

# Anthropogenic and natural radionuclides in soil of a tropical rainforest of Southern Costa Rica

## Radionúclidos antropogénicos y naturales en el suelo de un bosque lluvioso tropical del sur de Costa Rica

Peter BOSSEW, Alexander HUBMER & Friederike STREBL

**Abstract:** Radiometric investigation in a rainforest of the Golfo Dulce region of southern Costa Rica has shown the presence of the anthropogenic radionuclides caesium-137, strontium-90 and the plutonium isotopes 238, 239 and 240 along with the naturally occurring radionuclides  $^7\text{Be}$ ,  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$ , and respective progenies.  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and Pu can be attributed to global fallout; no other sources could be identified. Soil cores were taken from five locations in 1996 and 2007. The mean inventory of  $^{137}\text{Cs}$  was determined to be 530 Bq/m<sup>2</sup> (decay corrected 1 Jan 2007), the ratio  $^{239+240}\text{Pu} : ^{137}\text{Cs}$  equals 4.5%, the  $^{238}\text{Pu} : ^{239+240}\text{Pu}$  ratio, was 3.4%. All values are in accordance with expectations for global fallout in the region. Vertical migration rates of  $^{137}\text{Cs}$  were found to be 0.9-1.7 mm per year. Mean concentrations were 169 Bq/kg dry matter of  $^{40}\text{K}$ , 13 Bq/kg of  $^{238}\text{U}$ , 11 Bq/kg of  $^{226}\text{Ra}$  and 9 Bq/kg of  $^{232}\text{Th}$ , all of which are quite low values. There are very few radio-ecological data available for tropical ecosystems. Therefore, our findings may serve as baseline information for further studies, e.g. on vertical migration of reactive (sorbing) substances in soil, or for erosion studies (horizontal translocation) using caesium-137 or plutonium as tracers.

**Key words:** radionuclides, global fallout, tropical rainforest soil, Costa Rica.

**Resumen:** Una investigación radiométrica en la selva tropical de Golfo Dulce, región al sur de Costa Rica, mostró la presencia de los radionúclidos antropogénicos cesio-137 y estroncio-90, los isótopos 238, 239 y 240 del plutonio y los radionúclidos naturales  $^7\text{Be}$ ,  $^{40}\text{K}$ ,  $^{238}\text{U}$  y  $^{232}\text{Th}$ , con sus respectivas progenies.  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  y Pu pueden atribuirse al "fallout" global, ya que no fue identificada ninguna otra razón para su presencia. Se tomaron muestras de suelo en 5 lugares, en 1996 y 2007. La contaminación media de  $^{137}\text{Cs}$  fue de 530 Bq/m<sup>2</sup> (desintegración corregida a 1 de enero de 2007), la proporción  $^{239+240}\text{Pu} / ^{137}\text{Cs}$  fue del 4.5%, y del 3.4% la proporción  $^{238}\text{Pu} / ^{239+240}\text{Pu}$ . Estos valores están de acuerdo con lo esperado para el "fallout" global de la región. Fueron encontradas tasas de migración vertical en el suelo de 0.9-1.7 mm / año para el  $^{137}\text{Cs}$ . Y concentraciones medias en materia seca de  $^{40}\text{K}$ : 169 Bq/kg,  $^{238}\text{U}$ : 13 Bq/kg,  $^{226}\text{Ra}$ : 11 Bq/kg y  $^{232}\text{Th}$ : 9 Bq/kg, las cuales pueden considerarse bastante bajas. La literatura científica sobre radio-ecología en ecosistemas tropicales es muy escasa, por lo cual estos resultados pueden servir como referencia preliminar para futuros estudios, por ejemplo sobre migración vertical de sustancias reactivas (absorbentes) en el suelo, o sobre erosión (traslación horizontal), usando como trazadores  $^{137}\text{Cs}$  o Pu.

**Palabras clave:** radionúclidos, fallout global, suelo de selva tropical, Costa Rica.

## Introduction

### The radioactive environment

Radioactivity, or ionising radiation, is ubiquitous in all environments. Due to the particular physical features of ionising radiation, the study of environmental radioactivity has become a distinct discipline within the environmental sciences. From a technical point of view, this is because specific techniques must be applied in assessing environmental radioactivity, partly quite different from the methods used in other disciplines. However, the main reason for the interest in environmental radioactivity is that radiation can be a hazard which has to be surveyed, modelled, predicted and, if

possible, contained. It has also turned out, on the other hand, that radionuclides can be used as quite easily measurable tracers for environmental transport processes, thus contributing to our understanding of how nature functions.

This article presents and discusses some results on environmental radioactivity in the Esquinas forest of southern Costa Rica. It gives by no means an exhaustive picture of environmental radioactivity of that region; rather, it concentrates on inventories of natural and anthropogenic radioactivity in rainforest soils.

In section 1, an overview of sources of environmental radioactivity is given, and the role and possible pur-

poses of radio-ecology is discussed shortly. In section 2, two small radio-ecological research projects carried out in the Esquinas forest are presented and their methods discussed. Results are given in section 3, and finally conclusions are drawn in section 4 which give an overview of possible future research work.

## Sources of environmental radioactivity: natural sources

Sources of environmental radioactivity may be classified into natural and anthropogenic sources. Natural sources, in turn, can be divided into cosmogenic and geogenic sources.

**Cosmogenic** radionuclides are products of interaction between cosmic rays and atoms of the atmosphere; among the more important ones are  $^3\text{H}$ ,  $^{14}\text{C}$  and  $^7\text{Be}$ .  $^{14}\text{C}$  has a well known application for chronological dating, while  $^7\text{Be}$  is used as a tracer for atmospheric processes and for certain soil-related phenomena like erosion and sedimentation, because it is continuously deposited on the soil surface.

**Geogenic** radionuclides: The elements building up planet Earth were produced in stars by nuclear reactions more than 5 billion years ago, and were dispersed through the universe when these stars underwent supernova explosions. Eventually such "litter" condensed and clustered, and formed our solar system. Among the nuclides produced in these lost stars were some unstable, i.e. radioactive ones. Those with sufficiently long half-lives have survived until today, and are consequently contained in our environment. About 50 so-called primordial radionuclides are known altogether. The most important of these radionuclides are  $^{40}\text{K}$  (potassium,  $1.3 \times 10^9$  years half life),  $^{238}\text{U}$  (uranium,  $4.4 \times 10^9$  y),  $^{235}\text{U}$  ( $7 \times 10^8$  y) and  $^{232}\text{Th}$  (thorium,  $1.4 \times 10^{10}$  y). While no stable isotopes of uranium and thorium exist, natural potassium consists mainly of stable isotopes,  $^{39}\text{K}$  and  $^{41}\text{K}$ , but the 0.0117%  $^{40}\text{K}$  make it an important ubiquitous radioactive source. Uranium and thorium isotopes do not decay into stable nuclides immediately, but yield decay chains consisting of different radionuclides of radium, radon, bismuth, polonium and lead among others, until they end up in stable isotopes of lead. Some of these so-called progenies or daughters are themselves quite long lived, like  $^{226}\text{Ra}$  with 1620 years half life, which is long enough to be separated from the precursor by ecological processes, due to their different chemical properties, notably different solubility in water. To complicate matters further, these chains contain radionuclides which are volatile noble gases, namely different isotopes of radon. Radon can escape from rocks and soils into the atmosphere, and can cause radiological problems, e.g. indoor radon accumulation, which is the second most important source of lung cancer after smoking when inhaled

by people. As a gas, radon behaves completely differently to the original radionuclide sources (uranium and thorium). The decay products of radon are again heavy metals, which can attach to aerosols and settle onto the soil surface (or the lung tissue), effectively being a kind of natural radioactive fallout. For this reason  $^{210}\text{Pb}$ , a long-lived (22.3 years half life) radioactive member of the  $^{238}\text{U}$  chain and progeny of  $^{222}\text{Rn}$ , is also used as tracer for sedimentation and erosion processes.

The activity concentrations of geogenic natural radionuclides are strongly controlled by geology, making radiometry an important tool for geological exploration. Granites often show high uranium contents, while limestone usually contains little uranium; there are however exceptions and even within geological units, large variability is possible.

## Anthropogenic sources

Human activities have resulted in the release of radioactive substances. Apart from several events and nuclear accidents which have resulted in local or regional contamination (e.g. GOIÂNIA 1987), three events have led to global, or continental-range contamination: fallout from atmospheric nuclear bomb tests between 1945 and 1980, the disintegration of a nuclear-powered satellite in 1964 and the Chernobyl accident in 1986.

On 21 April 1964, the US satellite SNAP-9a which was powered by a thermoelectric aggregate fuelled by ca. 1 kg  $^{238}\text{Pu}$  disintegrated in the high atmosphere, dispersing its plutonium inventory world-wide. Most of the global  $^{238}\text{Pu}$  inventory stems from this event.

The fallout of Chernobyl affected the northern hemisphere almost exclusively, while southern and equatorial zones remained almost entirely uncontaminated, because only a very small fraction of the radioactive cloud has entered the stratosphere, which is a condition for global mixing. This was different for the large atmospheric thermo-nuclear bomb tests between 1954 and 1962 (mainly in Novaya Zemlya, Soviet Union, and the Pacific test area, USA) whose fallout was subsequently distributed around the globe, including Costa Rica. The resulting so-called global fallout is roughly evenly distributed longitudinally but shows a distinct latitudinal gradient. The most recent data on global-scale distribution of global fallout  $^{137}\text{Cs}$  can be found in AOYAMA et al. (2006).

On a local and regional scale, however, the fallout deposition density is strongly correlated with mean rainfall intensity, because "wash-out" from the atmosphere is the most efficient mechanism of deposition. For Costa Rica, this correlation is suggested by data presented in SALAZAR & MORA (1996); for Venezuela, a similar effect is reported by LABRECQUE et al. (2002), where the

authors explain exceptionally high local levels of  $^{137}\text{Cs}$  fallout in mountainous cloud forest with high frequency of fog.

From figures given in UNSCEAR (1982, p. 229; details of the estimate in BOSSEW & STREBL 1996) a mean  $^{137}\text{Cs}$  deposition density of 0.54 kBq/m<sup>2</sup> can be derived for Costa Rica. The interpolation of figures given by AOYAMA et al. (2006) yields 1.04 kBq/m<sup>2</sup>, i.e. almost twice as much (both figures have been decay corrected to 1 January 2007).

A number of soil samples from different parts of Costa Rica have been taken for radiometric analysis by SALAZAR & MORA (1996). However, the results on  $^{137}\text{Cs}$  are reported as gravimetric activity concentrations only (Bq/kg), rather than as inventory (Bq/m<sup>2</sup>). Therefore, they are only of limited use as baseline data for fallout distribution.

## Radio-ecology of tropical environments

The objectives of radio-ecology as a scientific discipline are twofold:

Since radioactivity is a potential hazard, its sources must be identified, its inventories assessed, and possible dispersion through the environment followed and, where necessary, contained. Particularly sensitive areas or environmental compartments must be identified and contamination pathways described in order to develop countermeasures against pollution. Obviously this requires knowledge not only about the inventories, but also about the ecological roles of radionuclides and transfer processes involved.

Since radionuclides are subject to ecological processes, like all substances, they can be considered as tracers of these transport and distribution processes. Being relatively easy to measure with great sensitivity, they are often good candidates as tools to study such processes. For example,  $^{137}\text{Cs}$  is widely used in studying erosion, since it is very strongly fixed on soil particles and is therefore redistributed together with them.

Very little radio-ecological research has been done in tropical environments, as opposed to temperate environments. The reasons are that until now, the tropics have been much less affected by human activities involving radioactive contamination than parts of Europe, Asia or North America. Thus, there was not much need for research aimed to contain or minimise hazards from radioactivity, compared with the latter regions. Secondly, the tropics belong, in most cases, to less affluent countries, where research funds are scarce and mostly limited to areas where urgent action is required, like health and hygiene, agricultural productivity, development of natural resources, etc.

However, exactly the latter objective has initiated radio-ecological research in a series of tropical countries. To name a few examples, considerable radio-ecological research has been carried out in Brazil and India in regions with naturally enhanced radioactivity (due to geological peculiarities). Uranium mining and processing has initiated radio-ecological research in tropical Australia and Brazil. In India and Taiwan, the increase in use of nuclear power has motivated the acquisition of radio-ecological baseline data for reference in case of contamination. On some Pacific islands, radio-ecology gained importance when the damage resulting from nuclear bomb tests had to be assessed and remedied as far as possible. In several tropical countries,  $^{137}\text{Cs}$  has been used as tracer in erosion studies. As a very recent example, a radio-ecological study in tropical Africa (Nigeria) aimed to acquire base information on fallout distribution on soil (AJAYI et al. 2007). To conclude, accidents leading to radioactive contamination can have global impact and more and more sub-tropical and tropical regions gain importance for (both regional and global) food production. Therefore, a sound radio-ecological knowledge base to cope with radioactive contamination events seems necessary for each ecological region.

Nonetheless, little is known about the cycling and transport properties of radionuclides in tropical ecosystems. We quote a passage from our article from 2001, which is still true six years on (BOSSEW & STREBL 2001, section 1.1):

“In a recent review about radio-ecology in tropical and subtropical ecosystems, FRISSEL (1997) stated that [soil-plant-transfer-] values of tropical and subtropical areas were completely missing up till 1989. From the findings collected in the last few years, the conclusion is drawn that there exists no systematic difference between soil-to-plant transfer factors in temperate, subtropical and tropical environments. Nevertheless, in tropical environments, there exist – as in temperate environments – ecosystems with a relatively high or low uptake. Relative mean TF-values can be higher or lower than average values by a factor 10 or even 100. According to FRISSEL (1997), it seems that in tropical systems the abundance of deviating systems is much higher than in temperate climates. One reason for this is the poor nutrient status of soils in many tropical systems and a low level of soil organic matter to buffer changes of pH and nutrient availability. Certain soil types seem especially prone to deviating uptake behaviour: coral soils, soils with high Fe-Al-oxide contents, acid soils and possibly volcanic soils. The same report concludes that, because only a small number of ecosystems have been investigated, more deviating systems must be expected.”

One of these statements has been more recently confirmed by WASSERMAN et al. (2005), who found particularly high TF in Brazilian soils with high iron content, and in acidic soils. An attempt to apply the so-called Absalom model for predicting TF from soil properties, originally developed for European soils (ABSALOM et al. 2001), to (sub-) tropical Asian soils (Japan, Taiwan, Bangladesh) showed gross agreement with measured TF for some typical crops, but also that tuning of the model to the peculiarities of the respective systems is necessary (RAHMAN & VOIGT 2004). A recent literature survey (AYUB & VELASCO 2006) presents 2700 single data entries for soil-plant transfer from radioecological studies (281 publications) in tropical and sub-tropical environments world-wide. However, only 246 data entries come from South American tropics (Brazil), and 30 values are reported from South America and sub-tropical Cuba. The coverage of radio-ecological information for soils in the South-American tropical region is even more sparse, with only one citation.

## Materials and methods

### Sampling

The results presented in this article are derived from samples which were taken in January 1996 and January 2007. The first survey was mainly aimed at acquiring basic data on activity concentrations of natural  $^{137}\text{Cs}$ , and inventories of anthropogenic  $^{137}\text{Cs}$  in top soil, as well as data on the horizontal and vertical variabilities. In the second sampling campaign, a finely resolved soil profile was taken, aimed at getting better data on the vertical distribution of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and plutonium, from which transport properties of these nuclides can be derived, as well as getting first data about  $^{90}\text{Sr}$  and plutonium inventories. Furthermore, long-term gamma-spectrometric measurements provide information about the possible presence of other anthropogenic radionuclides.

In 1996, four forest locations in the vicinity of the Esquinas lodge were sampled, along with two sites on nearby meadows for comparison. The soil samples were taken with a metal sampler as 15 cm deep cores with 25 cm<sup>2</sup> cross section. The cores were divided into top, middle and bottom layers, respectively (0-5 cm; 5-10 cm; 10-15 cm). At every location, five cores were taken in order to be able to assess the small-scale local variability. Further details about sampling and sample processing are given in BOSSEW & STREBL (2001) and in the original report (BOSSEW 1996).

The sampling site of 2007 is located near site 2 of 1996, in primary forest close to the “sendero fila” path, at an altitude of about 210 m above sea level. The site is located on the crest of a ridge, with almost no inclination. It can therefore be anticipated that no substantial erosion with rainwater has taken place, so that essentially no fallout radionuclides were carried away with surface erosion. The geographical location is approx. 83°12'W, 8°41'N (determined with Google Earth). The soil was taken according to the following scheme:

After clearing the site of vegetation (small shrubs), the organic layer, thickness ca. 1 cm, was removed in a rectangle of 60×80 cm<sup>2</sup> size. From the mineral soil surface, a 30×40 cm<sup>2</sup> rectangle was excavated 1 cm deep, yielding soil layer A. Then the second layer, B, was excavated, again 1 cm deep, as a 30×36 cm<sup>2</sup> rectangle, and so on. Layer thicknesses are 1 cm down to 5 cm, then 2 cm down to 15 cm, and finally two layers 7 cm and 8 cm thick, respectively, were excavated. The total depth of the resulting pit is 30 cm. Thirteen layers (including the organic layer) were taken altogether, the sizes of the rectangles chosen such as to produce samples of approximately equal mass. The pit after excavating down to approximately 15 cm is shown in figure 1a.



**Fig. 1:** (a) Excavation of a stratified soil sample, Jan. 2007. Dashed line: approximate shape of the first layer; (b) approximate map of sampling site 2, 1996. Circles denote the locations of trees.

## Analytical procedures

The samples were shipped to Austria and analysed at the laboratory of the Austrian Institute of Applied Ecology (1996) or the University of Salzburg (2007). For gamma analysis, standard HPGe measurement methods were applied.  $^{238}\text{U}$  was evaluated using the 63 keV double line of  $^{234}\text{Th}$ , in 1996, whereas in 2007 the 1001 keV line of  $^{234\text{m}}\text{Pa}$  was used, together with the most recent available value (to our knowledge) of its photon emission probability (YÜCEL et al. 2003). For  $^{90}\text{Sr}$  and plutonium analysis radiochemical pre-treatment is required, before beta counting (LSC) and alpha spectrometry. The procedure is described in detail, e.g., in BOSSEW et al. (2007), and references quoted therein. The  $^{90}\text{Sr}$  analyses could not be finished in time for this article, unfortunately, and will be published later. Also, detailed analyses of  $^{241}\text{Am}$  and other radionuclides are not yet complete.

## Statistics and calculations

### Horizontal variability:

The parameter for quantifying horizontal variability of a quantity used in this study is the “coefficient of variance” (CV), a measure of relative dispersion of data. Following the idea of analysis of variance (ANOVA) which decomposes the variance of grouped values into the mean variance within the groups, and the variance between the groups, we define the mean variability within the groups as

$$CV_{(\text{within})} := \sqrt{[SQ_{(\text{within})}/(n-1)] / AM_{(\text{all})}}$$

and the variability between the groups:

$$CV_{(\text{between})} := \sqrt{[SQ_{(\text{between})} * k / (n * (k-1))] / AM_{(\text{all})}}$$

where

(within) denotes variability of soil cores taken within one sampling site

(between) denotes variability between different sampling sites within the investigation area; in our case,

$n$  = number of cores = 21,

$k$  = number of sites = 5.

$SQ_{(\text{within, between})}$ : sum of squares following standard variance analysis.

### Critical interval:

As critical interval to significance  $p$ ,  $CI(1-p)$ ,  $0 < p < 1$  (or  $0 < p < 100\%$ ) we understand, given a random variable  $x$  distributed as  $f$ ,

$CI(1-p) := (a, b)$  such that  $\text{prob}(x < a | f) < p/2$  &  $\text{prob}(x > b | f) < p/2$ ,

in other words, the area of the histogram (or the probability density function) of  $x$  above the interval

$(a, b)$  equals  $1-p$  (or  $100\% - p\%$ ) of the total area. This means that with  $(1-p)$  probability a sample falls within  $(a, b)$ .

Here we use  $p = 0.05$ , that is, calculate CI95-intervals. Since not enough data are available for distribution-free estimates of the  $CI(1-p)$ , the interval limits are estimated by assuming a distribution. The CI concept applied here is different from the confidence interval of the estimated mean.

### Vertical migration:

The physico-chemical laws which control the migration of contaminants are known and can serve as basis for the development of migration models. One model which is often used for describing the migration of

$$C(x,t) = J_0 \left\{ \frac{1}{\sqrt{\pi Dt}} \exp\left(-\frac{(x-vt)^2}{4Dt}\right) - \frac{v}{2D} \exp\left(\frac{vx}{D}\right) \text{erfc}\left(\frac{v}{2}\sqrt{\frac{t}{D}} + \frac{x}{2\sqrt{Dt}}\right) \right\}$$

certain reactive contaminants in soil, like caesium and plutonium, is the convection-dispersion model (CDE), according to which the concentration  $C$  at depth  $x$  and at time  $t$  after fallout equals

with  $v$  = effective convection velocity and  $D$  = effective dispersion constant. (For details see e.g., BOSSEW & KIRCHNER 2004.) Radioactive decay would add a factor  $e^{-\lambda t}$ , which has been omitted for simplicity.

We fitted this model to the empirical distributions of  $^{137}\text{Cs}$  and plutonium of the 2007 soil sample in order to estimate the transport parameters  $v$  (apparent convection velocity) and  $D$  (dispersion coefficient). The empirical distribution parameters are,

$$AM := \int x c_0(x) dx;$$

$$SD = \sqrt{Var} \text{ where } Var = \int x^2 c_0(x) dx - AM^2;$$

$c_0(x) := c(x) / \int c(x) dx$ , where  $c_0(x)$  = empirical profile (Bq/cm<sup>3</sup>).

Integrals are approximated as sums. For the CDE fit a migration time of  $t = 42$  years is assumed (i.e. fallout at 1.1.1965, this date effectively marks the end of fallout from atmospheric bomb tests).

## Results and discussion

### Activity concentrations and inventories

The following radionuclides were identified in the soil samples:

Natural radionuclides:  $^7\text{Be}$ ,  $^{40}\text{K}$ , progenies of the  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  series.

Anthropogenic radionuclides:  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$ ,  $^{238}\text{Pu}$ ,  $^{241}\text{Am}$ . The plutonium isotopes  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  cannot be separated with the analytical means available at our laboratory.

**Table 1:** Inventories of radionuclides in the soil samples

	$^{137}\text{Cs}$ Bq/m <sup>2</sup>	$^{40}\text{K}$ Bq/kg	$^{226}\text{Ra}$ Bq/kg	$^{232}\text{Th}$ Bq/kg	$^{238}\text{U}$ Bq/kg
n	21	21	20	20	20
AM	530	169	10.6	8.6	12.9
CV%	37%	84%	19%	15%	29%
Med	512	107	9.9	8.4	13.2
CI95	255-953	23.5-552	7.6-14.4	6.7-10.8	7.1-21.1

n = number of soil cores used for the analysis; AM = arithmetic mean; CV = coefficient of variation; CI95 = 95% critical interval, log-normal frequency distribution anticipated; reference date for  $^{137}\text{Cs}$  activity: 1.1.2007. Bq/kg data refer to dried (105°C) soil samples.

**Table 2:** Ratios  $^{239+240}\text{Pu} : ^{137}\text{Cs}$  and  $^{238}\text{Pu} : ^{239+240}\text{Pu}$ . Reference date 1 Jan 07.

	$^{239+240}\text{Pu} : ^{137}\text{Cs}$	$^{238}\text{Pu} : ^{239+240}\text{Pu}$
this work, sample 2007:	0.0445	0.034*
BUNZL & KRACKE 1988	0.0290 ± 0.0039	
PERKINS 1980		0.035
MITCHELL et al. 1990, and references there	0.034 – 0.041	

\*) lowest layer (22-30 cm) removed as outlier

The results for important radionuclides are summarised in tables 1 and 2. Each soil core is considered as one sample for the statistics of table 1. The  $^{137}\text{Cs}$  inventory (530 Bq/m<sup>2</sup>) is thus very similar to the value predicted for global fallout by UNSCEAR (540 Bq/m<sup>2</sup>), but considerably lower than the one estimated from AOYAMA et al. (2006) of 1040 Bq/m<sup>2</sup>. In our investigation, the soil profiles were cut at 15 cm depth (samples of 1996), or 30 cm depth (sample 2007). Therefore our results under-estimate the true inventory systematically, since a certain fraction of the total  $^{137}\text{Cs}$  inventory may have migrated to deeper soil depths already. For the 1996 samples, this systematic error has been estimated at about 10%; for the 2007 sample (which was taken to greater soil depth for this reason), it is much smaller. The natural radionuclides  $^{40}\text{K}$ ,  $^{234}\text{Th}$  and  $^{226}\text{Ra}$  (from

the  $^{238}\text{U}$  series) and  $^{232}\text{Th}$  are roughly homogeneously distributed in the soil column, so mean activity concentrations are quoted in the table. This is of course not true for anthropogenic ( $^{137}\text{Cs}$ , plutonium) or natural ( $^{210}\text{Pb}$ ,  $^7\text{Be}$ ) fallout radionuclides.

Some of the variables can be considered approximately normally or log-normally distributed. The cumulative distributions of the  $^{137}\text{Cs}$  inventories (Bq/m<sup>2</sup>) and mean (over the soil profiles)  $^{40}\text{K}$  concentrations (Bq/kg) are shown in figure 2 together with the fitted log-normal distributions. The log-normal fits were used to estimate the critical limits (CI95) shown in table 1.

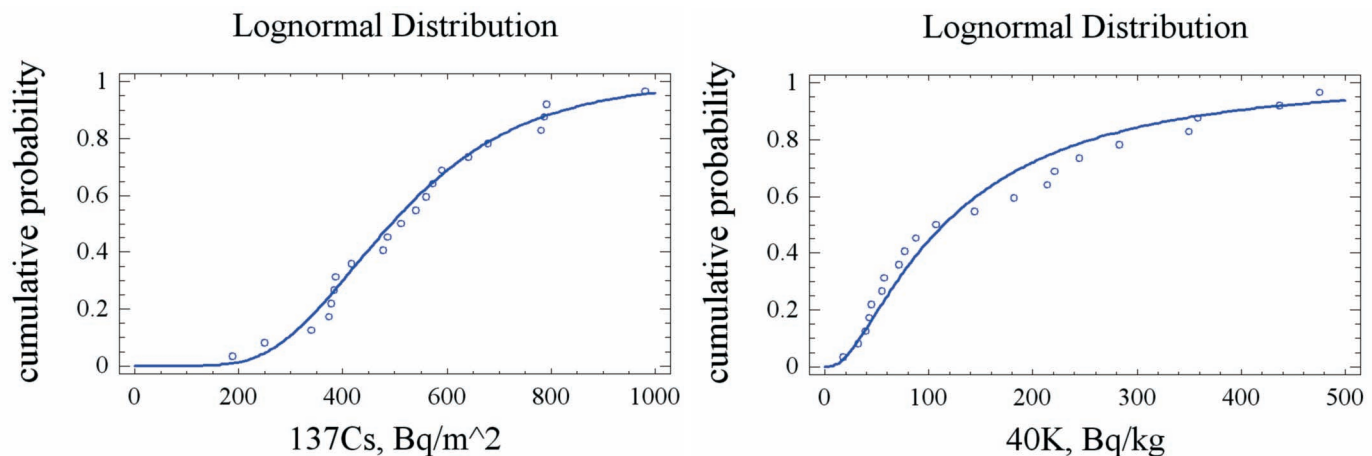
We believe that the  $^{137}\text{Cs}$  is more or less entirely derived from global fallout. Based on existing literature, it is estimated that the Chernobyl contribution is only about 1.5% (for details see BOSSEW & STREBL 1996).

Table 2 shows the ratios (ratio between inventories = weighted means of layers) of  $^{239+240}\text{Pu} : ^{137}\text{Cs}$ , and the plutonium isotopic ratio. The latter appears to be perfectly in line with literature data, while the plutonium:caesium ratio is somewhat higher than expected. For the vertical distribution of this ratio and further discussion see also chapter 3.3 below.

### Horizontal variability

The diameter of the sampling sites of 1996, i.e. of the area within which the 5 individual soil cores were taken, is between 10 and 30 m, while the diameter of the entire sampling area is approximately 1 km. The variabilities of the  $^{137}\text{Cs}$  inventories within the sampling sites, the variability between all soil cores, the mean variability within the sites and the variability between the sites are plotted against the diameters of the respective sampling sites in figure 3.

The results are similar with results found in literature for  $^{137}\text{Cs}$  inventories on grassland sites in Europe; no ref-



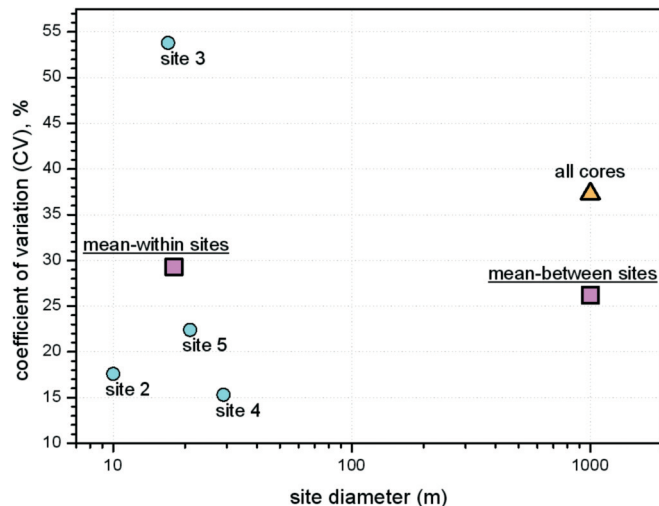
**Fig. 2:** Cumulative frequency distributions of  $^{137}\text{Cs}$  inventories and  $^{40}\text{K}$  concentrations, and fitted log-normal distribution. Each point represents one soil core.

erence data are available for tropical rainforests. However, for pure global fallout one would have expected a “smoother” distribution, i.e. more evenly distributed values and lower coefficients of variance. Three qualitative reasons for the observed variability may be given:

- Site 3 is located on the bottom of a narrow gorge, where soil redistribution must be expected to lead to a redistribution of deposited  $^{137}\text{Cs}$ , resulting in enhanced spatial variability. The site was deliberately chosen for examining whether such an effect can be observed. Sites 2, 4 and 5 are located on flat terrain on hill ridges, where no significant redistribution is anticipated.
- Deposition in forests is controlled not only by the mean rainfall intensity, which can be assumed to be rather uniform over small distances, but also by interception by vegetation canopy. Depending on the characteristics of the vegetation cover, deposition of fallout radionuclides can show considerable local variability at the soil surface: dense canopy may shield off fallout, while stem run-off could lead to increased wet deposition with collected rainwater around tree trunks.
- Finally, the sampling and measuring procedure involves some uncertainty which we estimate at about 27% per soil layer (details in BOSSEW 1996) and 16% per soil core, for the samples of 1996. The mean pure spatial variability within sites is therefore,

$$\sqrt{(\text{mean CV}_{(\text{within})})^2 - 16^2} = \sqrt{[29.3^2 - 16^2]} = 24.5\%$$

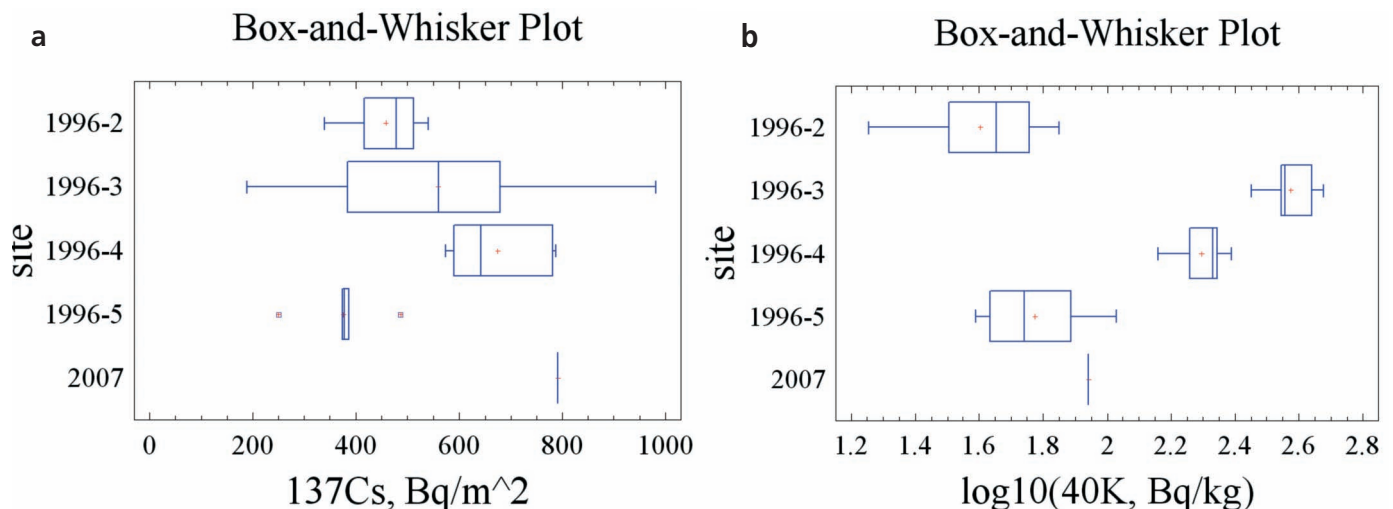
Knowing about spatial variability of a  $^{137}\text{Cs}$  soil inventory at different scales is important if this radionuclide is to be used for erosion studies. In such studies, the differences between inventories at two locations (e.g. top and bottom of a hill) are interpreted as the result of soil (incl. radionuclide) loss processes.



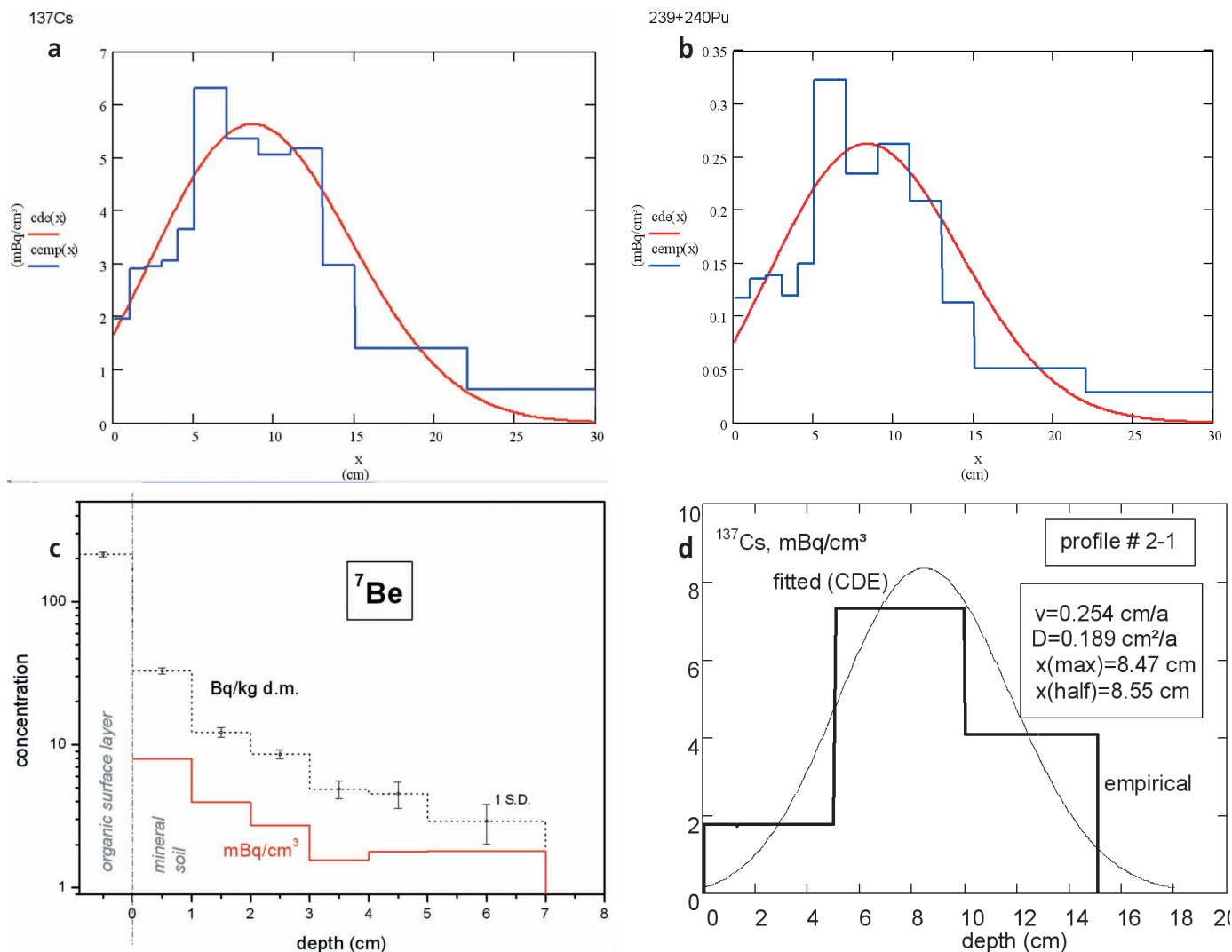
**Fig. 3:** Variabilities of  $^{137}\text{Cs}$  inventories within and between sampling sites.

Our data are not sufficient, however, to decide if differences between  $^{137}\text{Cs}$  inventories per site are significant, even with a mean CV between sites of 26.2% (figure 3). On the other hand, the Kruskal-Wallis test for medians results in a significant ( $p = 0.033$ ) difference between the sites. Further investigations would be necessary to decide on this but given the strongly structured topography of the region, and possibly micro-climatic variability resulting in a variability of mean rain intensity, a significant variability of  $^{137}\text{Cs}$  deposition within a 1 km sized area does not appear implausible.

For  $^{40}\text{K}$ , the sampling sites are significantly (figure 4b) different (ANOVA for  $\log_{10}(^{40}\text{K})$ :  $p < 0.05$ ). The reason may be found in different nutrient status of the soils at investigated sites. Also in respect to  $^{238}\text{U}$  and its progeny  $^{226}\text{Ra}$ , the sites seem to be different, although much less pronounced than for  $^{40}\text{K}$ . Finally, in respect to  $^{232}\text{Th}$ , the sites are homogeneous.



**Fig. 4:** Box-and-whisker plots to visualise the variability of (a)  $^{137}\text{Cs}$  inventories and (b)  $^{40}\text{K}$  concentrations within and between sites. + ... AM; box: lower / upper quartile, divided by the median; the whiskers extend to 1.5 interquartile distance.



**Fig. 5:** Vertical distributions (profiles) of radionuclides in soil: (a) profile 2007,  $^{137}\text{Cs}$ ; (b) profile 2007,  $^{239+240}\text{Pu}$ ; (c) profile 2007,  $^7\text{Be}$ ; (d) profile 2-1 of 1996,  $^{137}\text{Cs}$ .

**Table 3:** Parameters of the vertical distributions of  $^{137}\text{Cs}$  and Pu in the soil columns taken in 2007 and 1996.  $^{137}\text{Cs}$  decay corrected 1 Jan 2007.  $\pm 1 \text{ S.D.}$

	unit	$^{137}\text{Cs}$	$^{239/240}\text{Pu}$	$^{238}\text{Pu}$ (*)	$^{137}\text{Cs}$
sample 2007					1996
<b>empirical</b>					
inventory	$\text{Bq/m}^2$	791	35.2	1.1	$517 \pm 193$
maximum	cm	6	6	10	$5.3 \pm 2.9$
AM	cm	10.4	11.1	8.4	
SD	cm	6.3	7.0	4.9	
Median	cm	8.4	8.0	7.5	$7.4 \pm 1.3$
mean velocity	$\text{cm/yr}$	0.25	0.26	0.20	
dispersion	$\text{cm}^2/\text{yr}$	0.96	1.17	0.58	
median velocity	$\text{cm/yr}$	0.20	0.19	0.18	$0.24 \pm 0.04$
<b>CDE-estimates</b>					
$v$	$\text{cm/yr}$	0.17	0.16	0.13	$0.14 \pm 0.09$
$D$	$\text{cm}^2/\text{yr}$	0.51	0.47	0.65	$0.79 \pm 0.49$
$J_0$	$\text{Bq/m}^2$	791	35.2	1.2	

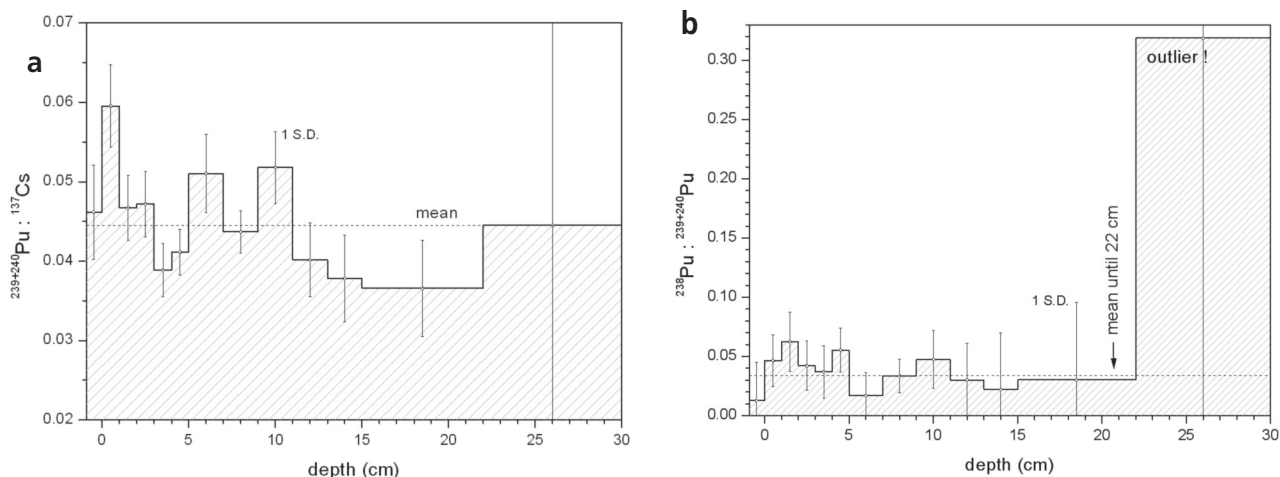
(\*)layer (22-30 cm) removed as implausible outlier for analysis.

## Vertical distribution

Substances which are deposited on the soil surface can be transported into the soil column by different processes, among which are percolation with rain water, diffusion in the soil solution, and soil redistribution by soil fauna (bioturbation). Caesium and plutonium are chemically reactive in most soil systems and tend to be fixed strongly on clay minerals which are ubiquitous in soils. Therefore, most of the deposited fallout radionuclides are still present in the upper soil column, in fact not far below the surface. Figures 5a, 5b and 5d show the distributions of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  in soil columns: although deposition occurred more than 40 years ago, the bulk of the contaminants still lies within the upper 15 cm of the soil column.

The curves in Figures 5a, 5b and 5d represent the result of fitting the convection-dispersion equation to measured radionuclide values for each soil layer.





**Fig. 6:** Depth distributions, (a) of the ratio  $^{239+240}\text{Pu} : ^{137}\text{Cs}$ ; (b) of the ratio  $^{238}\text{Pu} : ^{239+240}\text{Pu}$ , in the soil taken in of 2007.

In Table 3, the results of the CDE model for soil inventory and vertical migration parameters convection velocity  $v$  and dispersion coefficient  $D$  are compared to empirically measured values from the sampling campaigns in 1996 and 2007.

The migration velocities, 0.17 cm/yr as derived from the CDE fit, are similar to the ones derived from the samples taken in 1996,  $0.14 \pm 0.09$  cm/yr (0.09-0.16). Such slow migration of caesium and plutonium is typical for most soils (for a recent survey see, e.g., KIRCHNER et al. 2008).

Also, the general shape of the radionuclide depth profiles (Fig. 5a and 5b) is typical for undisturbed forest soils. Lower radionuclide values in superficial soil layers are the result of losses due to downward migration and root uptake of radionuclides. Since the deposition event has ended and no “new” caesium or plutonium is introduced to the soil surface, concentrations and respective radionuclide inventories slowly decrease in superficial soil layers.

The cosmogenic radionuclide  $^7\text{Be}$  has a relatively short half-life, 53.3 days. Its distribution in the soil column is approximately exponential (figure 5c), which is characteristic for nearly continuous fallout. The fact that it is detectable down to 7 cm depth points to efficient percolation with rain water.

From the measured data, integrated  $^7\text{Be}$  fluxes of  $3.9 \text{ Bq/m}^2/\text{d}$  or  $97 \text{ Bq/m}^2/\text{month}$  can be derived. These quantities are in agreement with data reported in literature.

Further, we investigate the depth distribution of ratios for the radionuclides  $^{239+240}\text{Pu} : ^{137}\text{Cs}$  and  $^{238}\text{Pu} : ^{239+240}\text{Pu}$ . The results are shown in Figure 6.

The ratio between  $^{137}\text{Cs}$  and Pu tend to become smaller with increasing depth. Although the availability of caesium for uptake via plant roots is rather limited,

because it strongly fixes to soil components (clay, organic matter), it is much higher than for plutonium (STREBL et al. 2007). If caesium were selectively extracted by plant roots, which tend to be denser in superficial soil layers, the Pu:Cs ratio would be higher.

Chernobyl fallout, which contained very little plutonium (except in the near zone around the NPP), arrived much later (in 1986) than the bomb fallout (1950-1960), in which the fraction of plutonium is higher, and the ratio to  $^{137}\text{Cs}$  is rather constant worldwide. If (additional) caesium had been deposited from the Chernobyl plume in our investigation site, ratios of Pu:Cs would therefore be lower in superficial soil layers than in deeper layers. This is evidently not the case, which leads to the conclusion that Chernobyl fallout, if any, is negligible in our investigation site, compared to global fallout from atmospheric nuclear weapon tests.

However, the observed variation in the Pu:Cs ratio is not statistically significant, so these considerations are rather theoretical.

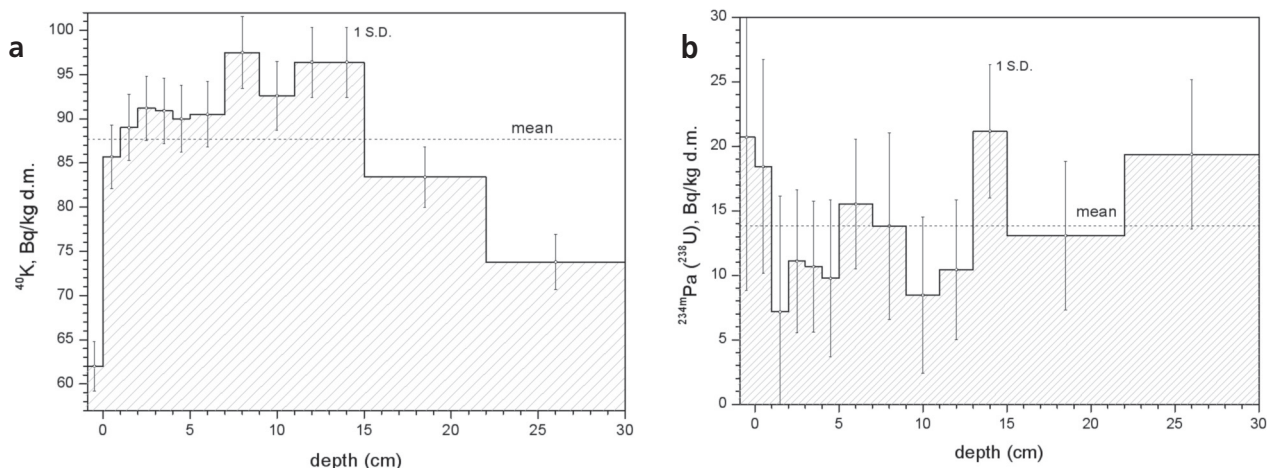
Finally, the depth distributions of the natural radionuclides  $^{40}\text{K}$  and  $^{238}\text{U}$  are shown in figure 7.

The mean (over the soil column)  $^{40}\text{K}$  and  $^{238}\text{U}$  concentrations are  $88 \pm 10 \text{ Bq/kg d.m.}$  and  $14 \pm 5 \text{ Bq/kg d.m.}$ , respectively. The results agree with the findings from the soil samples taken in 1996 (Table 1).

**Table 4:** Organic surface layer, statistical parameters

quantity	unit	AM	CV	Median	CI95
area density	kg d.m. / m <sup>2</sup>	1.10	57%	0.91	0.40-2.30
area activity concentration, $^{137}\text{Cs}$	Bq/m <sup>2</sup>	2.54	105%	2.07	0.48-6.99
mass activity concentration, $^{137}\text{Cs}$	Bq/kg d.m.	2.37	69%	1.89	0.59-6.14

d.m. = dry matter; AM = arithmetical mean; CV = coefficient of variation; CI95 = 95% critical interval, log-normal frequency distribution anticipated; reference date of activity: 1.1.2007.



**Fig. 7:** Depth distributions of natural radionuclides, (a)  $^{40}\text{K}$ , and (b)  $^{238}\text{U}$ .

### The organic surface layer

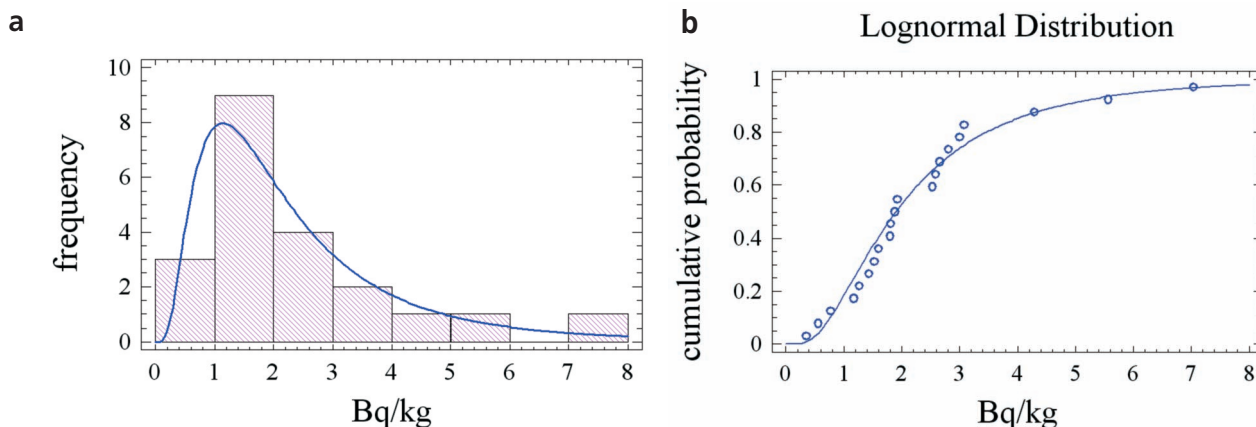
The organic surface layer consists of litter from the vegetation growing above. In a tropical rainforest, this litter is typically decomposed very quickly and recycled into the living biomass, so that practically no humus layer exists, unlike in temperate forests. Instead, the organic litter layer covers the mineral soil rather loosely. Its density is quite low, as we found a mean of only 1.1 kg dry matter / m<sup>2</sup> over the sampling sites.

Table 4 summarises the statistics of the area density of the organic layer and the  $^{137}\text{Cs}$  contained therein.

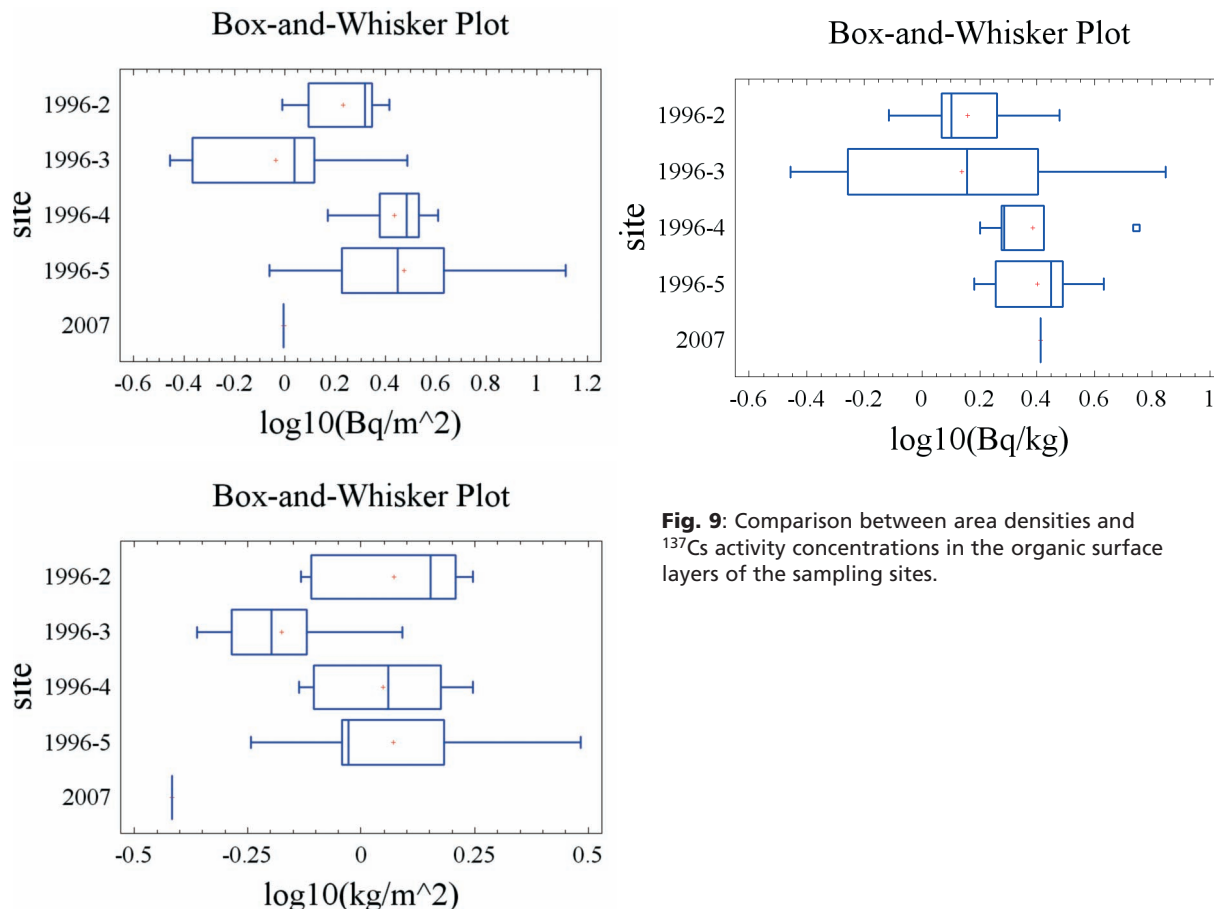
The frequency distribution of  $^{137}\text{Cs}$  in the surface layer is again approximately log-normal (figure 8). As shown in figure 9, area density of the surface layer and  $^{137}\text{Cs}$  concentrations appear quite different between the sampling sites; however the differences indicate no significant grouping, as can be shown with the analysis of variance (ANOVA) technique.

Interpretation of the findings is difficult, as long as little is known about the ecological role of the surface layer, its interaction with underlying soil, or recycling time of organic matter. The radionuclides found in the surface layer may have several sources:

- Redistribution of soil substance by animals like digging earthworms, or by mechanical translocation (re-suspension) due to rain fall (“soil splash”);
- Contamination of the organic material with soil matter by inaccurate sampling;
- Transfer of radionuclides from soil via roots to the plants above, from which the organic matter is derived;
- Traces of the original fallout onto the plant surface (i.e. the interception by the leaves), stored in or on the plant and subsequently contained in organic litter (this is only applicable for trees which are at least about 45 years old);



**Fig. 8:** (a) frequency histogram of  $^{137}\text{Cs}$  activity concentration (Bq/kg d.m.) in the organic surface layer; (b) cumulative probability and log-normal fit.



**Fig. 9:** Comparison between area densities and  $^{137}\text{Cs}$  activity concentrations in the organic surface layers of the sampling sites.

- Finally, trace substances like radionuclides may be transferred to the organic layer from the underlying soil by fungal mycelia.

In any case, more research about the notoriously complicated soil-plant interface would be necessary to clarify the subject.

## Conclusions and outlook

We have acquired information on the horizontal and vertical distributions of natural and anthropogenic radionuclides in the soil of the Esquinas rainforest area.

The results gained in these two zero-budget mini-projects can, at best, give first information on radionuclide inventories in an environment, where almost no radiometric data exist. Indeed, our results of the 1996 campaign have already been incorporated into a global fallout database (AOYAMA et al. 2006). There remains a lot to do in order to establish a robust radiological database of the region:

It is not known how representative the results are for the area, despite estimates of the horizontal variability. This would require more samples, also based on a better knowledge of the environment.

The area sampled so far extends to about  $1\text{ km}^2$ . Considering the high heterogeneity of the landscape in this region (or natural terrain in general), the representativeness of the results – if given at all – is locally restricted.

Virtually nothing is known about radio-ecology of agricultural soils in the region.

Finally, to our knowledge, nothing at all is known about the radio-ecology of plants in the region, for example, soil to plant transfer factors.

Since this would be basic research with limited immediate practical value, there is little chance of realisation; on the other hand, the existing good infrastructure of the site, and of Costa Rica altogether, would deem it perfect for that kind of research.

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### Addresses of authors:

Peter BOSSEW  
Institute for Environment and Sustainability (IES)  
European Commission Joint Research Centre (JRC)  
Via Fermi 1  
I-21020 Ispra, Italy  
E-mail: peter.bossew@jrc.it

Alexander HUBMER  
Division of Physics and Biophysics  
University of Salzburg  
Hellbrunner Straße 34  
A-5020 Salzburg, Austria  
E-mail: alexander.hubmer@sbg.ac.at

Friederike STREBL  
Division Radiation Safety and Applications  
Austrian Research Centers GmbH – ARC  
A-2444 Seibersdorf, Austria  
E-mail: friederike.strebl@arcs.ac.at

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