histosols (S12) and Chernozems (S17) and we have used only these predominant soils as examples during subsequent speculations and discussions. The most significant spatial distribution in the Asian part is connected with Cambisols and Podzols soils as well as with corresponding ecosystems. In the third region under study various Histosols were predominant.

2.2. Algorithm of critical loads calculations

The algorithm for computer calculations of critical load of nutrient nitrogen (CLnutr N) included the following equations:

$$CLnutr(N)={}^{*}Nu+{}^{*}Ni+{}^{*}Nde+{}^{*}Nl(crit)], \quad (1)$$

where * mean that each of the terms refer to the values at the actual total atmospheric precipitations at a side. Nu and Ni are permissible nitrogen uptake and soil immobilization, Nde is permissible denitrification and Nl(crit) is permissible critical nitrogen leaching.

Permissibe atmospheric nitrogen uptake (*Nu) was given as:

$$^{*}Nu=Nupt-Nu, \quad (2)$$

where Nupt-annual accumulation N in biomass
and Nu-annual uptake of N from soil.

Nupt was calculated accounting for the coefficients of annual biogeochemical turnover (Cb) the values of which values varied from < 0,1 up to >25. Annual Nu from soil was calculated on a basis of nitrogen mineralizing capacity (NMC) of soils which was determined experimentally or calculated using regression equations (Bashkin et al, 1993). So,

$$Nu=(NMC-Ni-Nde)Ct, \quad (3)$$

where

$$Ni=0.15\ NMC, \text{ if } C:N<10$$

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Ni=0.25 NMC, if 10<C:N<14
Ni=0.30 NMC, if 14<C:N<20
Ni=0.35 NMC, if C:N>20
Nde=0.145 NMC+6.477, if NMC>60 kg/ha/yr
Nde=0.145 NMC+0.900, if NMC<10 kg/ha/yr
Nde=0.145 NMC+0.605, if 10<NMC<60 kg/ha/yr

Permissible immobilization of atmospheric N deposition (*Ni) was found as:

*Ni=[(0.2 NH4+0.1 NO3)/Cb]Ct, if C:N<10 (4a)
*Ni=[(0.3 NH4+0.2 NO3)/Cb]Ct, if 10<C:N<14 (4b)
*Ni=[(0.35 NH4+0.25 NO3)/Cb]Ct, if 14<C:N<20 (4c)
*Ni=[(0.4 NH4+0.3 NO3)/Cb]Ct, if C:N>20, (4d)

where Ct - hydrothemical coefficient given as a relative part of St>50C from the annual sum.

Permissible denitrification from atmospheric deposition (*Nde) was found as:

*Nde=(Nde/NMC)Ntd Ct, (5)

where Nde/NMC - denitrification fraction, which depends on many features of soils and calculated on a basis of experimental data and Ntd - total N deposition.

Finally, permissible critical leaching of atmospheric nitrogen (*Nl crit) was given as:

Nl(crit)=QC_{Ncrit}, (6)

where Q - annual surplus of precipitation (run off) and C_{Ncrit} - permissible nitrogen concentration in surface waters.

Critical loads for maximum and minimum nitrogen, sulphur and acidity as well as exceedances for all studied parameters were calculated on a basis of Mapping Manual (Task Force on Mapping, 1993).

The calculations of sulphur and nitrogen depositions have been made on a basis of meteodata and emissions for 1991 yr regarding to the asian EMEP grid 150x150 km (Galperin et al., 1994). There are some definite ways of transition of pollutants from Europe to the northern Asia with their subsequent distribution under the influence of circumpolar air flows. This model calculation is not altered principally from previous results of K.Rahn and G.Shaw (Rahn and Shaw, 1982).

2.3. Uncertainty analyses

A contribution of input variables of the uncertainty of the model's results was estimated by using regression and correlation analysis of model outputs from corresponding inputs. For ex., in the case of the CLnut(N) model output is CL(N) and input variables are Nu, Nl, Nde, Ni. For different CLs, the analyses were carried out separately for various soil types. The uncertainty analyses were based on the assumption of a linear relationship between model outputs and input parameters. As a rule, the existing criteria are connected to an assessment of parts of the variance, influenced by any source in relation to the total variance value. Moreover, the measure of interrelation of input sources has an important meaning. Firstly the assessment of the uncertainty of input parameters based on the methods of regression analysis has been done. Since it is impossible to prove that input sources are not correlated absolutely, use of the criterium RTU (RooT of the Uncertainty contribution) is mostly correct (Kros et al., 1993). This critirium expresses the relative changes in the uncertainty of model outputs in relation to the relative changes in the uncertainty of input sources. The relative measure of sensitivity was assessed by criterium SRC (Standard Regression Coefficient) which

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characterized an alteration of computed outputs as a standard deviations from a corresponding alterations of input parameters.

For these purposes, the software package UNCSAM (UNCertainty analysis by Monte Carlo SAMpling techniques) has been developed by Janssen et al.(1992). This was used in our studies.

2.3.1 Monte Carlo method

Monte Carlo methods are widely used in the analysis of uncertainty and sensitivity. The methow which called Monte Carlo simulation was used in this stuty .

In the application of Monte Carlo simulation in the context of uncertainty and sensitivity analysis two main concepts are of impotance (Hettelingh, 1990):

1) sampling from parameter value ranges can basically be done in three ways, i.e. random sampling, stratified sampling and Latin Hypercube sampling;

2) The analysis of the relative importance of model parameters: the issie here is to quantify how much each of the parameters is contributing to the model responce.

The process of getting output coeffitients, that show the degree of sensitivity and uncertainty of input data, contents following stages.

The 0-th stage. The data can be imagined either as a set of experimental values or as a definite distribution function giving before. Whereas there are various distributions and, naturally, type, expected value, variance and, possibly, border values, then it needs to make samples of these distributions to obtain input matrice of values. For example, it needs to make N samples, and we have M distributios of input sources. Then applying, for instance, method of Latin HyperCube of M-degree N times we can obtain this matrice. Latin HyperCube is a one of the methods of sampling distributions, which can apply to non-correlative sources. Thus, there are M non-correlative sources xi, each of them is distributed by some distribution law. There is a row of the same repeating act for each xi: to obtain N values, the region of values is deviding on N non-intersected equalprobabititive intervals. One assidental value from each interval (with the corresponding law of distribution) is taken and, thus, N assidental values for each volume xi can be gotten. These acts are repeating M times for obtaining the assidental matrice M*N. Furthemore, the input sources and one or a few depended variables create , correspondingly, one or a few equations of linear regression

$$yj=xi*bi+b0$$

where: yi - depended variables
and xi - input sources.

The values of vectors yj (with the length eqial N, in this case) is getting only from experimental data. These speculations are correct for using the model of linear regression, because in the majority of cases this model expresses adequately the dependence between the sources xj and the outputs yj. In the case, when the linear model does not provide a good approximation, a bit-linear model can be used, i.e. the region of definition of xi is deviding on a few close regions, intersecting only on the borders. On every region own linear function (or a few yj) is determining. Such approximation allows an adequate description, practically, of any function and, correspondingly, it is possible to give a description of this function relatively correctly helping linear of bit-linear regression model, even when the exact kind of dependence yj from xi is inknown. In the case, if it is not possible to obtain qyantitative approximation by a simple linear model, the way of substitution ofinput values of xi on their ranking values often gives a success,

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i.e. the region of values of each xi is deviding on a few segments. Then the index of segment, that contain this value, is giving on the input instead of the value of xi. This method is called as the method of ranking regression and it works in the ajority of cases too. In this stage using the method of Monte Carlo simulation there is an opportunity to substitute the sampling of values by the input of the experimental data table.

The 1-st stage. Having the matrice of valuea M*N and one or a few vectors with a N-length on the input, M values of variance and expected values (the same prossess for yj) can be calculated and the values of correlation between the columns of matrice (between xi). These values being selfinterested are using for calculation of output coefficients in the second stage.

The 2-nd stage. The values of yj and xi are processing by the method of Monte Carlo or , in another words, by the method of statistical tests, which includes the determination of values (that necessary for following calculations) by the input sample (one statistical test) and their precisition with the use of subsequent vector. This processing is continueing upto the complete exhasting of all values (in this case N tests), i.e. in this case the method of Monte Carlo includes the modelling of accidental numbers of samples (vectors yj(k), xi(k), j=1,...,r; i=1,...,m)) with the following calculation of various coeffitients for the uncertainty analysis.

The 3-d stage. Using the values of correlation, various coeffitions are connected with the measures of sensitivity and uncertainty:

ORC - the measure of absolute sensitivity;
SRC - the measure of relative sensitivity, connected with a standard deviation;
RTU - the measure of uncertainty, indepentent from the volumes of correlation.

It is known that the abovementioned coefficients SRC and RTU are the most useable in analysis of sensitivity and uncertainty. These coefficiets have the largest area of application where almost all the studied cases are occuring (Golinets,1995).

3. RESULTS AND DISCUSSION

Table I shows the uncertainty (RTU) and sensitivity (SRC) contribution of input variables to the critical loads of sulphur, nitrogen and acididy for various ecosystems of the Asian part of Russia, the European part and the North-Western regions of Russia.

Regarding the Asian part, one can see the following. The strongest influential factor for the values of CLnut(N) is the parameter Nu. For the biggest part of Siberia and Far East territory this parameter has the first rank and only in the case of Humic Cambisols and Cryic Gleysols ecosystems is its rank second. Among others, Ni and Nde parameters, which are closely intercorrelated, are the weakest ones. The given parameters do not practically have any influence on the values of CLnut(N), except Dyctric Cambisol ecosystems. The influence of Nl values is decreasing in the following row of ecosystem forming soils: Regosols > Humic Cambisols > Cryic Gleysols = Gelic Podzols > Andosols > Dystric Cambisols > Eutric Cambisols > Luvic Phaerozems = Chernozems > Kashtanozems. These results reflect, in significant degree, the geographical change of ecosystems from north to south and correspondingly an alteration of relationship of temperature and moisture constituents in hydrothermic coefficient Ct. So, the main impact to the assessment of the influence of inputting parameters (Nu, Ni, Nde, Nl) on both uncertainty and sensitivity of outputting values of CLnut(N) belongs to Nu. It is connected firstly with a deficit of N as the main nutrient in all studied ecosystems as well

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as an existing spatial and temporal variability of this parameter that relates to a significance and correctness of experimental and computed values of Nu. In accordance with relatively better knowledge of hydrological picture and relatively homogenous values of critical concentration of nitrogen in surface waters, CLcrit, included in the calculation of Nlcrit values, the input of the given parameter into the uncertainty of CLnut(N) is expressed in a lesser degree. Furthermore, the runoff processes are practically not significant for ecosystems of Luvic Phaerozems, Chernozems and Kashtanozems due to an exceedance of evapotranspiration above precipitations. During the calculations of CLnut(N) for ecosystems of the Asian part, the values of critical immmobilazation and denitrification of N depositions both in relative and absolute meanings played a subordinate role that obviously reflects their minor contribution into uncertainty and sensitivity analysis of the computed output values (Table I,Fig 1-4).

In regards to the European part of Russia, one can see that the most important contribution in the uncertainty, based on this criterium for the majority of soils, is connected with Nu and the second most important rank varies for different conditions (Table II and III). The analogous conclusion can be made on a basis of SRC criterium which shows that the model is most sensitive to parameters Nu and Nl for all soil types and to parameters Nl and Nde for Luvisols.

During the assessment of eutrophying N parameters [CLmin(N), CLnut(N), CLmax(N)], the predominant influence is connected with the parameter of critical nitrogen uptake, Nu. It should be noted, however, that the application of different uncertainty/sensitivity and risk assessment must not be absolute, because a number of possibilities, whose correctness is not obvious, are suggested by this analysis. Nevertheless, the demonstration of the "importance" of the uncertainty of the source, Nu to the uncertainty of model outputs for various soils is scarcely likely to be accidental.

4.CONCLUSIONS

Use of various statistical approaches in an assessment of values of critical loads of N in terrestrial ecosystems of different regions of Russia allows for improvement in the interpretation of obtained results and the determination of sources of discrepancies related with Nu and Nl. To a large extent, the interpretation of data can be done through the application of uncertainty and sensitivity analyses which permit the estimation of both the uncertainty of input variables and the uncertainty of model outputs.

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Table I

Percentage of uncertainty contribution in input variables to the critical loads of nutrient nitrogen for various ecosystems of Asian part of Russia

ecosistem forming soils	sensitivity and uncertainty	Nu	Nl	Nde	Ni
Regosols	SRC	50	41	2	7
	RTU	30	27	21	22
Cryic Gleysols & Humic Cambisols	SRC	37	56	2	5
	RTU	22	37	21	20
Gelic Podzols	SRC	47	45	3	5
	RTU	40	39	10	11
Andosols	SRC	81	17	1	1
	RTU	61	30	1	8
Eutric Cambisols	SRC	41	0	30	29
	RTU	63	10	14	13
Distric Cambisols	SRC	96	2	1	1
	RTU	38	32	16	14
Luvic Phaerozems and Chernozems	SRC	96	1	1	2
	RTU	65	6	12	17
Cashtanozems	SRC	95	0	5	5
	RTU	61	5	19	15

SRC - Standard Regression Coefficient
RTU - RooT of the Uncertainty

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Table II

Uncertainty contribution of input variables to the critical loads of S, N and acidity for various ecosystems in different regions of Russia, percentage.

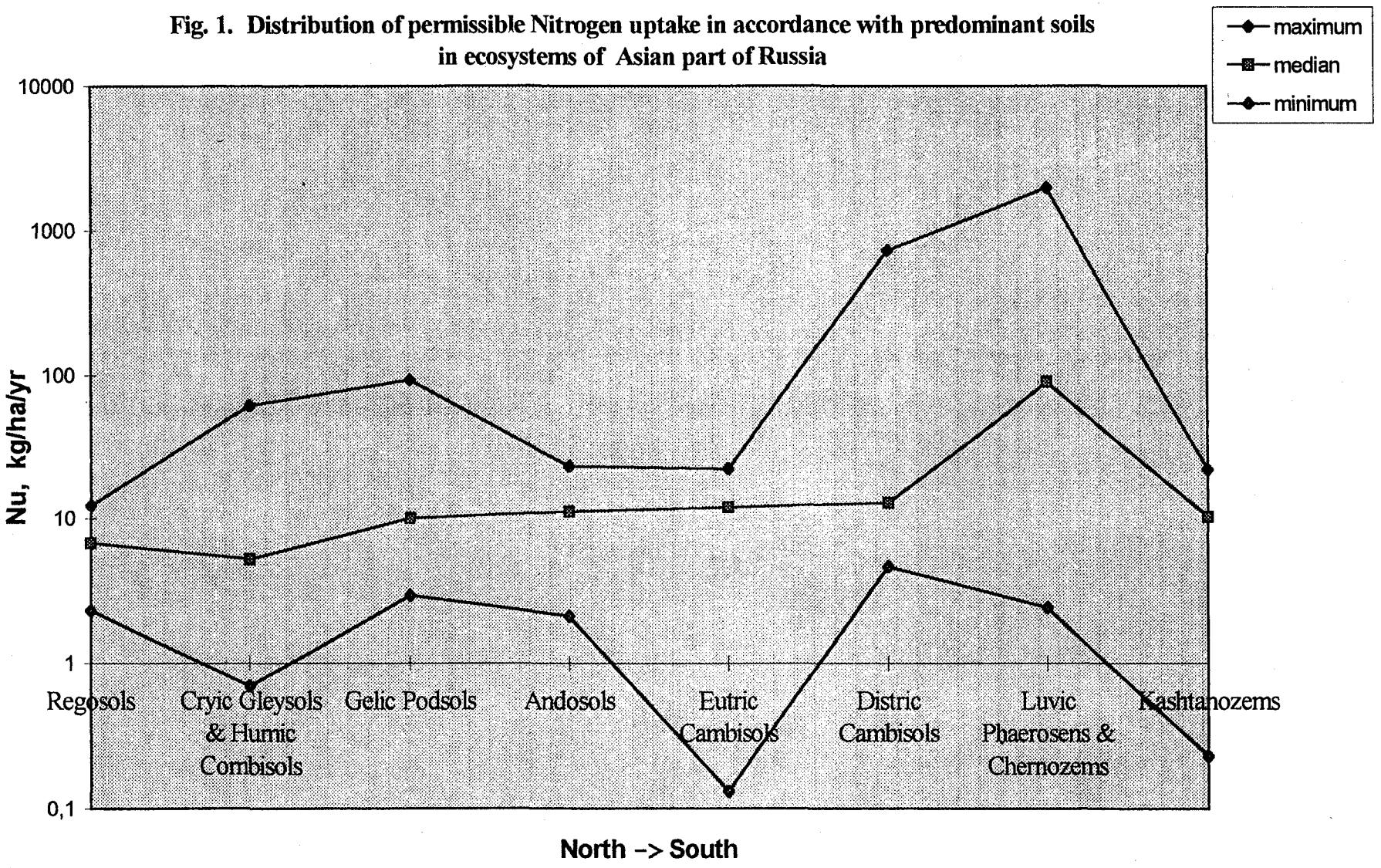
Uncertainty and Sensitivity			Variables Nu	Nl	Nde	Ni	ANCw	ANCl	(BCd-BCu)
Northern Asia (1° * 1°)	RTU	min	22	6	1	8	-	-	-
		mean	47	23	17	14	-	-	-
		max	64	39	21	22	-	-	-
	SRC	min	40	0	0	0	-	-	-
		mean	67	21	5	6	-	-	-
		max	97	55	30	29	-	-	-
European part of Russia (1° * 0.5°)	RTU	min	25	19	8	10	-	-	-
		mean	38	32	14	15	-	-	-
		max	49	46	18	22	-	-	-
	SRC	min	21	25	1	3	-	-	-
		mean	48	43	2	5	-	-	-
		max	61	71	3	10	-	-	-
North-Western part of Russia (15′ * 30′)	RTU	min	51	14	10	17	17	14	20
		mean	53	18	10	25	40	17	44
		max	57	21	10	42	84	21	60
	SRC	min	56	8	6	12	22	5	0
		mean	69	11	6	23	43	10	42
		max	84	13	6	43	85	14	65

Table III

The relative percentage of some input variables for various soils in the European part of Russia according to RTU

variables	S10	S11	S12	S17
Nu	37	14	39	49
Nl	32	46	19	31
Nde	16	25	20	9
Nim	15	15	22	11

Fig. 1. Distribution of permissible Nitrogen uptake in accordance with predominant soils in ecosystems of Asian part of Russia



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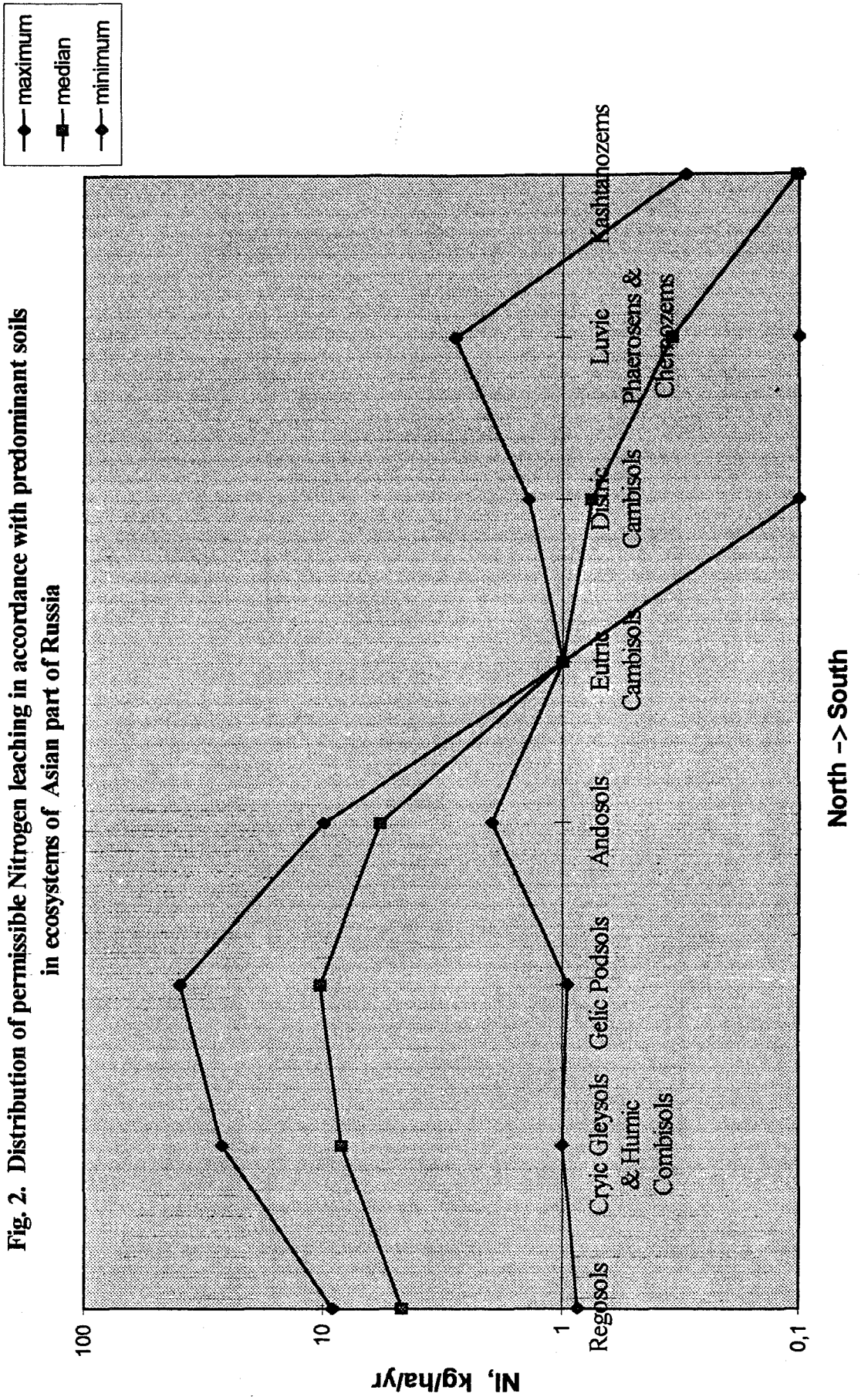


Fig. 2. Distribution of permissible Nitrogen leaching in accordance with predominant soils in ecosystems of Asian part of Russia

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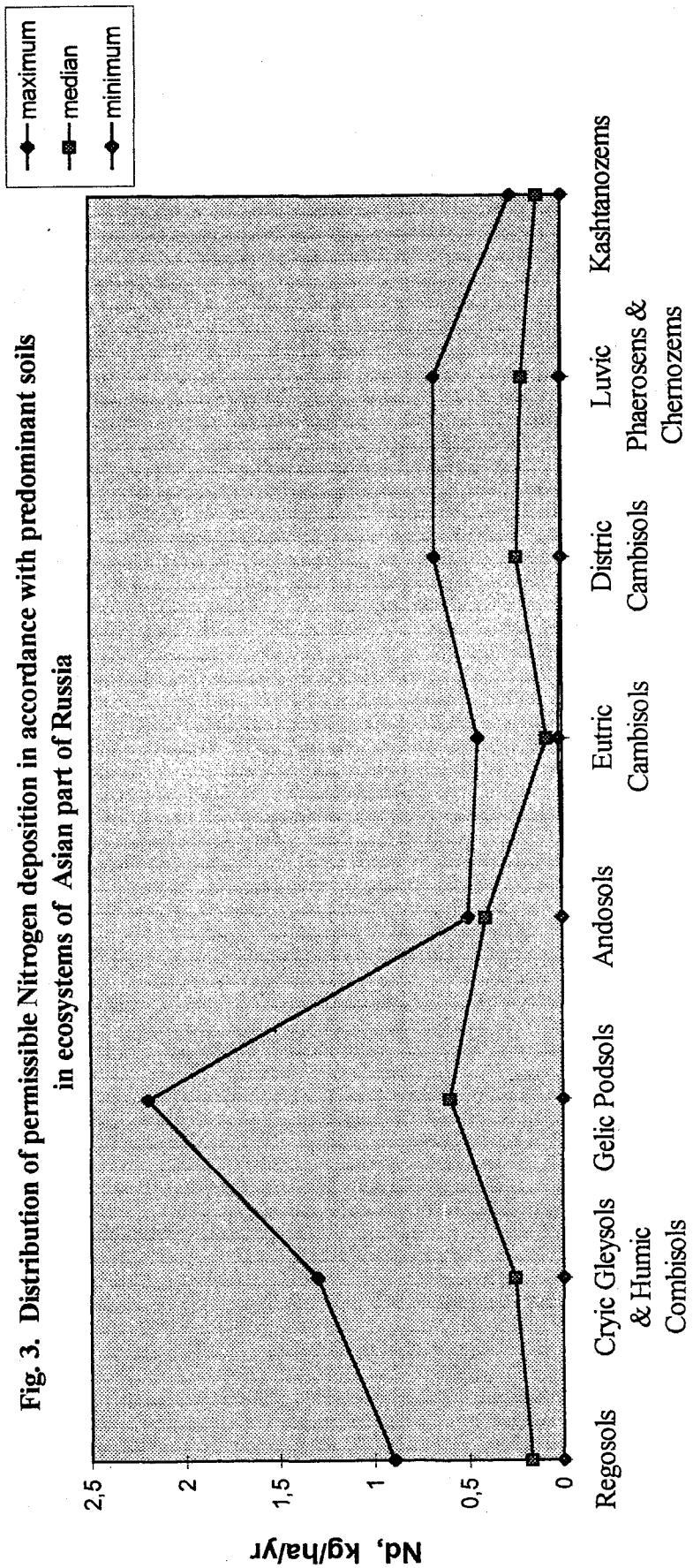
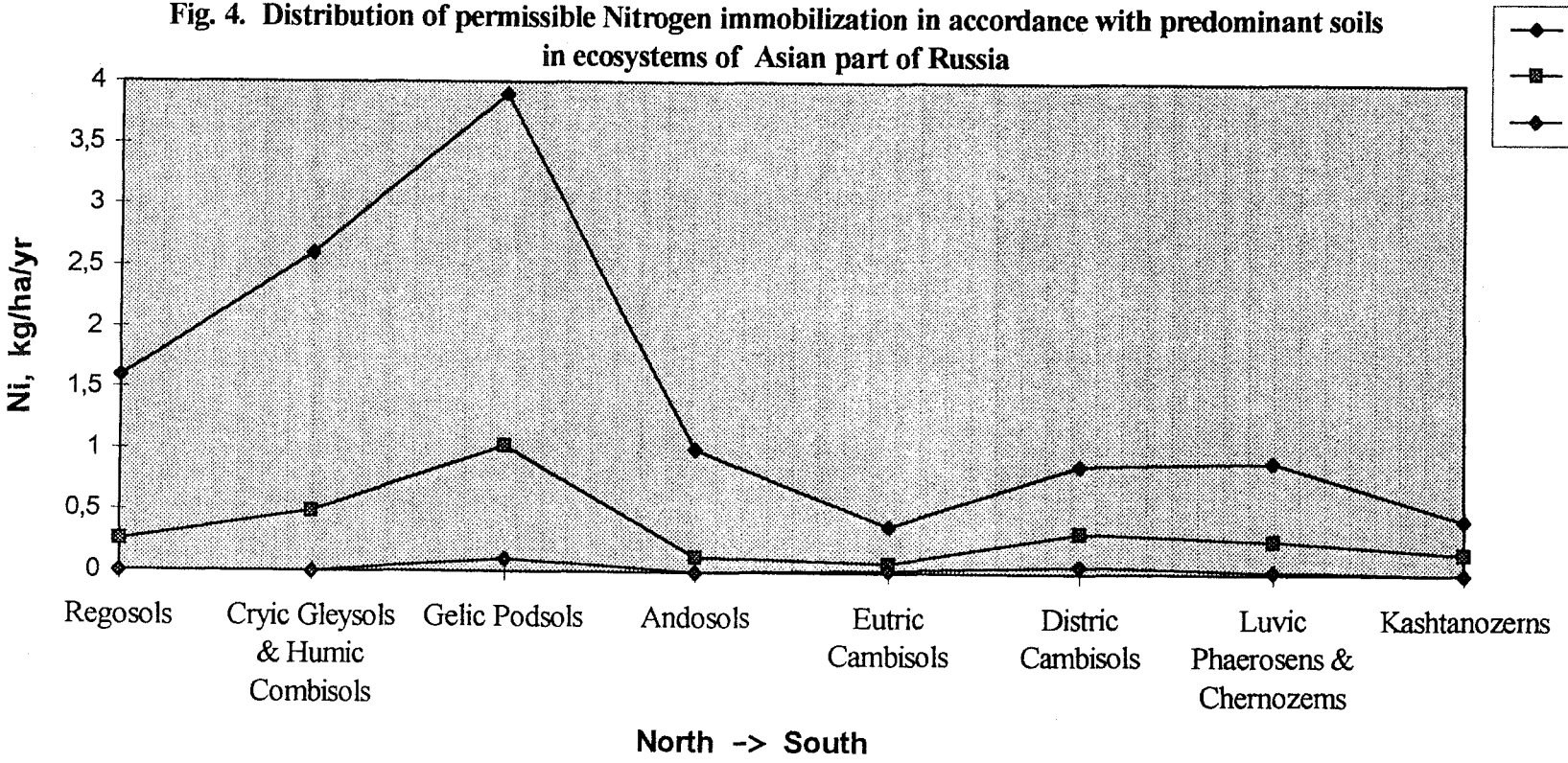


Fig. 3. Distribution of permissible Nitrogen deposition in accordance with predominant soils in ecosystems of Asian part of Russia

Fig. 4. Distribution of permissible Nitrogen immobilization in accordance with predominant soils in ecosystems of Asian part of Russia



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SSMB CRITICAL LOADS FOR NUTRIENT NITROGEN, THEIR EXCEEDANCE AND A TEST OF THEIR VALIDITY: A CASE STUDY IN NORTHERN GERMANY

A. Branding[1], E.W. Reiche[1], D. Nagel[2] & T. Spranger[3]

[1] Ecosystem Research Center, Kiel University, Schauenburger Str. 112, 24118 Kiel, Germany

[2] ONÜ GmbH, Müncheberger Str. 1, 15345 Prädikow, Germany

[3] Umweltbundesamt, FG II 1.2, Mauerstr.52, 10117 Berlin, Germany

Abstract

Critical loads for nutrient nitrogen are calculated for a beech ecosystem typical of Northern Germany. The study area is strongly affected by agricultural activities in the surroundings. The elements of the steady state mass balance critical load for nutrient nitrogen (estimated for this site based on the data available on the national scale from the German NFC for Mapping) are compared to more detailed site specific empirical data obtained from the Bornhöved ecosystem research project. Critical loads exceedances are estimated from 5 years' throughfall plus stemflow measurements, corrected for canopy uptake by using canopy budget models. High exceedances largely due to ammonia/ammonium input originating from agriculture are found. It is shown that inputs are systematically underestimated if uncorrected throughfall measurements or bulk deposition measurements multiplied with so called filter factors are used. Two scenarios are combined with a dynamic model and changes in fluxes and pools are observed during a 30 year simulation period. This aims at testing the basic assumption of steady-state critical load models that structure and function of the ecosystems should not be altered in the long term under critical loads. As well it is estimated what would happen to the ecosystem if the present high nitrogen loads continue.

Introduction

The steady state mass balance (SSMB) is used to establish critical loads for deposition to soils and to provide critical load maps on a European Scale. The concept of the SSMB is based on several simplifying assumptions and requires only a small dataset which is an important advantage on the one hand. On the other hand the use of this concept has serious disadvantages: only qualitative information is given about the effects of the critical loads exceedance and about the timescale of ecosystem changes as a result of elevated deposition (FERRIER et al. 1995). Since it is always questionable wether the soil system is under steady state conditions it is difficult to validate the SSMB approach and the critical loads established with this method. By using dynamic models it is possible to evaluate the quantitative impacts on the soil system. Dynamic models can be applied on different timescales and provide information about the behaviour of pools and fluxes under proposed environmental situations. Although they require larger datasets and are therefore mostly applicable to sites, they provide useful information to validate critical loads.

The objectives of this paper are 1) to apply the SSMB [National Critical Loads Data Set] to the Bornhöved beech forest 2) to compare the SSMB elements with the actual fluxes of the, beech forst on a site level 3) to estimate the critical loads exceedances using different data sets and methods and 4) to apply a dynamic model for a retrospective analysis, for a trend scenario under present deposition and for a critical loads scenario.

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Material and Methods

The field data used for the dynamic model were obtained from the network *Ecosystem Research Bornhöved Lakes Area*. Actual N-fluxes from 5 years of continuous measurements of nutrient cycles provided extensive experimental data of the nitrogen cycling within a beech forest.
The research area is sited in the weichselian-moraine landscape of Schleswig-Holstein about 30 km south of the city of Kiel and the westernmost outskirts of the glaciation. The small 95 year old beech forest (1st generation after former agricultural use) is surrounded by fields with intensive agricultural activities. The pronounced variability of ecosystems is typical for landscapes of Northern Germany. The parent materials of the Cambic Arenosols (FAO Classification) are highly waterpermeable glacial sands (see Tab.1). The eutrophic ground vegetation of the forest (*deschampsia flex.*, *urtica dioica*) reflects the nutrient status of the system.

Tab.1: Chemical properties of a Cambic Arenosol (German Classification: brown earth) under beech Forest in Northwest Germany; n.d. = not determined [Data from: SCHIMMING & WETZEL 1995]

Hor.	lower depth	Stones	Sand	Till	Clay	C_{org}	N_t	pH	CEC	H^+	Al^{3+}	basic cations
	cm	%					g/kg		$mmol_c$			
L	3,5-2,5											
Of	0.											
Aeh	5	11.6	78.50	16.0	5.50	3.40	2.0	3	66.20	15.90		20.90
Ah	28	26.0	80.0	16.90	3.10	0.90	1.0	3	24.10	3.20		2.20
AhBv	38	59.20	84.0	13.50	2.50	n.d.	n.d.	4	8.70	0.40	6.60	1.70
Bv1	60	56.40	92.90	5.0	2.10	0.40	0.40	4	6.10	0.10	4.90	1.10
Bv2	91	23.20	96.30	2.3	1.40	0.40	0.20	4	6.40	0.50	4.30	1.60
rGBv	110	11.30	97.90	1.5	0.60	0.10	0.10	4	9.0	0.30	2.40	6.30
Bv3	130	12.30	97.60	1.6	0.80	0.10	0.10	4	6.60	0.60	3.10	3.0
BbsC	154	2.0	99.20	0.5	0.30	0.10	0.10	4	3.60	0.50	1.20	1.90
BvC	165	21.70	98.0	1.2	0.80	0.10	0.10	5	11.10	0.20	1.40	9.50

The soil nitrogen dynamics were determined by measuring inorganic nitrogen concentrations of the soil solution (ceramic cup samplers). Net mineralisation was determined by breeding soil samples in polyethylene bags. Throughfall and stemflow were determined at 4 sites within the forest, wet deposition and bulk deposition at several sites outside the forest. The total atmospheric deposition to the forest was calculated by using the throughfall method combined with a canopy-interaction model established by ULRICH and van der MAAS (described in DRAAIJERS et al. 1996).

Comparison of the elements of the steady state mass balance and empirical data used for dynamic modelling

A comparison between the national critical loads data set provided by the German NFC for mapping and empirical data for the Bornhöved Lakes Area was made. Tab. 2 shows the elements of the mass balance of the national critical loads data set and the empirical data on site level. All actual fluxes are higher than the steady state mass balance, especially the nitrate leaching. The critical load is estimated at 13,7 kg ($ha^{-1}*a^{-1}$) and is most probably exceeded. The rather high critical load value is due to high uptake rates under critical loads (see soil characteristics Tab. 1).
Tab. 3 shows the N-deposition to Bornhöved beech forest calculated with different methods. There are large variations between the deposition estimates. The interpolated deposition from

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the national data set is much lower than the deposition calculated by using throughfall data in combination with the canopy-interaction model (DRAAIJERS et al. 1996). This canopy-interaction model considers canopy uptake as well as dry deposition of N-compounds determined at the site and is therefore more a more accurate way to estimate the total N-input than using e.g. filter factors. The critical loads exceedance is so high that reliable deposition data become more important than reliable critical loads data. This might be a general conclusion for those areas in Central Europe where the critical load for nitrogen is strongly exceeded.

Tab. 2: The elements of the steady state mass balance from the national critical loads data set as compared to empirical data on site level used for dynamic modelling (all values kg/($ha^{-1}*a^{-1}$)

	immobilsation organic matter N_i	net removal vegetation N_{up}	denitrification N_{den}	leaching N_l
steady state under critical load	1,5	11,5	<1	0,5
dynamic model with actual data (no steady state) (1989-1992)	2	20	1-2	17

Tab. 3: N-Deposition onto Bornhöved beech forest estimated with different methods

Method	N-Deposition kg/($ha^{-1}*y^{-1}$)
Interpolated Deposition from National Data Set Bulk Deposition x Filter Factor	26
Actual Data Bornhöved: Throughfall measurements 1989-1993	
Bulk Deposition x Filter Factor	21
Throughfall	24
Throughfall & Canopy Interaction Model (DRAAIJERS et al. 1996)	40

The dynamic model WASMOD

To understand the present system behaviour with respect to N-fluxes and to estimate future behaviour a dynamic model (REICHE 1994) was applied (see Fig. 1). The model system "WASMOD" integrates methods of landscape analysis and ecological process modelling in order to describe the water budget, the dynamics of soluble and adsorbed substances in the soil, as well as different processes of the soil nitrogen and carbon budgets. Model simulations can be done with a spatial resolution that varies from single sites to large catchments. The model describes in detail transport- and transformation processes in the upper soil horizon and the root zone. Modules calculating processes related to the vegetation and to the unsaturated/saturated zone have been considered with a higher degree of generalization. Usually data which characterize the water capacity and hydrological conductivity are not available. Therefore the model system includes transformation procedures which estimate those values as a function of soil texture and content of the organic matter. The interception of rainfall by the canopy is calculated as a function of precipitation intensity and the leaf area

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index. The rates of actual transpiration are computed with consideration of soil water potential and plant specific parameters like root depth and vertical root destribution. In order to characterize the nitrogen and carbon transformation processes, plant uptake and return via litter fall has to be calculated accurately. Growth stage dependent assimilate demands are estimated for different tree compartments for modelling the assimilate distribution. Water or nitrogen deficiency causes a reduction of biomass growth. The input parameters which characterize the phenological and physiological behaviour of a beech forest were modelled by HOFFMANN (1995) based on results of the Solling Project (ULRICH et al. 1986).
The simulation of nitrogen uptake by the tree depends on the transpiration rate, the amount of soluble nitrogen and a specific maximum uptake capacity, which is defined by the plant age and can be reduced in case of stress-conditions. The relocation of nitrogen and carbon is calculated by assuming a constant C/N ratio for the stem wood. The calculation of nitrogen turnover rates in the upper soil layer closely connected to the dynamics of organic carbon in the soil. Therefore the submodels describing the mineralization and immobilization are linked to a submodel which simulates the turnover of organic carbon (HANSEN et al. 1990). This part of the model is based on the concept of JENKINSON et al. (1987). The organic pool is divided into 3 subpools:

- added organic matter (AOM)
- dead native soil organic matter (SOM)
- microbial biomass (BOM)

The subdivision of each subpool into two other subpools enables the computation of the transformation rates by a first order kinetic. The idea of this concept is that the physical and chemical properties of the dead organic matter varies in a wide range. The two subpools represent this range by characterizing a fraction of slow turnover processes on the one hand and faster turnover processes on the other hand. As a consequence the quantitative relation between the two pools regulates the transformation rate. The microbial biomass also is divided into two subpools in order to represent the range of different organisms due to their dynamics of population development. On the one hand there are species which have adapted to not easily decomposable compartments, where the population changes over long periods. On the other hand there are organisms adapting very fast to changes in nutrient conditions. The decomposition rate can be calculated by the following equation.

$$\zeta = K_x C_x$$

ζ_x = decomposition rate of the pool x (kg C $m^{-3} s^{-1}$)
C_x = carbon concentration in soil of the pool x (kg C m^{-3})
k_x = decomposition coefficient for pool x (s^{-1})

In a first step the coefficient K is defined due to standard conditions (clay content= 0%, temperature= 10°C, water content= field capacity, pH value>= 5). For each of the soil compartments the coefficient varies due to changing values of the soil water content, temperature, acidity and clay content.
The quantification of the nitrogen mineralization is a result of all carbon turnover rates divided by their C/N ratios. In periods which are characterized by CO_2 production organic nitrogen will be transformed into ammonium. Otherwise the mineralization process begins after the addition of fresh organic matter and immobilization will be simulated because of the wide C/N ratios. Carbon transformation processes are reduced by NO_3^- and /or NH_4^+ deficiency.

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Fig. 1: Partial structure of the model WASMOD (REICHE 1994) describing nitrogen cycling of ecosystems

Tab. 4: Important subprocesses of the model system WASMOD (REICHE 1994)

A	for single site simulations
1.	simulation of soil water dynamics (incl. interception, evapotranspiration, infiltration) simulation of drain flow
2.	simulation of ground water flow
3.	simulation of vertical solute transport in soils
4.	simulation of carbon assimilation and distribution of assimilates
5.	simulation of N-uptake via plant roots and nitrogen return via litter
6.	simulation of ad- and desorption dynamics in soils
7.	calculation of vertical transport of soil temperature
8.	simulation of carbon turnover processes in the soil
9.	calculation of microbial controlled nitrogen turnover in soils and at the soil surface including
10.	gaseous N-losses
B	as a model on regional scale
1.	simulation of overland flow
2.	calculation of regional water and nitrogen budget (particulary for water catchments)
3.	calculation of water and nitrogen input into rivers and lakes with varying degrees of temporal and spatial resolution

Table 5: Input parameters of the dynamic model WASMOD for scenario modelling

compartment	parameter	source
soil properties	texture	actual measurement results
	physical properties	estimated on the basis of soil texture and organic matter content
	initial carbon and nitrogen content	estimated by taking into account, that the area of the beech forest has been used as arable land till 1900
	pH-values	estimated on the basis of progressing acidification
climate data	precipitation air humidity max. air temperature min. air temperature	measured values (German Metereol. Service Hamburg) period: 1960-1990 repeated 4 times 1900-1930 1930-1960 1960-1990 1990-2020
deposition data	nitrogen input as nitrate and ammonium	estimated with consideration of changing emission conditions

Results of the dynamic modelling

One objective of this case study was to compute the change of C- and N-dynamics, if the nitrogen inputs drastically reduced by accepting critical loads as an upper input limit.

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Therefore model simulation are computed for a period of 120 years in total (from 1900 to 2020). It was necessary to reconstruct present conditions related to the initial soil properties (starting point: 1900), the climate and nitrogen inputs by atmospheric deposition (see tab. 2).
Fig. 2 shows the present situation of N-fluxes and pools (as shown in Tab. 2). The critical load exceedance is high, there is a high N-turnover by N-uptake and litterfall. The nitrate leaching is high, the net immobilisation is relatively high as well. The *trend-scenario* shows the system behaviour under the present deposition loads. The nitrate leaching will increase and also the N-turnover will become higher whereas the net immobilisation will be relatively low. The reason for the latter is most likely the N-saturation of the system. Under the *critical loads* scenario (i.e. the scenario without an exceedance of the critical load during the next 30 years) the N-turnover will decrease as well as the leaching. The mineralisation is still larger than the immobilisation, which means there is no N-deficiency.
Fig. 3 shows the system behaviour more detailedly with respect to the pools of N and C in litter and organic soil horizon. Since 1900 the pools of N and C have been built up, especially since 1960. Under the *trend* scenario the pools still continue to increase slowly. Under the *critical loads* scenario there is a decrease of N-pools, but the level is still higher than 1960. The C-pool increases because of a slower mineralisation. This would mean that decreasing the N-input might increase the CO_2 binding capacity of this beech ecosystem which would be an important aspect within the framework of the global change discussion.

Discussion

The reason for focussing on the effects of nutrient N inputs was that very high N inputs are coupled with relatively low S inputs, a situation typical of large parts of rural NW Europe.
Especially in areas with high agricultural activity and small forests ammonia/ammonium deposition contributes strongly to eutrophication (and also acidification). The landuse history is an important aspect when one is trying to explain the system behaviour at the research site: the forest is the first generation after former agricultural use. The N-input originating from agriculture can be assumed to be relatively high before 1960. The recent change in agricultural practice to use slurry instead of manure supported the ammonia volatilisation and cause increasing deposition rates of reduced nitrogen compounds in the Bornhöved Lakes Area.
An interesting aspect within the simulation results is the tendency of increasing carbon amounts in the litter layer and the upper soil layer when nitrogen inputs are reduced. This effect can be explained by increasing C/N ratios in the litter caused by the reduction of nitrogen uptake, which is a consequence of less nitrogen which is available fo the trees. The C/N ratio is one of the functions which regulate the mineralization intensity. The question is, if this really is the dominant effect. For example, changes of root density and root distribution also could influence the contents of organic matter. Furthermore the development of the ground vegetation has to be considered. In order to decide about the correctness of the model results in this point, more data from field measurements describing the carbon and nitrogen accumulation over long periods are needed.

Conclusions

The Bornhöved beech forest is a N-saturated system. Continuation of present loads causes further destabilisation of the N-cycle (high leaching rates, vegetation changes etc.)
The steady-state situtation is not reached within 30 years of actual load = critical load. If critical values are to be met in 2020 then interim target loads for this forest ecosystem are much lower than critical load.
The decrease of N-input might increase the storage capacity for CO_2 in N-saturated ecosystems.

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The land-use history (the past agricultural use) and the spatial heterogenity (small scale variability in deposition or soil characteristics) is important for site-level analyses.
If the critical loads exceedance is high accurate depositon data are even more important for effect research and policy than accurate critical loads data.

Fig. 2: Dynamic modelling of N-Pools and fluxes with the model WASMOD

Fig. 3: N- and C-pools in litter and organic soil horizons: retrospective and future modelling under different assumptions of N deposition and soil acidity

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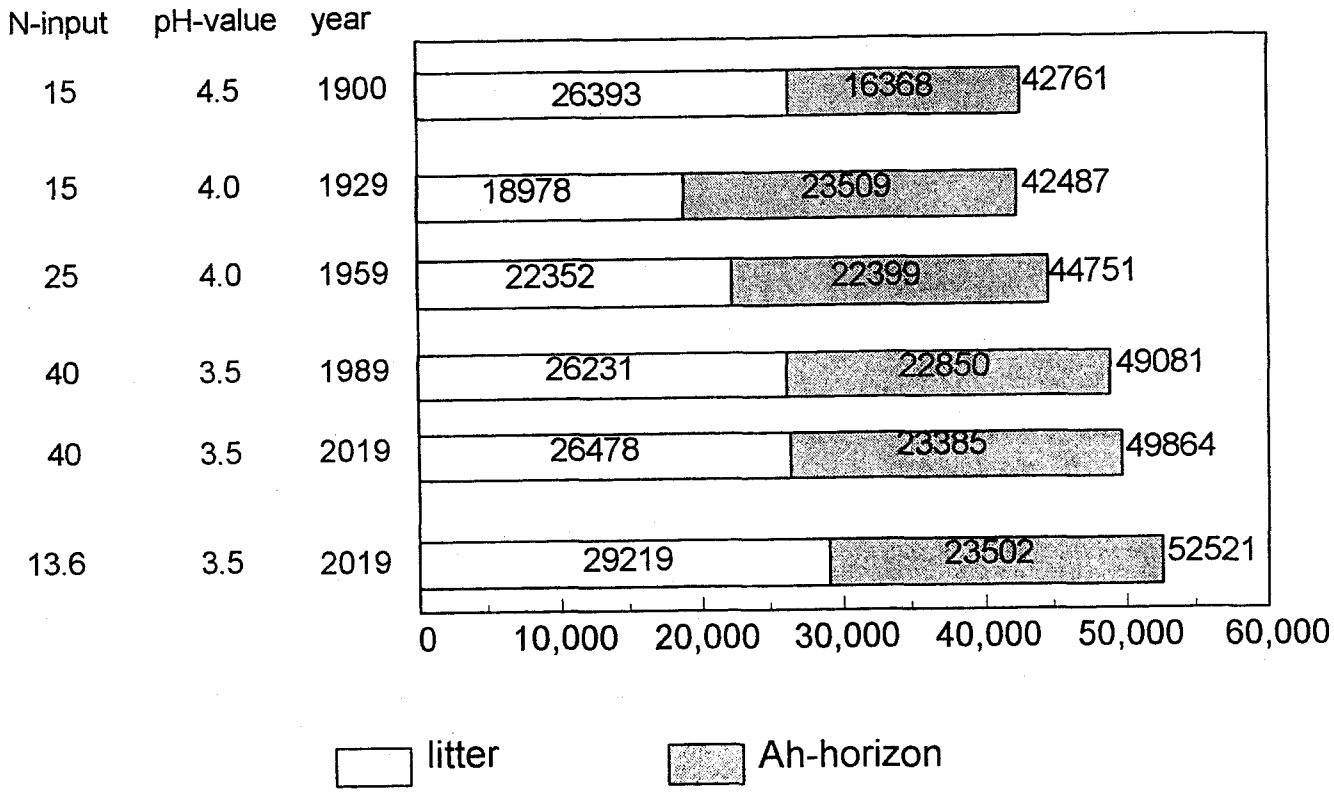
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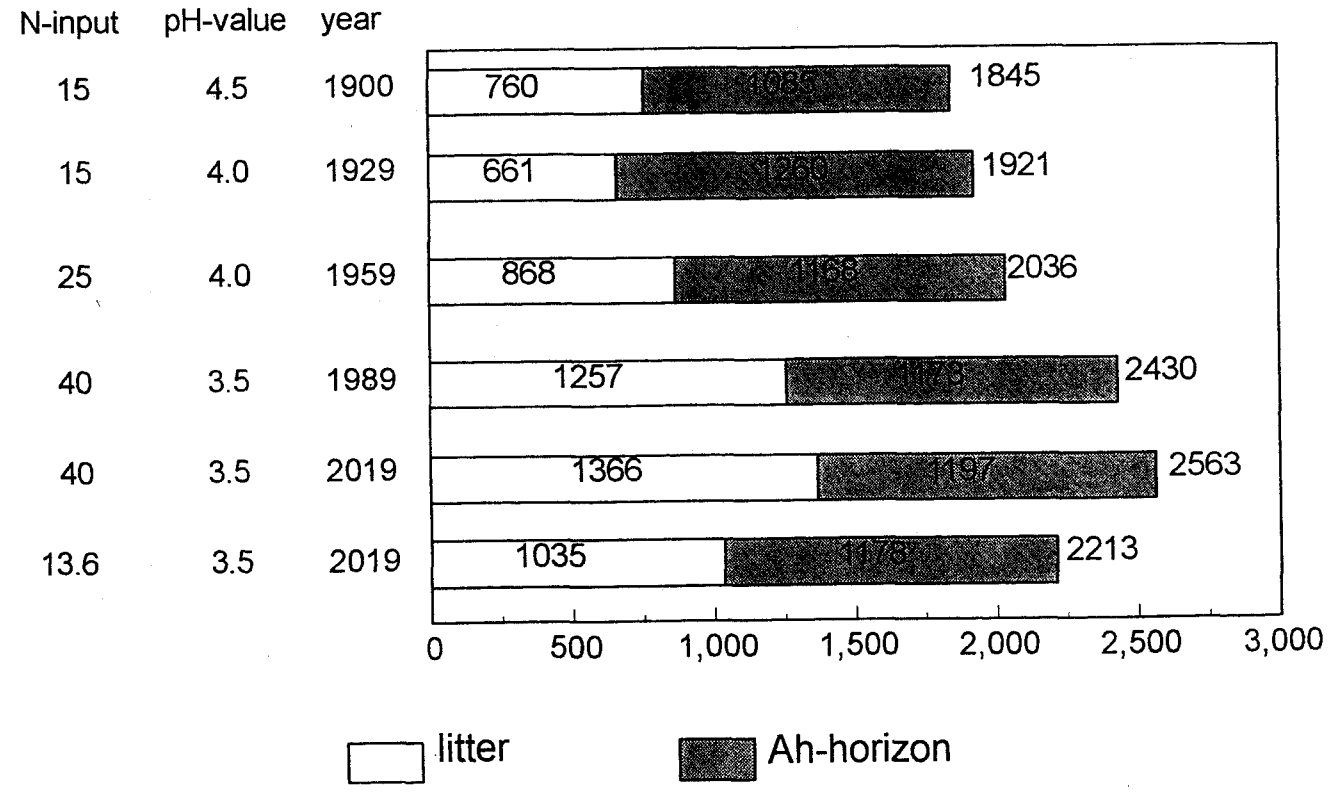
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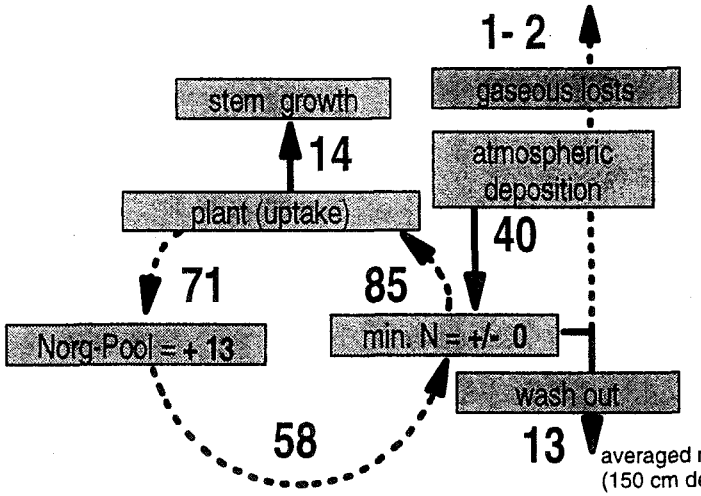


N-total



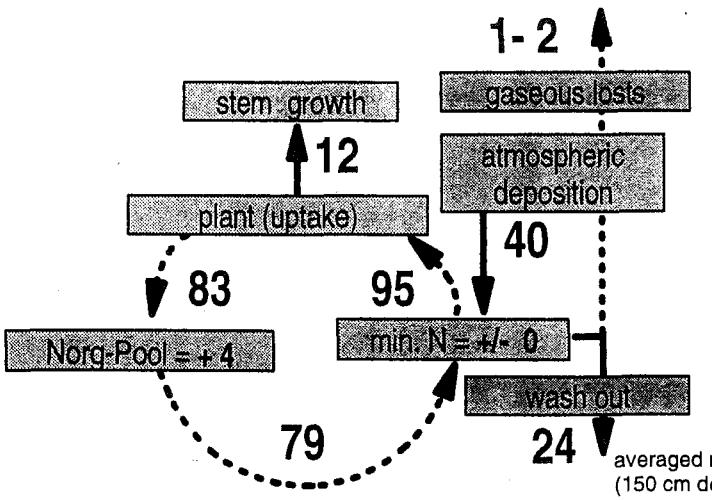
present situation

pH-value 3.5



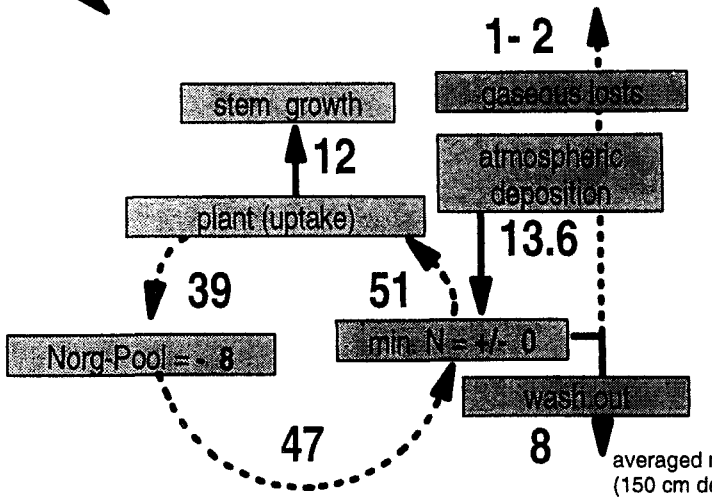
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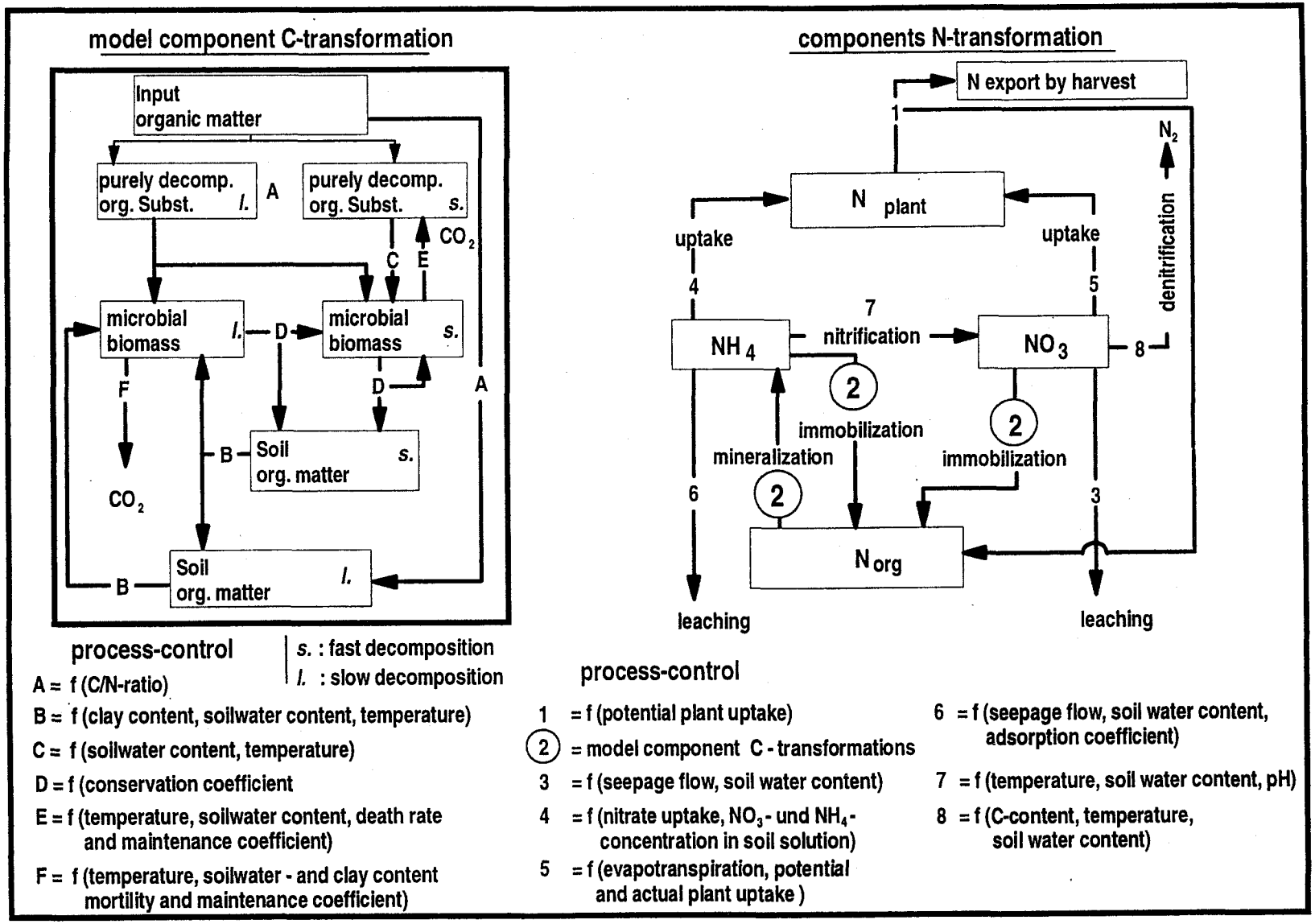
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SPATIAL CONSEQUENCES OF USING CRITICAL LOADS AS A DAMAGE FUNCTION

R. A. Skeffington and I. R. Rodgers

National Power Research & Engineering, Windmill Hill Business Park, Whitehill Way, Swindon, Wiltshire SN5 6PB, UK.

ABSTRACT

A critical load is essentially a threshold below which no adverse ecological effects occur. Critical loads for deposition of nitrogen, sulphur and acidity are currently exceeded over wide areas of Europe, and are likely to remain so for some time in the future. Indeed, in some high rainfall areas, acidity critical loads calculated according to current methodology are exceeded by background deposition. In order to minimise environmental damage in this situation, it is necessary to clarify and quantify the ecological meaning of critical load exceedance. If this can be expressed mathematically, it may be possible to use it in numerical optimisation approaches as part of an emission control policy. This mathematical expression of ecological damage we refer to as a *damage function.*

The choice of damage function is not obvious, as a number of them seem intuitively reasonable. In this paper we consider whether there is any ecological evidence which would enable any particular damage function to be selected. We also briefly explore the consequences of choosing different damage functions for the optimal distribution of SO_2 emissions in the UK. Dynamic modelling of ecological consequences is suggested as a way ahead.

INTRODUCTION

Critical loads are intended essentially to be the maximum deposition at which no adverse ecological effects occur (Nilsson and Grennfelt, 1988). The critical load concept leads logically on to a methodology for pollution control. First define the critical load: then adjust emissions in the most cost-effective manner until critical loads are not exceeded. This is an attractive approach for a number of reasons. It sets clear environmental objectives; it minimises the costs involved in attaining them; it apportions the costs towards those doing most damage; it is amenable to mathematical treatments such as optimisation; and it is reasonably comprehensible for decision makers. In practice, various problems have arisen in applying the technique. This paper considers just one of these: what should be the aim

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of emission control policy if critical loads cannot be attained? There might be two possible reasons for this: one is the need to set interim targets on the way to a complete attainment of critical loads, and the other is that, at least as they are defined at present, critical loads are in fact impossible to attain in certain areas.

THE PROBLEM OF UNATTAINABLE CRITICAL LOADS

Critical loads which can never be attained

As an example, of a critical load which could never be attained, it is instructive to compute the degree of critical load exceedance which would occur in the UK if human beings had not taken to burning fossil fuels. It is accepted that rain would be acidic even in the absence of anthropogenic combustion, owing to emissions of SO_2 and NO_x from natural sources such as marine algae, volcanoes and soils. Estimates of the amount of acidity not due to human activity have varied. Charlson & Rodhe (1982) estimated that the "natural" pH of rain would vary around the world between 4.5 and 5.5 with a median of 5.0. Skeffington (1995) compared the deposition of acidity resulting from rain at pH 5.0 with the (soil) acidity critical load, and found that natural deposition alone exceeds the critical load in 1,455 km^2 of the UK (Fig 1b). Exceedance occurs in most of the uplands, including Dartmoor, Wales, the Lake District, the North Pennines, Galloway, and the Scottish Highlands. Whelpdale *et al.* (1988) estimated the North Atlantic background concentration of non-marine *sulphate* to be 6-8 $\mu eq.L^{-1}$. Taking 7 $\mu eq.L^{-1}$ of acidity as a lower bound for background deposition leaves 86 km^2 of the UK still in exceedance, distributed over the Lake District, N. Wales and Scotland (Figure 1a). If as an upper estimate of background we take the acidity of rain approaching the Western seaboard of the UK from the Atlantic (RGAR, 1990), then 11,958 km^2 of the UK would be in exceedance, covering most of the UK uplands and some lowland western areas too (Fig 1c). Similar results would almost certainly be obtained for other high rainfall, sensitive areas such as Southern Norway.

These calculations will be conservative as they take no account of dry deposition, cloudwater deposition or seeder-feeder enhancement of SO_4^{2-} or NO_3^-, and deposition of other acidifying species such as NH_4^+; they use a critical load database where the value of the critical load



Figure 1: Areas where the Critical Load is Exceeded by Background Deposition.

Each black square indicates where the soil acidity critical load is exceeded by background deposition in rain, where this is assumed to be: (a) natural S emissions from the North Atlantic, but no N (7μeq.L^{-1}, pH 5.15); (b) estimated natural S and N emissions (10 μeq.L^{-1},pH 5.0); (c) current rain concentrations from North Atlantic airflows (18.6μeq.L^{-1}, pH 4.73).

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is increased if there is more than a certain proportion of agriculture in the area, and they use rainfall data on a 3km scale. The finding that natural deposition of S and N can exceed the critical load may appear surprising, but there is no intrinsic reason why all pieces of land should be able to resist natural acid deposition. Soils in these areas will steadily acidify over time, aided by their own intrinsic acidification processes, into the acid soils which are indeed characteristic of such areas. Once in a steady state with respect to atmospheric acidity, un-neutralised acidity will be exported to be neutralised further down the catchment. This illustrates also that critical load exceedance does not necessarily imply environmental disaster.

The above discussion illustrates that critical loads, if taken literally, are impossible to attain given present calculated values. The only prospect of attaining critical loads over Europe as a whole is if exceedance is allowable on a certain percentage of ecosystems or ground area - e.g. the pentile (5th percentile) approach described by Posch *et al.* (1995) - *and* deposition and critical loads or both are averaged over quite large areas, such as the 22,500 km^2 used in the EMEP model. But this approach tends to run into criticism because it can leave quite large areas "unprotected".

Interim targets

A further difficulty is that of "interim targets". It had long been recognized that "due to economic, technological and other constraints, such reductions [to meet critical loads] may not be achieved everywhere, or in one step" (UNECE, 1992). This gives rise to the necessity of defining the targets to be met at each step. In the negotiations which led up to the 1994 Sulphur Protocol, a target of "60% gap-closure" was used - i.e. the gap between 1990 deposition and critical loads was to be reduced by 60%. This retains the idea that more needs to be done to help the most sensitive areas, but is only one of a number of possible targets. Some others that have been suggested are:

- minimise the total exceedance of critical loads
- minimise the total exceedance of target loads
- minimise the total <u>percentage</u> exceedance
- minimise the <u>number</u> of squares exceeded

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- minimise the number of ecosystems exceeded
- minimise the deposition on exceeded squares
- use a higher percentile (such as 50%) as the target to be attained.
- multiply the critical loads by a factor to achieve complete attainment
- minimise exceedance up to a certain cost based on economic criteria

These targets, instead of reducing all deposition to the critical load, involve the minimization of some function of critical load exceedance. We call these *damage functions*. This is standard terminology in economics, and implies that critical load exceedance is being used as a surrogate for ecological damage.

THE EFFECTS OF CHOICE OF DAMAGE FUNCTION

The damage function which is selected should satisfy a number of criteria:

a) it ought to be derived from, and relatable to, current scientific understanding and evidence of the nature of impacts;
b) it should be clear what is to be protected from damage
c) it needs to be expressible mathematically in a form suitable for inclusion in numerical optimisation approaches.

The choice of damage function is by no means obvious, as a number of damage functions seem *a priori* reasonable, yet may have a significant impact on the optimal strategy chosen. We illustrate this below by examining the consequences of applying a number of damage functions to power station emissions within the UK using the MATADOR suite of models (Rodgers, 1994), as we do not have adequate access to the RAINS model used in UNECE.

This study focused on the atmospheric deposition of sulphur, and has employed published maps, on a 10km basis, of sulphur critical loads for soils (ITE, 1994). Consider the area of interest to be divided into N areas, labelled j. Denote by d_{ij} (gS m^{-2} yr^{-1}) the total deposition of sulphur to area j from source i, and by CL_j (gS m^{-2} yr^{-1}) the sulphur critical load. Let the total deposition from all sources to area j be denoted $d_j = \Sigma_i d_{ij}$. Then the following damage functions are defined:

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$$D_{1i} = \sum_i d_{ij}$$

$$D_{2i} = \sum_i d_{ij} H(d_j - CL_j)$$

$$D_{3i} = \sum_i (1 - \frac{CL_j}{d_j})\, d_{ij} H(d_j - CL_j)$$

$$D_{4i} = \sum_i \frac{d_{ij}}{CL_j}$$

$$D_{5i} = \sum_i \frac{d_{ij}}{CL_j} H(d_j - CL_j)$$

$$D_{6i} = \sum_i (1 - \frac{CL_j}{d_j}) \frac{d_{ij}}{CL_j} H(d_j - CL_j)$$

where

$$H(x) = 0 \quad for\ x<0$$
$$= 1 \quad for\ x>0$$

These damage functions have the following interpretations: D_{1i} is the total deposition to the area under study from source i; D_{2i} is the total deposition of sulphur from source i to exceeded squares. D_{3i} is the same as D_{2i} but with the contribution ascribed to a single source factored by the fractional contribution of that source to the total deposition on area j. D_{4i}, D_{5i} and D_{6i} are the same as D_{1i}, D_{2i} and D_{3i} respectively, but with the deposition divided by the critical load i.e. the effect caused by the deposition is weighted in favour of more sensitive areas which have low critical loads.

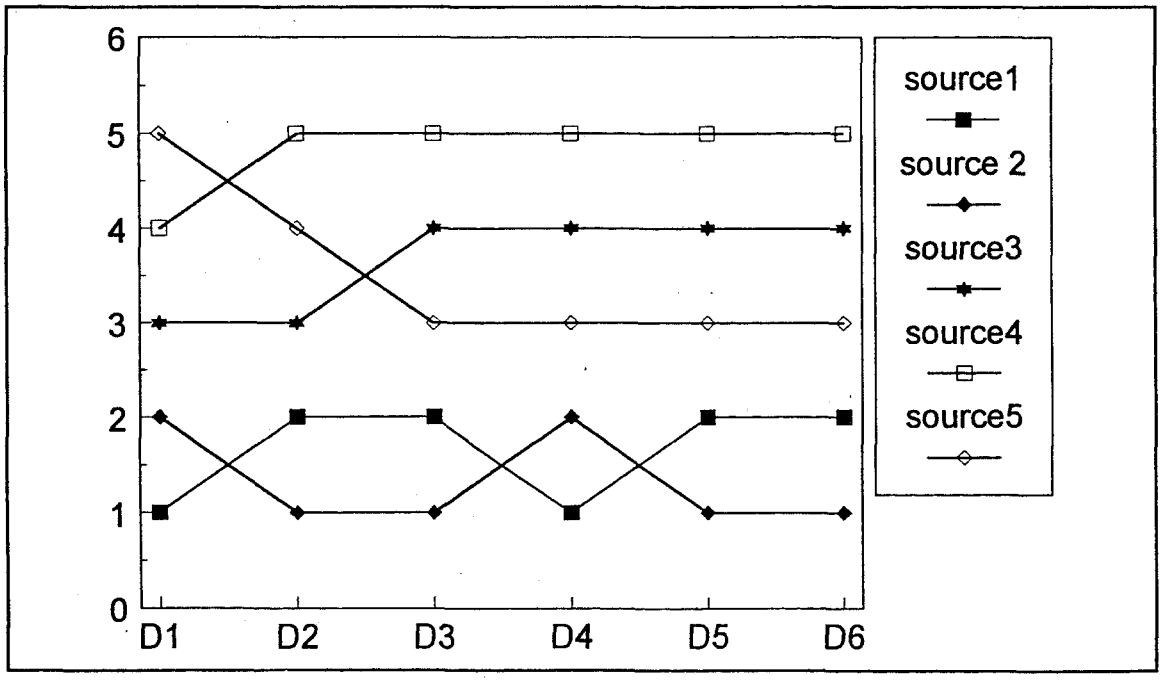


Fig.2. Rank order from 1 (least) to 5 (most) of calculated environmental damage for the damage functions (D1 to D6) defined above.

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Figure 2 above shows the effects 'rankings' found for 5 power stations in the UK, based on published 1992 emissions, where 1 has the most and 5 the least environmental effect. It is evident that the ranking order is rather dependent on the choice of damage function, though perhaps not very strongly in this case. A similar picture is seen if the actual damage score is compared instead of the ranking. This small illustration uses tools and data which are available to us, but a similar result (i.e. that the optimum pattern of emissions will differ depending on the damage function used) seems likely if Europe as a whole is considered using an integrated assessment model such as RAINS.

THE ECOLOGICAL MEANING OF CRITICAL LOADS EXCEEDANCE

Which damage function is the best one to use? To answer this question, it is necessary to quantify the *ecological* meaning of critical load exceedance. This has not yet been attempted, but the following discussion is intended as a contribution to this effort.

Methods of calculating critical loads

The methods most commonly used to calculate critical loads for the various receptors differ somewhat in the ecological meaning which can be attached to exceedance. Critical loads for **waters** are normally calculated by the Steady State Water Chemistry Method. This computes the deposition which will in the long term maintain a certain acid neutralising capacity (ANC) in the water. A value of ANC has to be selected as the damage criterion, and this is normally 0 or +20 μeq L^{-1} There is however a wide natural variation in the ANC of lakes and streams, and a continuous gradient of biological responses to the chemical conditions with which ANC is correlated. The method attempts to take this into account by incorporating a necessarily uncertain estimate of the pre-industrial base cation concentration. Critical loads for **forests** are normally calculated using some variant of the steady-state mass balance equation. The criterion for damage is usually that the base cation/Al ratio in the soil solution should not fall below some value (such as 1) thought to be characteristic for certain plant species. There is a wide natural variation in the base status of forest soils, and the method partly takes this into account by including an estimate of the base cation weathering rate in the calculations. The methods for forests and waters are thus formally similar in that there is a damage criterion (ANC or bc/Al ratio) and an estimate of intrinsic environmental

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resistance to acidification. The critical load is computed as the load which the water or forest soil can sustain without the ANC or base cation/Al ratio *eventually* falling below the criterion value. But because the response times of soils to deposition are very much longer than those of waters, soils are far more likely to have a critical chemical value which is currently better than the bc/Al criterion, whereas waters are more likely to be acidified to a state worse than the ANC criterion. One consequence of this is that if deposition is coming down but the critical load is still exceeded, then water quality is likely to be improving but soil is likely to be still acidifying. The ecological consequences of critical load exceedance will thus be different.

Critical loads for **soils**, as calculated by the original "level zero" method (Nilsson and Grennfelt, 1988), have different criteria. Here the critical load is the deposition which will *maintain the soil in its present state*. This method of setting critical loads has been largely replaced in European work by the steady-state mass balance method. The UK, however, has recently decided to revert to the level zero method, considering that the steady-state mass balance method needs further development (J. Hall, pers. comm.). Ecologically, the level zero method accepts the environment as it is, even if acidified, but protects it against further deterioration. In this it contrasts with the steady-state methods, which allow acidification provided the criterion species can continue to survive.

This raises another point, as different species will be protected by different criteria. A certain critical load may protect growth of Norway spruce or acid-intolerant brown trout, but not that of some more sensitive species. But raising the damage criterion to protect the most sensitive species will lead to a critical load being calculated to protect species in situations where they can never have existed naturally. An example is the adoption of an ANC = 20 μeq L^{-1} criterion for S. Norway lakes (Skeffington, 1995).

Dynamic modelling - the way ahead?

The above discussion should make it clear that it will not be easy to define the ecological meaning of critical load exceedance. It will depend on what method is used, what species are being considered, and what is the current state of the environment. This is not very helpful in choosing optimisation criteria. The one clear principle is that the higher the

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exceedance, the more damage is likely. Whether exceedance should be considered absolute or relative to the critical load is not obvious. There are also ethical issues, such as whether to spread damage widely but in small amount, or concentrate damage so some areas are affected but most left untouched. We suggest that the way to take discussion further is dynamic modelling of ecological consequences of different scenarios. Models that translate chemistry into the responses of real organisms are already available. These could be used to predict the ecological consequences of different optimisation criteria. This would at least make the effects of certain actions more explicit, and should lead to improved decision-making.

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DERIVATION OF DEPOSITION AND UPTAKE SCENARIOS

M. Johansson[1)], M. Alveteg[2)], C. Walse[2)] and P. Warfvinge[2)]

[1)]Finnish Environment Agency, Impacts Research, P.O.Box 140, FIN-00251 Helsinki
[2)]Lund University, Chemical Engineering II, P.O.Box 124, S-22100 Lund

ABSTRACT

Dynamic acidification models are used to assess the time development of chemical responses in ecosystems. The derivation method presented here is used to provide consistent deposition and forest growth uptake histories and future scenarios for three dynamic acidification models (MAGIC-WAND, SAFE and SMART), which are applied to selected integrated monitoring (IM) sites in Europe.

The deposition of each compound is divided into different components using measured bulk and throughfall depositions. Components originating from anthropogenic sources depend on emission histories. The dry deposition components are affected by forest filtering in correlation to canopy size. The future sulphur and nitrogen depositions base on EMEP transfer model and reported country-level emission data.

The forest growth uptake is based on the average biomass density and element content together with annual volume increment of different tree compartments. The representative potential growth curve parameters are calibrated to current measurements.

The method has been applied to selected IM sites in Europe for dynamic model simulations. A more complicated approach would call for data not available for all sites in the IM program. The current scheme illustrates one promising way of compiling scenarios at a level sufficient to dynamic acidification models.

1 INTRODUCTION

Both steady-state and dynamic models have been developed to predict the acidification of soils, lakes, streams and groundwater (e.g. Cosby et al. 1985, de Vries et al. 1989, Sverdrup et al. 1992). The steady-state methodologies are applied as a policy tool through critical load maps applied in europeanwide emission reduction strategies. For the timing of necessary emission controls, the dynamic models can be used to predict chemical responses of ecosystems to changing deposition and land use.

Three acidification models, MAGIC-WAND, SAFE and SMART, are being applied to selected IM sites to describe the chemical processes of the soils and lakes in the area concerned. Dynamic modelling is currently one of the key activities in the UN-ECE effects-oriented work. The models require deposition and uptake by forest growth as external input. The derivation method used for generation of this input originates from the work of the SAFE modelling group (Alveteg et al. 1995). Presently the derivation method and the three acidification models have been applied to four IM sites (Figure 1). The derivation of historical development and future scenarios for deposition and uptake is here explained with the case study of the Birkenes site in Norway (Kvindesland et al. 1994).

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2 DEPOSITION

The deposition of each compound is divided into different components using bulk and throughfall deposition measurements: originating from sea or anthropogenic sources, and these both in dry and wet fractions. Historical and future development and forest filtering effect are then allocated to proper components.

The total dry component for sulphur, chloride and sodium is assumed to be the difference between throughfall and bulk deposition, as they are mobile ions in the tree canopy. For base cations other than sodium, the minimum of sodium or chloride filtering is used. For nitrogen compounds the dry total, or anthropogenic, component is bulk deposition times minimum filtering factor of sulphur, sodium or chloride.

The wet marine components are estimated with seasalt correction factors from the sodium or chloride wet deposition. The dry marine component for sulphur is calculated using the same ratio to its wet marine component as between its total dry and total wet component. For other compounds, except for nitrogen which is completely of non-marine origin, the dry marine component is estimated with seasalt correction factors using the sodium or chloride dry deposition. The obtained wet marine value for any compound cannot be larger than its measured total wet deposition.

The wet anthropogenic components are calculated by subtracting the wet marine component from total wet deposition. The same applies to the dry anthropogenic component. The possibly remaining unallocated deposition is finally assigned to canopy exchange, serving as an indication of internal circulation in the forest. The wet anthropogenic component for both nitrogen compounds is the measured bulk deposition.

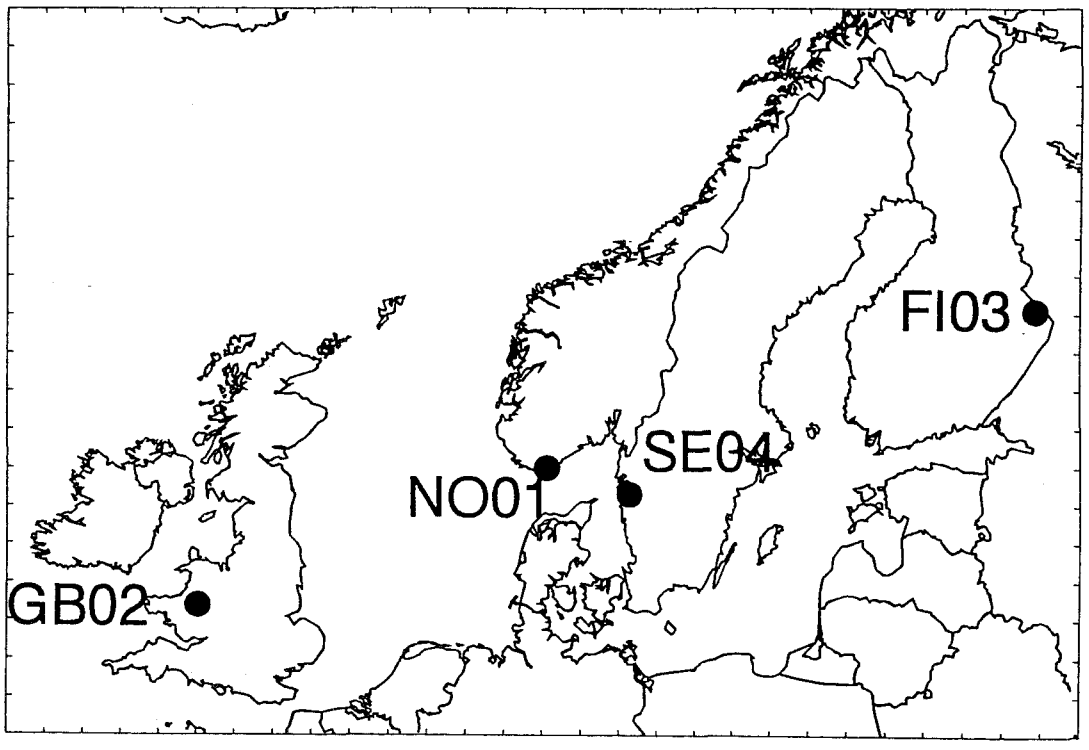


Figure 1 The location of IM sites where the derivation method has been applied (GB02 Afon Hafren, NO01 Birkenes, SE04 Gårdsjön and FI03 Hietajärvi).

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The wet marine component is assumed to remain constant in time. Other wet and dry anthropogenic components are dependent on deposition histories, calibrated to the present measurements of one year. Sulphur deposition history for 1880-1991 is extracted from data by Mylona (1993) for each EMEP grid cell separately. The earlier scaling is extended back to 1800 using an average estimate for Europe by Sverdrup et al. (1995). Average nitrogen histories for the whole of Europe are estimated on basis of work by Sverdrup et al. (1995) and Asman and Drukker (1988). The non-marine dry deposition component of base cations is assumed to be partly connected to anthropogenic activities reflected by measurement data during a period of large emission reductions (Hedin et al. 1994), and is here assumed to follow the historical sulphur deposition curve.

All dry deposition components are affected by forest filtering, which is assumed to depend linearly on the tree needle volume represented by the needle mass in internal calculation. The present filtering factor is calibrated to current canopy volume and dry deposition estimates.

The deposition to open land is assumed to equal the sum of wet components plus 20 % of the dry deposition. However, the open land deposition does not have a big effect in the mainly forested IM sites.

In the scenarios, base cation and chloride depositions are kept at present level. For acidifying deposition there are three future scenarios for the model application (Table 1). The deposition is calculated with the EMEP transfer matrices and reported official UN-ECE emissions for the target years 2000, 2005 and 2010. The alternatives for sulphur deposition are current reduction plans (according to second sulphur protocol, SP2) and maximum feasible reductions (Cofala and Schöpp 1995). For nitrogen, the NO_x and NH_y depositions are frozen at the present (1992) level. For NO_x, a reduction scenario of flat 30 % reduction in deposition is utilized to demonstrate the possible effects of such a measure.

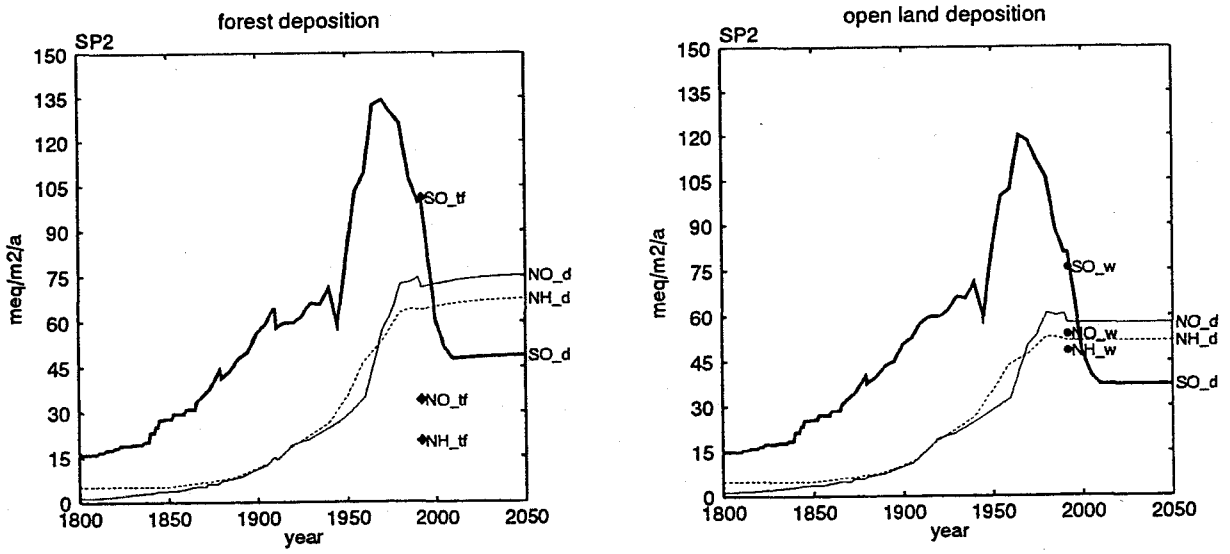


Figure 2 Simulated forest and open land depositions of sulphur and nitrogen compounds for scenario A for the IM site in Birkenes, Norway. Throughfall and bulk deposition measurements from the year 1992 are also indicated.

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Table 1 *Three scenarios employed in the acidification model runs, roughly representing the possible range of future acidifying deposition.*

scenario label	SO_2	NO_x	NH_y
A) best prediction	SP2	present level	present level
B) lower limit	MFR	-30%	present level
C) upper limit	present level	present level	present level

3 UPTAKE

The uptake by forest growth is based on the biomass density and element content together with annual increment of each tree compartment: stem over bark, branches and needles. The element concentrations used are the same as reported for Birkenes. The growth estimate is most proper for a temporally homogenous forest with one major tree species, or a well-known tree species distribution each of known age, after clearcut or forest fire.

Potential annual growth and standing volume are calculated from a logistic forest growth curve, representing the whole forest and initially based on conditions from southern Germany. The branch growth is assumed to be one fifth of stem over bark growth according to the applied Marklund (1988) biomass functions and Mälkönen (1975). The needle growth is described with a growth curve similar to that of stem over bark. All growth curve parameters are calibrated to current volume and growth (Kvindesland et al. 1994). In Birkenes, the median standing volume 177 m^{-3} ha^{-1} (stem over bark) of spruce with average age of 80 years including annual increment values for all compartments were reported.

The forest growth is assumed to be nitrogen limited. The growth of all above-ground compartments must be reduced if the nitrogen via atmosphere and mineralization is not enough to satisfy the estimated potential growth.

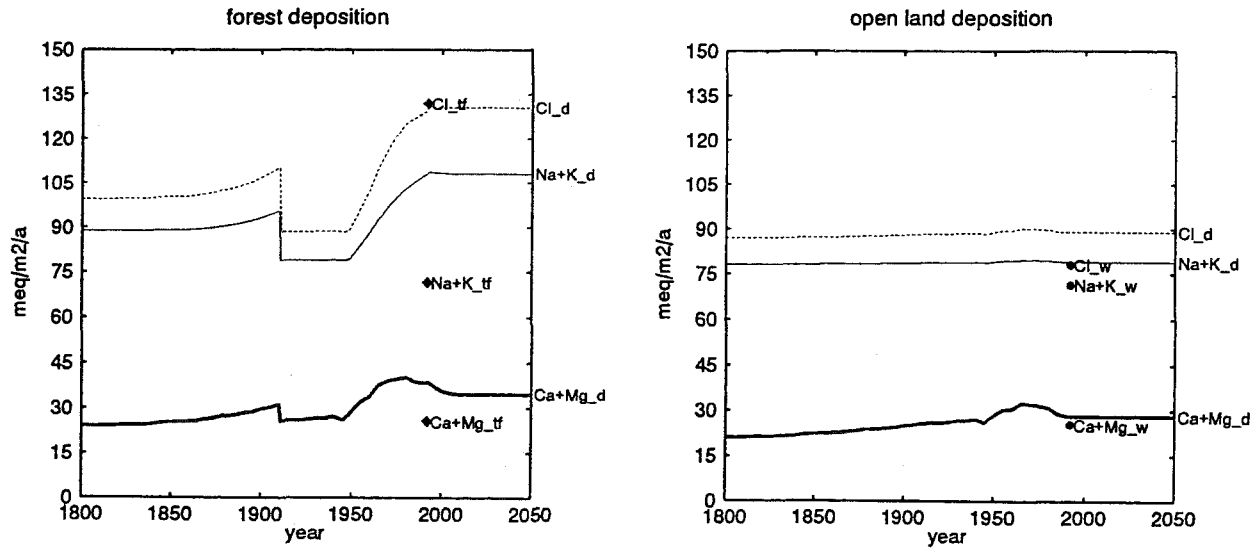


Figure 3 *Forest and open land depositions of base cations and chloride for IM site NO01 in Birkenes, Norway. Throughfall and bulk deposition measurements from the year 1992 are also indicated.*

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4 RESULTS

The derived scenarios for Birkenes IM site are presented in Figures 2 to 4. Figure 2 shows the forest and open land depositions of sulphur and nitrogen compounds for scenario A, a feasible estimate for probable future deposition development. Figure 3 gives the forest and open land depositions of base cations and chloride for scenario A. Measured deposition values are indicated in Figures 2 and 3. The forest filtering affects historical base cation deposition much stronger than sulphur or nitrogen compounds, due to the higher fraction of dry deposition components for base cations. Therefore the clearcut in 1910 in Birkenes results in a less drastic reduction in sulphur and nitrogen depositions as compared to reductions in base cation and chloride depositions.

Figure 4 shows the net uptake by forest, which is required as model input and obtained by extracting mineralized material flux from total forest uptake values. Figure 5 presents preliminary results of SAFE model simulations predicting the stream pH for all three deposition scenarios (Table 1).

5 DISCUSSION AND CONCLUSIONS

The ion balance in modelled deposition in Birkenes was calculated in the period under study (1800 - 2050). The calculated pH is slightly above four and even the historical pH has been rather low, dropping below five already in 1850, being thus slightly lower than currently measured pH. This may indicate an imbalance between estimated historical deposition compound values, since the calibrated models tended to overestimate acidification effects in comparison to measurements. The magnitude of historical deposition is sensitive to the measured deposition values, to which it is calibrated, calling for use of measurement data from several years. Further uncertainty is brought in by the relationship of the forest needle volume to the filtering effect, which may not be linear in all phases of the forest cycle.

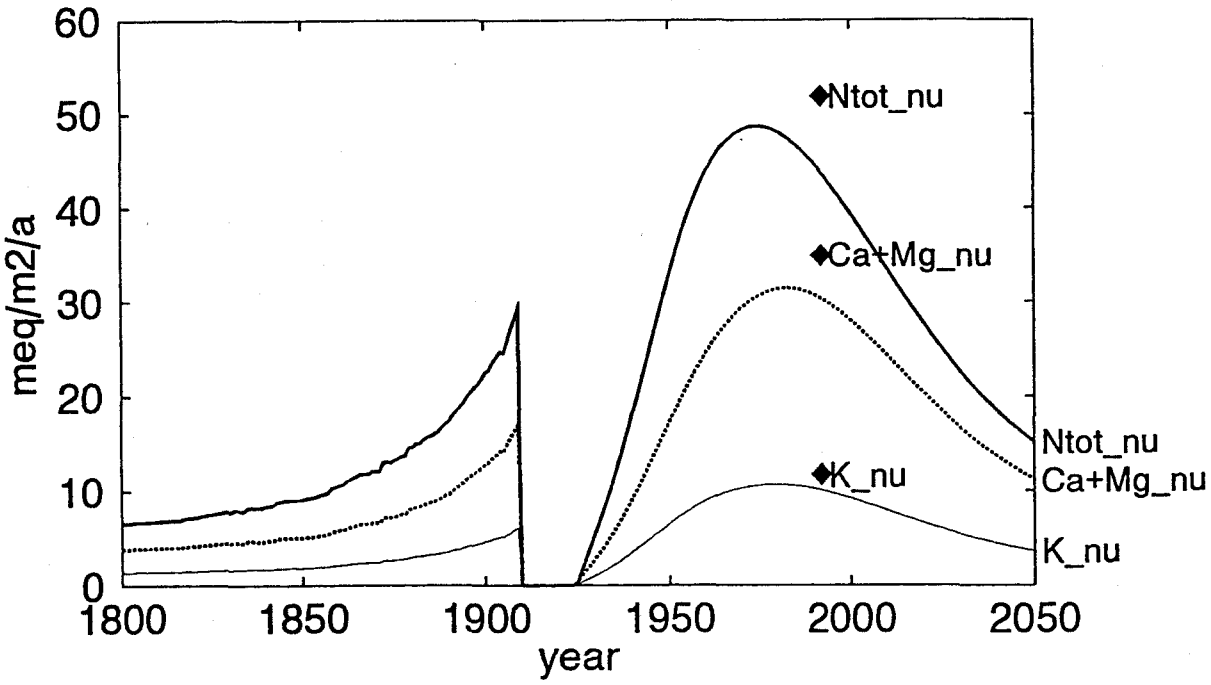


Figure 4 Net uptake by forest at IM site NO01 in Birkenes, Norway. Estimated uptake values for the year 1992 are also indicated.

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The nitrogen deposition histories are estimated averages for the whole of Europe. The difference between actual and derived history is likely to be largest at sites where the short-range transport of pollutants is significant, as for ammonia.

Growth estimates could be refined by using locally estimated growth curves for the region and tree species concerned. A more complete description would, however, call for data not available for all sites in the IM program. The growth curve calibration only to present volume and growth may lead to faulty predictions in the early and late phases of the rotation period. To depict more in detail the growth of all tree compartments various biomass functions (Marklund 1988) could be applied in the derivation method itself. However, the data available from IM sites restricts this approach.

In Birkenes, potential growth curve could be fitted to the present volume and growth only by allowing a rather high final forest standing volume. The future growth and uptake may thus be overestimation.

This derivation method was the first attempt to supply site-specific deposition and uptake scenarios for acidification model application at IM sites. A more straightforward approach would have been to estimate a modelled open land deposition with a constant filtering factor and historical scaling and a simple piecewise linear growth curve to estimate uptake. Instead, steps were taken into an already more complex description of hypothesized deposition breakdown and of a nitrogen-limited tree growth with a dynamic filtering capacity of dry deposition, based on earlier work on SAFE model applications.

The advantage of the current approach is the rather modest requirement of input data. The results, in turn, retain considerable uncertainty due to e.g. too few calibration values, although the uncertainties can be small when compared to potential errors involved in the whole modelling exercise. The method is refined with the experience on forecoming applications to IM sites, even at the cost of increasing input data requirements. Presently the scheme as such illustrates one promising way of compiling scenarios at a level sufficient to dynamic acidification models.

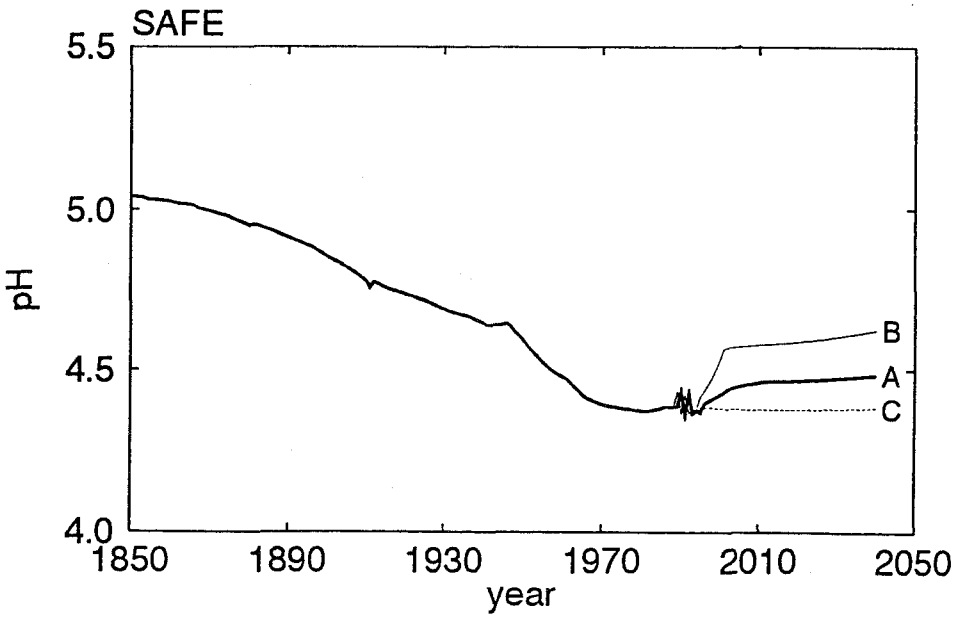


Figure 5 Preliminary simulation results of the SAFE model on stream pH at IM site NO01 in Birkenes, Norway, for all three deposition scenarios (Table 1).

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Framework for the combination of dynamic vegetation and soil geochemical models to assess the effects of air pollution on heathlands

Jesper Bak & Knud Tybirk

National Environmental Research Institute, Department of Terrestrial Ecology
Vejlsøvej 25, DK-8600 Silkeborg, Denmark

Abstract

Assessment of the influence of nitrogen deposition on heathlands involves quantification of the complex interaction between atmospheric deposition, soil geochemical changes, and vegetation competition under the influence of management, climate and pests. Existing soil geochemical models tend to lack a good description of plant growth and competition, while vegetation models tend to have a quite rudimentary description of the soil geochemistry. Instead of developing a new "complete" model, it has been decided to integrate existing models to calculate critical loads for Danish heathlands. The Dutch CALLUNA model has been chosen to model the competition between heather and grasses under different nitrogen deposition scenarios with the influence of regular management and stochastically occurring heather beetle attacks. The SAFE model has similarly been chosen to model the soil geochemistry as a function of atmospheric deposition, mineral weathering, plant growth, ion exchange and nitrification. The models are run in iteration under a common shell-program written in DCL with SAS programs for converting input and output files.

Initial model run indicates that the balance between heather and grasses is very delicate and the consequences of the exceeding the critical load even in a limited period of time might be drastic. More work is needed to adapt and combine the models, but the presented framework opens for better future critical load calculations and mapping for (semi-) natural ecosystems.

Introduction

During the last decade critical loads have been established as a concept to assess the influence of human activities to ecosystems. In the early eighties the focus was mainly on transboundary transport of sulfur emissions from industry and power plants, but later it has been accepted that also nitrogen and ozone may have serious effects on terrestrial ecosystems. As a consequence, international agreements on abatement of air pollution have been negotiated under the UN/ECE and laid down in the Geneva Convention on Long Range, Transboundary Air Pollution. The latest protocols under the convention have been effect-oriented based on the scientific work commenced in a series of workshops held at Skokloster (1988), Bilthoven (1990, 1991, and 1992), Lökeberg (1992), Katowice (1992), Madrid (1992), Grange-over-Sands (1994) and Wienna (1995).

In general critical loads can be set on three levels: Level 2 evaluation involves dynamic modeling; Level 1 evaluation involves steady state calculation of a chemical criterion; and Level 0 evaluation involves expert judgment based on field experiments, long term monitoring or model calculations. A critical load for heathlands was first set at the UN-ECE workshop in Lökeberg based on a level 0 evaluation (Bobbink et al. 1992, Grennfelt &

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Thörnelöf 1992). The recommended values have since been adopted by the UN-ECE Task Force on Mapping and included in the "Guidelines for Mapping" (UBA, 1993).

Lowland heathlands are primarily low-nitrogen ecosystems made by and maintained by man in earlier times throughout north western Europe. There exists, however, coastal heathlands, which are unmanaged and a result of natural succession. Heathland ecosystems are now threatened in many countries by a number of factors, one of which is believed to be deposition of nitrogen resulting in conversion of heathlands into grasslands (Heil & Bobbink 1993, Bobbink & Roeloefs 1995). In the Netherlands, intensive work during the 80ies and early 90ies have enabled the researchers to establish rather reliable critical loads for nitrogen on Dutch heathlands by combining ecosystem research and dynamic vegetation modeling (Aerts & Heil 1993, Heil & Bobbink 1993). This work has been a major step forward in the understanding of the relationship between air pollution and vegetation dynamic and the Dutch critical loads for heathlands is used for European heathlands in general (Bobbink et al. 1995). There are, however, many types of heathlands present on a range of different soil and climatic conditions throughout the distribution area, and there is no doubt that detailed studies will reveal different critical loads for nitrogen for different heathland types and in different regions.

The model used in the Dutch heathland research, the CALLUNA/ CALGIS/ HEATHSOL models, are basically vegetation competition models and have not been developed for detailed calculations of soil chemistry and hydrology (Heil & Bobbink 1993, van Deursen & Heil 1993, Bakema et al. 1994). To enhance the handling of the soil geochemistry, the model has been combined with the Swedish SAFE model, which is a dynamic version of the PROFILE model. Both models are developed by Sverdrup & Warfvinge in Lund (Warfwinge & Sverdrup 1992, Sverdrup & de Vries 1994). The PROFILE model have been widely used for mapping of critical loads in Europe (Downing et al. 1993, Bak & Løkke 1995). SAFE and PROFILE have, however, mainly been used on forested ecosystems and need validation on heathland ecosystems.

The intention of this paper is to present a preliminary framework for the combination of existing models for soil geochemistry, hydrology, heathland vegetation and management and to present an application on an intensively studied Danish heathland research site. The presented work is at a preliminary stage and the resultant calculations need to be refined and confirmed, but the presented framework is believed to provide a basis for improving critical load calculations on heathland and, in the longer run, other semi-natural ecosystems.

Models and methods

The basic idea of this work is to combine the SAFE and CALLUNA models iteratively so output from one model is used to generate input for the other and vice versa. The modeling is performed with data from the Danish heathland research site at Hjælm Hede. Later this platform should be combined with a Danish hydrological model used for the Hjælm site (Ladekarl in press) and the management method being used in CALLUNA should be modified and adapted to the local conditions, perhaps involving the Scottish HILL GRAZING MANAGEMENT MODEL (MLURI 1994, Grant & Armstrong 1993).

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Technically the integrated model consists of the two base models, ported to a WAX/VMS computer, a common shell program written in DCL, which calls the models iteratively. Two SAS programs, also called by the shell program, read output files from one model and generates input files for the other, adjusting the appropriate parameters. Later on the model complex will be ported to a UNIX platform and the shell and interface programs rewritten in C to achieve better performance.

The CALLUNA model
The core of the CALLUNA model is the vegetation competition part. The competition is driven by sunlight, water, and nitrogen availability. The intention of the model is to simulate how the competition between grasses and heather evolve under different nitrogen deposition scenarios including regular sodcutting and stochastic attacks of heather beetles (*Lochmaea suturalis*). The involved heathland species are the grasses *Deschampsia flexuosa/ Molinia coerulea* (depending of humidity of the site) and the heathers *Calluna vulgaris / Erica tetralix*, (depending on the site).

The input parameters to CALLUNA are the following (for details see Heil & Bobbink 1993, Bakema et al. 1994):

Deposition: NH_x, wet, dry, NO_y, wet, dry

and the output files are the following:

Fluxes: growth, C, N; mortality C, N
Masses: mass C, N; litter C, N
Mineral: mineral C, N; inflow C, N; outflow C, N
Soil: leaching, nitrification and denitrification of NH_4 and NO_3, soil content of NH_4 and NO_3.

The setup of CALLUNA consist of the following major parameters:

- C/N ratios, leaf, culm, branch, flower, root, plant and litter
- Canopy exchange, NH_4 and NO_3
- mortality rates of leaf, culm, branch, flower, root,
- redistribution
- competition, height, root length, Specific Leaf Area, patchiness
- growth of leaf, culm, branch, flower, root,
- mineralization of leaf, culm, branch, flower, root,
- sod cutting as management practice
- heather beetle attacks: chance and susceptibility

Details of how the model works and scenarios obtained by running the model under Dutch conditions can be found in Heil & Bobbink (1993).

For illustration we present two typical runnings of the CALLUNA using the setup files kindly provided by the authors. Under low nitrogen deposition scenario (under Dutch conditions this means 15 kg N ha^{-1} yr^{-1}) the balance between heather and grass favors the heather under conditions with sod cutting every 30 years and stochastic beetle attacks. When the system is kept low in nitrogen, heather is favored in the competition with more nitrophilous grasses. (Figure 3a). Under high deposition scenario (40 kg N ha^{-1} yr^{-1}) the balance tips in favor of the

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The first step is the combination of CALLUNA and SAFE. CALLUNA has a rather simplistic soil chemical module applying a one-layer or "stirred tank" approach. Deposition and mineralization are nitrogen inputs and vegetation uptake and leaching are nitrogen output in the model (Figure 1a). SAFE is a multi-layer model with a more complex calculation of soil geochemistry for each layer including, for nitrogen, immobilization, nitrification and denitrification as well as sources and sinks of base cations including mineral weathering. (Figure 1b).

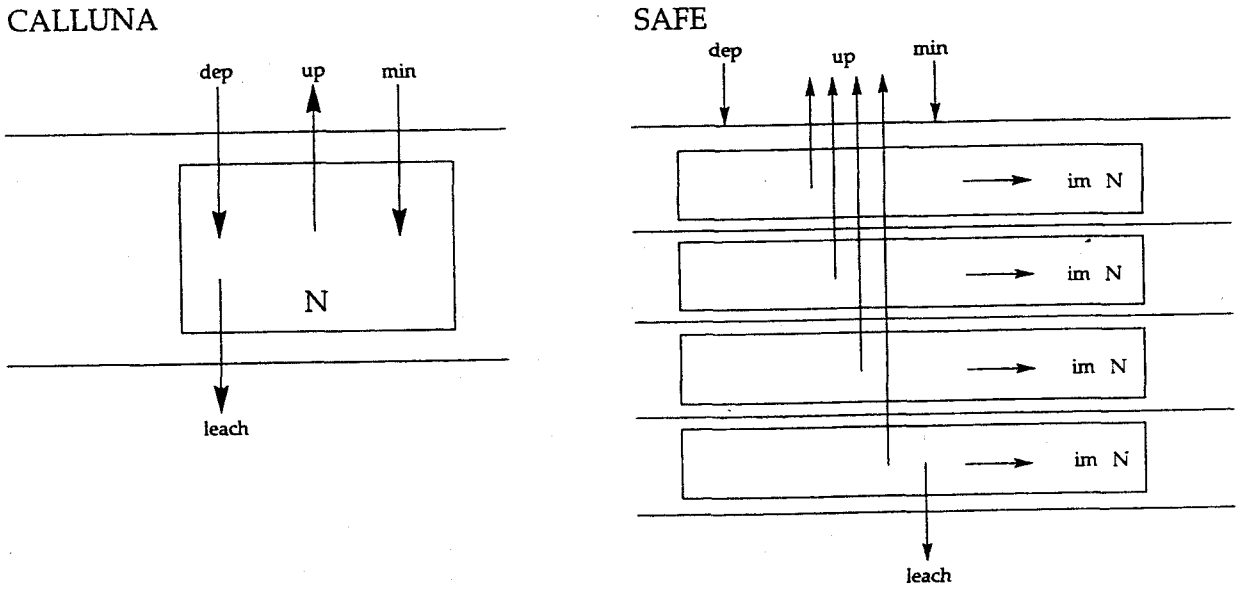


Figure 1. Illustration of the soil chemical calculations in the two used models. a. The "stirred tank" model for soil in CALLUNA. b. The four layered calculations in SAFE.

In the combination of the models, a basic scenario of vegetation development is calculated with the CALLUNA model based on a deposition scenario. The resultant vegetation uptake is used as input to the SAFE model and the resultant development of soil geochemistry calculated. This calculation is then used to recalculate the actual availability of nutrients and the result is input to CALLUNA. As the CALLUNA model uses a "stirred tank" approach, the availability of nutrients in the model can be adjusted by changing one source or sink. In practice it is easiest to change the nutrient availability by adjusting the deposition scenario as deposition is input to the model in a separate input file. The new deposition scenario is used to recalculate the vegetation development, and the process is iterated. (Figure 2).

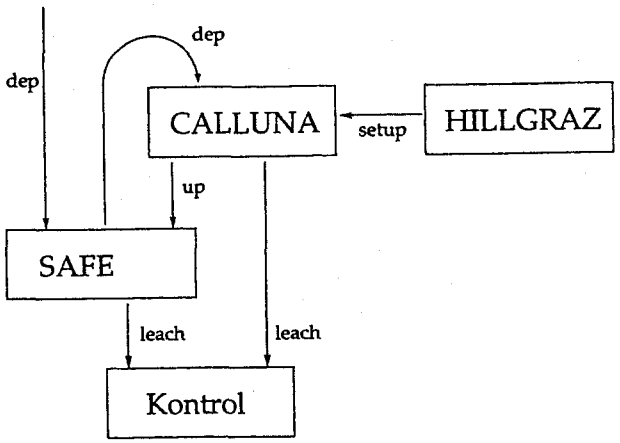
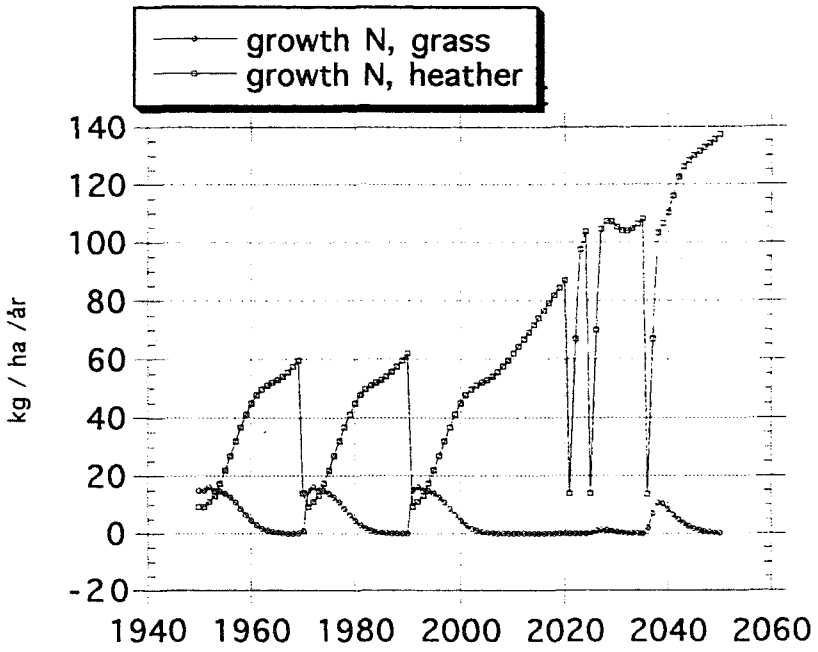


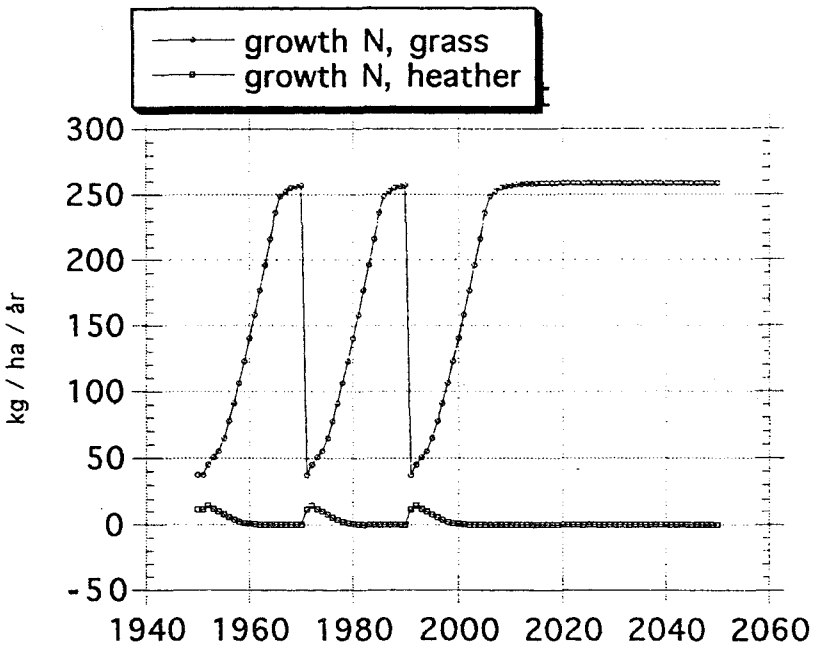
Figure 2. Interface between the SAFE and CALLUNA models. The intention is to integrate a grazing model and a hydrological model at a later stage.

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grasses and the only fluctuations seen are caused by sod cutting, after which heather is rapidly outcompeted by the grasses and the heather beetles do not have sufficient food for attacking (Figure 3b).



3a



3b

Figure 3a. CALLUNA running at the setup provided by the authors under low nitrogen deposition level (15 kg N ha^{-1} yr^{-1}). Heather stays as the dominant element and the regular sod cutting and stochastic heather beetle attacks can be seen as drop down of the heather growth curve. 3b. As 3a but with high nitrogen deposition level (40 kg N ha^{-1} yr^{-1}). Grass has taken over in dominance and the former heathland has been converted into grassland.

The SAFE model

The function of the SAFE model can be illustrated in a simplified way by Figure 4. The relationship of soil water to deposition, percolation and plant uptake are central as in CALLUNA, but the SAFE model also includes the complex interactions of soil water with ion exchange, mineral weathering, nitrification and denitrification, which in some areas are very important for a realistic picture of nutrient availability.

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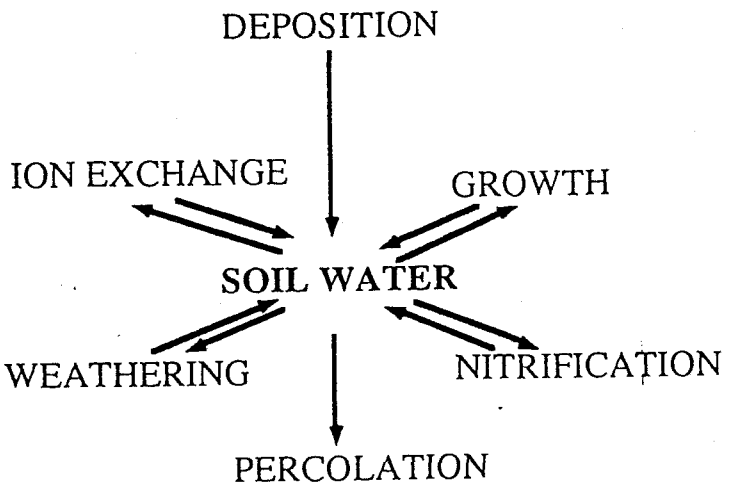


Figure 4. Diagram showing the principles of soil water interactions in the SAFE model.

Table 1. Summary of the units used in input files of the SAFE model.

Mac file name	Variable	Unit
site/site.init	Temperature	Kelvin or Celsius
	stream DOC	mg/l
	stream PCO2	in multiples of atmospheric pressure
	layer thickness	m
	water content	
	soil density	kg/m^3
	surface area	m^2/m3soil
	PCO2	in multiples of atmospheric pressure
	inflow	in percent of precipitation
	outflow	in percent of precipitation
	BC uptake	in percent of total uptake in profile
	N uptake	in percent of total uptake in profile
	DOC	mg/l
	Gibbsite constant	$\log_10$ of the constant in moll
	Cation Exchange Capacity	
	pH	
	Base Cation	Eq/l
	Base saturation	in fractions
	mineralogy	in percent of toal amount of minerals
site.dep	all	mEq/m^2, year
site.circ	all	mEq/m^2, year
site.bio	all	mEq/m^2, year
site.flow	prec	m/year
site.cal	base saturation	fraction

The input files to SAFE are the following (see also table 1):

Dep: SO_4, Cl, NO_3, NH_4, Ca, Mg, K, Na
Cir: canopy exchange, litterfall, mineralization, nitrogen, base cations
Bio: uptake of nitrogen and base cations
Flow: precipitation

The output files are:

de(n): input from the $(n-1)^{th}$ layer:
Acid Neutralizing Capacity (ANC), base cations (BC), N, S, Cl and Na
re (n): results from the $(n-1)^{th}$ layer:
pH, BC_{sat}, ANC, BC, BC_{wea}, BC/Al-ratio, N, S, Cl, Na, and Al

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Known levels and scenarios for deposition of S, N and base cations has been used for model calculations of SAFE applied to a sandy soil similar to Danish heath soil (Figure 5 and 6). The calculations show how the N-deposition is reflected in the soil chemistry. Acid neutralizing capacity (ANC, Figure 6) goes down parallel to the high acidifying deposition in Figure 5, and nitrogen availability is following the deposition level, which is relatively high at present (approx. 15 kg N ha^{-1} yr^{-1} is considered high for Denmark).

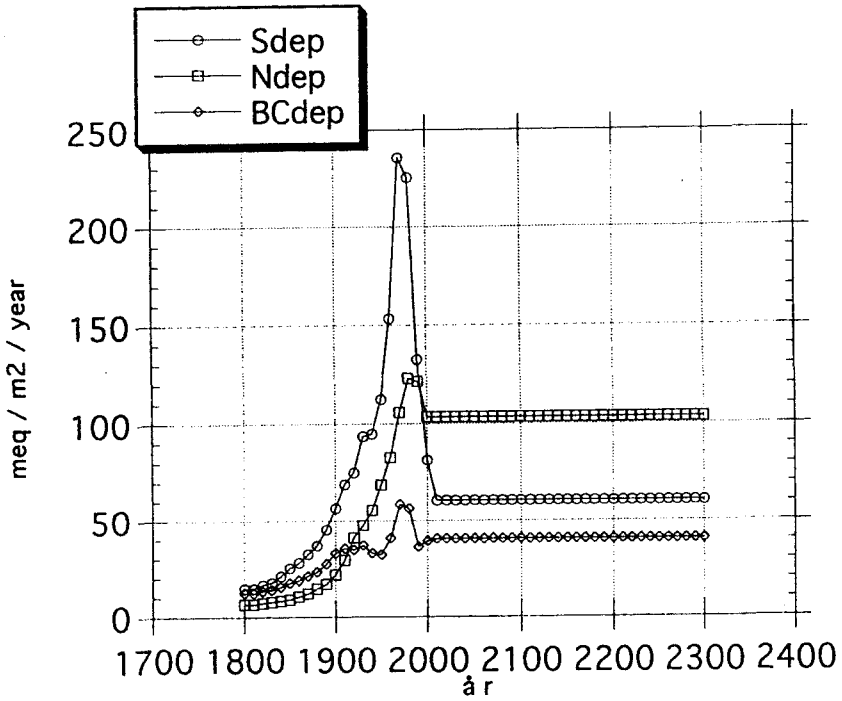


Figure 5. Deposition input to SAFE. The levels used for the future are assumed levels after reduction measures have been carried through for S and N.

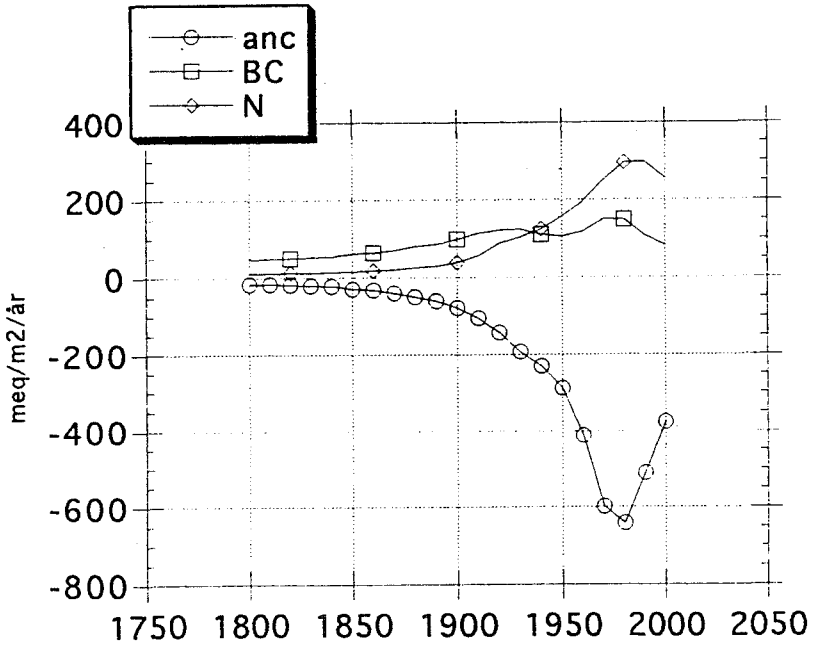


Figure 6. The modeled soil water chemistry using deposition levels as in figure 5.

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Results

The combination of the two models requires a lot of data and computational capacity and so far only very preliminary runnings can be shown to illustrate the framework for intensified work during the coming years.

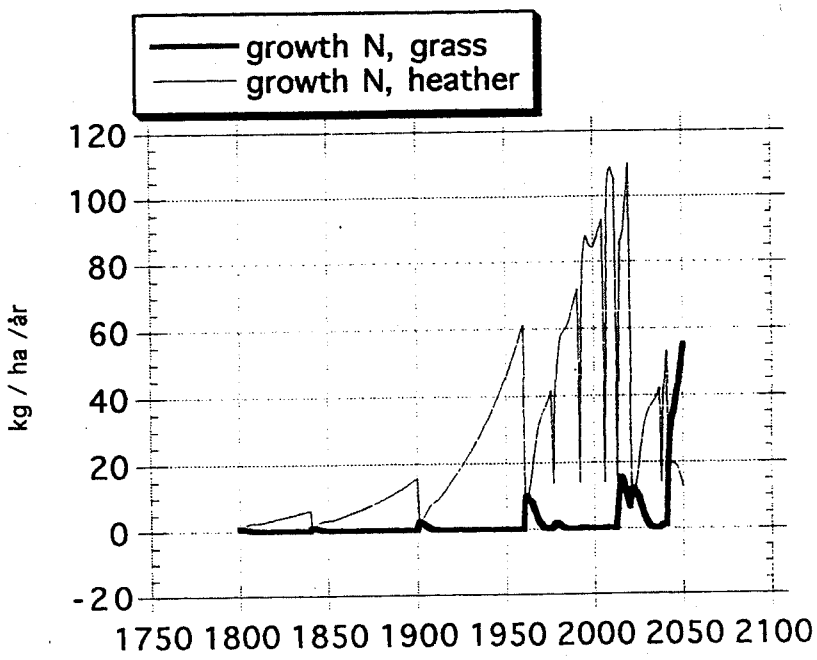


Figure 7. Preliminary CALLUNA modeling using SAFE soil chemistry calculations for a typical Danish heathland soil under the presented nitrogen deposition scenarios. From being heather dominated until year 2000, the combination of high nitrogen deposition and heather beetle attacks will change this balance and the site will end up being grass dominated from year 2040 onwards.

An initial model run with more or less default values for the model parameters have been performed. In this run only the soil parameters, climatic parameters, the deposition scenario and the management practice have been adjusted to fit the simulated Danish site at Hjælm Hede. The resulting simulation curve for growth of grass and heather is shown in figure 7. From 1960 onwards the beetle attacks severely influence the competition and increase in dominance resulting in a change into grass dominance from year 2040 onwards. This happens even if the reduction scenario for nitrogen deposition as shown in Figure 5 should be effectuated.

There are, however, evidence that the vegetation at the simulated site, at present deposition level, is not nitrogen limited, which is a presumption in the CALLUNA model. Instead the vegetation is believed to be limited by the level of phosphorous and maybe potassium at the site (Riis-Nielsen, pers. comm). This complicates the use of the models as they are. The proper way to handle this will be to include competition for other nutrients in the CALLUNA model and to include phosphorus in the SAFE model. This will be done in cooperation with the authors of the models. The importance of base cation deposition will increase in such future calculations.

At present the assumption of phosphor limitation is integrated in the model calculations by limiting the amount of available nitrogen in the input to the CALLUNA model, when the total growth rate exceeds the expected amount of available phosphorus. This will lead to a smaller accumulation of nitrogen in the ecosystem and is therefore not quite satisfactory. With the

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phosphorus limitation included in the system, the resulting curve for simulated growth shows that beetle attacks affects the heather growth and population dynamics, although heather stays the dominant vegetation within the time frame of the simulation (Figure 8). In this model run, nitrogen availability has only been reduced for the 6 years with highest growth rates, indicating a rather high sensitivity of the system being modelled.

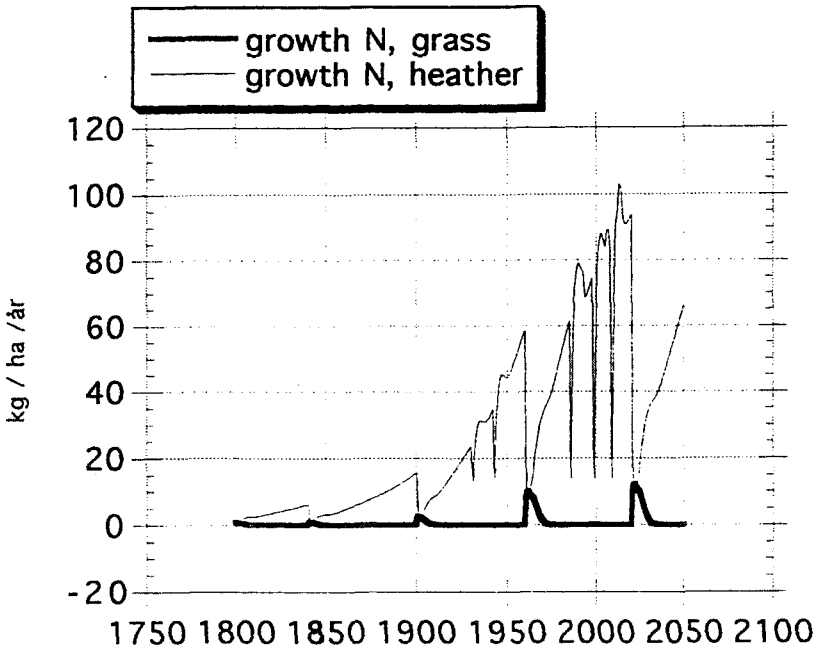


Figure 8. Preliminary CALLUNA modeling using SAFE soil chemistry calculations for a typical Danish heathland soil including growth limitation due to phosphorous limitation. Heather keeps on being the dominant vegetation element using the known and predicted nitrogen deposition levels.

Discussion

As stated in the introduction these calculations are preliminary and many model adaptations and refinement are to be done. As such, the ecological consequences of the resultant simulations should not be discussed in detail due to numerous uncertainties at present stage of development.

The major problems for the model integration and application for improved critical load values for Danish heathlands are at present:

1. The necessary data for the Danish site are not yet fully integrated (mineralogy, hydrology, growth, mortality and redistribution parameters under Danish conditions, the beetle attack intensity and frequency) but most of these data are expected to be available during 1996.

2. For realistic simulation at Danish heathland sites the specific conditions of natural succession under the applied management strategies should be integrated. Sod cutting is never used in Denmark nowadays and the model should be adapted to work with mowing and/ or grazing and/ or fire instead. Mowing should work rather much as sod cutting in the model, only removing less nutrients and influence the regrowth competition for the involved species. Grazing is somewhat more complicated as the con-

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tinuos removal of nutrients is expected to give a complex response for competition between heathland species (Grant & Armstrong 1993).

The natural succession in Danish heathlands involves other species than *Calluna* and *Deschampsia*. The boreal species *Empetrum nigrum* (crowberry) becomes dominant late in succession at most Danish heathlands, and this species has been very little investigated so far. Much work is needed to integrate this species in the model simulations, but for a rather intensively managed heathland as the Hjælm site, *Empetrum* will not become dominant and the inclusion in the model has not been found crucial.

3. Problems with duplicated processes in the two models in the nitrogen cycling and missing processes as related to natural conditions, e.g. the competition for phosphorous and base cations, have to be solved. In the present implementation, soil processes have been simulated by the SAFE model while nitrogen cycling in relation to plant growth has been simulated by the CALLUNA model. This choice can more or less be done by proper setup of the models and by adjustment of the deposition and uptake data exchanged between the models. At a later stage, canopy exchange of base cations will, however, have to be handled by the SAFE model.

4. Combining the rather complex SAFE model with CALLUNA and later other models require much calculations time or very fast processing. This is due to the stochastic processes involved by heather beetle attacks, requiring at least 20 or up to 100 runnings to obtain reliable scenarios for future development under various nitrogen deposition scenarios.

The results of these preliminary model simulations should be taken with precautions, but the principles and ideas provide the basis for an important step forward in combining existing models for ecosystem effects with soil chemical modeling and deposition scenarios. Such work is needed for improving the present critical load setting and mapping, but rather detailed data are needed for the exercise.

Acknowledgments

The work presented here is a further development of the work in the model developing groups at the universities in Lund and Utrecht and their open-minded discussions and provision of model versions is the fundament for this work. Data used for the models have and will be generated by colleagues in the Danish SMP-HEATH project including the work of Dept. of Earth Science, University of Aarhus and Botanical Institute, Univ. of Copenhagen. The national and international cooperation is greatly acknowledged.
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FOREST HEALTH MONITORING - A CANADIAN APPROACH

J. Peter Hall

Coordinator Forest Health Research, Science and Sustainable Development Directorate, Canadian Forest Service, Natural Resources Canada, Ottawa CANADA K1A 0E4

Abstract. The Canadian Forest Service (CFS) has established forest health monitoring networks; the Acid Rain National Early Warning System (ARNEWS) and the North American Maple Project (NAMP) to assess the state of health of forests. These monitoring systems evaluate the condition of tree crowns, determine what stress factors are affecting trees, and from this information, assess the state of health of forests.
The results from these monitoring systems show that the greatest stresses on forests are caused by insects, diseases, and weather extremes, all natural features of the environment. Specific cases of damage caused by air pollution have been observed. Systems to monitor the health of forests are an integral and essential part of the sustainable management of forests.

1. Introduction

The sustainable management of forests is a major goal of the Government of Canada. The Canadian Forest Service (CFS) of Natural Resources Canada as the forest research arm of the federal government, leads this initiative in cooperation with the provincial governments who are responsible for forest management and industrial partners who manage the forest for economic goals.

Classic pollutant effects on stands have occurred most frequently around strong point source emissions of heavy metals, fluorine, SO_2 and NO_x. Damage on a regional scale from pollutants transported over long distances is rare, but predicted losses of forest productivity have given rise to concerns that forests are at risk to regional pollution (Rennie 1987, Fraser et al. 1985).

Here, forest health is defined as the conditions of forest ecosystems that sustain their complexity and vitality. It is axiomatic that the state of health of forests must be understood if actions are to be taken to preserve it, or to act to restore it if it is endangered. Forest health monitoring systems have evolved from previous surveys including the well-known Forest Insect and Disease Survey which has been operating for 60 years.

[1] Background paper for the International Workshop - Spatial and Temporal Assessment of Air Pollutant Impacts on Ecosystems, Vienna, 22-24 November, 1995.

This paper describes two systems: the Acid Rain National Early Warning System (ARNEWS) and the North American Maple Project (NAMP).

2. Methods of assessing forest health

Monitoring biological responses to stresses in the field is difficult because biological systems respond to a complex of environmental factors rather than to a single factor such as `air pollution' (Bormann 1985). The approach taken by the CFS is to assess the state of health of the forest using a common set of measurements taken on a series of permanent sample plots (Hall and Addison 1991). The strategy of ARNEWS is to detect and determine the cause(s) of observed damage, and to monitor the long-term changes in vegetation and soils attributable to

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pollutants. The experience of field technicians trained to distinguish these symptoms from abnormal climatic conditions, nutrient deficiencies and the effects of insects and diseases is crucial to the functioning of ARNEWS.

ARNEWS is a national system and is used to assess forest health on a national scale. Other considerations led to the establishment of NAMP. Sugar maple is one of the most important hardwood species in eastern Canada, and concerns about its health were widespread during the 1980s. It was widely proposed that airborne pollutants were damaging the trees and at the same time, knowledge of tree condition was lacking (Millers et al. 1989). In 1988, the United States Forest Service and the CFS established a network of monitoring sites in the northeastern United States and Canada over the range of sugar maple (Millers et al. 1991). The principles used in ARNEWS formed the basis of NAMP.

The assessment of tree health in both systems is based on: (a) Mortality levels; are they normal or greater than normal?
(b) Crown condition; does it indicate poor health? and
(c) Symptoms of pollution damage; are there any present?

(a) Mortality

A rate of 1-2% annual mortality is considered normal in unmanaged forests; it is caused by natural thinning as the stands age. Mortality in excess of this rate could be caused by any number of natural factors, such as insects, diseases, windthrow or drought; or by anthropogenic factors such as air pollution. The cause of tree mortality is determined in the field and supplemented by laboratory examination as necessary.

(b) Crown Condition

The condition of tree crowns is used to evaluate forest health. Crown condition in hardwoods is assessed using measures of dieback and transparency. Defoliation is used as a measure of crown condition in conifers.

Dieback is assessed as the percentage of the outer crown that contains bare twigs and or bare branches. The system of classification is based upon the visible parts of the outer crown: the quantity and quality of foliage, and the percentage of the outer crown that contains bare twigs and dead branches. There is a direct link between the amount of crown dieback and the health of the tree. As crown dieback approaches 40%, the health of the tree begins to decline and it becomes susceptible to damage by secondary organisms. Recovery is possible but if dieback increases and persists, the possibility of recovery decreases (Lachance et al. 1995).

Transparency is defined as the amount of skylight visible through the foliated portion of the crown (NAMP program only) and thus is the opposite of foliage density (Millers et al. 1991). Crown transparency levels up to 25% are frequently observed and reflect the presence of drought, weather conditions at time of flushing, or insect defoliation. Generally, good growth the following year will bring back thick foliation (Lachance et al. 1995). However, if a stand has more than 25% transparency, there may be significant stress with the probability of dieback appearing the following year.

Defoliation of conifers is defined as foliage missing, for whatever reason, from the normal complement of foliage (D'Eon et al. 1994). The assessment considers the natural loss of needles as twigs mature. However, as defoliation approaches the 30-40% range growth is

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reduced and the trees become stressed. In this condition they are less able to resist damage from other organisms or to tolerate stresses and the probability of mortality increases.

The causes of damage symptoms are determined by the field staff whose observations are confirmed by laboratory examination. Insects and diseases are identified and their levels of damage are determined.

(c) Air Pollution

Air pollution is the third measure of forest health. Many symptoms of air pollution damage are known; descriptions of these conditions have been developed from laboratory fumigations or from field observations near point sources of pollution (Malhotra and Blauel 1980). Nevertheless, symptoms of air pollution damage are not highly specific and often resemble damage from other causes. Symptoms are recorded by field technicians trained to distinguish them from the effects of abnormal climatic conditions, nutrient deficiencies, insects, and diseases (Hall and Addison 1991). Damage from air pollution may lead to poor health since trees are not naturally adapted to stress from pollutants.

3. Program objectives

The objectives of the two monitoring programs are to provide the information to assess the state of health of forests in general and sugar maple in particular. The objectives of ARNEWS are to detect the possible damage to forest trees and soils caused by acid rain, by identifying the damage sustained by forests that are not attributable to natural causes or management practices; and to monitor vegetation and soils over the longterm.
The objectives of NAMP program are to determine the annual rate of change in crown condition, and to determine whether crown condition differs between levels of pollution deposition. Trees in stands managed for sap production and trees in unmanaged stands are monitored.

4. Data reliability

Visual estimation techniques for data collection are often considered to be somewhat unreliable because of observer bias and interpretation error (Burkman et al. 1990). Monitoring systems assess tree condition over large areas across Canada's forested ecozones (ARNEWS) or throughout a species range (NAMP). A common methodology is used, but the program is carried out by several regionally based teams operating independently. The differences that can exist among observers and among regions raises concerns as to the consistency and reliability of data on tree condition.
The field staff operate a quality control program to review the quality and reproducibility of data obtained from the field. The objectives of the program are to implement training of field staff on a national basis, to check critical variables annually, and to test the accuracy and reproducibility of methods.

Cross-checks are performed on plots within two weeks of measurements by field crews. Crown condition and crown damage are assessed during the cross-check and compared to the observations of the regular field crews, thus ensuring reproducibility of the data.

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5. Results and Discussion

Results from the monitoring systems are summarized based on the data collected to date (Hall 1995a, 1995b; Cooke et al. 1995). Data from the networks are supplemented by other surveys, all with the goal of assessing the health of forests.

(a) Mortality

In the ARNEWS system between 1986 and 1993, overall mortality was 1.1%, with conifers having 1.0% and hardwoods 1.4% mortality. These rates are within the normal range in maturing natural forests. Mortality varied among years because the amount of insect damage and drought conditions varied widely. Sugar maple in the NAMP system had an annual mortality rate of 0.6% between 1988 and 1994, similar to the rate (0.9%) observed in ARNEWS (Cooke et al. 1995). The overall rate was the same in sugarbushes and in unmanaged stands.

Most major species had annual mortality rates of less than 2% (Table 1). Mortality was in the normal range, and caused mostly by natural thinning as the stands aged. Mortality was caused by drought, frost, windstorms, winter storms, root rots, spruce budworms, and other defoliating insects (Hall 1995a).

However, three species had mortality rates at the high end of the scale: balsam fir, trembling aspen, birches including white birch, and mountain paper birch. The higher rates of mortality in balsam fir and trembling aspen resulted from damage caused by the eastern spruce budworm (*Choristoneura fumiferana* Clem.) and the large aspen tortrix (*C. conflictana* Wlk.) respectively, and from subsequent damage caused by secondary insects and diseases which killed many trees. Trembling aspen was also subjected to a number of unseasonably early and late frosts which caused added stress to the trees.

White birch near the Bay of Fundy in eastern Canada had extensive damage twig and branch dieback, and abnormally high mortality rates (Magasi 1989). Birches in other parts of the species range had normal mortality rates.

(b) Crown Condition

Nearly all the observed damage on trees was caused by insects and diseases. Insects caused defoliation on most species throughout Canada, the greatest damage occurring on conifers in the Maritimes and Ontario.

Damage varied widely from year to year, among species, and among regions. Occasionally damage was severe enough to cause mortality. The commonest defoliating insects, the eastern spruce budworm, and the large aspen tortrix caused extensive defoliation in balsam fir and trembling aspen respectively. Jack pine was damaged by a disease (*Endocronartium harknessii* [J.P. Moore] Y. Hirat.) to the extent of occasional mortality. The damage though widespread is part of the normal stresses in forests to which tree species are adapted. Recovery from damage was also observed (Hall 1995a, 1995b).

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Table 4. Mortality rates for selected species in ARNEWS plots, 1987-1993.

Species	Annual rate of mortality %
Eastern white pine *Pinus strobus* L.	0.8
Jack pine *P. banksiana* Lamb.	1.1
Lodgepole pine *P. contorta* Dougl.	0.8
Tamarack *L. laricina* (Du Roi) K. Koch.	0.5
Black spruce *Picea mariana* (Mill.) BSP	0.9
White spruce *P. glauca* (Moench) Voss	0.6
Douglas-fir[1] *Pseudotsuga menziesii* (Mirb.)Franco	0.9
Balsam fir *Abies balsamea* (L.) Mill.	1.8
Trembling aspen *Populus tremuloides* Michx.	2.3
Birches[2] *Betula* spp.	3.0
Red oak *Quercus rubra* L.	0.9
Sugar maple *Acer saccharum* Marsh.	0.9

[1] Includes Douglas-fir and Rocky Mountain Douglas-fir. *P. menziesii var. glauca* (Beissn.) Franco.

[2] Mixture of white birch (*B. papyrifera* Marsh.) and mountain paper birch (*B. cordifolia* Regel)

More than 90% of the sugar maple measured in NAMP were healthy (Cooke et al. 1995). The percentage of dieback on trees over the measurement period was consistently higher in sugarbushes than in unmanaged stands. The differences were not statistically different; however, both dieback and transparency were consistently higher in stands managed for sap production (sugarbushes). This may indicate slightly higher levels of stress in sugarbushes.

Crown transparency varied widely among stands and over time. Transparency was usually higher in larger, older trees and in sugarbushes than in unmanaged stands. Transparency varied from year to year in response to an infestation of an insect, pear thrips (*Taeneothrips inconsequens* Uzel.) and in response to drought. In both cases transparency diminished in a year or two (1990/1991) to lower levels. Thus transparency, unlike dieback, is not a good indicator of tree health but it is a variable that indicates rapid changes in tree condition (Lachance et al. 1995).

One of the objectives of NAMP is to evaluate the relationship between levels of dieback and transparency and deposition of pollutants. No consistent relationships were found between sulphate and nitrate deposition levels on the one hand and dieback or transparency on the other (Lachance et al. 1995). However, for nitrate deposition there was a tendency toward greater dieback at higher levels of deposition.

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Transparency was usually higher in zones of high deposition. Deposition of pollutants varies in intensity and location from year to year, so that individual plots may be in different deposition zones in different years. This further complicates the interpretation of the relationship.

(c) Damage caused by pollution

The primary incidence of air pollution damage observed in the ARNEWS plots was on birches near the Bay of Fundy in eastern Canada. Damage symptoms included leaf browning, and premature leaf senescence, followed by twig and branch dieback. Over several years many trees died (Magasi 1989). There has been some recovery as tree condition fluctuates with the frequency of acid fogs suggesting a cause and effect relationship (Cox et al. 1989). In areas outside the Bay of Fundy birches did not have these symptoms (Hall 1995a, 1995b).

Ozone-like symptoms were observed in eastern Canada on white spruce, black spruce, balsam fir, yellow birch (*Betula alleghaniensis* Britt.), sugar maple, red maple (*Acer rubrum* L.), and white birch. These symptoms were small yellow flecks on the leaves; investigations continue, to determine the cause. Similar instances of needle flecking have been observer on Douglas-Fir in British Columbia.

6. ARNEWS plots with symptoms of decline

Decline is defined as a continued and progressive deterioration of condition ending in the death of the tree (Manion 1981). In several ARNEWS plots, more than 15% of the dominant and codominant trees had greater than 50% damage to their crowns for three or more years. These plots are also in areas of acid-sensitive soils and have annual deposition levels of pollutants in excess of 20 kg/ha, a combination that raises the question whether pollutants may be adding stresses to trees already damaged. Pollution levels are low compared with those in most ARNEWS plots, but the natural acidity of the soils means that they have limited buffering capacity to neutralize any incoming acids. Damage on these plots has been caused by defoliating insects, diseases, and drought and recovery has been slower than in areas of soils with higher buffering capacity (Hall 1995a.) There is no direct evidence of pollution damage, however, trees may be further stressed by pollutants on these sensitive soils.

7. Soil sampling and analysis

One of the objectives of the ARNEWS program is to monitor the effects of pollution on soil chemical properties at each ARNEWS plot. The initial sampling and analysis were made between 1985 and 1987 with additional sampling in 1990. The results indicate that there are reductions in nutrients in areas of higher deposition of sulfate and nitrate pollutants suggesting that changes are taking place in the soils under the ARNEWS plots (Hall 1995a). The changes are greatest in the zones of higher deposition, leading to the suggestion that pollutants are the cause.

8. Conclusion

Results from ARNEWS and NAMP show that mortality has generally been in the normal range for unmanaged forests, with 1-2% of the trees dying annually as a result of natural

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thinning. Higher mortality levels were caused by insects and/or diseases. Some increased mortality on birches, however, was caused by pollutants.

Tree condition data indicate that trees were generally healthy; the crowns were damaged by a variety of insects, diseases, and abiotic factors which commonly damage forest trees.
Sugar maples were generally healthy, with normal levels of mortality. Dieback declined over the measurement period and was consistently higher in sugarbushes than in unmanaged stands. Transparency fluctuated from year to year in response to short-term stresses. Areas of higher pollutant deposition were often accompanied by increased levels of dieback and transparency. Other stresses, including drought and insect damage, also affected crown condition.

Investigations of damage symptoms on birch concluded that the damage was caused by pollutants. Other damage symptoms resembling those caused by air pollution were also observed in eastern Canada on pines, spruce, and maples, and in western Canada on Douglas-fir. Investigations continue to determine the cause of the damage.

Constant monitoring is necessary to explain the causes of forest decline. It is necessary to monitor forest conditions to know what is happening if we are to make conclusions on the state of the health of our forests.

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EPIPHYTIC LICHEN VEGETATION AS A BIOMONITORING SYSTEM IN FOREST DECLINE STUDIES IN SLOVENIA

BATIČ, F[1] and KRALJ, A.[2]

[1]Agronomy Department, Biotechnical Faculty, University of Ljubljana
61000 Ljubljana, Jamnikarjeva 101, SLOVENIA
[2]Slovenian Forestry Institute, Večna pot 2, 61000 Ljubljana, SLOVENIA

ABSTRACT

Epiphytic lichen vegetation has been used as a bioindicator of the air quality of forest sites in Slovenian forest decline inventories since 1985. On the base of an assessment of the cover and frequency of all three lichen thalli types, an index of atmospheric purity (IAP) was calculated for each assessed tree and together for the plot. High values of the index indicate rich epiphytic lichen vegetation and clean air, whereas small values indicate poor epiphytic lichen vegetation and presumably polluted air. The results of an inventory carried out in 1995 are presented, when the grid of the inventory was 4 x 4 km. Values of the index show that air pollution in Slovenia is rather high, especially around the major known air pollution sources but also in some areas without known local emissions of air pollutants. Apart from indicating air pollution in comparison with measured air pollutants at the nearest measuring site, the condition of epiphytic lichen cover and to lesser extent the species composition were analysed according to climatic parameters, the elevation of the forest site, the stand structure and the history of the stand. There are indications that apart from the air pollution and climate of the forest site, the type of forest management may play an important role in the survival and flourishing of lichens. It was also proved that there is no single relationship between the condition of epiphytic lichen and the forest damage at a particular site. As already stated, the state of epiphytic lichens reflects well the air quality at the site, and partly also the forest stand structure and history, but cannot explain damage to forest trees. It is only one of the parameters which can contribute data for stress analysis of the tree damage at a particular forest site.

INTRODUCTION

According to present knowledge forest decline is a multistress dependent process in which air pollution plays an important role (Schütt et al. 1986, Cowling 1989). Measurement of air pollutants are not possible for several reasons in all forests, and therefore there is a need for the use of bioindicators, already present in the forests, in order to assess air pollution at the forest site under consideration.

Epiphytic lichen vegetation has been used as an air quality bioindicator at forest sites in Slovenia since 1985 when forest die-back inventories first started (Batič & Kralj 1989, Batič 1991a, Batič 1991b, Batič & Kralj 1995). Epiphytic lichens were chosen for this purpose because they were well proven indicators of air pollution, especially of " classic " air pollution in which sulphur compounds and dust were the major air pollutants (Skye 1968, Hawksworth & Rose 1976, Ferry

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et al 1973, Deruelle 1978, Richardson 1988, etc.). This type of air pollution is still todays the prevailing pollution type, besides pollution caused by traffic and agricultural activities (Anonymous 1994), and the use of epiphytic lichens as an air pollution bioindicator monitoring system also seems a reasonable tool according to experience in other countries (Wittmann & Türk 1988, Gliemeroth 1990, Hoffman 1994)

METHODS

Two different methods of using epiphytic lichens as bioindicators of air quality have been used in the forest decline studies in Slovenia.
In forest decline inventories a very simple method of mapping the three most common thalli types of epiphytic lichens, namely crustose (C), foliose (F) and fructicose (R) was implemented by assessing the frequency (a) and the cover (c) of the each thalli type on a group of six trees, selected from 24 trees on which decline symptoms were observed. The group of six trees for epiphytic lichen overgrowth assessment was chosen according to the following criteria: climatozonal tree species, suitable age of the trees, and favourable light conditions of the trees for epiphytic lichen growth. Lichens were observed on trees in three separate strata: the tree trunk base up to 0.5 m height (1); the tree trunk from 0.5 m to 2.5 m height (2), and the tree crown (3). Frequency and cover of all three lichen thalli types were assessed within each tree trunk stratum on a very simple scale:

frequency (a): 1 up to 5 thalli per observed trunk surface
2 6 - 10 thalli per observed trunk surface
3 more than 10 thalli per observed trunk surface

cover (c) : 1 thalli cover 1 to 10 % of observed trunk surface
2 thalli cover 11 to 50 % of observed trunk surface
3 thalli cover more than 51% of observed surface

From these observed and assessed lichen parameters a special index of atmospheric purity (IAP) was calculated using the following equation:

IAP = C(a+c) + F(a+c) + R(a+c); (IAP= index of atmospheric purity; C=crustose thalli, F=foliose thalli, R=fructisose thali; a, c = frequency and cover).

The IAP can be calculated separately for the observed tree strata (IAP_1- tree trunk base, IAP_2- tree trunk at "breast" height, IAP_3 - tree crown; span: 0 - 18), or for the whole tree (IAP or IAP_t; span: 0 - 54); $IAP=IAP_1 + IAP_2 + IAP_3$.
For better transparency of data, the values of IAP can be classified in similar groups like forest damage classes, representing the state of the epiphytic lichen overgrowth and air quality at the forest decline inventory plot. Calculations of IAP are possible on a tree species basis as well as on a tract (inventory plot) base. The most reliable data are obtained when the results of observations on the same species are compared, or on the group of tree species with similar ecological circumstances for epiphytic lichen growth, e.g. type of crown, bark properties, etc.).

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For a very rough presentation, data can be computed on a plot basis, but otherwise species basis presentations are much more precise.

This method is used for assessment of air pollution by epiphytic lichens on a very broad scale, at a state level. It is very simple and cheap and can be carried out by a trained team responsible for the whole forest decline inventory. In all case studies of forest decline, the results of this method are only the starting point and further mapping of lichen species is necessary.

RESULTS AND DISCUSSION

The results of the epiphytic lichen assessment carried out in the forest decline inventory of 1995 are presented in Figs 1-1c. Forest decline inventory was carried out on a 4 x 4 km grid. The values of IAP are classified in groups, calculated for the whole tree on the tract (IAP), or for observations for separate strata (base of the tree trunk, trunk at breast height and for the crown) and displayed on a map of Slovenia.

Comparison of IAP values with measurements of air pollutants in Slovenia and measurements of total sulphur content in Norway spruce needles shows that epiphytic lichen vegetation is a relevant bioindicator of air pollution, even when such a simple method is used. All known air pollution point sources are well expressed by small IAP values found around major towns, industrial plants and thermal power plants. Additionally, there are some areas where lichen overgrowth is quite poor but there are no known local sources of air pollution (Julian Alps). In these cases transport of air pollutants from remote areas is probably involved. The lowest values of IAP on the presumably polluted sites were determined when IAP was calculated only on the basis of lichen observations in crowns, (IAP_3 ; Fig. 1c), followed by observations on trunks at breast height (IAP_2 ; Fig. 1b), and the highest, when obsevations were carried out on the base of tree trunks (IAP_1 ; Fig. 1a). All these localities are apparent also when IAP was calculated for the whole tree (IAP; Fig. 1), but less distinctly. Calculations of IAP, presented in Figs. 1-1c were carried out regardless tree species. When comparing the results of observations on a tree species basis, data are not presented, it is clear that data differ significantly. Most noticeable are observations carried out on common beech which shows the worst epiphytic lichen overgrowth due to its particular crown and bark characteristics. These findings are similar to the results of 1991 forest decline inventory (Batič & Kralj 1995) in which common beech and hornbeam had also the worst epiphytic overgrowth due to above mentioned characteristics, e. g. smooth bark, and centripetal type of crown according to precipitation distribution (Barkman 1958).

Comparison of the results of the inventories carried out in 1991 and 1995 in (Fig. 2) on a tract basis shows that air pollution remained quite similar. That is consistent with the overall situation of air pollution in Slovenia, which has not been significantly reduced and such a situation is expected (Anonymous 1994).

Comparison of IAP values with the forest site parameters shows a slight increase of values with the altitude of the site (Fig. 3). This is consistent with the more favourable climatic conditions for lichen growth at higher altitudes and smaller air pollution due to the location of pollution sources. Nevertheless, there are also some opposite findings where lichen overgrowth decreases with altitude (Julian Alps, Karavanke, Snežnik, Kočevsko). In this cases transboundary air pollution transport or temperature inversions are probably involved. Comparison of IAP data with other site parameters such as exposure, relief, etc. have not yet been evaluated. Climatological data

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such as rainfall, wind speed, temperature and moisture regime, and insolation are known for several of the plots where forest decline inventories have been carried out. Comparison of the IAP map of Slovenia with maps of the above quoted parameters (Petkovšek & Trontelj 1987) show that there is the greatest coincidence of high IAP values with the lowest July temperatures of the site. In regard to the nature of the lichens as poikilohydric plants this coincidence seems logical, but the reference data bases are not the same and the match of these data might also be accidental.

Comparison of the IAP values with the forest stand characteristics has not yet shown any relevant differences. There is a tendency of increase in the IAP values with the increase of tree trunk diameter (Fig. 4) of the tree on which lichen overgrowth was assessed. This roughly corresponds to increase of IAP with the age of tree. Comparison of the IAP values with defoliation of trees on the plot (Fig. 5) shows that up to 40 percents of defoliation quite different state of lichen overgrowth was detected, but at higher defoliation of trees on the plot the IAP values were extremely low. This comparison showed that there is better to compare IAP values with defoliation of trees on the plot than with overall assessed damage of the stand as we did in the past (Batič & Kralj 1989).

On the basis of ten years lasting epiphytic lichen observations in forest decline inventories in Slovenia we can conclude that epiphytic lichens are suitable bioindicators of air pollution, even when such a simple method is used. Lichen data can provide inview into the air quality in remote forest areas where measurements of air pollutants are not expected to be carried out, in other more accessible areas, where measurements of air pollutants are available they can help in the interpretation of the effects of air pollutants on different levels of forest ecosystem.

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Fig. 1: Forest decline inventory in 1995. The state of epiphytic lichen overgrowth on the tracts (IAP), presentation for whole tree.

Fig. 1a: Forest decline inventory in 1995. The state of epiphytic lichen overgrowth on the tracts (IAP_1), presentation for tree trunk basis.

Fig. 1b: Forest decline inventory in 1995. The state of epiphytic lichen overgrowth on the tracts (IAP_2), presentation for tree trunk at the breast height.

Fig. 1c: Forest decline inventory in 1995. The state of epiphytic lichen overgrowth on the tracts (IAP_3), presentation for tree crowns.

Fig. 2: Comparison of IAP frequency between forest decline inventories carried out in 1991 (3286 trees) and in 1995 (4014 trees).

Fig. 3: Forest decline inventory in 1995; relationship between the IAP values and altitude of the plot.

Fig. 4: Forest decline inventory in 1995; relationship between the IAP values and stem diameter of at breast height of the trees on the plot.

Fig. 5: Forest decline inventory in 1995; relationship between the IAP values and defoliation of trees on the plot.

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Fig. 1: Forest decline inventory in 1995. The state of epiphytic lichen overgrowth on the tracts (IAP), presentation for whole tree.

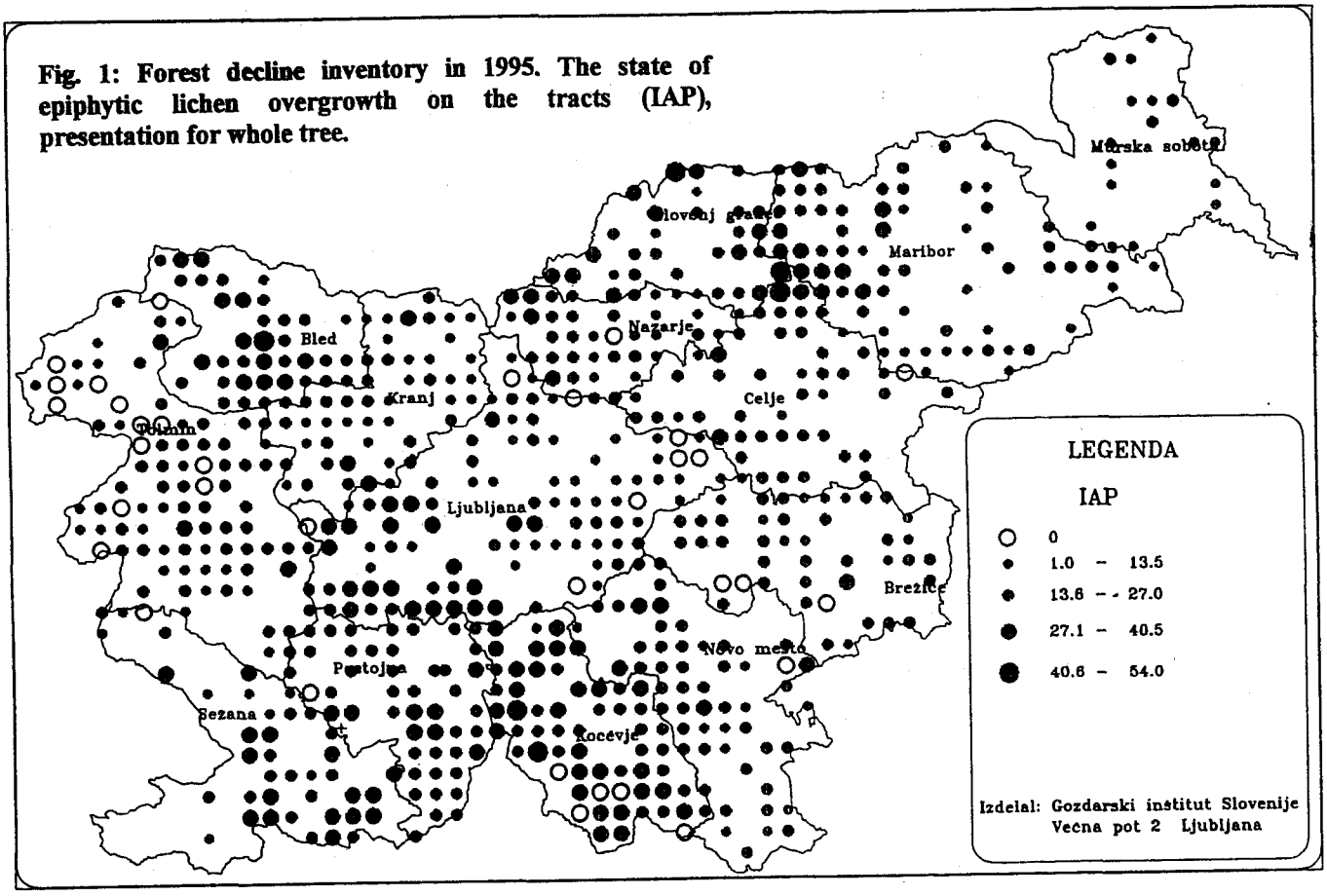
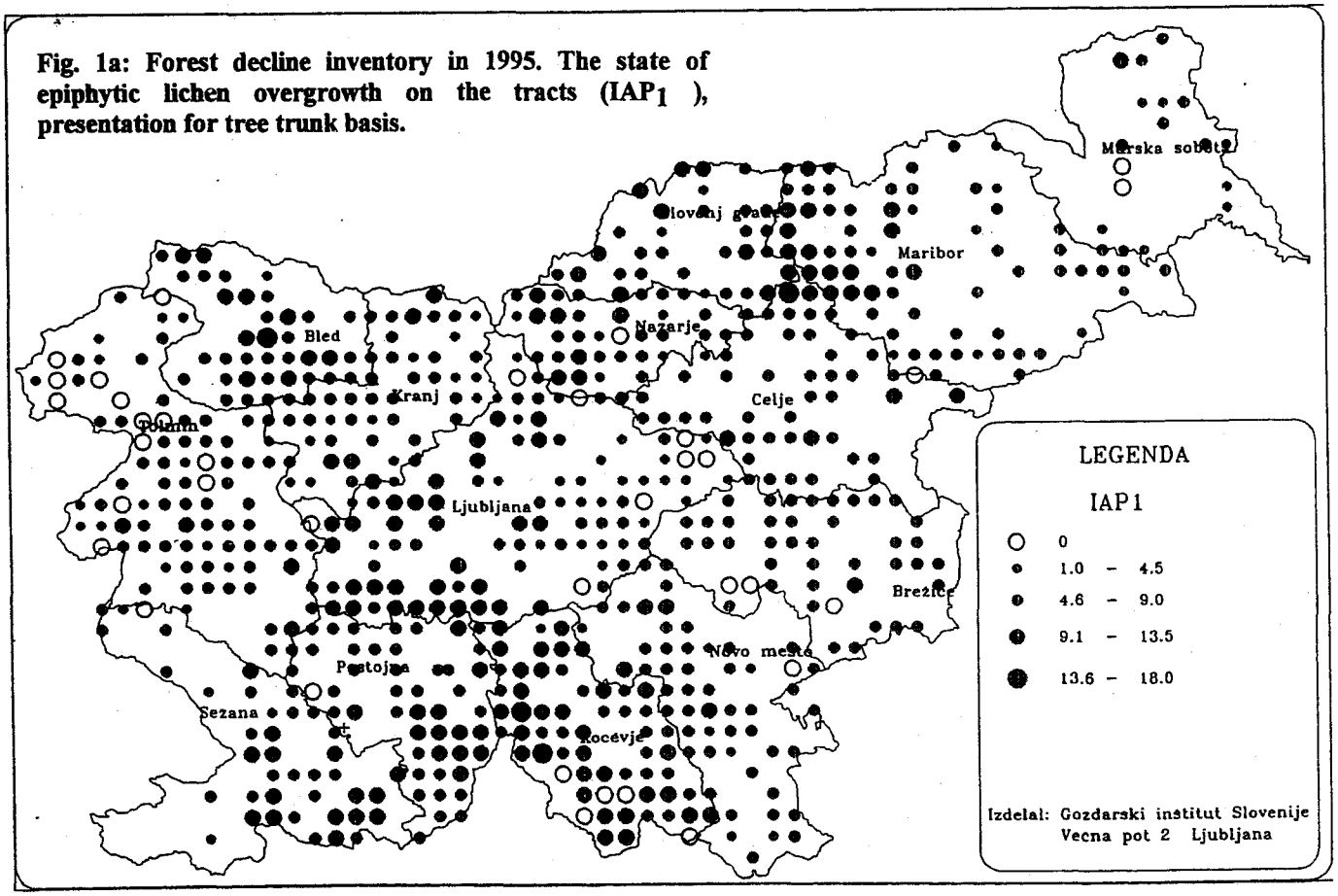


Fig. 1a: Forest decline inventory in 1995. The state of epiphytic lichen overgrowth on the tracts (IAP_1), presentation for tree trunk basis.



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Fig. 1c: Forest decline inventory in 1995. The state of epiphytic lichen overgrowth on the tracts (IAP_3), presentation for tree crowns.

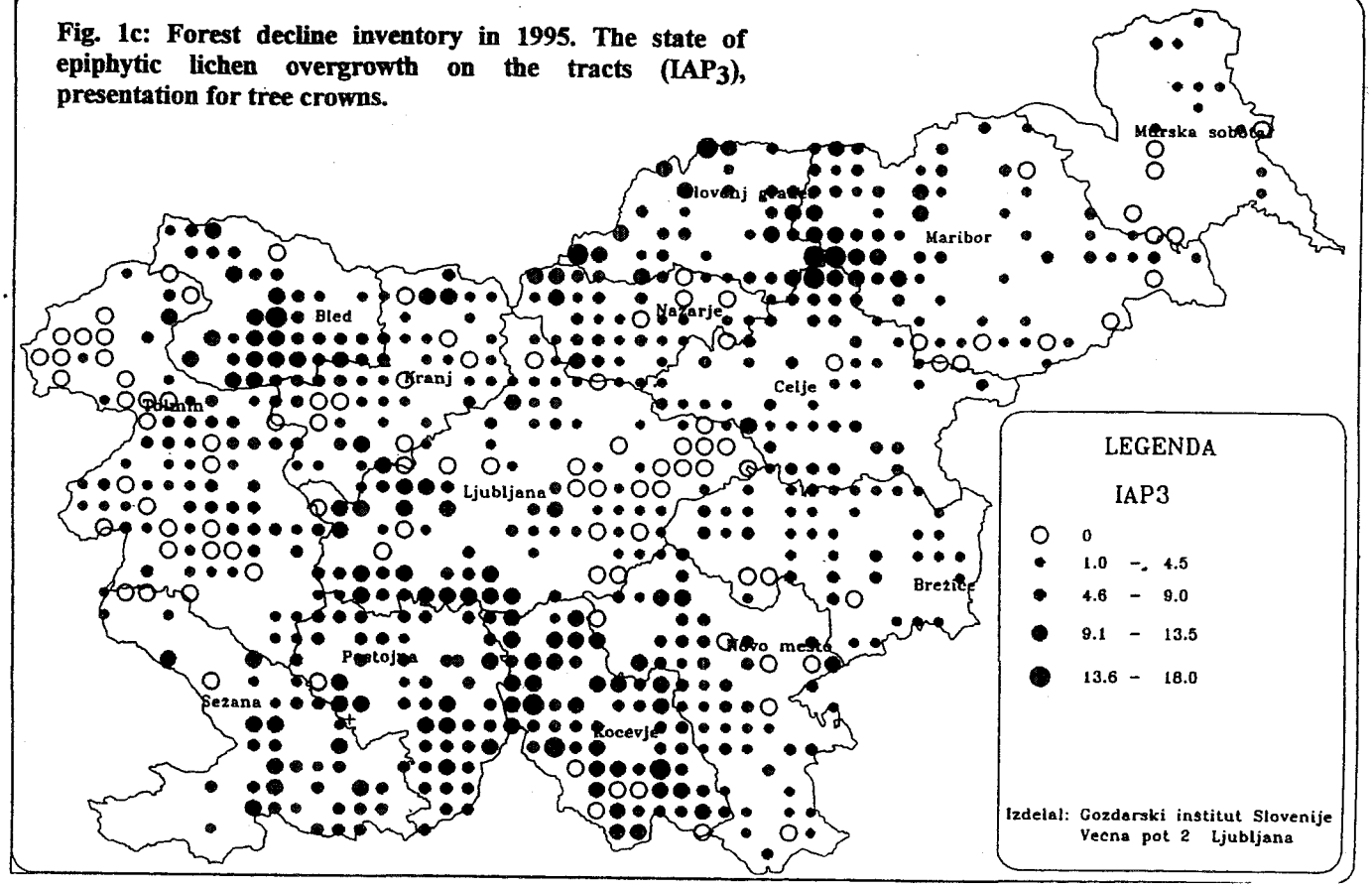
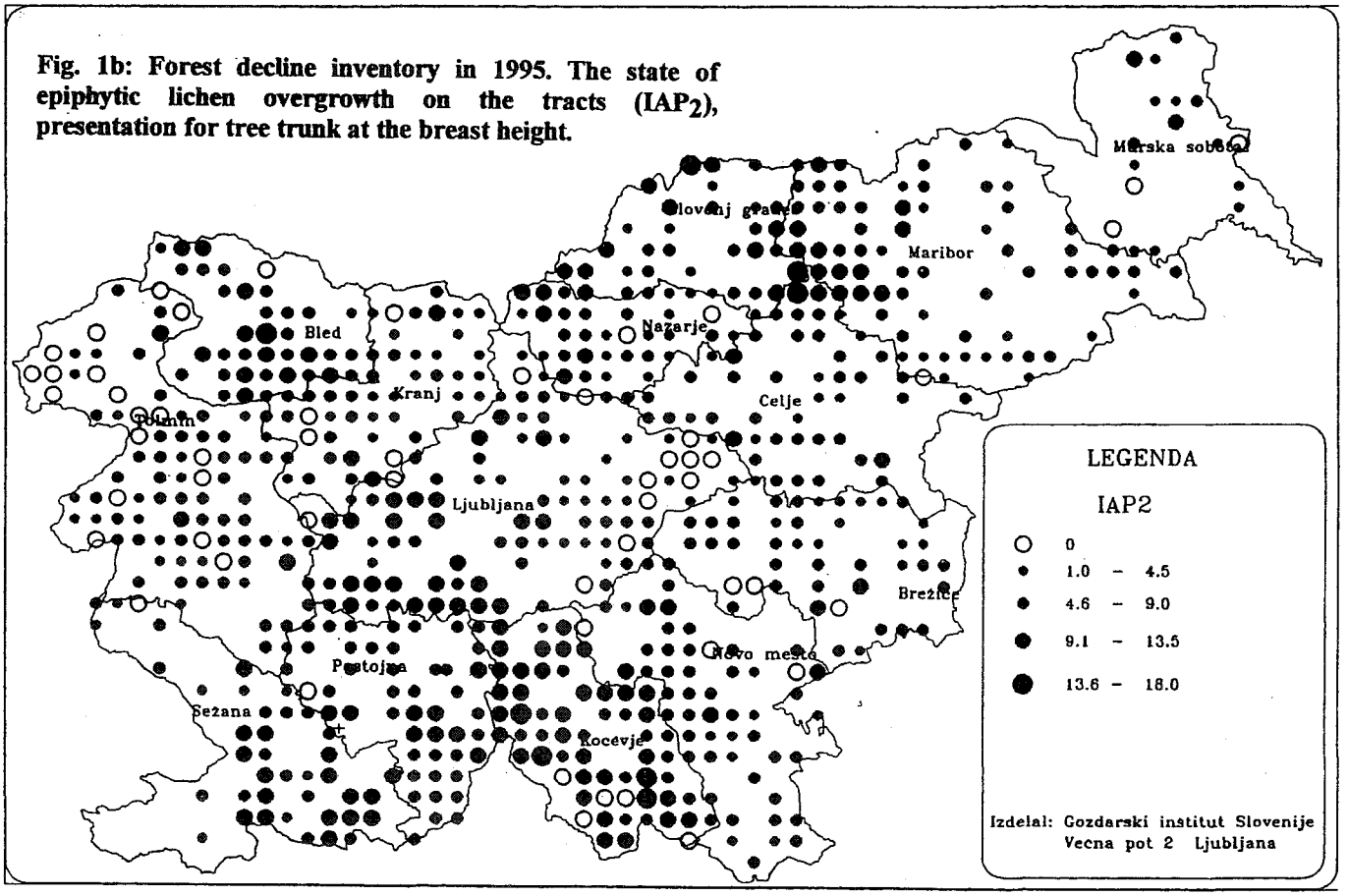


Fig. 1b: Forest decline inventory in 1995. The state of epiphytic lichen overgrowth on the tracts (IAP_2), presentation for tree trunk at the breast height.



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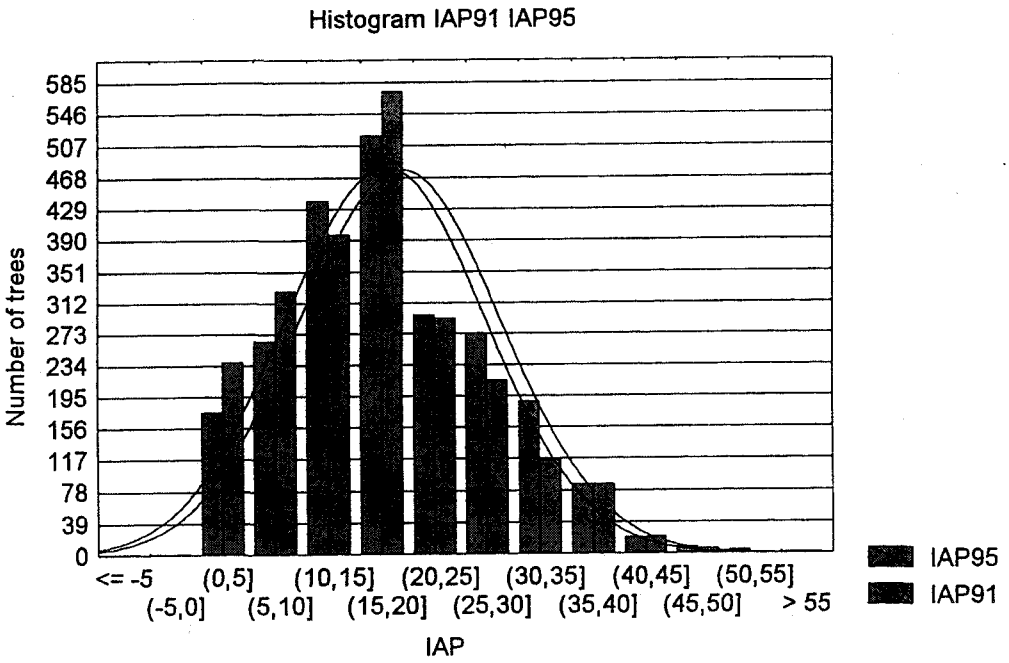


Fig. 2: Comparison of IAP frequency between forest decline inventories carried out in 1991 (3286 trees) and in 1995 (4014 trees).

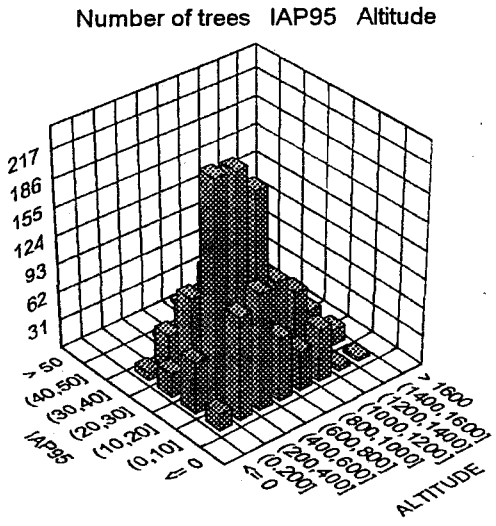


Fig. 3: Forest decline inventory in 1995; relationship between the IAP values and altitude of the plot.

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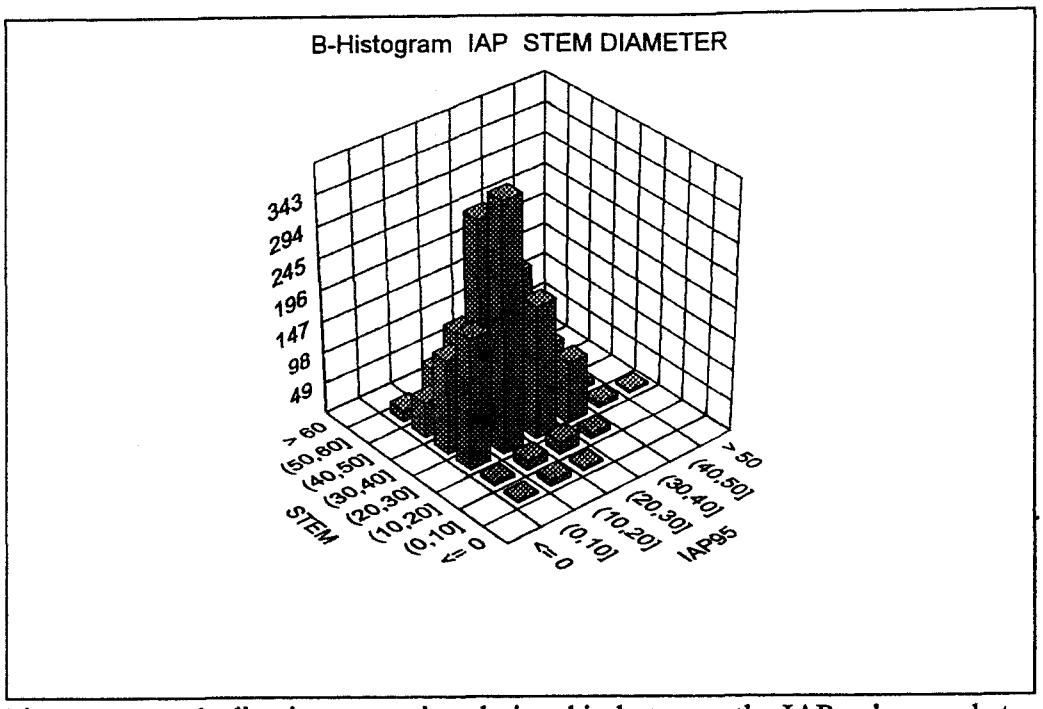


Fig. 4: Forest decline inventory in relationship between the IAP values and stem diameter of at brest height of the trees in the plot.

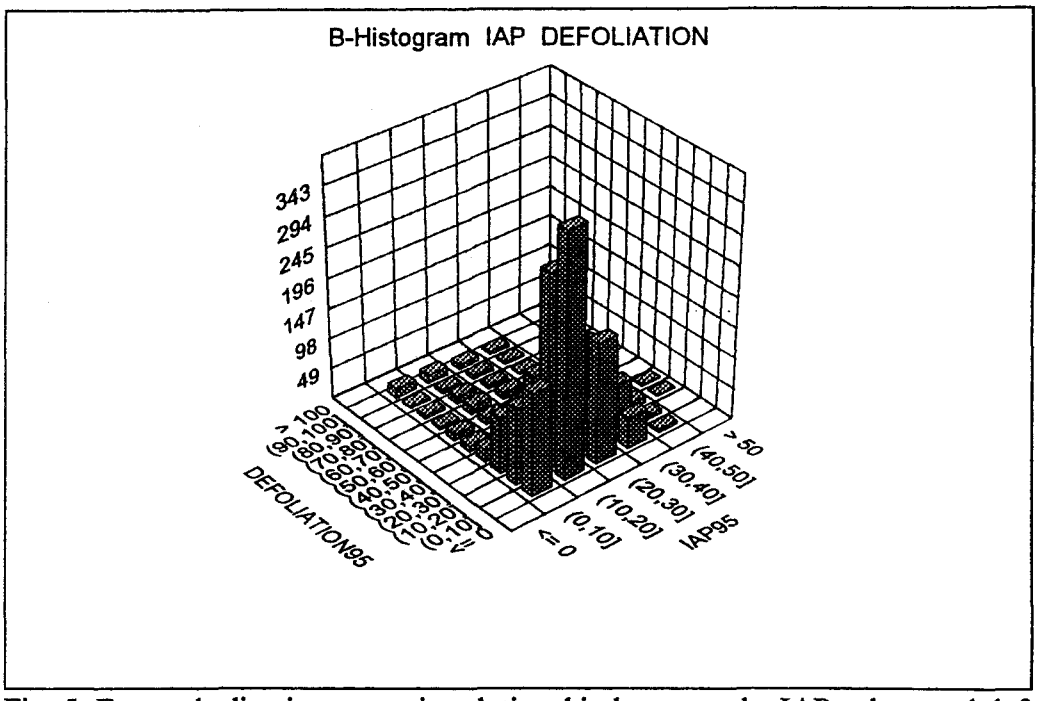


Fig. 5: Forest decline inventory in relationship between the IAP values and defoliation of trees on the plot.

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THE EFFECT OF COPPER, THE HEAVY METAL POLLUTANT, ON PHOTOSYNTHETIC GAS EXCHANGE AND ELECTRON TRANSPORT PROCESSES

H.R.Bolhàr-Nordenkampf[1]), G.Horváth[2]), M.Droppa[2]), J.I.Haumann[1]), L.I.Horváth[3]), M.H.Meister[1]), W.F.Postl[1]) and B.Szalontai[3]).

[1])Institute of Plant Physiology, University of Vienna, Biocentre, Althanstrasse 14, POB 285, A-1091 Vienna, Austria; [2])Department of Plant Physiology, University of Horticulture and Food Industry, Ménesi út 44, POB 53, H-1502 Budapest, Hungary and [3]) Institute of Biophysics, Biological Research Centre, POB 521, H-6701 Szeged, Hungary.

ABSTRACT

Spinach (*Spinacea oleracea L.*) was cultivated in a hydroponic system of a growth cabinet. Copper was taken up by roots from the nutrient solution containing 50 or 150 μmol copper provided via copper sulphate. *In vitro* experiments with copper polluted chloroplasts and thylakoid membranes clearly showed the effect of copper. Beside of a decline in maximum PSII chlorophyll-fluorescence a modification in the protein-lipid phase of chloroplast membranes was observed. FTIR spectra showed clearly that a band around 1620 cm^{-1} (amid I region), which arises in controls during denaturation of proteins by heat was missing after copper treatment. ESR spectra of membranes with spin labelled phospholipids led to the conclusion that the relative amount of solvation lipids was significantly increased.

These results indicate that under copper stress conditions the inhibition of the photosynthetic electron transport is related to copper induced alterations of protein conformation which consecutively leads to modifications of lipid-protein interactions within the chloroplast thylakoid.

In contrast to these results in vivo measurements traced up rather small changes in photosynthetic metabolism referring to nutrient solutions containing 150 μmol Cu. Reductions in photochemical capacity (Fv/Fm) and quantum yield (Φapp) measured by chlorophyll-fluorescence and light response curves were smaller than losses in light saturated photosynthetic rates (Asat). This could be caused primarily by a reduced activity of the Calvin cycle. The reduction in dark respiration was thought to be an additional reason for reduced growth. As copper concentrations finally found in leaves were not too high, it is assumed that copper was in part excluded from uptake by roots and molecules transported into leaves might have been stored by complexation in the vacuole of mesophyll cells. It has to be questioned that the reduction of growth followed by the death of plants (150μmol Cu) is primarily caused by a direct influence of copper on the photosynthetic processes as it is derived from in vitro experiments.

INTRODUCTION

The sources of heavy metal pollution include metal industries, mines, car fumes, power stations, households burning fossil fuels, waste water sludge, paints, tyres (Cd), organic manuring (pig manure contains high amounts of Cu and Zn) and certain chemicals used in

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agriculture such as phosphate fertilisers and fungicides (Tuba and Csintalan, 1992). Copper accumulated in soil is taken up by plants through their root system. Passing the endodermis copper molecules transported in the xylem water are able to reach the mesophyll tissue of leaves. From the apoplast copper can be incorporated into the cytoplasm. Some copper will permeate further on into the vacuole and is thought to be stored by complexation. If copper molecules enter the chloroplast they should be able to interact with many sections of the photosynthetic electron transport chain. Though, copper was shown to inhibit both the light and dark reactions of photosynthesis in chloroplasts from bacteria, algae and higher plants. Regarding the repressive effect on photosynthetic electron transport, photosystem II was more susceptible to copper pollution than photosystem I. In addition the key enzyme in the Calvin cycle, RuBisCO can be strongly inhibited by copper. Although *in vitro* experiments suggested various targets in electron transport for copper inhibition these results are not necessarily confirmed *in vivo* (Droppa and Horvath 1990).
The main goal of this study was to find out about how the decrease of plant growth and biomass production, generally observed in areas polluted with copper, can be related to the copper effect on electron transport found *in vitro*.

MATERIALS and METHODS

Spinach (*Spinacea oleracea L.*) was cultivated in a hydroponic system of a growth cabinet. Half strength Hoagland III was used as nutrient solution, which was changed every tenth day. During the photoperiod of 11 hours spinach plants were exposed to a photon flux of 75 μmol.m^{-2}.s^{-1} (equal to 6 700 Lux) combined with a temperature of 20°C. Night temperature was set to 17°C. Day and night relative humidity was kept around 80% corresponding a saturation deficit equal to a dew point of -3.0°C below ambient temperature.
The Hungarian group used very young and small plants (10 days old) from growth cabinets for growth analyses. *In vitro* experiments were performed with market spinach. In contrast the Austrian group used plants from growth cabinets only. Plants 45 days old with several large leaves (20cm^2) were taken for *in vivo* gas exchange and chlorophyll fluorescence measurements.
For the *in vitro* experiments chloroplasts were isolated by grinding leaves in a buffer containing 0.4 M sorbitol, 10 mM NaCl, 5 mM $MgCl_2$, 2 mM EDTA, 1 mM $MnCl_2$ and 50 mM MES pH 6.5. Consequently twice washed chloroplasts were stored in a solution where MES was replaced by 50 mM HEPES pH 7.5 (Reeves and Hall, 1973). Before measurements chloroplast or thylakoid membrane suspensions were polluted by copper.
Fourier transformed infrared spectroscopy (FTIR) was used to study changes in protein conformation (amide bands I and II) and C-H stretching of fatty acyl chains. For this purpose thylakoid membranes were resuspended in a D_2O based buffer. After centrifugation the pellet was placed between two CaF_2 windows separated by a 15 μm aluminium spacer. Spectra were recorded on a Philips PU9800 FTIR spectrometer and evaluated by a software with fitted Lorentzian components. The spectral resolution was 4 cm^{-1} and 128 interferograms were summed for each spectrum. Temperature was step wise increased by 2°C and after 10 minutes thermal equilibration spectra were recorded.
For spin labelling, the thylakoid membranes (0.5 mg chlorophyll a) were suspended in HEPES (pH 7.5) buffer and spin-labelled solution in ethanol was added while vortexing (Arrondo et al 1993). After 60 minutes of incubation the ESR spectra were recorded using a

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Brucker ECS 106 (9GHz) spectrometer with rectangular TE102 cavity and equipped with a computer controlled nitrogen gas flow thermostat (ER 411VT). ESR spectra of C-14 positional isomer of spin labelled phosphatidylglycerol in membrane fractions were recorded. Typical spectrometer settings: microwave power 10 mW, scan range 100 Gauss, modulation amplitude 1.2 Gauss, gain 2 x 105, A/D resolution 16 bit.

Attached leaves were used for *in vivo* experiments. Plants 45 days old were treated with copper for 6 and 12 days. Copper was taken up by roots from a half strength 'Hoagland III' nutrient solution containing 50 μM or 150 μM copper provided via copper sulphate.

Non modulated chlorophyll fluorescence was used to determine the photochemical efficiency of PSII in dark adapted leaves (Fv/Fm; called photochemical capacity, PHOCA; Bolhàr-Nordenkampf et al 1991). Measurements were performed with a PEA from Hansatech. Fluorescence signals generated by a modulated light source were determined by means of a modulated fluorescence measuring system (MFMS, Hansatech). This instrument was part of a set up especially designed to perform fluorescence measurements under controlled conditions.

According to the quenching of the fluorescence signal the formation of heat deactivating centres (PSII) can be related predominantly to the parameter q_{NP} (non photochemical quenching) which equals the amount of photochemically inactive PSII centres. The efficiency of linear electron transport (Φ_e, yield of non cyclic electron transport) was calculated from the fluorescence signals after about 5 min of illumination by actinic light. Recovery after 90 seconds of dark red light (720nm) was measured to study the relation between amounts of down regulated and non functional PSII centres (Bolhàr-Nordenkampf et al 1989, Bolhàr-Nordenkampf and Öquist 1993, Bolhàr-Nordenkampf et al 1994).

In leaves light induced absorbance changes around 830 nm can be used to monitor the formation and destruction of P-700$^+$ ('oxidised PSI'). The MFMS equipped with emitter - detector combination for dark red (830nm) and a backup system to record the signal served for this purpose as well (Bolhàr-Nordenkampf et al 1993).

To perform measurements for CO_2-response curves (A/Ci) and light-response curves (A/Q) setups partly developed in the Austrian laboratories were used. Portable and lab equipment (LCA II and 225-MK3 with PLC and LSC-2) was combined with gas mixing units, variable light sources and dew point bathes and dew point mirror. The amount of active RuBisCO was estimated from the carboxylation efficiency (CE) determined by measuring an A/Ci response under saturating light conditions. The dimensionless value quantum yield (Q.Y.) results from the fraction mol CO_2 fixed divided by mol photons incident on (or absorbed by) leaves. These data are gained at the steepest part of the light response curve (40-100μmol.m^{-2}.s^{-1}) where photosynthesis is limited by light. At high light intensities the Calvin cycle is optimally supplied with NADPH.H$^+$ and ATP and the activity and the amount of the enzyme RubisCO and other Calvin cycle enzymes become limiting. Light saturated photosynthetic rates (A_{sat}) are restricted by the inorganic phosphate supply into the chloroplast and they are is also sensitive to changes in the leaf conductance (Long et al 1989, Postl et al 1992 Long and Hällgren 1993).

RESULTS

Young (10 days old) and old (45 days old) spinach plants treated for nearly 14 days with copper showed in average a reduction in growth (dry matter accumulation) of 20 to 30%

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with 50 μM Cu and of 50 to 70% with 150 μM Cu respectively. Older plants seem to be less sensitive to copper poisoning by 30%. These plants accumulated remarkable amounts of copper only when exposed for 40 days to a nutrient solution containing 150 μM Cu reaching values of 124 mgCu.gDM^{-1} and of 645 mgCu.gDM^{-1} in young and old leaves.
Plants cultivated in 50 μM Cu seem to be able to exclude significantly copper molecules from uptake (young leaves 17 mgCu.gDM^{-1}, old leaves 20 mgCu.gDM^{-1}) when compared to controls (young leaves 8 mgCu.gDM^{-1}, old leaves 14 mgCu.gDM^{-1}). In nutrient solution with 150 μmol Cu added roots got a dark colour and the isoenzyme-pattern of peroxidases was changed compared to controls (data not shown).
The intensity of chlorophyll fluorescence was higher on the upper (adaxial) side of dorsiventral spinach leaves by 13% with nearly no effect on Fv/Fm (PHOCA). Leaves from controls had PHOCA values of 0.816 ± 0.002 on the upper side and of 0.820 ± 0.002 on the lower side. Even when growing in 150 μM Cu changes induced by copper were comparably small (fig.1a). On the upper side of leaves higher fluorescence signals, combined with a prolonged reduction time of the system (Tfmax +12%) resulted in lower values for PHOCA and for the efficiency of linear electron transport (Φ_e) by less than 10%. These losses were estimated to be caused by down regulated (65%) and non functional (35%) PS II reaction centres. The response described above correlated with a small increase of non photochemical (q_{NP}) and F_O quenching (q_O). In addition PS I is stronger oxidised in plants treated with 150 μM copper. This rise in the capacity of P_{700} results in a steep but rather normal correlation with Φ_e (fig. 1b). All the other data (controls, 50 μM Cu) are clustered around 0.35 (P_{700}) and 0,72 (Φ_e). In comparison with controls it could be argued that slightly more electrons from PS II would be needed to reduce P_{700} in copper treated plants.

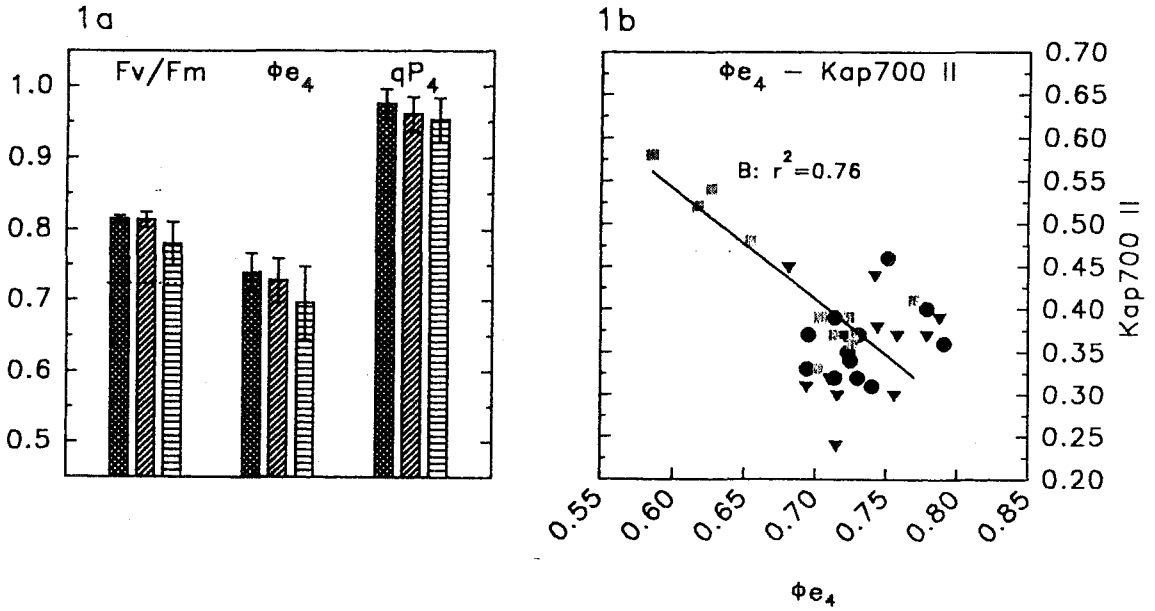


FIG1: Spinach, last fully developed leaf; modulated fluorescence and P700 absorption changes.

● Control (K) n=12
▼ Copper 50μmol (A) n=14
■ Copper 150μmol (B) n=15

Confidence interval was calculated for i=95%.

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These results were thought to be rather a consequence of a copper caused reduction in CO_2 fixation rate (Calvin cycle) than a direct effect of copper on the electron transport. It could be concluded that in this case electron transport was hindered perhaps due to the action of an increased delta-pH, which could give rise to higher F_0 and Fm fluorescence without changing Fv/Fm (PHOCA).

Apparent quantum yield (QY) and light saturated photosynthesis (Asat) start to become lower after a few days of copper treatment. The reduction of quantum yield in leaves of plants grown in 150 μM Cu was comparable with that of PHOCA and reached 14% after 11-12 days of treatment (fig. 2a), whereas Asat was lower by -22% (fig. 2b).

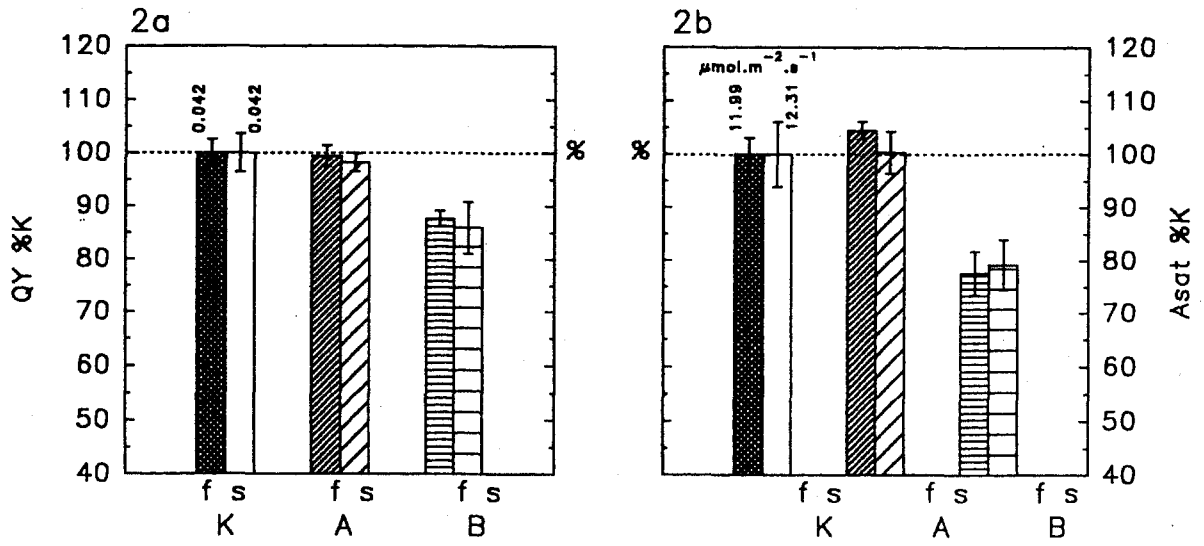


FIG2: Spinach, last fully developed leaf; quantum yield (QY) and light saturated photosynthesis (Asat) derived from light response curves.

Fig2a: apparent QY as a percentage of control
Fig2b: Asat as a percentage of control

Control (K)
Copper 50µmol (A) n=7
Copper 150µmol (B)

f after 4–6 days copper treatment; s after 11–12 days copper treatment;
Error bars give standard error.

If Calvin cycle activity is thought to be reduced by copper primarily carboxylation efficiency (CE) should be reduced strongly. Plants grown in 150 μM Cu developed a significant lower CE after 6 and 12 days with -14% and -47% respectively (fig. 3a).

Comparing the average values of reduction in QY (-14%) with that of CE (-32%) it was concluded that Calvin cycle activity could be a target for the action of copper in photosynthetic metabolism before the electron transport chain is influenced. Together with lower dark respiration rates (fig. 3b) the decrease in CO_2 fixation is thought to be the reason for reduced plant growth on copper polluted soils.

In contrast to these findings *in vitro* experiments with chloroplasts and thylakoids clearly showed a copper induced inhibition of electron transport. In copper poisoned chloroplasts variable chlorophyll fluorescence drastically decreased resulting in PHOCA values smaller than 0.5.

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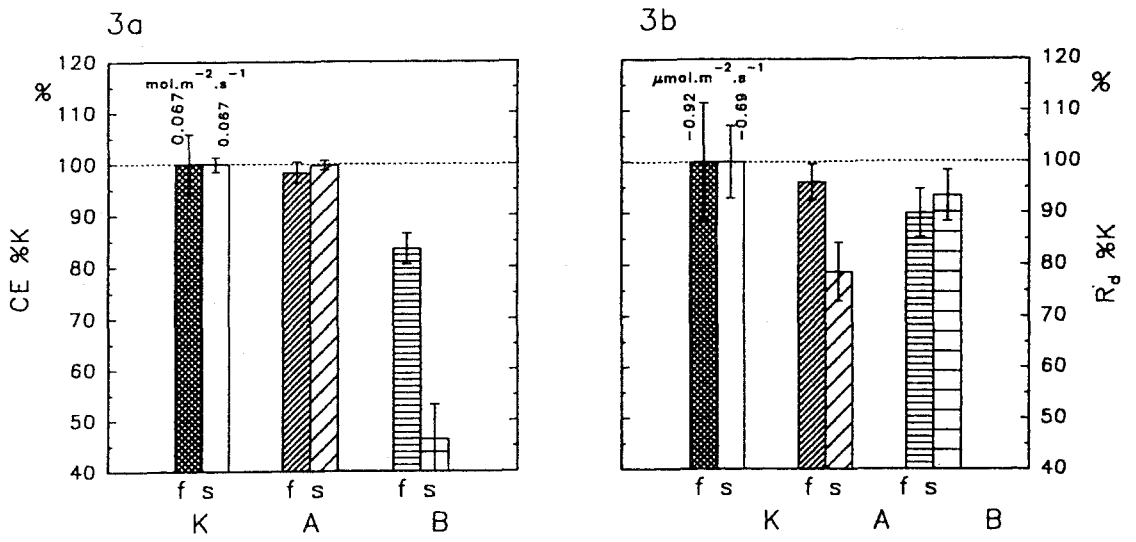
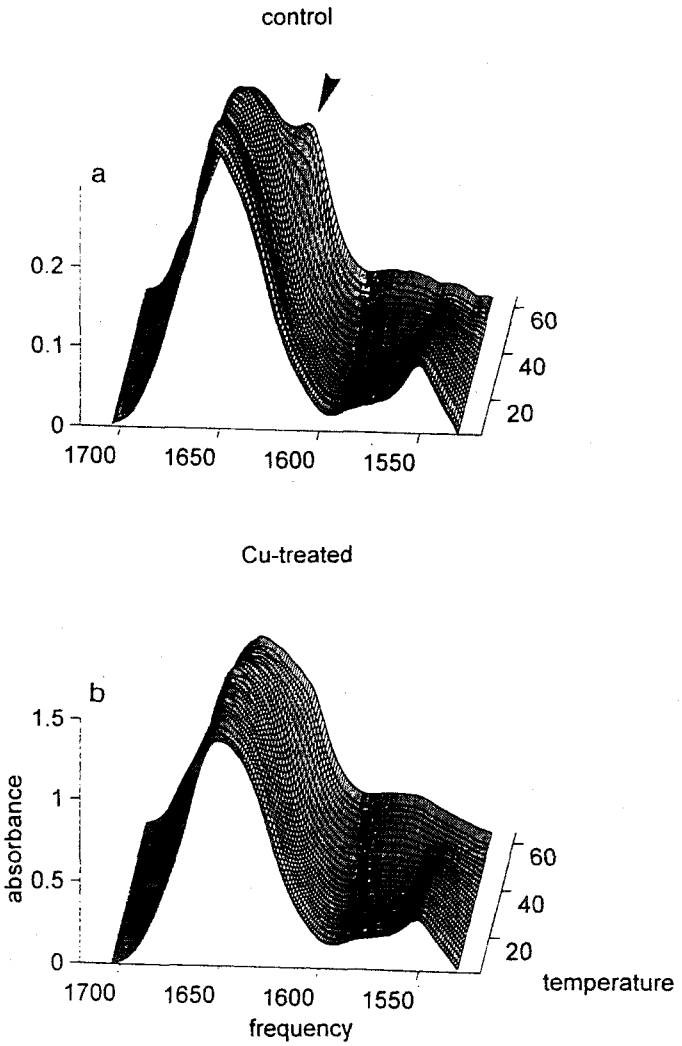


FIG3: Spinach, last fully developed leaf; carboxylation efficiency (CE) and dark respiration (R_d) derived from CO_2 response.
a: CE as a percentage of control; b: R_d as a percentage of control.

Control (K)
Copper 50µmol (A) n=6
Copper 150µmol (B)

f after 4–6 days copper treatment; s after 11–12 days copper treatment;
Error bars give confidence intervals 95% (3a) and standard error (3b).



FTIR spectra of thylakoids provided information about changes in protein conformation in the region of amid I vibrations of the constituent proteins. These small copper induced differences in the amid I region were studied by perturbing the membrane structure by increasing temperature from 5° to 65°C.
Considering ongoing tendencies in the 30 spectra recorded with increasing temperature a new band emerged around 1626 cm^{-1} during denaturation in one or more protein components of the thylakoid membrane (fig. 4a).

Figure 4: The amide regions of control and copper treated thylakoid membranes. Arrow indicates the emerging band characteristic for heat-denaturation in control thylakoids (amind I band). a) control, b) copper treated.

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The characteristic temperature of this change at 42°C correlates well with the usually observed loss of photosynthetic activity. The band in amid I region vanished after copper treatment probably as consequence of an interaction which alters the starting conditions giving rise to a denaturation following a different path (fig. 4b). These changes indicated a serious alteration in the protein organisation, which is thought to cause changes in the β-sheath content of proteins in copper treated thylakoids (van Stokkum et al 1995).

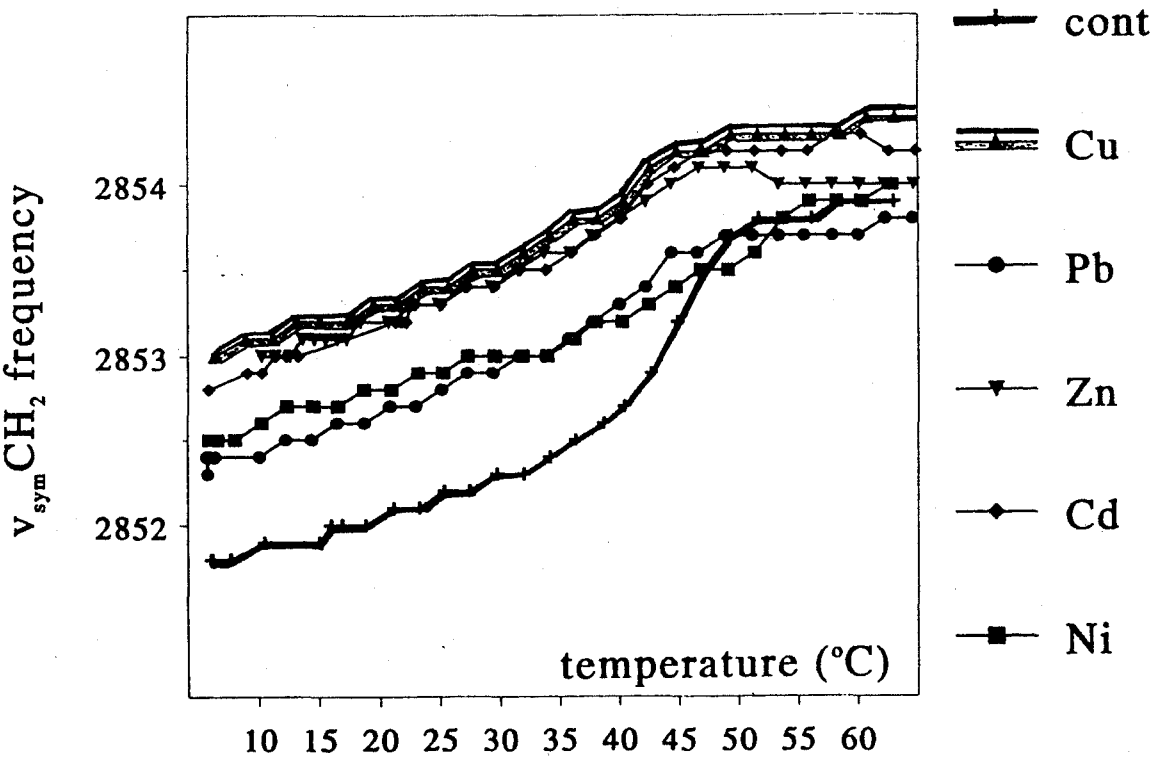


Figure 5: The shift of $\nu_{sym}CH_2$ frequencies upon changing the temperature in control and copper treated thylakoids. Each point represents a spectrum and a band fitting. The accuracy of band frequency determination was always better than 0.1 cm^{-1}.

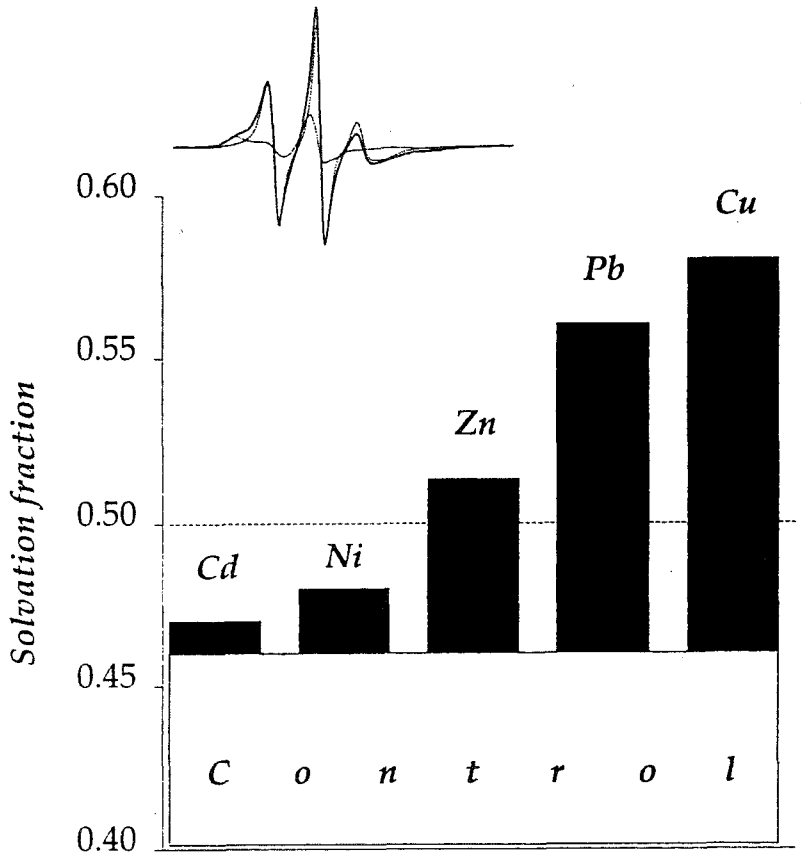
The C-H stretching region of thylakoid membranes which predominantly originates from the fatty acyl chains of membrane lipids was altered by copper as well. The band $\nu_{sym}CH_2$ (and $\nu_{sym}CH_3$) is known to reflect phase properties of membrane lipids. An increase of its frequencies can be correlated with an elevated disorder in the lipid part of biological membranes. If $\nu_{syn}CH_2$ frequencies were plotted versus temperature copper poisoned thylakoids developed a shift around 42°C which was primarily thought to be caused by an increase in lipid disorder (fig. 5). The comparison of plants grown with different temperatures and light regimes in the field (spring, summer) proved that the upshift in frequencies was strongly dependent on growing conditions (data not shown).

Therefore it was concluded that changes in the structure and behaviour of thylakoid membranes after copper poisoning were predominantly related to conformational modifications of membrane proteins (van Stokkum 1995).

Phase transitions of the lipid part of thylakoid membranes have to be seen as a consequence of the copper-protein interactions. Low mobility proteins of biomembranes carry a shell of motionally restricted lipids. This solvation layer at the hydrophobic surface of proteins can be studied by spin labelled ESR spectroscopy (Ivanich et al 1995). A complete series of ESR spectra of spin labelled phophatidylglycerol incorporated into chloroplasts following *in vitro*

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treatment with various copper concentrations was recorded. Pronounced spectral changes were obtained with 1 mM Cu(II) chloride.



Excessive amounts of copper ions led to lipid-lipid phase separation in addition to the changing solvation to fluid fraction. The spectral effects were quantified by subtracting the two composite components and in addition inter subtractions were performed between the spectrum pairs recorded prior to and after the addition of copper ions (fig.6).

Figure 6: The fraction of solvation lipid components in chloroplasts treated with various heavy metal ions as determined by spin label ESR spectroscopy in conjunction with computer-assisted difference spectroscopy. A typical two-component ESR spectrum of spin labelled phosphatidyl-glycerol is shown in the insert.

Clearly the solvation fractions after copper increased almost by 50% as compared to the control. Results gained *in vitro* indicated that under copper stress conditions the inhibition of photosynthetic electron transport is related to the stress induced alterations of the protein conformation which causes modifications of the lipid-protein interaction within the chloroplast thylakoid as well.

DISCUSSION

Findings from *in vitro* and *in vivo* copper stress experiments proved on one hand the inhibition of electron transport as a consequence of conformational changes of membrane proteins, on the other hand especially older plants developed the ability to exclude copper from uptake by roots. In addition the incorporation of copper molecules in chloroplasts of mesophyll cells was thought to be controlled by the capacity to store copper via complexation in the vacuole. *In vivo* gained results led to the conclusion that copper ions were able to disturb CO_2 uptake by interaction with the plasmalemma and / or by reducing the

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carboxylation efficiency of RuBisCO. In both cases copper stress would give rise to a reduction in Calvin cycle activity, which was observed to be the first reaction after copper treatment *in vivo*. Enhanced dark respiration could be caused by stress compensation mechanisms such as repair and complexation of copper ions.

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CORRESPONDENCE

Prof. Dr. Harald R. Bolhàr-Nordenkampf, Division of Horticultural Plant Physiology, Institute of Plant, Physiology, Biocentre, University of Vienna, Althanstrasse 14,
POB 285, A-1091 Vienna, Austria.
Fax --43-1-31-33-6-799; e-mail: bolhar@pflaphy.pph.univie.ac.at

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PERSISTENT CHLORORGANIC COMPOUNDS IN THE ECOSYSTEM OF THE BALTIC SEA (1976-1995)

Ott Roots

Ministry of the Environment of Estonia, Environment Information Centre,
Mustamäe tee 33, EE0006 Tallinn, Estonia

Abstract. The aim of this article is to use polychlorinated biphenyls (PCB) and chlororganic pesticides (COP) as tracers for nonpolar organic contaminants cycling in the Baltic Sea. If we compare the chlororganic contents of herring in different areas of the Baltic Sea, we can state, that when it comes to the distribution of chlororganic compounds, the northern part of the Riga Bay is the reference area for the Baltic Sea.

The sum up we can say, that such levels of organochlorines in the organism of the Baltic herring in the coastal area of Estonia would not constitute any human health risk.
Key words:Baltic herring, PCB, pesticides, Baltic Sea, Health risk.

INTRODUCTION

Environmentally hazardous substances such as PCB and COP, are acutely toxic, persistent and bioaccumulative (i. e. they become concentrated in food chains to reach toxic levels).

Following the decision of the Baltic Marine Environment Protection Commission, a systematic monitoring of harmful substances , in selected species of the Baltic Sea biota was carried out in Estonia (Roots 1992; 1994; Roots et. al., 1993).

While studing PCB compounds, one has to consider both the partition coefficient (between 1-octanol and water) and stereochemistry of compounds. Separate PCB congeners determination is necessary for a number of reasons:

- congeners differ in their toxic properties
- their half-life time in organisms can differ considerably.

The methods described in this article have been utilized for analysing a number of biological environmental samples (Jensen et al., 1983; Bergman et. al., 1992; Roots et al., 1993).

The joint FAO/WHO Expert Commitee on Food Aditores (JECFA) generally sets the ADI (the acceptable daily intake) of a food additive on the basis of the highest "no-observed-effect" (NOEL) level in animal studies. ADI and NOEL values used in our work are based upon the following literature data (FAO/WHO; 1971; 1978; 1985; Reports and Studies, 1991).

RESULTS AND DISCUSSION

Risk assessment integrates hazard identification, exposure assessment and dose-response assessment.

For each level of seafood consumption, the carcinogenic risk from marine pollutants will vary according to the concentration present, which will vary from place to place and from time to

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time. As the same time it is known, that marine foodstuffs are not the only route (Table 2) of exposure of the human population and any overall risk assessment must take this into account (Roots, 1986).

The cancer incidence in Estonia is below the European average. Estonia ranked 47th for male and 51st for female.

However, the incidence rates of stomach cancer and lymphocyclic leukemia in both sexes and cancer of the pancreas in males were higher in Estonia than those in other European countries on average. According to the pattern of these cancer sites, Estonia ranked 4-6th in Europe and to the top 20 when 166 world populations were compared. The incidence rates of cancer of the thyroid and tests in males, non-Hodgkin's lymphoma in females and breat and bladder in both sexes in Estonia were low compared with other European countries. Estonian site distribution of cancer is similar to Latvia and several other Eastern European countries (Thomson, 1994).

When selecting a catchment area for fish as the Estonian coastal zone, we proceeded from 2 principles:

- the part of fish in the food of the people living in the region is relatively big
- the air pollution from local sources inside Estonian boudaries is small (Table 2).

Appropriate regions would be the northern part of Riga Bay (Tables 1a and 1b) and the western part of the Gulf of Finland (Roots et al., 1993).

The following questions will need a proper response:

- how big is the amount of fish consumed in a day, that we could take as a base for our calculations?
- what species of fish will we select to study?
- the effect of which chlororganic compounds will we observe?
- from which standards we will proceed, when evaluating the danger of toxicants?

When considering the possible effects of consuming seafood, it necessary to characterize communities and individuals according to the amounts they consume, although there are likely to be large differences between communities in different parts of the world in their consumption of seafood and between those, who eat fish or shellfish. For example, based on FAO food balance data, the European reginal average consemption of seafood (fish and shellfish) is 60 g/person/day and in East Asia 79 g/person/day. 150 g/day is the figure used to represent high consumption (Reports and Studies, 1991).

The species chosen as test organisms and the sampling procedures recommended for monitoring harmful substances in biota are intended to provide a picture of the levels of harmful substances in the studied organisms and to determine trends in their levels over time. HELCOM has selected herring and cod as study organisms in the Baltic Monitoring Program (BMP), because:

- they can be caught in all parts of the Baltic (this is important for comparison)
- they are easy to collect
- they are of suitable size for preanalitical sample treatment
- their biology is fairly known
- they are in important commercial species.

The monitoring program is based on measurements of obligatory (mandatory) and tentitative determinants (Haarich, 1994). We selected Baltic herring and toxicants presented in (Haarich, 1994) to serve as a base for our studies. In 1994, 45416 tons of fish were caught from the Baltic Sea by Estonian fishermen; from that amount 34494 tons were herring and 9079 tons were sprat. In connection with the decrease of salinity in the northern part of the Baltic Sea, the catching of cod has noticeably decreased, consisting of 227 tons, that is 0,7% from the overall amount of caught fish (Estonian Environment 1994, 1995).

Trends of environmental contaminants have to be studied in defined areas (same place

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every year).

The important role is played by the time and place of catching, i. e. population location, when different regions of catching are compared (Roots et al., 1978; Roots, 1992).

The period in late summer selected for herring is relatively stable for the organisms in terms of migration and physiology of these species. The prespawning age classes of herring seem to be local or regional species at young age (Parmanne, 1990; Bignert et al., 1993). In all probability, samples of younger fish originate from the same stock and are thus representatives of the local environment (Roots et al., 1990; 1994; Bignert et al., 1993; Haarich, 1994). (Figure 1).

The average bodyweight of herring, which had been relatively stable during the years 1940-1960, started to increase in most of the regions of the Baltic Sea in the late 1970ies. (Figure 2). High average value of bodyweight and - length was observed in all age groups of herring until the second half of the 1980ies, when it started to fall unexpectedly. In the 1990ies, one can also observe the drop concerning the average bodyweight of sprat. The studies concerning the feeding of herring and sprat, that were carried out during the years 1982-1992 in the north-eastern part of the Baltic Sea showed changes in the diet of the fish and also the rising number of fish with an emptly stomach (Figure 3). These changes are probably connected with the hydrological conditions prevailing in the Baltic Sea from the mid - 1980ies (Lumberg, Ojaveer, 1992). These conditions are at least partially responsible for the rapid deceleration in the growth of sprat and (specially) herring (Lankov, 1992; Raid et al., 1989; 1995).

Along side of herrings' age, length, weight, sex, fat percentage and degree of maturity, we recommend to bring forth the percentage of empty stomachs (and the content of different food in stomachs) as an additional parameter. One can only assume, that the rising number of fish with an empty stomach has contributed to the stability of chlororganic compound contents in the northen part of the Baltic Sea. (Figures 1-3).

Our herring data from the Gulf of Finland (Roots, Aps, 1993) and from Riga Bay (Tables 1a and 1b) in 1991-1992 showed lower DDT levels than in the samples collected by Haahti and Perttilä in 1986 (Haahti et al., 1988). By the data of Haahti and Perttilä (1988) in 1986, the summary DDT concentrations in the herring muscle ranged from 0,3 mg/kg in the Bothnian Bay to 0,7 mg/kg in the Gulf of Finland (in extractable fat). The mean summary DDT concentration in the muscle tissue of the Riga Bay herring was 0,28 mg/kg in extractable fat.

Also HCB and HCH isomers have been analysed with the monitoring substances. If we compare the present HCH values (Table 1a) with the ones analysed by Haahti and Perttilä in 1986 (Haahti et al., 1988) and by Blomkvist et al., 1992 (Blomkvist et al., 1993). We can see that they are all at the same level.

The present results (Blomkvist et al., 1993) show that the area studied in Estonia has a very small influence on local contamination. Samples of perch were collected during August and September, 1992 at three different localities along the coast of the Baltic Republcs. The localities were south of Hiiumaa in Estonia, Daugavgriva (the delta of River Daugava) in Latvia and Kuronian Bay in Lithuania.

Corresponding concentrations in perch from the Swedish reference area, Kvädöfjärden, in the Baltic proper west of Gotland were 0,53 (sDDT) and 2,08 (sPCB) ppm respectively. It is obvious that the Kuronian Bay results indicate a strong eutrophication since the relative amount of DDD in fish. There is a similar tendency, although not as strong, in the Daugavgriva material.

The PCB pesticides analyses made by the Lund University on the samples taken in the three Baltic air-research stations in Vilsandi and Lahemaa in Estonia and near Riga in Salaspils in Latvia showed that the air and rain water samples taken in Estonian stations were relatively cleaner compared with the samples taken in Latvia (Larsson et. al., 1991). This refers either to the long-range transportation or to the local waste centre situated near the Gulf of Riga.

Air and precipitation concentrations at the Latvian Salaspils station (delta of River

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Daugava) were 7,6 and 8,7 times and at the Lithuaninan (Curonian Lagoon) station 1,3 and 2,6 times higher, respectively in 1991-1992 than the same concentrations at the Estonian Lahemaa station at the same time (Larsson et al., 1993; Roots, 1993). The same situation could be seen when Blomkvist et al., (1993) analysed the fish. Although the contaminant analyses were made only on samples from one year and should be verified by additional studies, the results suggest that the most important risk substances, PCBs and DDTs, occur in comparatively low concentrations, also in areas receiving waste from such large city as Riga (Blomkvist et al., 1993). The results from Hiiumaa (the northern part of the Riga Bay) are very promising and strongly support the opinion that this is a high class reference area.

The joint FAO/WHO Expert Commitee on Food Additives (JECFA) generally sets the ADI (the acceptable daily intake) of a food additive on the basis of the highest "no-observed-effect" (NOEL) level in animal studies.

ADI - is the daily dosage of a chemical which, during an entire lifetime, appears to be with out appreciable risk on the basis of all the facts known at the time. In cases where JECFA considered that a provisional tolerable weekly intake (PTWI) was a more appropriate concept than an ADI, PTWI has been considered in the context of exposure to a particular material from seafood. Where an ADI has been established for a particular pollutant it will be noted whether the intake from seafood is likely to be a substantial part of the ADI (Reports and Studies, 1991).

NOEL - the risk that may be incurred by especially susceptible groups, such as children and old people.

ADI and NOEL values used in our work are based upon the following literature data (FAO/WHO, 1971; 1978; 1985; Reports and Studies, 1991).

DDT. DDT and its metabolites are some of the most freguently reported organochlorine contaminants in the environment. An overall no-effect level (NOEL) for toxicity of DDT of 0,25 mg/kg/bodyweight/day in humans and an ecceptable daily intake (ADI) of 0,02 mg/kg bodyweight was set by the Joint Meeting on Pesticide Residues (JMPR) in 1984 (FAO/WHO; 1985; Reports and Studies, 1991).

If a 60 kg person would consume 150 g/day of fish containing 20 µg/kg of DDT and its metabolities, the daily intake would be 0,05 µg/kg bodyweight. This would constitute 1/400 of the ADI.

HCH - Hexachlorocyclohexane. Technical HCH containe a number of isomers of which gamma-HCH (Lindane) is commonly used as an insecticide. In the Baltic Sea the concentrations of all three isomers (alpha, beta and gamma) are similar (Roots et al., 1993).

If a 60 kg person would eat 150 g/day of fish containing 60 µg/kg (alpha-, beta- and gamma-HCH), this would constitute of 1/53 of the ADI (for gamma-HCH of 0,008 mg/kg bodyweight was set by the JMPR). No ADI was established for the other isomers or for technical HCH.

Aldrin+Dieldrin. This is not a problem for the Baltic Sea. If a 60 kg person would consume 150 g/day of fish containing 0,5 µg/kg (max) of aldrin plus dieldrin, the daily intake would constitute of 1/80 of the ADI (0,0001 mg/kg bodyweight).

Chlordane. The main components are alpha- and gamma-chlordane and the most toxicologically significant metabolite is oxy-chlordane.

If a 60 kg person would eat 150 g/day of fish containing 4 µg/kg gamma-chlordane plus oxy-chlordane (Roots et al., 1993), this would constitute of 1/50 of the ADI (0,0005 mg/kg bodyweight).

PCB - Polychlorinated biphenyls. Human health studies of "Yusho"disease in Japan (1968) and in Taivan (1979) have shown, that contamination of food with PCB form an industrial source can be a cause of severe human diseases. JECFA found it is possible to establish, for PCB's, a tolerable intake value for humans.

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If a 60 kg person would eat 150 g/day of fish containing 20 µg/kg PCB, this would amount to 0,05 µg/kg bodyweight per day. This is about 1/3000 of the estimated average daily intake in the accident in Japan in 1968 (Reports and Studies, 1991), which involved about 1000 people, and 1/800 of the no-effect level of 0,04 mg/kg bodyweight per day for monkeys.

To sum up we can say, that such levels of organochlorines in the organism of the Baltic herring in the coastal area of Estonia would not constitute any human health risk.

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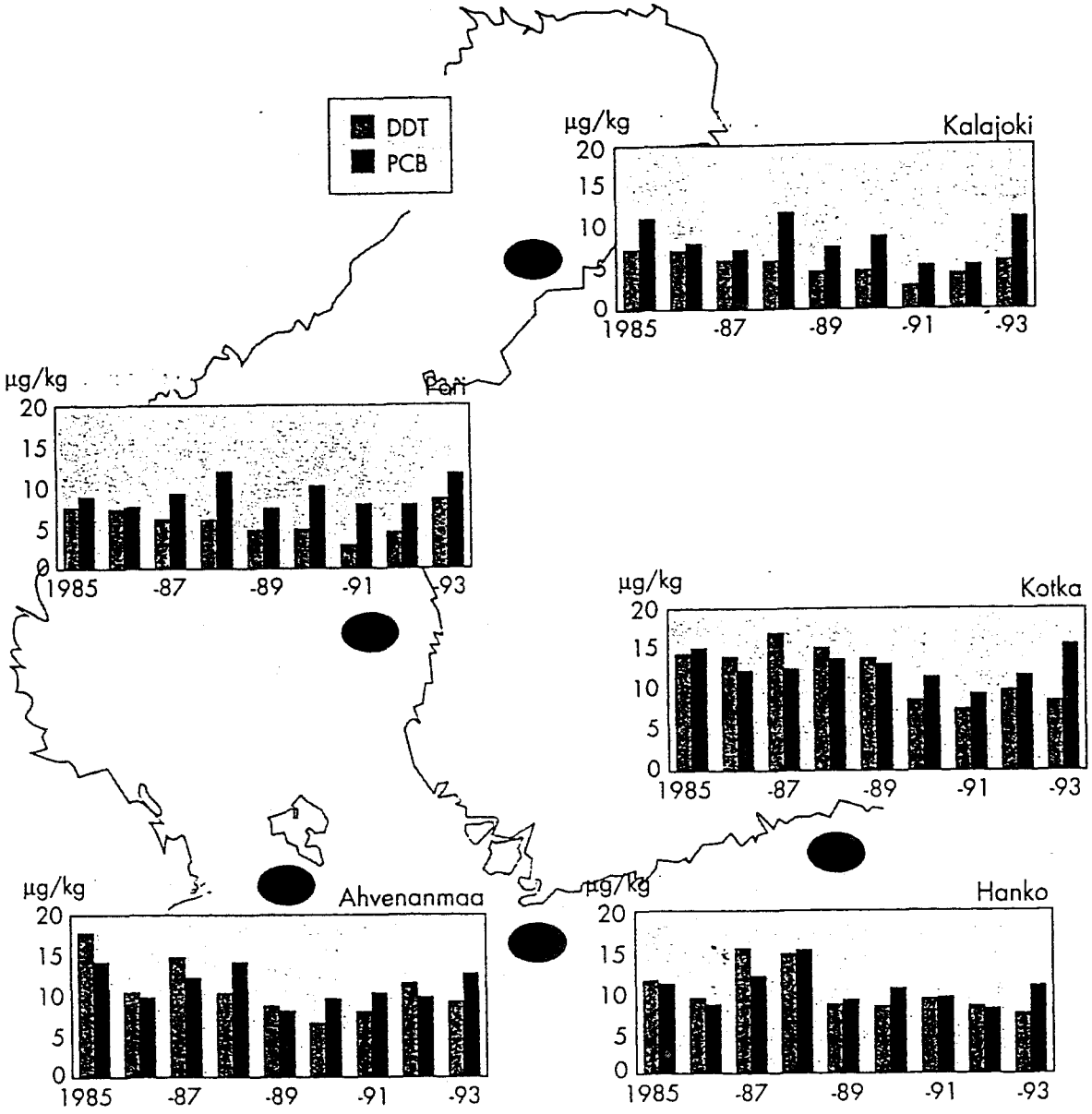


Figure 1. Concentrations of summary DDT and PCB in the muscle tissue of herring (µg/kg wet weight) 1985-1993 (Haahti, 1994).

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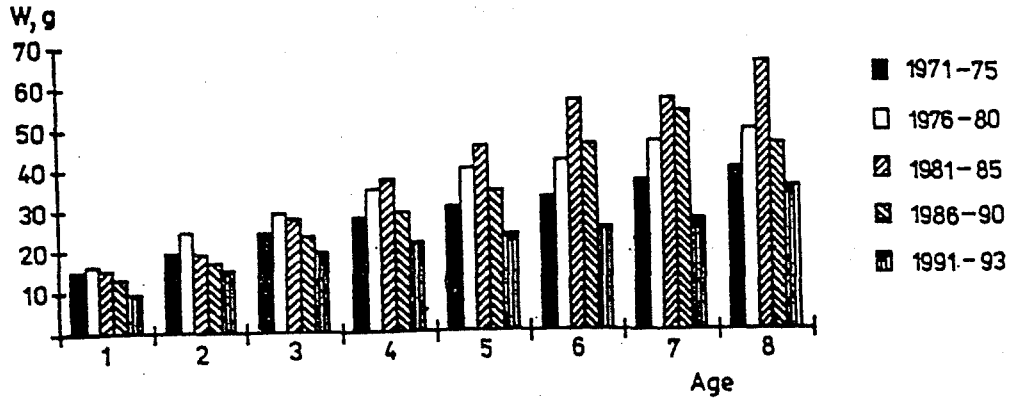


Figure 2. Mean weight at age of herring in the Gulf of Finland in 1971-1993 (Raid et al., 1995).

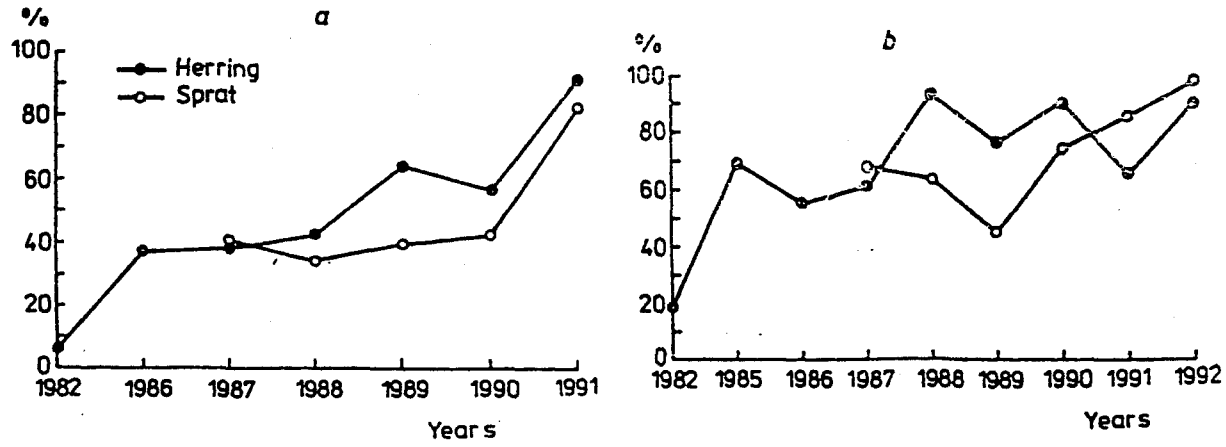


Figure 3. Proportion of herring and sprat with empty stomachs in the northern Baltic in April 1982-1991 (a) and in November-Detsember 1982-1991 (b) (Raid et al., 1995).

Table 1a. Concentrations (ng/g fat) of chlororganic compounds in the muscle tissue of the Baltic herring (Gulf of Riga)

Nº	Length, cm		Weight, g	Age	Sex		Matu-rity	Fat %	Concentration ng/g (fat)									
	L	L_s			♂	♀			α-HCH	HCB	β-HCH	γ-HCH	δ-HCH	Aldrin	p,p′DDE	p,p′DDD	p,p′DDT	sum.DDT
1.	12.3	10.3	10.3	2	+		III	3.8	16.3	17.7	10.0	6.7			99.3	81.4	10.6	191.3
2.	13.6	12.2	12.3	2		+	IV	2.3	2.0	42.0		0.8			155.2	189.8	46.5	391.5
3.	12.4	11.3	10.0	2		+	IV	2.6							32.0	58.8	24.4	115.2
4.	13.0	11.4	12.2	2		+	IV	3.9	4.0	15.5	29.9	8.1	2.6		39.8	216.6	73.3	329.7
5.	12.2	10.8	9.1	2	+		III	3.2		15.4		4.3			195.0	126.1	64.6	385.7
6.	13.6	12.3	12.8	2		+	III	0.8	1.8			9.1			170.0		48.7	218.7
7.	11.7	10.4	9.0	2	+		III	0.9	2.9	8.9		19.9			363.0		141.2	504.2
8.	13.3	11.0	12.5	2	+		IV	1.7	19.4				1.2		168.8		21.7	190.5
9.	12.1	10.7	9.6	2		+	IV	1.6	6.3	6.2		11.2			77.9		24.4	102.3
10.	12.1	10.7	9.4	2		+	IV	4.2	0.6	6.4		3.0			81.6		25.8	107.4
11.	13.4	12.2	12.2	2	+		III	1.2	9.3	23.7	9.9	11.4		2.5	186.7		50.6	237.3
12.	12.0	10.6	9.4	2	+		IV	1.3	4.0	9.9	10.6	6.4			177.3	7.8	80.1	265.2
13.	11.6	10.3	8.1	2		+	III	3.5	4.9	12.1		6.5			166.4	21.5	61.8	249.7
14.	12.6	11.3	11.1	2		+	IV	1.2	1.3	19.4		2.9		1.1	235.7		58.4	294.1
15.	12.9	11.4	12.0	2		+	III	2.2							113.9		44.0	157.9
16.	12.3	11.0	10.3	2	+		III	3.8	26.8	11.9	7.8	26.1		2.5	88.0	110.5	47.2	245.7
17.	13.6	12.2	12.3	2		+	IV	2.3	8.2	34.7		10.8			302.5	169.9	112.4	584.8
18.	12.4	11.0	10.0	2		+	IV	2.6	14.3	36.5		8.2			183.2	62.1	45.4	290.7
19.	13.0	11.4	12.2	2		+	IV	3.9	21.5	13.6	13.8	25.9	8.4		168.3	200.3	48.8	417.4

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Table 1b. Concentrations (ng/g fat) of chlororganic compounds in the muscle tissue of the Baltic herring (Gulf of Riga)

Nº	Length, cm		Weight, g	Age	Sex		Maturity	Fat %	concentration PCB ng/g (fat)							
	L	L_s			♂	♀			49	52	101	118	138	153	180	105
1.	12.3	10.3	10.3	2	+		III	3.8			26.0	53.0	27.9	49.7	13.2	12.8
2.	13.6	12.2	12.3	2		+	IV	2.3	31.7	67.6	162.5	390.6	119.7	226.6	101.9	64.2
3.	12.4	11.8	10.0	2		+	IV	2.6	19.1	45.9	46.9		68.0	134.0	26.4	17.2
4.	13.0	11.4	12.2	2		+	IV	3.9	7.5	18.9	35.0	83.2	61.5	102.7	12.1	18.1
5.	12.2	10.8	9.1	2	+		III	3.2	10.2	34.6	43.5	96.3	63.0	108.6	18.1	20.3
6.	13.6	12.3	12.8	2		+	III	0.8			31.8	295.8	236.9	421.7	135.2	73.3
7.	11.7	10.4	9.0	2	+		III	0.9			145.5	170.4	183.1	363.7	99.9	23.1
8.	13.3	11.0	12.5	2	+		IV	1.7	52.1		87.2	130.7	107.7	215.8	58.9	7.2
9.	12.1	10.7	9.6	2		+	IV	1.6	62.0		70.7	100.4	66.1	118.3		7.3
10.	12.1	10.7	9.4	2		+	IV	4.2	15.5		35.4	51.9	62.4	87.8	99.8	3.9
11.	13.4	12.2	12.2	2	+		III	1.2	76.9		99.7	118.0	87.1	165.3	5.2	5.9
12.	12.0	10.6	9.4	2	+		IV	1.3	54.9		74.2	103.3	77.0	149.4	37.6	
13.	11.6	10.3	8.1	2		+	III	3.5	9.7		64.1	108.2	69.8	125.9	21.9	6.2
14.	12.6	11.3	11.1	2		+	IV	1.2	28.9		139.0	153.4	159.1	256.7	240.4	14.5
15.	12.9	11.4	12.0	2		+	III	2.2	15.3		34.4	73.0	79.6	122.0	101.1	6.1
16.	12.3	11.0	10.3	2	+		III	3.8		9.4	30.5	57.7	40.4	46.6	7.0	
17.	13.6	12.2	12.3	2		+	IV	2.3	27.6	55.4	159.9	204.5	227.0	269.7	70.6	
18.	12.4	11.0	10.0	2		+	IV	2.6	34.4	19.1	59.4	97.1	83.9	112.3	19.4	
19.	13.0	11.4	12.2	2		+	IV	3.9	17.9	19.0	47.3	92.6	75.7	83.3	10.0	

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Table 2. Levels of chlororganic compounds in the air (analysed 'by Lund University Exotoxicological laboratory)

Domain No. *	Chloro-biphenyls No. (IUPAC)	Concentr. in the air (ng/m3)x10-2 Lahemaa	Vilsandi
2	7,9	0.063	
3	6	0.082	
6	15,17,18	0.465	0.257
7	24,27	0.105	0.066
8	16,32	0.212	0.023
11	26	0.035	0.034
12	25	0.178	0.102
13	28,31	0.382	0.303
14	20,33,53	0.285	0.197
15	22,51	0.080	0.039
16	45	0.150	0.148
19	52	0.531	0.334
20	49	0.276	0.172
21	47,48,75	0.275	1.655
22	35		
23	44	0.358	0.258
24	37,42,59	0.171	0.094
25	41,64	0.346	0.195
27	40	0.213	
29	63	0.154	0.333
30	74	0.137	0.112
31	70	0.231	0.189
32	66,95	0.472	0.500
36	92	0.233	0.256
37	84	0.220	0.345
38	90,101	0.325	0.267
39	99	0.226	0.041
42	97	0.123	0.099
43	87,115	0.095	0.126
46	77,110	0.331	0.417
47	82,151	0.098	0.174
50	118,149,123	0.162	0.158
53	146	0.023	0.124
54	105,132,153	0.168	0.214
55	141,179	0.037	0.110
58	138,158,160	0.225	0.324
61	187	0.039	
62	183	0.021	
63	128	0.039	
66	174	0.034	0.056
67	177	0.028	0.084
68	156,171,202	0.029	
72	180	0.029	
75	200		0.706
77	170,190	0.027	
79	199	0.064	0.042
80	196,203		
84	194	0.017	0.049
	Chlororganic pesticides		
	α-HCH	1.288	1.281
	HCB	0.553	0.580
	γ-HCH	0.211	0.076
	p,p'DDE	0.111	0.562
	p,p'DDT	0.063	0.594

Lahemaa 25.02. - 09.03.1992
Vilsandi 28.03. - 11.04.1992
* (Shultz et al.,1989)

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LIST OF PARTICIPANTS

Dr. Mike R. Ashmore
Imperial College of Science, Technology and Medicine
Centre for Environmental Technology
prince's Gardens
SW7 2PE 48 London
U.K.
& +44 171 5949291
fax: +44 171 5810225

Dr. Jesper Bak
Ministry of the Environment
Department of Terrestrial Ecology
P.O. Box 314
Vejsovej 25
DK-8600 Silkeborg
Denmark
& +45-89 20 14 00
fax: +45 89 20 14 14

DI Rolf Becker
ÖNU-GmbH
Münchebergerstr. 1
D-15345 Prädikow
Germany
& +49 33 436281
fax: +49 33 436281

Univ.Prof.Dr. Harald R. Bolhár-Nordenkampf
Institut für Pflanzenphysiologie
Althanstraße 14
A-1091 Wien
Austria
& +43 1 31336 362
fax: +43 1 31336 799 (776)

Dr. Maurizio Badiani
D.A.B.A.C. - Universitä di Viterbo
Via S.C. de Lellis
I-01100 Viterbo
Italy
& +39-761-357368
fax: +39 761 357242

Prof.Dr. Franc Batic
University Ljubljana
Agronomy Department, Biotechnical Faculty
Jamnikarjeva 101
61000 Ljubljana
Slovenia
& 386 61 123 11 61
fax: 386 61 123 10 88

Dr. J. Benton
Nottingham Trent University, UK
Dep. of Life Science
Clifton Lane
NG11 8NS Nottingham
U.K.
& +44 115 9486683
fax: +44 115 9486636

Dr. Mike Brown
Institute of Terrestrial Ecology
Monks Wood Abbots Ripton
PE 17 2LS Huntingdon
U.K.
& +44 1487 773381
fax: +44 1487 778467

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Dr. Keith Bull
ITE Monks Wood
Abbots Ripton
PE17 2LS Huntingdon
U.K.
& +44 148 777381
fax: +44 148 777467

Dr. Radovan Chrast
ECE / UN
Palais de Nations
CH-1211 Genève
Switzerland
& +4122 917 2358
fax: +4122 907 0107

Dr. G.P.J. Draaijers
RIVM
Air Research Laboratory
Antonie van Leeuwenhoklaan 9
NL 3720 BA Bilthoven
The Netherlands
& +31 30 2742824
fax: +31 30 2287531

Dr. Lisa Emberson
Imperial College of Science, Technology and Medicine
Centre for Environmental Technology
prince´s Gardens
SW7 2PE 48 London
U.K.
& +44 171 5949283
fax: +44 171 5810245
e-mail: l.emberson@ic.ac.uk

Dr. Andreas Fangmeier
Institut für Pflanzenökologie
Heinrich-Buff-Ring 38
D-35392 Gießen
Germany
& +49 641 702 5865
fax: +49 641 702 5866

Dr. Susanne Flachner
Ministry for Environment and Reg. Policy
Fö u. 44-50
H-1011 Budapest
Hungary
fax: +361 201 2361

PD Dr. Jürg Fuhrer
FAC
Schwarzenburgstraße 155
CH-3097 Liebefeld-Bern
Schweiz
& +41 31 323 8371
fax: +41 31 323 8415

Dr. I. Fumagalli
ENEL CRAM
via Rubattino, 54
I-20134 Milano
Italy
& +39 27224 3921
fax: +39 27224 3915

Dr. Thomas Gauger
Univ. Stuttgart
Inst. f. Navigation
Keplerstr. 11
D-70174 Stuttgart
Germany
& +49 711 1212166
fax: +49 711 1213740

Dr. Ludger Grünhage
Institut für Pflanzenökologie
Universität Giessen
Heinr.-Buff-Ring 38
D-35392 Gießen
Germany
& +49 641 7025865
fax: +49 641 7025866

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Dr. Karl-Heinz Häberle
LMU München
Lehrstuhl für Forstbotanik
Hohenbachernstraße 22
D-85354 Freising
Germany
& +49 8161 714795 od. (714948)
fax: +49 8161 714576

Dr. J. Peter Hall
Canadian Forest Service
St. Joseph Blud. Hull
K1A 165 351 Quebec
Canada
& +1 819 9971107
fax: +1 819 9943389

Dr. Chris Heyes
IIASA Laxenburg
Schloßplatz
A-2361 Laxenburg
Austria
& +43 2236 807 417
fax: +43 2236 71313

Dr. Sylvia Hofer
Universität für Bodenkultur
Inst. für Chemie
Gregor Mendel Straße 33
A-1180 Wien
Austria
& 47 654 6502
fax: 47 654 6542

Dr. Helmut Hojesky
Bundesministerium für Umwelt
Abteilung I/9
Radetzkystraße 2
A-1030 Wien
Austria
& 43 1 71158 4889
fax: 43 1 71158 4245

Dr. John Innes
Eidgenössische Forschungsanstalt für Wald,
Schnee und Landschaft
Zürcherstraße 111
CH-8903 Birmensdorf
Switzerland
& +41 1 7392216
fax: +41 1 739 2215

Dr. Hans-Jürgen Jäger
Institut für Pflanzenökologie der Justus-Liebig
Universität Gießen
Heir.-Buff-Ring 38
D-35392 Gießen
Germany
& +49 641 702 5865
fax: +49 641 702 5866

Dr. Matti Johansson
Water and Environment Research Institute
P.O. Box 250
FIN-00101 Helsinki
Finland
& +358 0 4028 347
fax: +358 0 4028 338

Dr. Markus Knoflacher
Austria Research Centre Seibersdorf
Dept. Environmental Planning
A-2444 Seibersdorf
Austria
& +43 2254 3874
fax: +43 2254 3888

Dr. Renate Köble
Univ. Stuttgart
Inst. f. Navigation
Keplerstr. 11
D-70174 Stuttgart
Germany
& +49 711 1213742
fax: +49 711 1213740

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Dr. Barbara Köllner
Landesumweltamt Nordrhein-Westfahlen
Postfach 102363
D-45023 Essen
Germany
& +49 201 79950
fax: +49 201 2995574

Dr. Erik Leeuwen van
RIVM
Air Research Laboratory
BA Antonie van Leeuwenhoeklaan 9
NL 3720 Bilthoven
The Netherlands
& +31 30 2743421
fax: +31 30 2287531

Dr. Allan H. Legge
Biosphere Solutions
1601 - 11th Avenue N.W.
T2N 1H1 Calgary, Alberta
Canada
& +403 282 4479
fax: +403 282 4479

Dr. Wolfgang Loibl
Austria Research Centre Seibersdorf
Dept. Environmental Planning
A-2444 Seibersdorf
Austria
& +43 2254 3875
fax: +43 2254 3888
e-mail: loibl@zdfzs.arcs.ac.at

Dr. Gun Lövblad
IVL
P.O. Box 47086
S-40258 Göteborg
Sweden
& +46 31 46 00 80
fax: +46 31 48 21 80

Dr. Iraida Lyulko
Latvian Hydrometeorological Agency
165 Maskavas Str.
LV-1019 Riga
Latvia
& + 371 7112 046
fax: +371 7145 154

Dr. Agnes Martos-Bartha
Ministry for Environment and Reg. Policy
Fö u. 44-50
H-1011 Budapest
Hungary
fax: +361 201 2361

Dr. Wojciech Mill
Inst. for Ecology of Industrial Areas
Kosslitha 6
PL-40-833 Katowice
Poland
& +48 3 1546031
fax: +48 3 1541717

Dr. Christian Nellemann
Norwegian Inst. of Land Inventory
Drobakveien 11
N-1430 As
Norway
& +47 64 949700
fax: +47 64 949786

Dr. Katinka Ojanperä
Agricultural Research Centre
FIN-31 600 Jokioinen
Finland
& +358 16 4188390
fax: +358 16 4188396

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Dr. László Pásztor
Herman u 15
H-1022 Budapest
Hungary
& +36 1 1558839
fax: +36 1 1558839

DI. Jaroslav Pekárek
Czech Hydrometeorological Institute
Observatory Kosetice
CZ-39422 Kosetice
Czech Republic
& + 42 365 98183
fax: +42 365 98015

Dr. Hakan Pleijel
IVL
P.O. Box 47086
S-40258 Göteborg
Sweden
& +46 31 460080
fax: +46 31 482180

Dr. Maximilian Posch
RIVM / CCE
P.O. Box 1
NL-3720 BA Bilthoven
The Netherlands
& +31 30 743529
fax: +31 30 250740

Dr. Ernst Walter Reiche
Projektzentrum Ökosystemforschung Kiel
Schauenburger Straße 112
D-24118 Kiel
Germany
& +49 431 880 4081
fax: +49 431 880 4083
e-mail: ernst@oekosys.uni-kiel.d400.de

Dr. Thomas Reichenauer
Austrian Research Centre Seibersdorf
Dept. Agricultural Research
A-2444 Seibersdorf
Austria
& +43 2254 3542
fax: +43 2254 740 60
e-mail: reichenauer@zdfzs.arcs.ac.at

Dr. Cuetka Ribarik Lasnik
Institut za Ekoloske Raziskave Erico Velenje
Koroska 64
63320 Velenje
Slovenia

Ph. Dr. Ott Roots
Ministry of Environment of Estonia
Environmental Information Centre
Kotua Str. 2
EE-0013 Tallinn
Estonia
& 372 2 55 22 41
fax: 372 2 55 22 41

Dr. Leo Saare
Estonian Environment Information Centre
Mustamäe Tee 33
EE OOO6 Tallinn
Estonia
& +372 639 4151
fax: +372 639 4071

Dr. Benjamin Sanchez-Gimeno
CIEMAT
Avda. Complutense 22
E-28040 Madrid
Spain
& +34 1346 6711
fax: +34 1346 6121

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Dr. Jürgen Schneider
Umweltbundesamt
Spittelauer Lände 5
A-1090 Wien
Austria
& +43 1 313045863
fax: +43 1 313045400

Dr. R.A. Skeffington
National Power Research & Engineering
Windmill Hill Business Park
SN5 6PB Wiltshire
U.K.
& +44 1793 896237
fax: +44 1793 896251

Dr. Christian Smoliner
Bundesministerium für Wissenschaft und Forschung
Rosengasse 4
A-1010 Wien
Austria
& +43 1 53120 6353
fax: +43 1 53120 6480

DI Wolfgang Spangl
Umweltbundesamt
Abt. für Lufthygiene
Spittelauer Lände 5
A-1090 Wien
Austria
& +43 1 313045 861
fax: +43 1 313045 400

Dr. Jòszef Szabó
RISSAC
Herman u 15
H-1022 Budapest
Hungary
& +36 1 1558839
fax: +36 1 1558839

Dr. Lena Skärby
IVL Swedish Environmental Research Inst.
P.O. Box 47086
Dagjämningsgatan 1
S-402 58 Göteborg
Sweden
& +46 31 46 00 80
fax: +46 31 48 21 80

Dr. Stefan Smidt
Forstliche Bundesversuchsanstalt
Tirolergarten
A-1131 Wien
Austria
& +43 1 87838 124
fax: +43 1 8775907

Dr. Gerhard Soja
Austrian Research Centre Seibersdorf
Dept. Agricultural Research
A-2444 Seibersdorf
Austria
& +43 2254 3542
fax: +43 2254 740 60
e-mail: soja@zdfzs.arcs.ac.at

Dr. Till Spranger
Umweltbundesamt
Mauerstraße 52
D-10117 Berlin
Germany
& +49 23145 842
fax: +49 23145 638
+49 308903 2798

Mag. Michael Tausz
Inst. of Plant Physiology
Schubertstraße 51
A-8010 Graz
Austria
& +43 1 316 380-5633
fax: +43 1 316 38 68 15

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Dr. Mette G. Thomsen
Norwegian Inst. of Forest Research
Hogskoleveien 12
N-1432 As
Norway
& +47 6494 9013
fax: +47 6494 8971

Prof.Dr. H.J. Weigel
Institute of Ecotoxicology
Bundesallee 50
D-38116 Braunschweig
Germany
& +49 531 596 315
fax: +49 531 596 366

Dr. Rupert Wimmer
BOKU Wien
Zentrum für Umwelt- und Naturschutz
Gregor Mendel Str. 33
A-1180 Wien
Austria
& +43 1 47 654-2300
fax: +43 1 47654-2304

Dr. Wilfried Winiwarter
Austria Research Centre Seibersdorf
Dept. Environmental Planning
A-2444 Seibersdorf
Austria
& +43 2254 3868
fax: +43 2254 3888
e-mail: winiwarter@zdfzs.arcs.ac.at

Dr. Artur R. Wojcik
Inst. for Ecology of Industrial Areas
Kosslitha 6
PL-40-832 Katowice
Poland
& +48 3 1546031
fax: +48 3 1541717