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The Behavior of Selected Trace Elements in Alpine Soils Developed on Black Shales in the upper part of the "Hauptdolomit" (Seefeld Area, Tyrol, Austria)

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6 Text-Figures, 2 Tables and 1 Plate

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Blatt 117

Tyrol
Karwendel Mountains
Black Shales
Hauptdolomit Formation
Seefeld Member
Alpine Soils
Trace Elements
Environmental Hazards

Contents

Das Verhalten ausgewählter Spurenelemente in alpinen Böden über Schwarzschiefern (Seefelder Schichten) des oberen Hauptdolomits im Gebiet von Seefeld in Tirol, Österreich

Zusammenfassung

Die Untersuchung des Verhaltens von ausgewählten Spurenelementen in alpinen Böden über Schwarzschiefern des oberen Hauptdolomits zeigte, daß die Mobilität der Elemente vom Bodentyp abhängig ist. Die Anreicherung zahlreicher Elemente (As, Co, Cu, Hg, Mo, Ni, Pb, V und Zn) in den obersten Bodenschichten aller untersuchten Profile ist von der Sorptions- und Komplexierungs-Kapazität der Humussubstanzen abhängig.

Abstract

The study of the behavior of selected trace elements in Alpine soils developed on black shales in the upper part of the "Hauptdolomit" formation in the Seefeld area, Tyrol, Austria has shown that the mobility of elements differs according to the soil type. The enrichment of many elements (As, Co, Cu, Hg, Mo, Ni, Pb, V and Zn) in the upper part of all studied soil profiles is due to sorption and complexation capacity of humus substances.

1. Introduction

Behavior of trace elements during the weathering of Norian (Upper Triassic) black shale intercalations in the upper part of the "Hauptdolomit" formation (Seefeld member) and soil profile formation has been studied in the Seefeld area, Tyrol, which extends from 1180 m to 2240 m in altitude. Vegetation is mostly represented by various types of grass at higher altitude (Nördlinger Hütte) and mixed forest, mostly coniferous at lower altitudes (Rudolfstollen locality).

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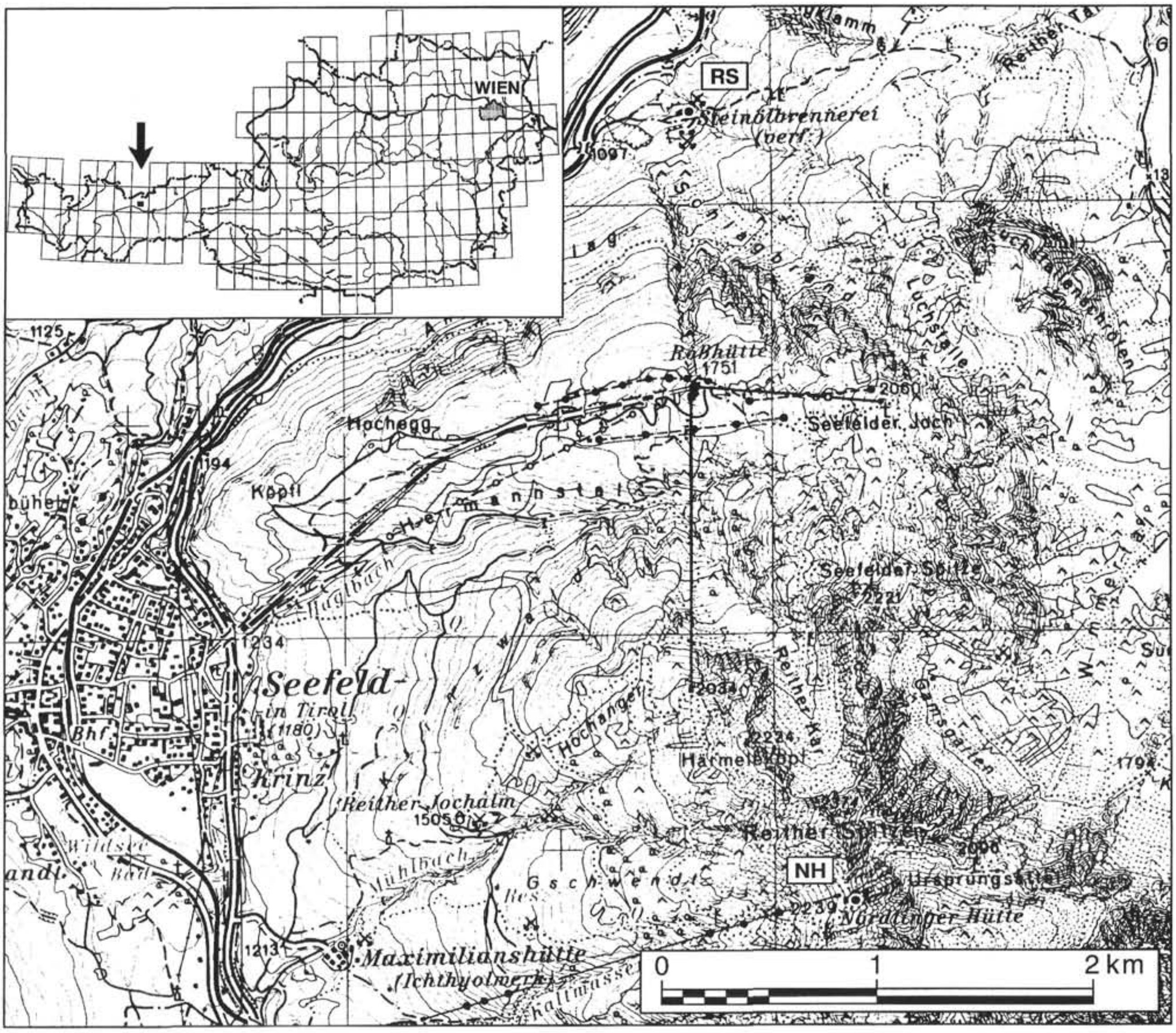
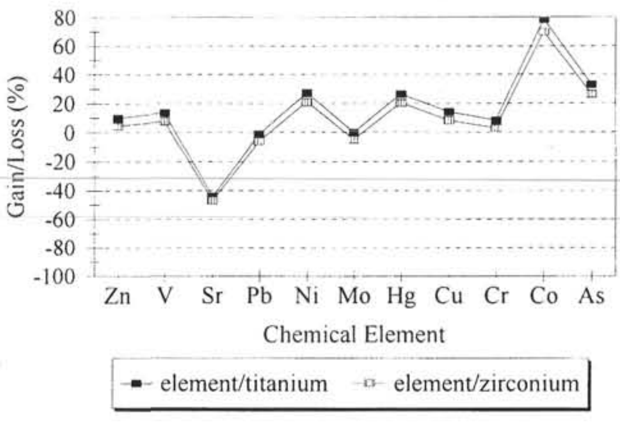


Text–Fig. 1.
Location of the studied soil profiles in the Seefeld area: "RS" = Rudolfstollen in the northern part of abandoned Ankerschlag oilshale mining district. "NH" = Nördlinger Hütte-surroundings.

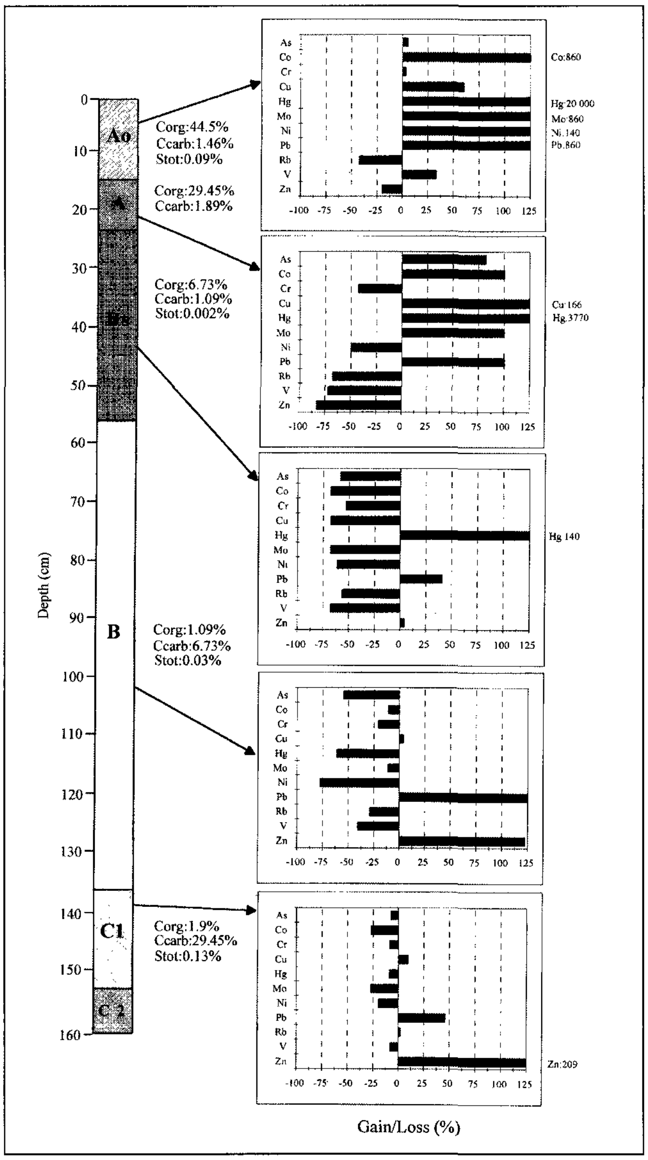
The main goal of this study was to characterize variations in the distribution of selected trace elements in different types of Alpine soils developed on black shale bedrock and to evaluate possible environmental impacts of these bedrock facies.

2. Geology and Mining History

In the Karwendel region the Upper Triassic (Norian), "Hauptdolomit" formation reaches a thickness of almost 2000m. The upper part of the section (Upper Norian), locally shows bituminous development with up to few hundred meters thick organic matter-rich dolomites and marls, known as the Seefeld member of the "Hauptdolomit" formation. It has been mined since more than 1000 years. The first historic document dates from the year 1350 (HRADIL & FALSER, 1930), where the recovery of oil shale is documented. Until 1964, oil shale was economically dry distilled from organic-rich marly layers, which exhibit $C_{org.}$ contents between 5 and 45 wt. % (BITTERLI, 1962). The sulfur-rich oil shale was successfully



Text–Fig. 2.
Compositional change (in %) for chemical elements in the A/C horizon of the Nördlinger Hütte No. 1 profile (lithosol) in relation to the fresh rock source. Calculations are based on titanium and zirconium as immobile elements.



Text–Fig. 3.
Compositional change (in %) for chemical elements in the Seefeld-Rudolfstollen cambic podzol profile. Titanium was used as immobile element. The description of soil horizons is given in the text.

used for pharmaceutical products, which were well marketed under the brand name "Ichtyol".

Research on these "fish-shales" has already started in the last century with descriptions of abundant fish fauna (e. g. KNER, 1866). Well preserved gymnosperm flora and even very scarce findings of saurians have also attracted great attention of paleontologists.

From the sedimentological and genetic point of view, the organic-rich Seefeld member was the scope of early basic studies by AMPFERER & HAMMER (1905), and especially by SANDER (1921, 1922) and TRUSHEIM (1930). Later papers introducing results of modern geochemical and stable isotope studies as well as actuosedimentological aspects mostly dealt with the palaeoenvironmenal and diagenetic history of the Seefeld organic-rich sediments (e. g. FISCHER, 1957 and A. G. FISCHER, 1964; MÜLLER & JUNGBLUTH 1968 and 1970; FRUTH & SCHERREIKS, 1984; BRANDNER & POLESCHINSKI, 1986; FRIES et al., 1987; KODINA et al., 1988; LOBITZER et al., 1988 and 1994; KÖSTER, 1989; POLESCHINSKI, 1989; IANNACE & KÖSTER, 1990; SCHENZLE & OSCHMANN, 1998 and others).

Genetically, two following different palaeoenvironmental settings are assumed for the formation of bituminous Seefeld member (BRANDNER & POLESCHINSKI, 1986; FRIES et al., 1987; IANNACE & KÖSTER, l.c.):

a) Restricted, shallow subtidal lagoonal setting ("lagoonal type"). Organic-rich dolomite sequences are represented by Mg-rich dolomite similarly as in recent evaporitic environments and showing well completed dolomitization process. Moreover, molds of the former evaporite minerals, e.g. of gypsum "knive stiches", can be found in some dolomite sequences of this type. Also, the highest $\delta^{18}O$ isotope signatures (0.9 ‰, PDB), correspond to the "lagoonal type" Seefeld member, indicating a higher salinity environment.

b) In contrast to the "lagoonal type", organic-rich Hauptdolomit sedimentation of the "small basin type", took place in restricted, tectonically controlled special basins. The organic-rich sequences consist of dolomite depleted in Mg and $C_{org.}$-rich marls. Slumping structures and endogenic brecciation, as well as the presence of normal marine biota, including ammonites, conodonts and *Heterastridium conglobatum* REUSS (BRANDNER & POLESCHINSKI, 1986 and POLESCHINSKI, 1989), are typical for the "small basin type" of the Seefeld member sequences.

Soil samples were taken from localities, where the underlying Seefeld member is of the "small basin type". Geochemical analyses of calcareous facies from Nördlinger Hütte prove that they consist of dolomitic limestone, showing higher SO_3 values (a. 5 wt. %) and about 100 ppm Ni (LOBITZER et al., 1994). Elevated values of Mo, V and Ti were detected especially in the marly samples (KODINA et al., 1988). In carbonate of the Nördlinger Hütte section the bitumen yield (Soxhlet extraction) ranges from about 1500 ppm up to about 18 800 ppm in dark brown soft laminated bituminous marl (LOBITZER et al., 1988). The organic matter can be classified as sulfur-rich "oil-prone" type-II kerogen for most of the local oil shales (KÖSTER et al., 1987).

3. Materials and Methods

Four soil profiles have been studied in the Seefeld District. Three profiles were sampled in the high-altitude environment (Nördlinger Hütte Site, altitude: 2240m), while the last one is located at lower altitude of approximately 1180 m, within the abandoned black-shale mining district (Seefeld, Rudolf Stollen). The location of studied soil profiles is given on Text-Fig. 1 and detailed description of these soil profiles is schematically shown in Text-Figs. 3–6.

The soil profile sampled close to the Rudolf Stollen (Rudolf Gallery) in Seefeld can be classified as the Alpine cambic podzol according to FAO/UNESCO (1974) classification. It consists of (1) a parent rock (unweathered calcareous black shale, C2-horizon), (2) a 20 cm thick layer of gray, slightly weathered, mechanically disintegrated parent rock layer (C1-horizon), (3) a rusty-brown, 80 cm thick illuvial horizon formed by sandy clay with numerous fragments of parent rock (B horizon), (4), a rusty, 40 cm thick horizon formed by clayey sand with sign of polyedric structure (Bs horizon), (5) a brown-black, 10 cm thick layer of a mature humus with rare fragments of the parent rock (A horizon) and, (6) a layer of raw humus with numerous roots of vegetation (Ao horizon).

Soils developed on black shales at the Nördlinger Hütte can be classified as Alpine lithosol-pararendzinas or rendzinas (FAO/UNESCO, 1974). From the bottom to the top, soil profiles consist of : (1) a slightly weathered, partly disintegrated parent rock (C1 horizon), (2) a layer of mature humus, 5–10 cm thick (A horizon) and, (3) an horizon of raw humus, 5 - 10 cm thick, with numerous roots of Alpine vegetation (Ao horizon). In the Nördlinger Hütte No. 1 profile, the humus material was relocated along cracks and frost-related structures downward, into the horizon of partly disintegrated parent rock layer (A/C horizon).

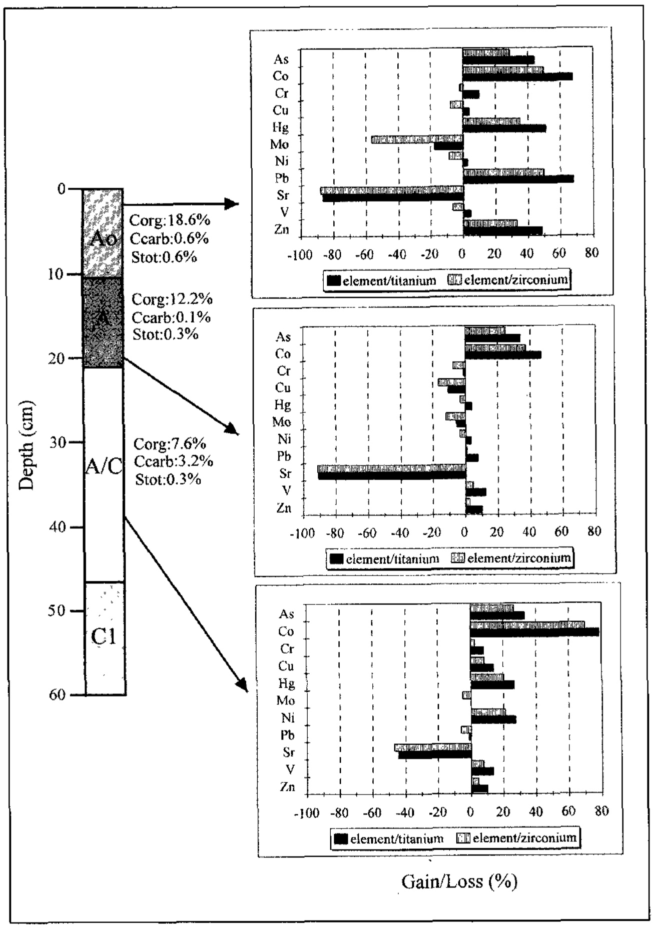
Samples were air-dried, pulverized and analyzed for Cr, Co, Cu, Mo, Ni, Pb, Rb, V, Zn and Ti using FAAS. Arsenic was determined using HGAAS and Hg by AMA technique. Sr and Ti were detected by ICP-AES. Concentrations of the organic carbon (C_{org}) and total sulphur (S_{tot}) were determined using IR method, carbonate carbon (C_{carb}) coulometrically.

To determine changes in the concentration of elements in various horizons of a soil profile relative to those in parent rocks, an immobile element concept was used. Within this concept, the ratio between the studied element and immobile element concentration (zirconium or titanium) in soil samples was compared with the same ratio in the parent rock according to the following equation (BRAUN & PAGEL, 1990):

$$\% \text{ change} = ((\chi/i)/(\chi_o/i_0) - 1) * 100,$$

where:
χ = content of the x element in the soil sample
χ_o = content of the x element in the parent rock
i = content of the immobile element in the soil sample
i_o = content of the immobile element in the parent rock.

Because the concentration of zirconium in many samples is below the detection limit of analytical techniques employed, titanium is preferably used as the immobile element in our calculations. Results obtained from the Nördlinger Hütte 1 profile, where both Ti and Zr are present in high concentrations show, however, a very high degree of correlation between relative changes in concentrations of elements determined using Ti and Zr as immobile elements (Text-Figs. 2 and 4).



Text–Fig. 4.
Compositional change (in %) for chemical elements in the Nördlinger Hütte No. I profile (lithosol-pararendzina). Titanium and zirconium were used as immobile elements. The description of soil horizons is given in the text.

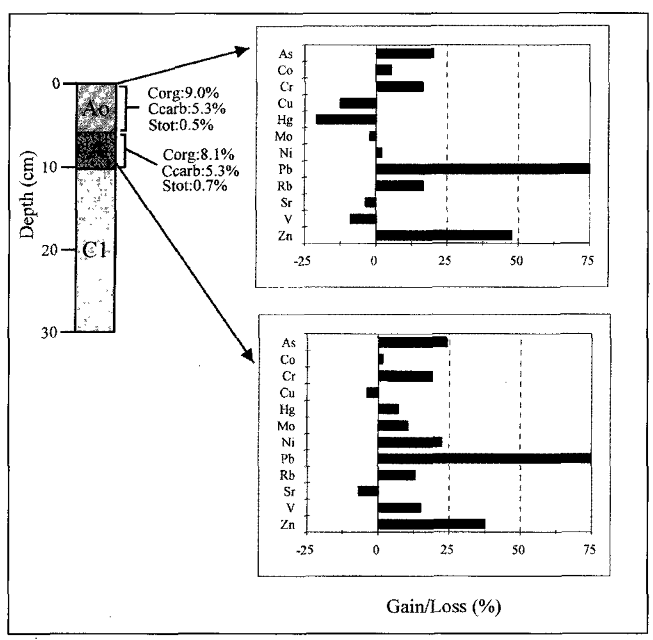
4. Results and Discussion

Results of trace elements concentrations in parent rocks and in overlying soil horizons are summarized in Table 1, concentrations of the C_{org}, C_{carb} a S_{tot} are given in Table 2.

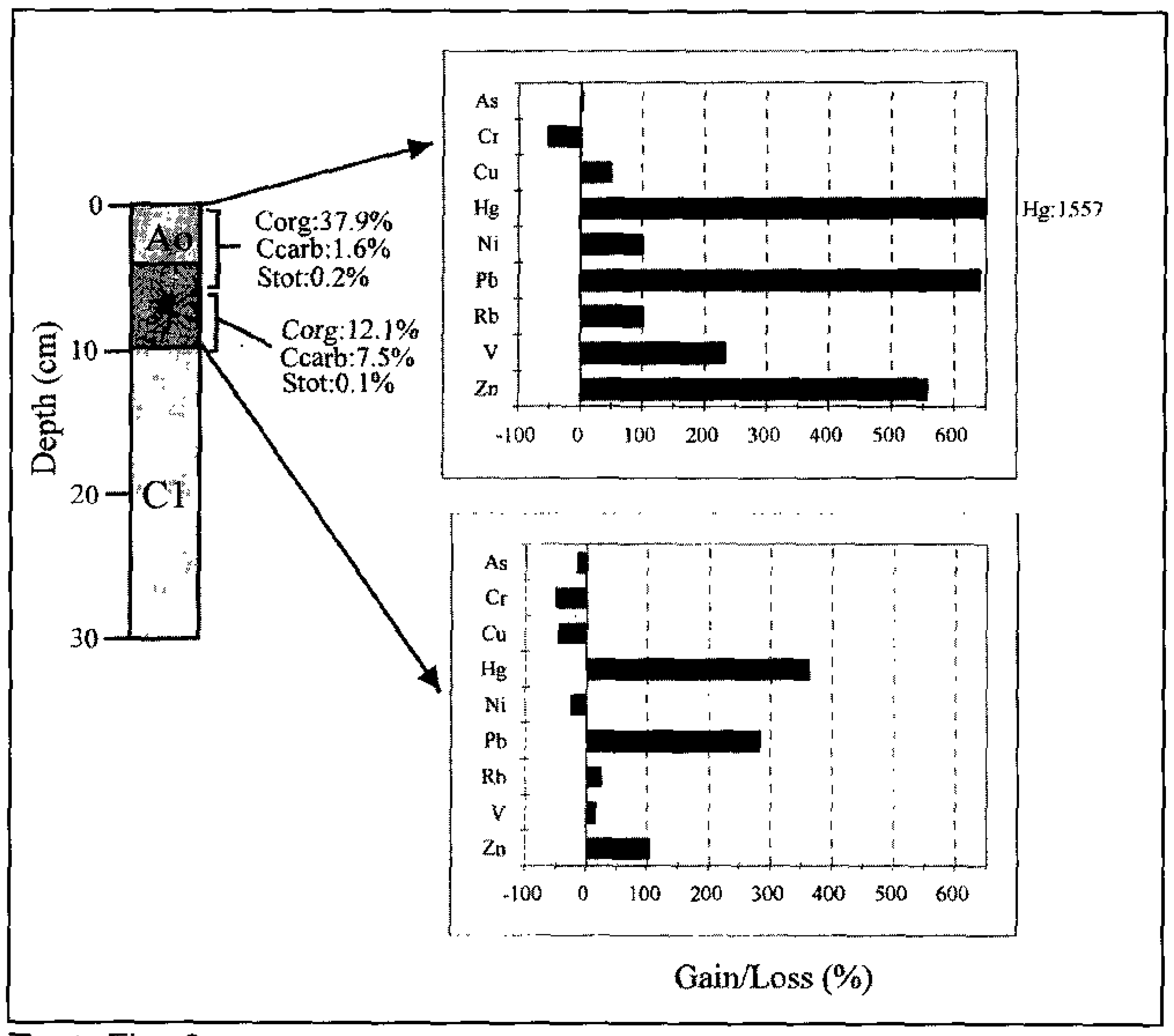
Changes in concentrations of trace elements (relative to titanium) in soil profiles are shown at Text-Figs. 3 to 6 and discussed in following paragraphs:

Soil profile Seefeld (near Rudolf Stollen)
Soil type: Alpine Cambic Podzol

Compared with parent rock (unweathered calcareous black shale, C2 horizon), the horizon of slightly weathered and mechanically disintegrated rock (C1) is up to 25% depleted in trace elements with exception of Zn, Pb and Cu. The enrichment in above elements is probably due to very low mobility of these metals in carbonate-saturated pore solutions (C_{carb} = 29.45%) or due to their downward migration and precipitation. Upward, in the B horizon, difference between Pb, Zn and Cu and other elements behavior is even more significant. The maximum depletion in many elements, with exception of Pb and Hg, is reached in the Bs horizon formed by a highly permeable clayey sand with low concentration of carbonate carbon (1.09%) and low concentration of organic carbon (6.73%). Mercury, which was depleted in un-



Text–Fig. 5.
Compositional change (in %) for chemical elements in the Nördlinger Hütte No. 2 profile (lithosol-rendzina). Titanium was used as immobile element. The description of soil horizons is given in the text.



Text–Fig. 6.
Compositional change (in %) for chemical elements in the Nördlinger Hütte No. 3. profile (lithosol- pararendzina). Titanium was used as immobile element. The description of soil horizons is given in the text.

derlying horizons starts to be enriched in this horizon, probably due to sorption or complexation by humus matter.

Relative enrichment of many metals (As, Co, Cu, Hg, Mo, Pb, V) in the Ao and A horizons can be related to the sorption and complexation by the humic organic matter and/or uptake by metals by living vegetation. The relative enrichment of metals in both horizons is not simple function of the C_{org} content. Enrichment in As, for example, is higher in the A horizon, in spite of lower content of C_{org} (29.5%), when compared with the Ao horizon (44.5% C_{org}). Differences in the enrichment of metals in the A and Ao horizons can be attributed to the different chemical composition of humus material or to the different rates of percolation by surface waters.

Remarkable enrichment in Hg relative to the parent rock in the A horizon (3770 %) and in the Ao horizon (20 000 %) may be tentatively explained as due to an anthropogenic contamination related to in-situ past retorting of black shale. It should be noted that the results of a pilot study from eastern Finland suggested a spatial correlation between the distribution of Hg-sulfide-rich black shales and the prevalence of coronary heart disease (LOUKOLA-RUSKEENIEMI et al., 1998).

Soil profile Nördlinger Hütte 1
Soil type: Alpine Lithosol-Pararendzina

Increased intensity of weathering is manifested by steady relative depletion of strontium upward the profile which can be related to the gradual decrease of carbonate component. Relative enrichment in many elements can be attributed to the extreme slow rate of chemical weathering and to the fixation of metals by humus. The humus fixation ability is demonstrated by a higher degree of enrichment in numerous elements in the Ao horizon (C_{org}: 18.6) compared with the A horizon (C_{org}: 12.2%). Enrichment of As and Co in the A/C horizon can be tentatively explained by the relocation of an humus material along cracks and joints into the mechanically disintegrated layer of the parent rock. The character of this soil profile at Nördlinger Hütte with typical bedding of host rock represented by alternating thinner black shales and thicker dolomitic limestones and dolomites are shown on Plate 1, Figs. 1 and 2. Deatiled black shale lithological features are given on Plate 1, Figs. 3 and 4. These sediments belong to the upper part of the Hauptdolomite formation designated as Seefeld member.

Soil profile Nördlinger Hütte 2
Soil type: Alpine Lithosol-Rendzina

The soil profile is characteristic of very low content of humus in the Ao and A horizons ($C_{org} < 10\%$). Very low rate of chemical weathering is demonstrated by negligible depletion of soil in highly mobile strontium. In comparison with lithosols with humus rich-A horizons (profiles NH1 and NH3), the enrichment of soil in other elements is relatively low, with exemption of Pb and Zn which tend to accumulate in both, the Ao and A horizons. The explanation can be seen in relative

Table 1
Concentration of some elements in soil profiles (Ao to B) and in parent black shales (C) in the Seefeld District

Element (in ppm) Soil Profile	Soil Horizon	As	Co	Cr	Cu	Hg	Mo	Ni	Pb	Rb	Sr	Ti	V	Zn	Zr
Rudolfstollen	Ao	0.6	<5	3	<2	0.340	<5	<5	<10	<3	<2	50	<15	<2	<10
	A	5.0	<5	8	8	0.310	<5	<5	<10	4	<2	240	<15	<2	<10
	Bs	7.1	<5	41	6	0.120	<5	12	22	34	<2	1499	54	39	<10
	B	2.8	<5	25	7	0.007	<5	<5	18	20	<2	540	36	30	<10
	C1*	7.0	<5	35	9	0.020	<5	11	10	35	<2	659	68	51	14
	C2**	5.5	<5	28	6	0.016	<5	10	<10	25	<2	480	54	12	27
Nördlinger Hütte 1	Ao	20.4	11	68	35	0.170	51	52	44	106	<2	1858	535	163	41
	A	27.5	14	89	44	0.170	85	76	41	123	<2	2698	834	175	57
	A/C***	22.5	14	80	46	0.170	74	77	31	103	5	2218	694	144	46
	C1*	15.1	7	66	36	0.120	66	54	28	82	8	1978	546	117	39
Nördlinger Hütte 2	Ao	22.8	9	66	36	0.140	76	58	43	90	3	1679	600	139	58
	A	24.4	9	70	41	0.190	86	72	30	90	3	1739	785	134	53
	C1*	24.4	11	73	53	0.220	100	73	15	99	4	2158	848	121	61
Nördlinger Hütte 3	Ao	2.5	<5	7	6	0.100	<5	5	37	10	<2	240	25	74	<10
	A	6.8	<5	24	7	0.090	<5	6	62	20	<2	779	28	59	<10
	C1*	1.9	<5	15	4	0.006	<5	<5	<10	5	<2	240	<15	9	<10

* – slightly weathered parent rock, ** – fresh parent rock, *** – slightly weathered parent rock with humus admixture

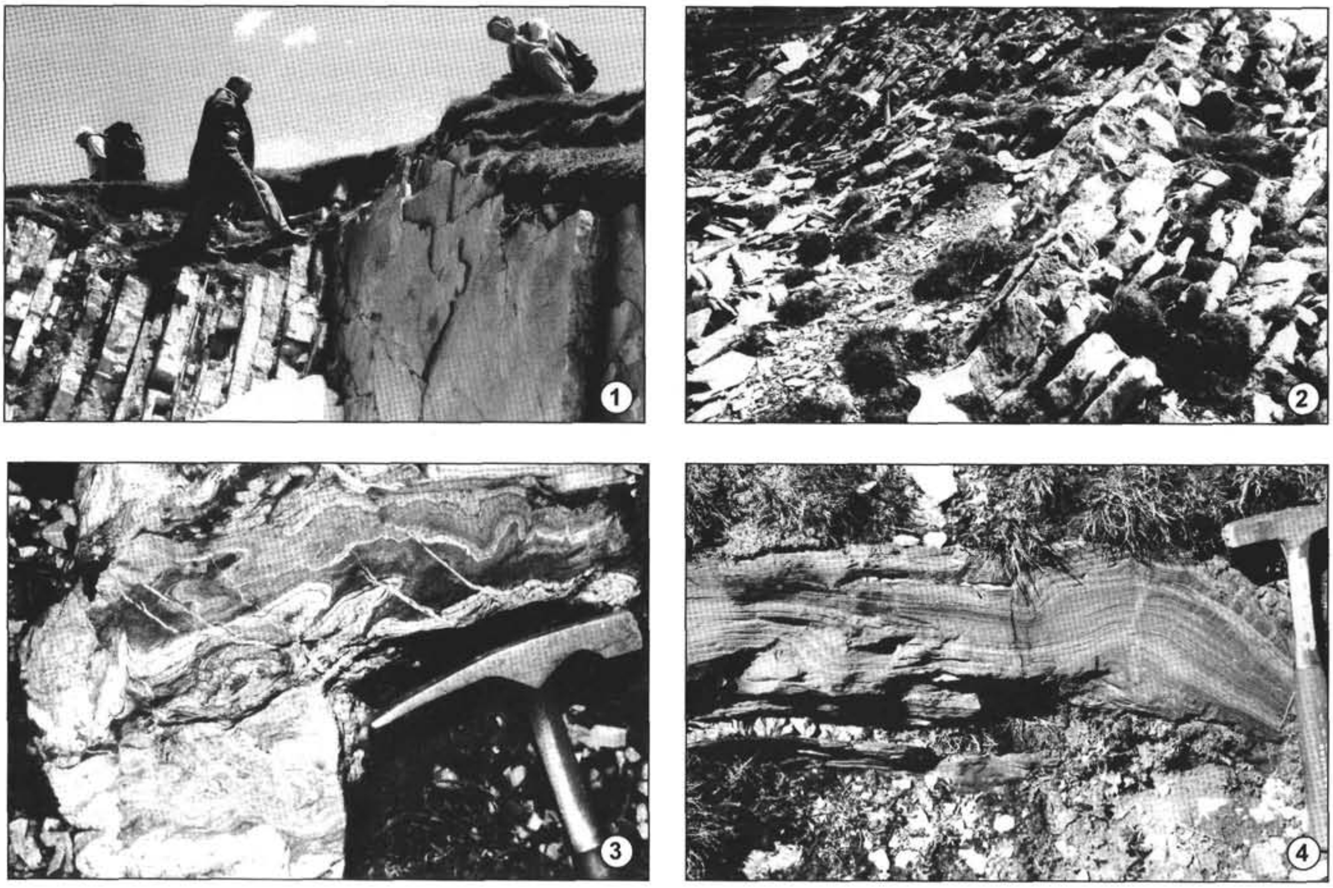


Plate 1

Fig. 1. The location of the soil profile and character of the host rock (Seefeld member of the upper "Hauptdolomit"), represented by alternating thinner black shales with thicker dolomitic limestones and dolomites at Nördlinger Hütte.

Fig. 2. Slightly bituminous dolomitic limestones and dolomites (right part of the picture) and calcareous and dolomitic black shales (left part of the picture), of the upper part of the "Hauptdolomit" – Seefeld member, about 100 m east of Nördlinger Hütte.

Fig. 3. Thinly laminated black shale of the upper part of "Hauptdolomit" – Seefeld member with slumping texture overprinted by (? early) diagenetic folding/shearing about 30 m north of Nördlinger Hütte in the upper part of tourist trail to Ursprungsattel. The thin light-coloured dolomitic interlayers are fractured.

Fig. 4. Thinly laminated slightly folded black shale (organic-rich calcareous dolomite) of the upper part of "Hauptdolomit" Seefeld member, underlain by organic-rich marls about 30 m north of Nördlinger Hütte in the upper part of tourist trail to Ursprungsattel.

high amount of carbonate (C_{carb}: 5.3%) which constraints the solubility of both metals.

Soil profile Nördlinger Hütte 3
Soil type: Alpine Lithosol-Rendzina

The soil profile is characteristic of very high content of humus in the A and mainly in the Ao horizon. The significant role of humus organic matter in the enrichment of soil profile in many elements is manifested by the substantially higher enrichment in the Ao horizon (C_{org}: 37.9%) when compared with the A horizon (C_{org}: 12.1%). Increased amounts of Pb and Zn in the A horizon are probably due to their low solubility in carbonate-saturated soil environment. Similarly as in other studied soil profiles, Hg tends to be substantially enriched in the humus-rich layer.

5. Conclusions

The present study shows that, in the Seefeld area, mobility of elements in Alpine soils developed on black shales differs according to the soil type. In cambic podzol soils, the majority of trace elements (As, Co, Cr, Cu, Hg,Ni, Rb and V) is strongly depleted in illuvial, sandy-clay and clayey-sand B and Bs horizons. In high-altitude lithosols, only strontium depletion as a consequence of carbonate dissolution, has been detected in soil horizons due to low rate of chemical weathering. Relative enrichment in Zn and Pb observed in lower soil horizons of both, the cambic podzol and lithosols is probably due to low mobility of these metals in carbonate-rich soil environment.

The enrichment of many elements (As, Co, Cu, Hg, Mo, Ni, Pb, V and Zn) in the upper part of all studied soil profiles is due to the sorption and complexation capacity of humus substances. The degree of enrichment is not, however, a simple function of the organic matter content. Differences in the relative enrichment of elements in the Ao and A horizons may be probably assigned to the different chemical properties of raw and mature humus. Remarkable enrichment in Hg in raw humus layer of the cambic podzol (20 000% relative to the parent rock), sampled in the abandoned black shale mining district near Seefeld, can be most likely explained as the result of anthropogenic contamination (in-situ retorting of black shales).

Table 2
Concentration of organic carbon (C_{org}), carbonate carbon (C_{carb}) and total sulphur (S_{total}) in soil horizons (Ao to B) and in parent black shales (C) in the Seefeld District

Soil Profile	Soil Horizon	C_{org} (%)	C_{carb} (%)	S_{total} (%)
Rudolfstollen	Ao	44.5	0.1	0.20
	A	29.5	0.6	0.13
	Bs	6.7	5.4	0.03
	B	1.1	11.0	0.01
	C1*	1.9	10.5	0.09
	C2**	1.5	11.2	0.09
Nördlinger Hütte 1	Ao	18.6	0.6	0.48
	A	12.2	0.1	0.54
	A/C***	7.6	3.2	0.34
	C1	7.6	4.6	0.29
Nördlinger Hütte 2	Ao	9.0	5.3	0.51
	A	8.1	5.3	0.70
	C1	7.2	4.5	0.69
Nördlinger Hütte 3	Ao	37.9	1.6	0.16
	A	12.1	7.5	0.14
	C1	1.1	12.3	0.01

* – slightly weathered parent rock, ** – fresh parent rock, *** – slightly weathered parent rock with humus admixture

It should be noted that, from the environmental safety point of view, high concentrations of metals in black shales represent potential environmental hazard only in regions with a weak vegetation cover and substantially decarbonatized soil profiles.

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