

# Forest Soil Development as investigated by Radioisotope Distributions

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## 1 Introduction

Soil can be considered as a set of layers which differ essentially in their physical and chemical properties. During the process of forest soil evolution, organic matter is supplied as litter and further decomposed by biologic activity. After structural destruction of plant remnants humic matter migrates via the soil solution into weathered mineralic subsoil. This results in a typical sequence of soil horizons: litter (L), fermented (Of) and humified (Oh) organic layers and mixed horizons (A) containing both mineralic and humic material. Occurrence and thickness of the organic horizons are governed by the decomposition rates and depend on environmental conditions like climate, availability of nutrients, litter composition etc., thus determining the appearing humus type. As a consequence of the occurring physicochemical conditions the upper part of the mineralic soil undergoes weathering processes leading to destruction of mineral structures and release of major and minor elements. Especially the lower organic and mixed horizons of forest soils are exposed to the impact of organic acids at low pH values down to pH 3.

Dynamic structures like forest soils can be advantageously studied by radiotracer methods. Experiments using the addition of artificial radionuclides are limited for obvious reasons to special investigation areas. Instead of this, the transuranic and fission nuclides distributed globally by the atmospheric nuclear weapons tests until 1963 and in Europe by the Chernobyl accident in 1986 may serve for the same purpose. The supply of those radionuclides took place in a limited time interval and for some of the nuclides

data are available about the deposited activity per area (EUROPEAN COMMISSION 1998; HARDY et al. 1973; BUNDESAMT FÜR STRAHLENSCHUTZ, FACHBEREICH STRAHLENSCHUTZ 1992).

Nuclides from the natural decay series are able to provide additional information because of their differences in the chemical properties. Dynamic processes in varying chemical environments lead to accumulation and depletion of the individual nuclides. The resulting radioactive disequilibria are suited as fingerprints of the occurring differentiation processes.

In the present paper the investigations should be focused on the following topics:

- (i) are the distributions of artificial radionuclides in forest soils stable enough to serve at present as markers of the time of their input?
- (ii) is it possible to establish a quasicontinuous time scale by applying <sup>210</sup>Pb dating on soil horizons?
- (iii) how do weathering processes influence the distribution of radionuclides in soil?

## 2 Sampling and Analytical Methods

All soil samples were taken *in situ* as slices of 1 - 2 cm thickness using a steel frame of 30 x 30 cm<sup>2</sup> area or a ring of 40 cm diameter to sample a defined area. This thin layer method allows the detection of small scale variations in the nuclide distributions within the profile. The material was dried at 40 °C for at least 48 h and after that it has been homogenised. Roots and stones larger than 5 mm were removed.

Table 1: Summary of the radionuclides detected and analysed by low level  $\gamma$ -spectrometry in soil samples from Saxony.

| Radionuclide   | Half life          | Origin  |
|--|--------------------|---|
| $^{238}\text{U}$ (via $^{234}\text{Th}$ , $^{234\text{m}}\text{Pa}$ )              | $4.5 \cdot 10^9$ y | geogenic, $^{238}\text{U}$ -series  |
| $^{230}\text{Th}$  | $7.5 \cdot 10^4$ y | geogenic, $^{238}\text{U}$ -series  |
| $^{226}\text{Ra}$ (via $^{214}\text{Pb}$ , $^{214}\text{Bi}$ )                     | $1.6 \cdot 10^3$ y | geogenic, $^{238}\text{U}$ -series  |
| $^{210}\text{Pb}$  | 22 y               | geogenic, airborne input ( $^{222}\text{Rn}$ -daughter), $^{238}\text{U}$ -series |
| $^{227}\text{Ac}$ (via $^{227}\text{Th}$ , $^{223}\text{Ra}$ , $^{219}\text{Rn}$ ) | 22 y               | geogenic, $^{235}\text{U}$ -series  |
| $^{228}\text{Ra}$ (via $^{228}\text{Ac}$ )   | 5.8 y              | geogenic, $^{232}\text{Th}$ -series   |
| $^{228}\text{Th}$ (via $^{212}\text{Pb}$ , $^{212}\text{Bi}$ , $^{208}\text{Tl}$ ) | 1.9 y              | geogenic, $^{232}\text{Th}$ -series   |
| $^{40}\text{K}$  | $1.3 \cdot 10^9$ y | geogenic  |
| $^7\text{Be}$  | 53 d               | cosmogenic  |
| $^{125}\text{Sb}$  | 2.8 y              | artificial, Chernobyl fallout   |
| $^{134}\text{Cs}$  | 2.1 y              | artificial, Chernobyl fallout   |
| $^{137}\text{Cs}$  | 30 y               | artificial, Nuclear weapon tests and Chernobyl fallout                            |
| $^{207}\text{Bi}$  | 32 y               | artificial, Nuclear weapon tests  |
| ( $^{241}\text{Pu} \Rightarrow$ ) $^{241}\text{Am}$                                | 430 y              | artificial, Nuclear weapon tests  |

For the radionuclide analysis two low-level- $\gamma$ -spectrometry systems containing a 36 % p-type and a 38 % n-type HPGe detector were used, respectively. The sample material was filled into gas proof measuring containers of cylindrical shape or into Marinelli beakers. In most cases, long measuring times up to 48 h were necessary. The radionuclides detected in soil samples are given in Table 1 together with their origin. All radioisotopes could be determined simultaneously. Note that some members of the natural decay series were analysed via their  $\gamma$ -emitting daughter nuclides.

### 3 Sampling Sites

The radionuclide profiles in this paper were recorded at four sites in Saxony (Germany) intensively studied in the frame of the Level II European monitoring program (*Olbernhau 1*, *Laußnitz*, *Colditz* and *Bad Schandau*) (RABEN et al. 2000), at a site in the vicinity of a monitoring station of the Leipzig university (*Leipzig*) and in a larch forest near *Nassau* in the Erzgebirge mountains. Characteristics of the locations and of the investigated soils are summarised in Table 2. The chosen soils are typical for the Saxon area (SÄCHSISCHE LANDESANSTALT FÜR FORSTEN 2000) as well as the humus types which represent a broad field of degradation conditions ranging from fast decomposition of plant residues during one year (mull) to hindered disintegration in acidified soils (raw humus).

Because of the long measuring times for the  $\gamma$ -spectrometry only one soil section per sampling

site could be taken. To get a widely representative profile of the sampled forest soil, preferably smooth and only little sloped sampling points at a minimum distance of 1.5 m from stems and centred between several trees were carefully selected.

## 4 Results and Discussion

### 4.1 General features of the investigated profiles

Fig. 1 illustrates typical activity distributions of radionuclides as found in similar shape in many of the observed soil sections. The great advantage of the thin layer sampling for the identification of accumulation and migration processes is clearly visible. Some features of the curves were generally found and can be summarised as follows:

#### 4.1.1 naturally occurring nuclides:

- The radioactive equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  is disturbed in the upper layers, i.e. in the O- to B-horizons. Below that it is balanced and represents the activity level of the geological background
- $^{210}\text{Pb}$  supplied as Rn-daughter mostly from the atmosphere to the soil surface does reach its equilibrium to  $^{226}\text{Ra}$  first in the mineralic layers. The excess in the organic and mixed layers is enormous and amounts to some hundreds of Bq per kg.

Table 2: Description of the locations and of the sampled forest soils in Saxony (Germany). The soil type is given according to the German classification (ARBEITSGEMEINSCHAFT BODENKUNDE 1994).

|                          | <b>Olbernhau 1</b>           | <b>Laußnitz</b>             | <b>Colditz</b>                    | <b>Bad Schan-<br/>dau</b>     | <b>Leipzig</b>                  | <b>Nassau</b>                   |
|--------------------------|------------------------------|-----------------------------|-----------------------------------|-------------------------------|---------------------------------|---------------------------------|
| geographic position      | top of eastern Erzgebirge    | 30 km north from Dresden    | 40 km south-east from Leipzig     | 30 km south-east from Dresden | southern part of Leipzig        | crest of eastern Erzgebirge     |
| altitude above sea level | 720 m                        | 170 m                       | 185 m                             | 260 m                         | 140 m                           | 705 m                           |
| geological background    | orthogneiss                  | aqueoglacial deposits       | porphyry                          | basalt                        | sediment                        | Muscovite gneiss                |
| vegetation               | spruce trees, grass          | pine trees, grass           | oak trees, grass                  | beech trees                   | mixed flood plain forest, grass | larch trees, grass              |
| soil type                | silty loam-braunerde-podzol  | sand-podzol                 | fine-sand-similigley              | braunerde                     | vega-gley                       | Braunerde                       |
| humus type               | raw humus - moder            | raw humus                   | moder                             | moder                         | mull                            | raw humus                       |
| sampling date            | July 26 <sup>th</sup> , 1999 | July 1 <sup>st</sup> , 1999 | September 20 <sup>th</sup> , 1999 | May 29 <sup>th</sup> , 2001   | June 13 <sup>th</sup> , 2000    | October 10 <sup>th</sup> , 2000 |

- The nuclide  $^{40}\text{K}$  is contained in natural potassium. Its distribution thus depicts the fraction of mineralic constituents in the soil matter. The sharp increase in the uppermost layers results from uptake by plants.

#### 4.1.2 artificial nuclides:

- $^{137}\text{Cs}$  is detectable over the whole profile and represents the main activity of the artificial nuclides. The total activity contains contributions both from the weapons tests and from Chernobyl.
- The activity of the other caesium nuclide  $^{134}\text{Cs}$  is about two orders of magnitude lower due to its shorter half life. Therefore, it falls at present below the detection limit in most of the samples.  $^{134}\text{Cs}$  was only supplied by Chernobyl fallout. The  $^{137}\text{Cs}/^{134}\text{Cs}$  ratio at the time of the Chernobyl accident amounted to 1.7 - 2.0 according to the literature (HÖTZL et al. 1987; WINKELMANN et al. 1986), own measurements on air filter samples gave a value of 1.9. This fact allows the separation of the total  $^{137}\text{Cs}$  activity into the fractions from the weapons tests and from Chernobyl, respectively.
- In the soil sections the determination of  $^{125}\text{Sb}$  now becomes difficult due to its short half life, comparable to that of  $^{134}\text{Cs}$ .
- The fallout from atmospheric weapons tests before 1963 is represented by  $^{241}\text{Am}$  and  $^{207}\text{Bi}$ . The former is a daughter nuclide of the  $\beta$ -emitter  $^{241}\text{Pu}$  (half life 14.4 y). Due to radioactive decay the activity of  $^{241}\text{Am}$  grows continuously and will reach a maximum in the year 2037 (APPLEBY et al. 1991).  $^{207}\text{Bi}$  is assumed to be produced by (d,xn)-reactions in the lead tamper or shield of thermonuclear weapons. Up to now, only little is known about the deposited  $^{207}\text{Bi}$  activity in the environment.

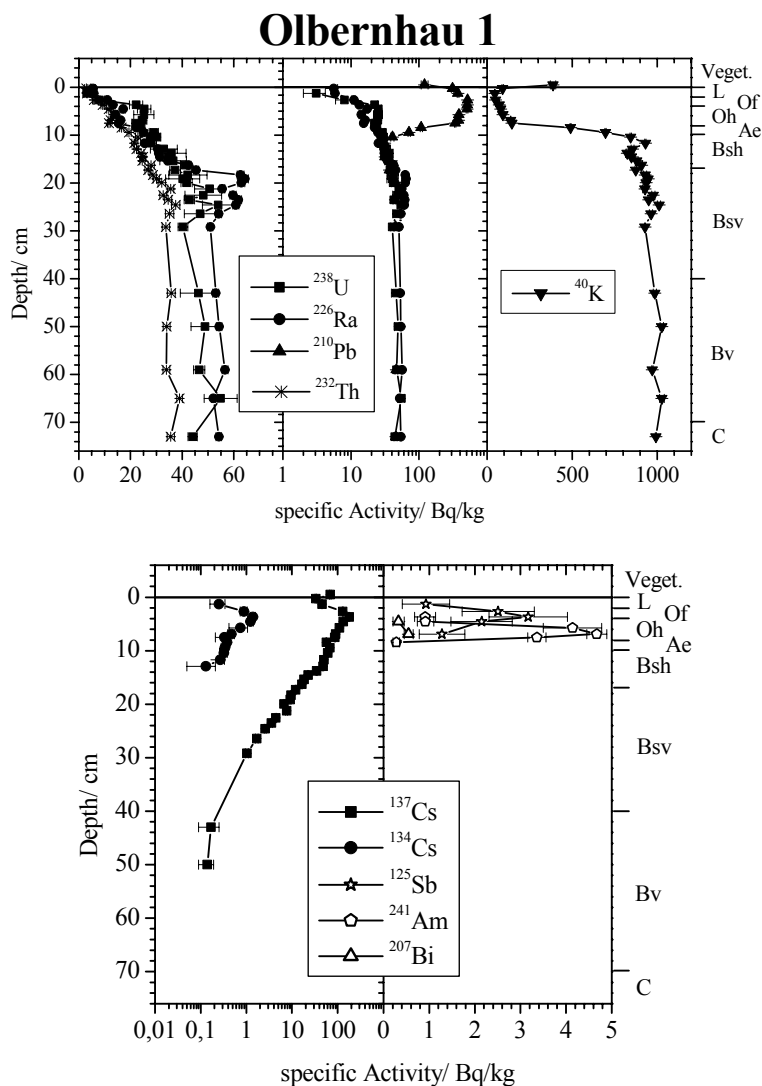


Figure 1: Distribution of natural (above) and artificial (below) radionuclides at sampling site Olbernhau 1. The soil horizons are classified at the right scale.

## 4.2 Distributions of artificial radionuclides and the time of deposition

Activity profiles of artificial nuclides at sites showing different humus and soil types are compared in Fig. 2.

Since the sampling method allowed the determination of the layer volumes, the results were given in terms of an activity concentration, i.e. in Bq per m<sup>2</sup> sampled area and cm layer thickness. This quantity illustrates more plastically the activity distribution in the sections as the mass-based specific activity in Bq/kg does.

The graphs of the <sup>137</sup>Cs activities are double peaked for the soils at *Olbernhau 1*, *Laußnitz* and *Colditz*. Whereas the first maximum appears in the organic Of/Oh horizons the second one is situated in mixed or mineralic layers (A(e,h)-Bsh). As calculated from the <sup>134</sup>Cs activities the upper peak contains mainly <sup>137</sup>Cs from Chernobyl accident, the lower one includes both contributions. Although the currently low <sup>134</sup>Cs activities cause large errors for the determination of the Chernobyl fraction, it is clearly visible that in layers containing the <sup>241</sup>Am peak, the Chernobyl <sup>137</sup>Cs concentration is low whereas a residue of <sup>137</sup>Cs from the weapons fallout is detected there. In the soil at *Leipzig* <sup>137</sup>Cs is broadly distributed in the Ah horizon with a large part of Chernobyl Cs at the maximum position.

Caesium as an alkali metal shows properties similar to potassium (SCHALLER et al. 1993) i.e. it is taken up by plants and fungi and strongly sorbed to clay minerals. Cs is not known to tend to complexation with organic matter. Therefore a two-stage-process is proposed for the interpretation of the  $^{137}\text{Cs}$  profiles: during the first step Cs is incorporated into plants and remains in the dead material until it is decomposed. After this Cs moves via the soil solution downwards and is sorbed onto the surfaces of clay minerals. So the

upper (Chernobyl-) $^{137}\text{Cs}$  peak is assigned to 15 year old litter debris. The retention of  $^{137}\text{Cs}$  from nuclear weapons tests in older organic horizons may be a consequence of direct transfer from the humus phase to the mineral content of this layer.

In general, in the forest soils except for the *Leipzig* site  $^{125}\text{Sb}$  is detected in those layers which also contain the first maximum of Chernobyl  $^{137}\text{Cs}$ . So we assume a migration behaviour similar to that of  $^{137}\text{Cs}$ .

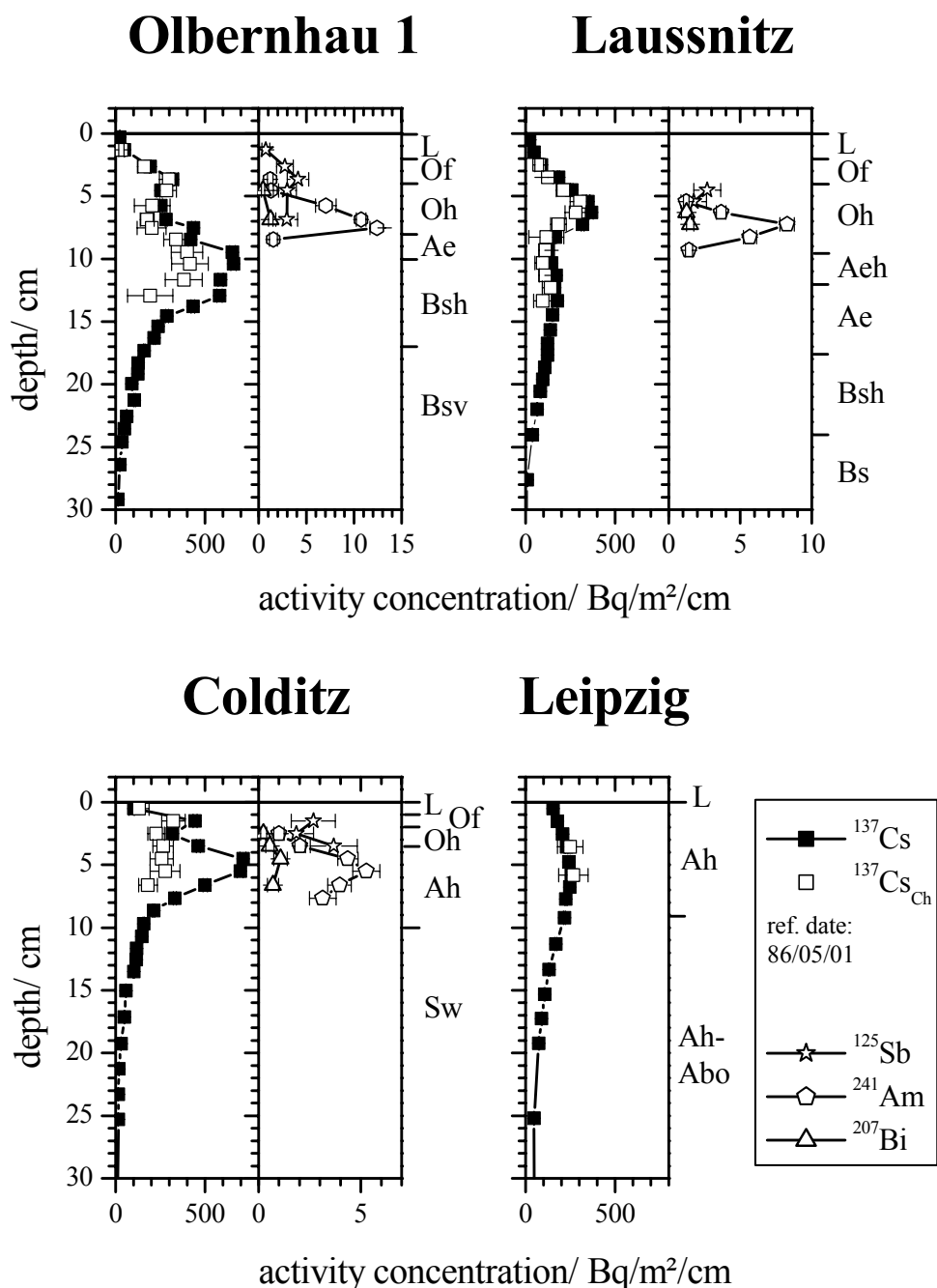


Figure 2: Distributions of artificial radionuclides at various sampling sites in Saxony. Values for  $^{137}\text{Cs}$  are decay-corrected to May 1st, 1986. The Chernobyl fraction ( $^{137}\text{Cs}_{\text{Ch}}$ ) of the total activity was calculated using a  $^{137}\text{Cs}/^{134}\text{Cs}$  ratio of  $(1.9 \pm 0.1)$  for fresh fallout.  $^{241}\text{Am}$  and  $^{207}\text{Bi}$  fell below detection limit at site *Leipzig*. The soil horizons are classified at the right scale.

The measured profiles and the above discussion specify the upper  $^{137}\text{Cs}$  maximum as a rather transient time marker for the Chernobyl accident.

Completely different conclusions can be drawn for the bomb nuclides: The  $^{241}\text{Am}$  distributions do not differ substantially between the sites *Laußnitz* and *Olbernhau 1*. The activity is concentrated at these similar sites in a single peak of 3 - 4 cm thickness located at the lower edge of the organic Oh horizon. At *Colditz* the Am-peak is found in the mixed Ah horizon with a rather blurred bottom side. In contrast to this,  $^{241}\text{Am}$  disappears totally at site *Leipzig*, i.e. its activity falls down below the detection limit (about 1 Bq/m<sup>2</sup>/cm) in all layers.

Our measurements show  $^{207}\text{Bi}$  distributions in the soils parallel to that of  $^{241}\text{Am}$ . Its activity concentrations are unfortunately close to the detection limit so that detailed comparisons to the Am profile are not feasible. Nevertheless,  $^{207}\text{Bi}$  is only included in layers also containing  $^{241}\text{Am}$ . If an organic layer is missing (*Leipzig*)  $^{207}\text{Bi}$  is again not detectable.

Americium exists in the environment in the oxidation state III and forms - just as its parent nuclide  $^{241}\text{Pu}$  - stable complexes with organic ligands (KIM et al. 1989). It can be expected that the migration of the heavy metal nuclide  $^{207}\text{Bi}$  is governed also by organic complexes as observed i.e. for lead and uranium. For this reason, it is reasonable to assume that, if  $^{241}\text{Am}$  and  $^{207}\text{Bi}$  were complexed in an initial state of litter decomposition, their plots reflect the distribution of organic material from the period of the nuclear weapons tests. Two facts support this hypothesis:

(i) From the data published in (HARDY et al. 1973; KREY et al. 1976) one can estimate  $^{241}\text{Am}$  inventories between 16 and 40 Bq/m<sup>2</sup> at January, 1st, 2000 for central Europe. The total  $^{241}\text{Am}$  inventories (Table 3) agree well with that data; i.e. at least a significant part of the primary input remained strongly fixed in the humus layer.

Table 3: Actual  $^{241}\text{Am}$  inventories in the sampled forest soils.

|                    | Total $^{241}\text{Am}$ inventory in Bq/m <sup>2</sup> |
|--------------------|--|
| <b>Laußnitz</b>    | 20.0 ± 1.7   |
| <b>Olbernhau 1</b> | 29.4 ± 3.4   |
| <b>Colditz</b>     | 21.0 ± 3.0   |

(ii) The nuclide distributions are narrow in Oh horizons but blurred in the Ah horizon of *Colditz* and disappear in the *Leipzig* profile. This con-

forms to the fate of plant residues - stacked as deposited in the O horizons but displaced as humate into the A horizons after further decomposition.

According to this discussion the  $^{241}\text{Am}/^{207}\text{Bi}$  peaks are well suited i.e. stable time markers for the beginning of the 60's. With their aid, the age of the lower part of the humus layer in the *Laußnitz* and *Olbernhau* sections can now be estimated to about forty years. At *Colditz* the humus turnover is accelerated and obviously finished after this period.

### 4.3 Dating of soil horizons by $^{210}\text{Pb}$

The observed  $^{210}\text{Pb}$  excess in the organic layers gives reason to the attempt of dating the organic horizons by means of the constant-rate-of-supply (CRS) model (APPLEBY & OLDFIELD 1978) which is successfully applied e.g. for lake sediments.

Lead as a heavy metal is well known for its ability to form stable organic complexes. So it is justifiable to presume as for  $^{241}\text{Am}/^{207}\text{Bi}$  a complexation with organic material deposited at the same time like  $^{210}\text{Pb}$  containing aerosols. In contrast to the fallout nuclides this supply is continuously with an at least approximately constant rate. Applying the CRS model to the soil system, its correctness can be controlled by comparison with the  $^{241}\text{Am}/^{207}\text{Bi}$  peaks ( $\approx 40$  y before sampling) and possibly with the upper (Chernobyl-) peak of the  $^{137}\text{Cs}$  distributions ( $\approx 15$  y before sampling).

The results in Fig. 3 for the sites *Olbernhau 1*, *Laußnitz* and *Colditz* show a good agreement between the age determination using  $^{210}\text{Pb}$  and the time markers. Only the Cs-peak at site *Laußnitz* does not fit to the results which is probably related to the low clay content at this site. No  $^{210}\text{Pb}$  excess was observed at site *Leipzig* where the organic layer is only a few millimetres thick.

Using these data, one can obtain conclusions about the turnover dynamics of the organic matter in the investigated soils. Tab. 4 summarises the ages at the base of the individual organic horizons. The lower decomposition rate of organic matter in Oh horizons of raw humus (*Olbernhau 1*, *Laußnitz*) compared to moder (*Colditz*) is clearly visible and emphasises the hampered microbial activity in acidified forest soils.

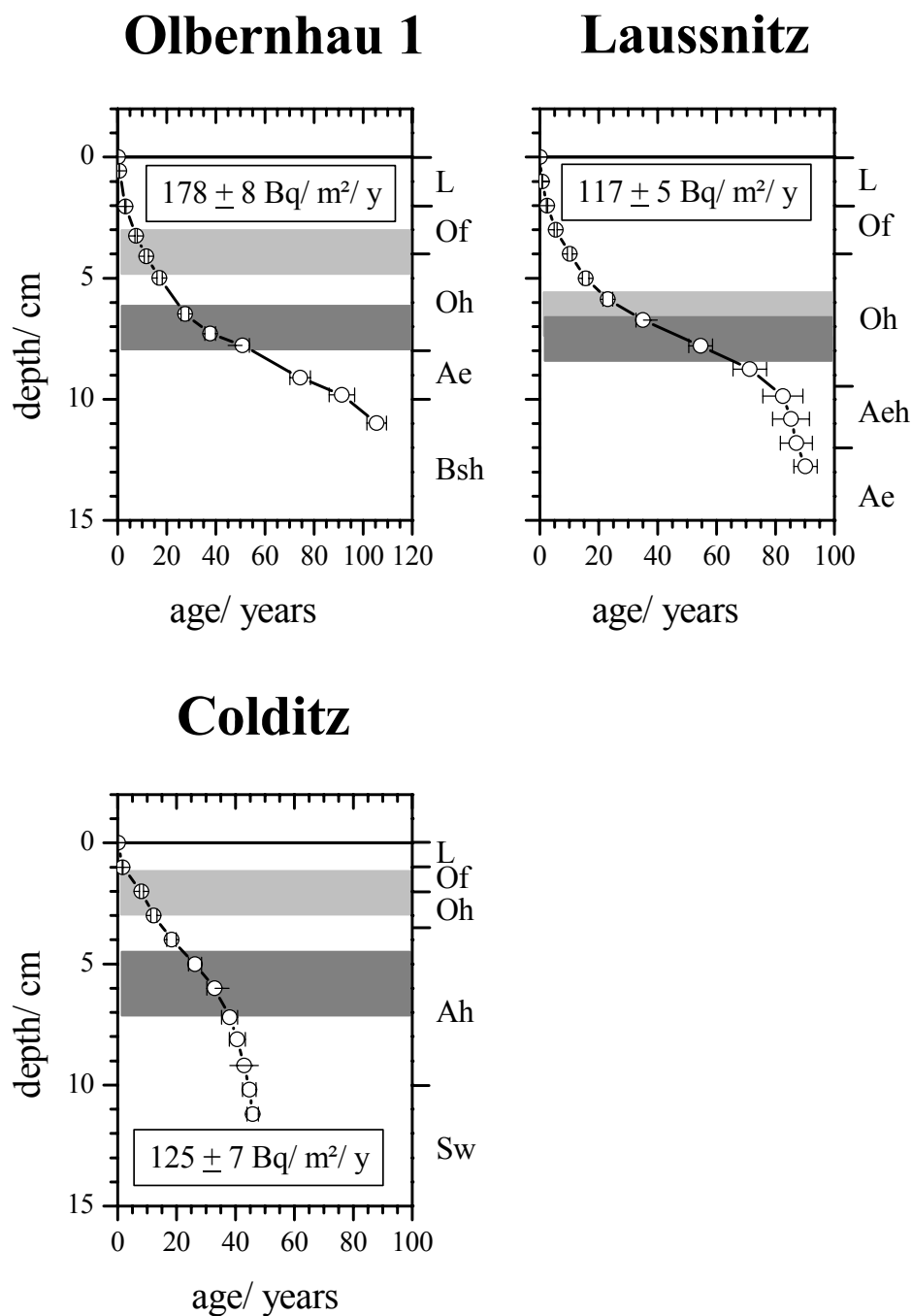


Figure 3: Results of  $^{210}\text{Pb}$  age determination on soil layers using the CRS model. The mean activity supply per area and time as calculated from the total excess of unsupported  $^{210}\text{Pb}$  is given in the boxes. Light and dark grey bars indicate the position of the upper (Chernobyl-)  $^{137}\text{Cs}$  peak ( $\approx 15$  y before sampling) and of the  $^{241}\text{Am}/^{207}\text{Bi}$  peak ( $\approx 40$  y before sampling), respectively.

Table 4:  $^{210}\text{Pb}$  ages at the lower border of the organic horizons. The humus type at *Olbernhau 1*, *Laußnitz* and *Colditz* was raw-humus-moder, raw humus and moder, respectively. Dating failed at site *Leipzig* (humus type: mull) because of vanishing  $^{210}\text{Pb}$  excess.

| Horizon | Olbernhau 1 | Laußnitz | Colditz |
|---------|-------------|----------|---------|
| L       | 3 y         | 2 y      | 2 y     |
| Of      | 11 y        | 9 y      | 8 y     |
| Oh      | 54 y        | 50 y     | 15 y    |
| A(e,h)  | 94 y        | 80 y     | 46 a    |

#### 4.4 Radioactive disequilibria and weathering

The compilation of  $^{226}\text{Ra}/^{238}\text{U}$  ratios in a number of soils (Fig. 4.) shows in some cases remarkable disequilibria. Generally, three different behaviours can be distinguished:

- **Olbernhau 1 & Nassau:** obvious U-accumulation in Oh horizons; in mineralic horizons Ra-accumulation
- **Laußnitz & Colditz:** U-accumulation to a little extent in Oh horizons; in general no disequilibrium
- **Bad Schandau & Leipzig:** little overall Ra-accumulation

These effects are currently not completely understood. A possible explanation results from the presence of organic acids and low pH values (down to  $\approx$  pH 3) in Oh layers of raw humus which may lead to enhanced weathering of min-

eralic particles. Traces of the discharged heavy metal Uranium are able to form organic complexes whereas Radium migrates to deeper layers and sorbs onto clay particles. Weathering should be less intensive in soils with higher quartz content as found in *Laußnitz* and in soils where an accelerated humus decomposition is connected with higher pH values (*Colditz*, *Bad Schandau*, *Leipzig*). This could explain the vanishing disequilibria in those sections. Further systematic work is necessary to clarify the effects leading to radioactive disequilibria in the upper soil layers.

## 5 Conclusions

The processes of soil evolution affect especially in the organic and mixed horizons the migration behaviour of natural and artificial radionuclides. Their distributions along a soil section can be successfully used for the determination of turnover rates of the soil organic matter. The degree of weathering processes is reflected by radioactive disequilibria in the  $^{238}\text{U}$  decay series. Acknowledgement

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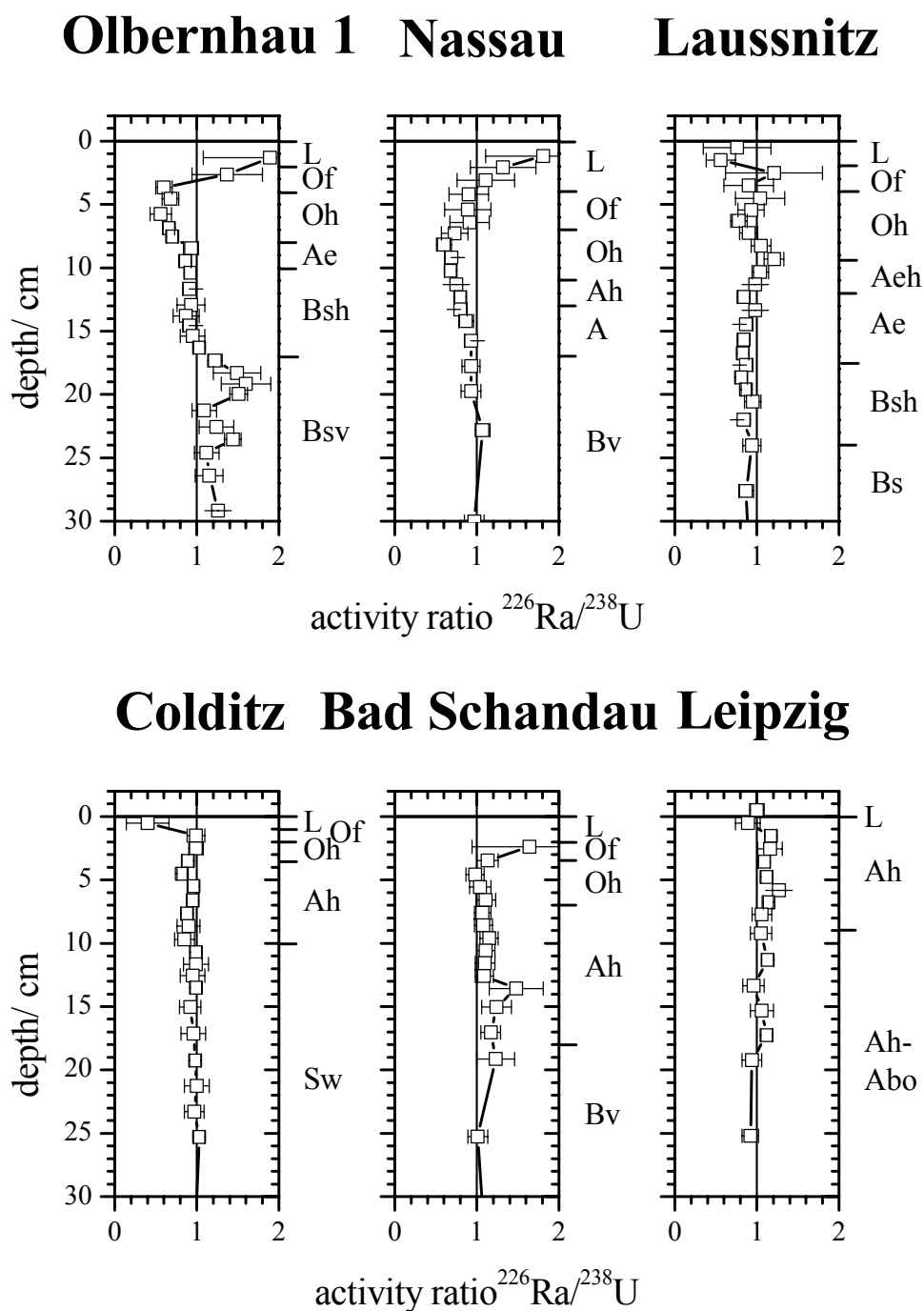


Figure 4: Distribution of the activity ratio  $^{226}\text{Ra}/^{238}\text{U}$  in the investigated soils.

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