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Vertical distribution of ^{238}Pu, $^{239(40)}$Pu, ^{241}Am, ^{90}Sr and ^{137}Cs in Austrian soil profiles

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Summary. The occurrence of ^{238}Pu, $^{239(40)}$Pu, ^{241}Am, ^{90}Sr and ^{137}Cs in the region of Nassfeld (Salzburg, Austria) is discussed at four different locations by evaluating a depth profile for each radionuclide. Furthermore, the plutonium separated from the soil samples was measured by Accelerator Mass Spectrometry (AMS) to get information on the isotope ratio ^{240}Pu/^{239}Pu. The radiochemical procedure consisted of a Pu separation step from Am and Sr by anion exchange in 8 M HNO_3 after oxidation state adjustment to Pu(IV). Am and Sr were coprecipitated with calcium oxalate. Am was separated from Sr by extraction chromatography using TRU-resin. The Sr separation was performed also by extraction chromatography using Sr-resin; after coprecipitation as oxalate, ^{90}Sr was measured in a gas proportional counter. For the determination of $^{239(40)}$Pu and ^{241}Am by α-spectrometry thin sources were prepared by microprecipitation with NdF_3.

With the respective Pu isotope ratios, it was possible to identify the global fallout as source of the contamination. This was confirmed by the ratio ^{241}Am/$^{239(40)}$Pu. From the activity ratio ^{90}Sr/^{137}Cs, it could be shown that most of these radionuclides stem from the reactor accident in Chernobyl.

1. Introduction

The global fallout from atmospheric weapons testing during the 1950s and 1960s of the last century and releases from nuclear power plants and spent fuel reprocessing plants are the main sources for anthropogenic nuclides in the environment. After the reactor accident at Chernobyl on April 26th, 1986, Austria and especially some of its alpine regions also received atmospheric deposition from this source [1–3]. Soil or sediment samples from these regions can thus be expected to hold contributions both from weapons test fallout and the Chernobyl accident.

Concerning the transuranium elements, the investigators attention is usually focused on the α-emitting Pu isotopes ^{238}Pu, ^{239}Pu and ^{240}Pu because of their high radiotoxicity; ^{239}Pu and ^{240}Pu cannot be separated by α-spectrometry due to their very similar α-energies, so the sum activity of $^{239(40)}$Pu is measured. The β-emitting ^{241}Pu, however, is much more abundant in environmental samples and, furthermore, its decay product ^{241}Am is a highly radiotoxic α-emitter with a half-life of 433 years. Nowadays, a little bit more than 40 years after the deposition of the maximum bomb-fallout Pu activities, the ingrown ^{241}Am has reached 85% of its maximum value and can be measured conveniently. So, a retrospective determination of the originally present fallout ^{241}Pu is possible.

The isotopic signature of the various Pu isotopes depends on the production process. So, from the derived activity ratios, a decision concerning the source term of Pu should be possible as the ratios vary with reactor type and nuclear fuel burn up [4]; the ^{238}Pu/$^{239(40)}$Pu activity ratio of the Chernobyl releases is known to be about 0.48 [4, 5]. With respect to weapons test fallout samples, the activity ratios depend on the fissionable material used and the type of weapons test [6, 7]; from global fallout samples on the northern hemisphere, it was derived between 0.025 and 0.04 [8–11]. For the measurement of the ^{240}Pu/^{239}Pu atom ratio which can give direct information on the source ^{239}Pu content, mass spectrometric methods like ICP-MS and accelerator mass spectrometry (AMS) are necessary [12–14]. In this work, the atom ratio ^{240}Pu/^{239}Pu was measured by accelerator mass spectrometry (AMS).

The typical setup of an AMS (Accelerator Mass Spectrometer) consists of two mass spectrometers coupled by a particle accelerator and, among the various methods of mass spectrometry, achieves the best background suppression. This can be attributed to the destruction of molecular isobars (molecular ions with the same mass as the ion of interest, *e.g.* $^{238}U^{16}OH^-$ for $^{239}Pu^{16}O^-$) and to the high ion energy (MeV) in the second spectrometer [4]. Whether mass spectrometry or decay counting is the method of choice for a certain isotope depends not only on the half-life. For instance, ^{240}Pu/^{239}Pu cannot be measured by alpha-spectrometry, since the alpha-energies cannot be resolved. On the other hand, ^{238}Pu cannot be detected by mass spectrometry at environemental levels due to the unavoidable

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contamination by natural ^{238}U. Generally, AMS measurements impose a much larger effort.

Considering the fission products, ^{90}Sr and especially ^{137}Cs are important environmental contaminants. Again their origin is the bomb fallout as well as the reactor accident at Chernobyl and nuclear fuel reprocessing. Both nuclides enter the food chain at least when freshly deposited; when they migrate into the soil they become strongly attached to soil particles and are no longer available for uptake by plants [15–17]. However, the migration velocity is strongly dependent on the soil species and can be considerably slower in alpine regions compared to regions at lower heights. While ^{137}Cs is easily measured γ-spectrometrically and therefore international data banks exist [18], there is still a gap in the data with regard to ^{90}Sr as its determination requires more effort. ^{90}Sr data from roe deer exist from pre- and after-Chernobyl times [19–21] while in Austria after the Chernobyl accident ^{90}Sr and other nuclides were determined in surface air [3] and ^{90}Sr/^{137}Cs and also $^{239(40)}$Pu/^{137}Cs ratios derived; however, ^{90}Sr soil data are scarce.

The aim of our work was to measure the Pu isotopes ^{238}Pu and $^{239(40)}$Pu as well as ^{241}Am in four soil profiles from the Salzburg province, Austria; also the fission products ^{137}Cs and ^{90}Sr were determined. Variations of deposited activities as well as of migration velocities of the respective nuclides in the soil column with these site parameters were investigated. The calculated ^{238}Pu/$^{239(40)}$Pu and ^{241}Am/$^{239(40)}$Pu activity ratios and the measured ^{240}Pu/^{239}Pu atom ratios enabled us to derive the source term of Pu; also the origin of ^{137}Cs and ^{90}Sr could be identified by the ^{90}Sr/^{137}Cs activity ratio.

2. Materials and methods

The samples were collected on an alpine pasture along the slope from the Schareck summit (3122 m a.s.l.) down to the Nassfeld near Böckstein (Salzburg province, Austria); the sites differed in elevation and also with respect to their orientation (see Fig. 1). Soil samples were collected at four different locations (T2, T5, T8 and T9) on August 25th, 1999.

Table 1. Altitude and coordinates of sampling sites.

Location	Altitude [m] right/high [m]	Coordinates BMN (M31):
T2	2530	427 260/212 370
T5	2280	427 660/211 850
T8	1850	428 330/212 850
T9	1790	428 380/211 990

Table 1 gives the height above sea level and the coordinates of the respective sites. T2 and T5 are from sites with clearly higher elevation compared to sites T8 and T9. T2 and T8 are situated on the outer bending of the slope facing to north or north-east, while T5 and T9 are situated near the indent between the “sample-hill” and the next hill, facing to south and south-east.

2.1 Sample preparation and measurement

The certified reference materials NIST SRM 4334G and NIST SRM 4332D, were obtained from the National Institute of Standards & Technology (Gaithersburg, USA). A stock solution of ^{236}Pu was obtained from the University Helsinki and calibrated against SRM 4334G by alpha spectrometry.

Roots and stones were separated from the air-dried soil samples by sieving and the ^{137}Cs activity concentration was measured with a Germanium detector Canberra BeGe 3830. Canberra DSA 2000 and measurement software Genie 3.1 (Canberra, USA) were used for spectrum evaluation. The soil samples were measured in a standard geometry, a beaker with 6.3 cm diameter and 150 mL volume. Depending on the activity level of the samples, the acquisition time was in the range from 30 000 to 150 000 s.

Two reference samples (IAEA-135 and IAEA-soil-6) were investigated with respect to Pu in order to check the validity of our results. After drying in porcelain dishes to constant weight in an oven set to 105 °C, samples (20 g) were ashed in porcelain dishes in an electric muffle furnace at 420 °C. The ashes were weighed and dissolved in 200 mL

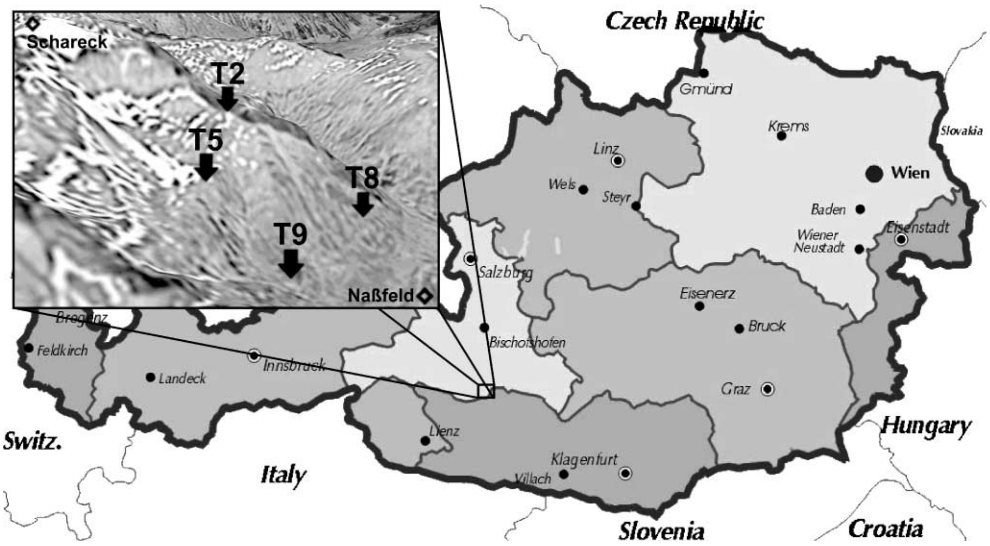

Fig. 1. Sampling locations.

8 M HNO_3 and ^{236}Pu and ^{243}Am tracer solutions and Sr^{2+} carrier were added. The chemical separation of the respective nuclides followed the procedure given in [22, 23].

2.2 Alpha and beta counting

Alpha spectrometry was performed with an ORTEC PIPS ULTRA-AS-450 detector with an active area of 450 mm^2. The counting time was 80 000 s. The software Genie 2.1 (Canberra, USA) was used for the evaluation of the spectra.

^{90}Sr was determined by gas proportional counting (OG 50, Tracerlab, Germany). The samples were measured for 10 000 s. Sample purity was checked by the increasing countrate due to the ingrowth of ^{90}Y.

2.3 Preparation and measurement of AMS samples

After alpha spectrometry measurements, the filter carrying the Pu sample was reprocessed for AMS. It was dissolved in 5 mL 65% HNO_3 and 2 mL 30% H_2O_2 in a Teflon beaker and evaporated for three times with the same reagents. The residue was fumed with 5 mL 37% HCl (3×) and then taken up in 3 mL solution containing 0.5 mg Fe/mL 37% HCl. This solution was transferred to a 50 mL vial, the Teflon beaker was washed with 8 mL deionised water and this solution was also added to the vial. By adding 25% NH_3 drop by drop, Pu was coprecipitated with $Fe(OH)_3$. The solution was heated softly on a hot plate for about 15 minutes, the pH was reduced to 9. After centrifuging, the precipitate was dried at 100 °C in an electric oven for about 2 h. The dried precipitate was combusted at 800 °C in a porcelain cup. A few mg of silver powder were added as a binder and the resulting samples were pressed into aluminium sample holders suitable for the ion source of VERA.

VERA, the Vienna Environmental Research Accelerator, is based on a 3-MV tandem accelerator [24]. The mass spectrometers are especially designed to allow the transport of ions of all elements, from the lightest to the heaviest. Molecular PuO^- ions are extracted from the solid sample in a caesium sputter source. After the first mass selection, the PuO^- is accelerated to the positive accelerator terminal and the molecules are broken up by gas stripping, which also efficiently removes any molecular isobars. Pu^{7+} (24 MeV) ions are selected in the second mass spectrometer. The ions of interest and interfering ions which pass all beam filters are identified with a high-resolution time-of-flight system with 3 m flight path and a final ionization chamber. The preference of the 7+ charge state over the more abundant 5+ (1% *vs*. 5% stripping yield, respectively) is based on the fact that $^{144}Nd^{3+}$ (entering in the co-precipitation step) with the same mass-over-charge ratio as $^{240}Pu^{5+}$ cannot be distinguished in the mass spectrometer and would overload the detector. For the 7+ charge state, no such interference is possible.

VERA can only count one ion species at a time, so six alternating counting intervals of typically 1 to 5 min on $^{240}Pu^{7+}$ and $^{239}Pu^{7+}$ are performed before proceeding to the next sample. Measurements on the sample set are repeated until the material is completely sputtered (about 1 mg/h) or the required measurement precision is reached. The total detection efficiency achieved is 1 Pu^{7+} event detected per 5×10^4 atoms of plutonium in the ion source.

3. Results and discussion

Table 2 shows our results of IAEA-135 and IAEA Soil-6 reference materials in comparison to the certified values. As the activity concentrations of the investigated samples are in the order of magnitude of the Soil-6 activity, the excellent agreement with this standard indicates the accuracy of our procedure.

Results of the plutonium and americium measurements are summarized in Table 3. For $^{239(40)}$Pu, the activity concentrations are given, while ^{238}Pu results are shown as ^{238}Pu/$^{239(40)}$Pu activity ratios, calculated for the year 1986. The yield of the chemical procedure estimated *via* the ^{236}Pu recovery was in the range of 50%–87%; about two thirds of the samples had yields larger than 65%. Americium results are given as activity ratios ^{241}Am/$^{239(40)}$Pu as measured in 2006. The chemical yield for the americium procedure was 60%–100%; 15 out of 21 samples showed values higher than 70%.

The Pu depth profiles show marked differences in activity concentrations and in depth of the activity maximum depending on site situation as well as on site elevation. Generally, the samples T2 and T8 show higher activity concentrations than samples T5 and T9. $^{239(40)}$Pu activity concentration maxima of 24 and 32 Bq/kg are found on sites T2 and T8 compared to 10 and 8.5 Bq/kg on sites T5 and T9. In these findings, the situation of the sites is reflected: T2 and T8 are situated on the outer bending of the slope facing to north or north-east, the weather-side, while T5 and T9 are situated near the indent between the “sample-hill” and the next hill, facing to south and south-east. So the “sample-hill” is shielding the indent, reducing the deposited activity by a factor of about 3. The plutonium maximum is found in 2.0–2.5 cm and 2.0–3.0 cm below the surface on sites T5 and T2, whereas it is 2.8–5.2 cm and 3.7–4.5 cm depth on sites T8 and T9. T8 and T9 are at 1850 and 1790 m a.s.l., whereas T5 and T2 are clearly higher at 2280 and 2530 m a.s.l.; obviously, the Pu penetration depth is due to the nature of the soil which is differing with site elevation.

The ^{238}Pu/$^{239(40)}$Pu activity ratio derived from the α-spectrometric measurements is between 0.02 and 0.05 in the soil layers where the larger portions of the Pu activities are found, this corresponds to global fallout where a ^{238}Pu/$^{239(40)}$Pu activity ratio of 0.025–0.04 is given in literature [8–11]. Higher values of 0.21–0.23 are also found in the uppermost layers of site T8, giving hints of a contribution from the Chernobyl accident: in Chernobyl fuel particles, a ^{238}Pu/$^{239(40)}$Pu activity ratio of 0.48 was measured [4, 5], while in surface air from Austria, values between 0.25 and 1.14 were found when the Chernobyl emissions reached Austria; about 50% of the air samples showed activity ratios between 0.4 and 0.6 [3]. The AMS results

Table 2. Activities of $^{239(40)}$Pu standards: comparison of certified values and measured values.

Sample	95% Confidence interval [Bq/g]	Measured value [Bq/g]	$\pm 1\sigma$
IAEA-135	0.205–0.226	0.224	0.009
IAEA-Soil-6	9.6×10^{-4}–1.11×10^{-3}	1.09×10^{-3}	1.70×10^{-4}

Table 3. Analysis results of plutonium and americium measurements. The $^{238}\text{Pu}/^{239(40)}\text{Pu}$ activity ratio is calculated for 1986, while $^{241}\text{Am}/^{239(40)}\text{Pu}$ is given as measured in 2006, $^{240}\text{Pu}/^{239}\text{Pu}$ is given as atom ratio, measurement uncertainties are given at $\pm 1\sigma$.

T2 Depth [cm]	$^{239(40)}\text{Pu}$ [Bq/kg]	$\frac{^{238}\text{Pu}}{^{239(40)}\text{Pu}}$	$\frac{^{241}\text{Am}}{^{239(40)}\text{Pu}}$	$\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$
6.6–8.1	0.34 ± 0.03	< 0.10	–	0.27 ± 0.10
5.5–6.6	0.51 ± 0.04	< 0.05	0.37 ± 0.08	0.19 ± 0.02
4.5–5.5	1.12 ± 0.07	0.044 ± 0.009	0.13 ± 0.02	0.16 ± 0.03
3.7–4.5	2.9 ± 0.2	0.028 ± 0.004	0.19 ± 0.02	0.26 ± 0.10
3.0–3.7	12.6 ± 0.7	0.020 ± 0.002	0.23 ± 0.02	0.22 ± 0.05
2.0–3.0	31.7 ± 1.9	0.025 ± 0.002	0.21 ± 0.02	0.16 ± 0.01
1.1–2.0	15.5 ± 0.9	0.034 ± 0.004	0.19 ± 0.01	0.21 ± 0.02
0–1.1	–	–	–	–

T5 Depth [cm]	$^{239(40)}\text{Pu}$ [Bq/kg]	$\frac{^{238}\text{Pu}}{^{239(40)}\text{Pu}}$	$\frac{^{241}\text{Am}}{^{239(40)}\text{Pu}}$
7.6–9.0	0.14 ± 0.02	< 0.25	–
6.4–7.6	0.16 ± 0.05	< 0.15	–
5.4–6.4	0.27 ± 0.09	< 0.12	–
4.3–5.4	0.56 ± 0.18	0.084 ± 0.034	–
3.3–4.3	1.21 ± 0.39	0.058 ± 0.027	0.20 ± 0.07
2.5–3.3	3.0 ± 1.0	0.031 ± 0.016	0.53 ± 0.18
2.0–2.5	9.9 ± 3.2	0.019 ± 0.009	0.47 ± 0.16
1.0–2.0	3.8 ± 1.2	0.052 ± 0.025	1.68 ± 0.54
0–1.0	5.9 ± 1.9	0.028 ± 0.006	–

T8 Depth [cm]	$^{239(40)}\text{Pu}$ [Bq/kg]	$\frac{^{238}\text{Pu}}{^{239(40)}\text{Pu}}$	$\frac{^{241}\text{Am}}{^{239(40)}\text{Pu}}$	$\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$
8.2–9.5	0.46 ± 0.03	< 0.12	–	0.19 ± 0.04
7.3–8.2	1.53 ± 0.09	0.059 ± 0.018	0.27 ± 0.03	0.17 ± 0.02
6.4–7.3	3.8 ± 0.2	0.044 ± 0.009	0.37 ± 0.03	0.18 ± 0.01
5.2–6.4	–	–	–	–
3.9–5.2	22.1 ± 1.3	0.038 ± 0.003	0.63 ± 0.04	0.17 ± 0.01
2.8–3.9	23.7 ± 1.4	0.044 ± 0.004	0.55 ± 0.04	0.14 ± 0.01
1.5–2.8	8.5 ± 0.5	0.062 ± 0.007	0.41 ± 0.03	0.16 ± 0.03
0.8–1.5	1.07 ± 0.08	0.214 ± 0.030	–	0.22 ± 0.04
0–0.8	0.88 ± 0.07	0.232 ± 0.043	–	0.51 ± 0.13

T9 Depth [cm]	$^{239(40)}\text{Pu}$ [Bq/kg]	$\frac{^{238}\text{Pu}}{^{239(40)}\text{Pu}}$	$\frac{^{241}\text{Am}}{^{239(40)}\text{Pu}}$
5.6–7.0	1.5 ± 0.3	0.062 ± 0.020	–
4.5–5.6	3.0 ± 0.6	0.043 ± 0.015	0.83 ± 0.17
3.7–4.5	8.4 ± 1.6	0.031 ± 0.008	–
2.6–3.7	6.2 ± 1.2	0.032 ± 0.010	0.44 ± 0.09
2.0–2.6	5.3 ± 1.0	0.049 ± 0.014	0.45 ± 0.09
1.1–2.0	3.4 ± 0.7	0.059 ± 0.016	0.27 ± 0.06
0–1.1	2.3 ± 0.5	0.043 ± 0.010	–

support our conclusions drawn from the activity ratios: the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio measured in samples from the sites T2 and T8 is between 0.14 and 0.22 for most of the soil layers, corresponding to the global fallout value of 0.175–0.19 from the literature [8–10]. Exceptions with higher values are the deepest layer of T2 and a layer in the middle of the core, both with low $^{239(40)}\text{Pu}$ activity concentration, and a high uncertainty of the atom ratio and the surface layer of T8. This last value of 0.51 ± 0.13 can be attributed to depositions from the Chernobyl accident.

^{241}Am has grown in from the much more abundant plutonium isotope ^{241}Pu since its deposition during the bomb test phase (a much smaller part might stem from the Chernobyl accident, see above). The activity ratio $^{241}\text{Pu}/^{239(40)}\text{Pu}$ was 4.2 in 1986 [25]; correcting for the ^{241}Pu half-life of 14.4 a, the value for 1963 was 12.6. The $^{241}\text{Am}/^{239(40)}\text{Pu}$ activity ratio is calculated according to the radioactive decay equation of a short living mother- and a long living daughter-nuclide by:

$$(^{241}\text{Pu}/^{239(40)}\text{Pu}) = \left(^{241}\text{Am}_{\text{t}}/^{239(40)}\text{Pu}\right)\,(432/14.4)\,f_{\text{corr}}\,,$$

where $1/f_{\text{corr}} = 1 - \text{e}^{-\ln 2(t/14.4)}$, a correction factor for the build-up of ^{241}Am after a period of t years since ^{241}Pu deposition. $^{241}\text{Am}/^{239(40)}\text{Pu}$ values of 0.28 and 0.37 are calculated for 1986 and 2006, respectively. From the Chernobyl fallout $^{241}\text{Pu}/^{239(40)}\text{Pu}$ activity ratio of 75 [2] a corresponding $^{241}\text{Am}/^{239(40)}\text{Pu}$ value of 1.5 can be calculated for the year 2006. From Table 3, it can be seen that our values are quite near to the global bomb fallout numbers, verifying again the

Table 4. Analysis results of ^{137}Cs and ^{90}Sr measurements (reference date: May 1st, 1986), measurement uncertainties are given at $\pm 1\sigma$.

T2 Depth [cm]	^{137}Cs [Bq/kg]	^{137}Cs [Bq/kg]	^{90}Sr/^{137}Cs
6.6–8.1	113 ± 7	< 80	< 0.7
5.5–6.6	142 ± 9	< 80	< 0.6
4.5–5.5	230 ± 14	53 ± 1	0.229 ± 0.015
3.7–4.5	394 ± 24	57 ± 4	0.145 ± 0.013
3.0–3.7	743 ± 45	58 ± 3	0.078 ± 0.006
2.0–3.0	1780 ± 110	87 ± 3	0.049 ± 0.004
1.1–2.0	4380 ± 219	97 ± 2	0.022 ± 0.001
0 –1.1	4320 ± 216	195 ± 13	0.045 ± 0.004

T5 Depth [cm]	^{137}Cs [Bq/kg]	^{137}Cs [Bq/kg]	^{90}Sr/^{137}Cs
9.0–10.2	49 ± 3	< 80	< 1.6
7.6– 9.0	64 ± 4	6.3 ± 0.2	0.097 ± 0.007
6.4– 7.6	85 ± 5	6.9 ± 0.2	0.081 ± 0.006
5.4– 6.4	108 ± 7	8.9 ± 0.2	0.082 ± 0.005
4.3– 5.4	157 ± 10	14.5 ± 0.6	0.092 ± 0.007
3.3– 4.3	295 ± 18	18.1 ± 0.3	0.061 ± 0.004
2.5– 3.3	622 ± 38	36 ± 1	0.058 ± 0.004
2.0– 2.5	1404 ± 85	48 ± 2	0.034 ± 0.002
1.0– 2.0	2810 ± 170	76 ± 2	0.027 ± 0.002
0 – 1.0	3790 ± 230	126 ± 1	0.033 ± 0.002

T8 Depth [cm]	^{137}Cs [Bq/kg]	^{137}Cs [Bq/kg]	^{90}Sr/^{137}Cs
8.2–9.5	614 ± 38	34.7 ± 0.7	0.056 ± 0.004
7.3–8.2	1131 ± 68	80.4 ± 1.3	0.071 ± 0.005
6.4–7.3	1496 ± 91	163 ± 3	0.109 ± 0.007
5.2–6.4	2330 ± 140	176 ± 3	0.075 ± 0.005
3.9–5.2	2960 ± 180	208 ± 3	0.070 ± 0.004
2.8–3.9	3630 ± 220	187 ± 2	0.052 ± 0.003
1.5–2.8	5910 ± 360	108 ± 3	0.018 ± 0.001
0.8–1.5	7760 ± 470	69 ± 2	0.009 ± 0.001
0 –0.8	6010 ± 360	88 ± 3	0.015 ± 0.001

T9 Depth [cm]	^{137}Cs [Bq/kg]	^{137}Cs [Bq/kg]	^{90}Sr/^{137}Cs
5.6–7.0	400 ± 24	24.2 ± 0.3	0.061 ± 0.004
4.5–5.6	711 ± 43	25.4 ± 0.5	0.036 ± 0.002
3.7–4.5	1427 ± 86	32.9 ± 0.5	0.023 ± 0.001
2.6–3.7	2140 ± 130	47 ± 2	0.022 ± 0.002
2.0–2.6	3210 ± 190	67 ± 1	0.021 ± 0.001
1.1–2.0	3220 ± 190	93 ± 1	0.029 ± 0.002
0 –1.1	2910 ± 180	123 ± 2	0.043 ± 0.003

fact that Pu contamination as consequence of the Chernobyl accident was nearly negligible in Austria.

In Table 4, the results of the ^{137}Cs and ^{90}Sr measurements are listed together with the ^{90}Sr/^{137}Cs activity ratios; the data are decay corrected with reference date of May 1st, 1986. Referring to the activity concentrations, we find a pattern similar to the plutonium isotopes: samples from sites T2 and T8 show higher levels than samples from T5 and T9. The T8 activity concentrations are clearly higher than those from T5 and T9, again reflecting the sample sites situations. The same can be said for ^{90}Sr: the activities summed over the entire soil core in T8 are three times as high as they are in T5 and T9 and also values from T2 are higher by factors 1.6 and 1.35, respectively. However, the positions of the activity concentration maxima are markedly different: the ^{137}Cs maxima are found in the surface layer (0.5 cm depth) in T5, in 1.15 cm depth in T8 and in two layers at depth 1.55 and 2.3 cm in T9. The ^{90}Sr maxima were found in the surface layer with the exception of T8; here the maximum is broader, covering 4 layers between 3.35 and 6.85 cm depth. The observation, that the ^{137}Cs and ^{90}Sr maxima are nearer to the soil surface compared to the plutonium isotopes, can be due to different behaviour of the respective nuclides in the soil and/or reflects the fact that the ^{137}Cs and ^{90}Sr deposition took place later than the plutonium deposition: while the Pu isotopes stem from the global bomb fallout in the 1950ies and 1960ies, the source of ^{137}Cs and ^{90}Sr was the Chernobyl accident in1986. This is verified by the ^{90}Sr/^{137}Cs activity ratios: while in bomb fallout the ^{90}Sr/^{137}Cs activity ratio is 0.61 [26], after Chernobyl, ratios between 0.012 and 0.017 were found in surface air [3]; the ratio in soil samples is expected to be higher by a factor of 2 [26], giving $(2.4–3.4) \times 10^{-2}$. This is comparable to our values presented here; the higher values especially in the deeper layers reflect an older contribution from the global fallout.

4. Conclusions

We found marked differences in the amount of deposited activity depending on the orientation of the sampling sites. The sample activities from the "weather-side" of the hill were much higher than the activities of samples from a "shielded" site; this holds for all investigated nuclides. With respect to the plutonium isotopes and to ^{241}Am, the maximum activities were found in layers nearer to the surface when investigating samples from higher elevations; in the downhill samples, the penetration depth of the nuclides was clearly larger. These findings reflect the differing of the soil types with height.

The ^{137}Cs and ^{90}Sr activity concentration maxima were found nearer to the soil surface compared to the Pu and Am maxima; often the maximum was detected in the uppermost layer. These findings suggest a different date of deposition for the Pu isotopes and the fission products, respectively. The same conclusion was drawn from the respective activity and atom ratios: the ^{238}Pu/$^{239(40)}$Pu and ^{241}Am/$^{239(40)}$Pu activity ratios as well as the ^{240}Pu/^{239}Pu atom ratios enabled us to derive the global weapons test fallout as the source of the Pu isotopes while the Chernobyl origin of ^{137}Cs and ^{90}Sr could be verified by the ^{90}Sr/^{137}Cs activity ratio.

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References

1. Bossew, P., Ditto, M., Falkner, T., Henrich, E., Kienzl, K., Rappelsberger, U.: Cäsiumbelastung der Böden Österreichs. In: *UBA Monographien Band 60*. Bundesministerium für Umwelt, Wien (1996).
2. Irlweck, K., Wicke, J.: Isotopic composition of plutonium immissions in Austria after the Chernobyl accident. J. Radioanal. Nucl. Chem. **227**, 133 (1998).
3. Irlweck, K., Khademi, B.: Radionuclide ratios of ^{90}Sr/^{137}Cs and $^{239(40)}$Pu/^{137}Cs in contaminated surface air after the Chernobyl accident in Austria. J. Radioanal. Nucl. Chem. **203**(1), 79 (1996).
4. Oughton, D., Day, P., Fifield, K.: Plutonium measurement using accelerator mass spectrometry: methodology and applications. In: *Plutonium in the Environment*. (Kudo, A., ed.) Elsevier, Amsterdam (2001).
5. Kutkov, V. A., Arefieva, Z. S., Murav'ev, Yu. B., Komaritskaya, O. I.: Unique form of airborne radioactivity: Nuclear fuel "hot particles" released during the Chernobyl accident. In: Environmental Impact of Radioactive Releases, Proceedings of an International Symposium on Environmental Impact of Radioactive Releases, IAEA, Vienna (1995).
6. Irlweck, K., Hrnecek, E.: ^{241}Am concentration and ^{241}Pu/$^{239(40)}$Pu ratios in soils contaminated by weapons-grade plutonium. J. Radioanal. Nucl. Chem. **242**, 595 (1999).
7. Yamamoto, M., Tsumura, A., Katayama, Y., Tsukatani, T.: Plutonium isotopic composition in soil from the former Semipalatinsk Nuclear Test Site. Radiochim. Acta **72**, 209 (1996).
8. Buesseler, K. O., Sholkovitz, E. R.: The geochemistry of fallout plutonium in the North Atlantic: II. Plutonium-240/plutonium-239 ratios and their significance. Geochim. Cosmochim. Acta **51**(10), 2623 (1987).
9. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), Report to the General Assembly with Scientific Annexes, UN-E.94.IX.2, United Nations, New York (1993).
10. Beasley, T. M., Kelley, J. M., Maiti, T. C., Bond, L. A.: ^{237}Np/^{239}Pu atom ratios in integrated global fallout: a reassessment of the production of ^{237}Np. J. Env. Radioact. **38**(2), 133 (1998).
11. Irlweck, K., Friedmann, Ch., Schönfeld, T.: Die Plutoniumkonzentration der bodennahen Atmosphäre in Wien in den Jahren 1962–1979. Mitteilungen der Österreichischen Sanitätsverwaltung **82**, 81 (1981).
12. Povinec, P. P., La Rosa, J. J., Lee, S. H., Mulsow, S., Osvath, I., Wyse, E.: Recent developments in radiometric and mass spectrometry methods for marine radioactivity measurements. J. Radioanal. Nucl. Chem. **248**, 713 (2001).
13. Hrnecek, E., Aldave de las Heras L., Betti M.: Application of microprecipitation on membrane filters for alpha spectrometry, liquid scintillation counting and mass spectrometric determination of plutonium isotopes. Radiochim. Acta **90**, 721 (2002).
14. Hrnecek, E., Steier, P., Wallner, A.: Determination of plutonium in environmental samples by AMS and alpha spectrometry. Appl. Radiat. Isotopes **63**, 633 (2005).
15. Bachhuber, H., Bunzl, K., Schimmack, W., Gans, I.: The migration of ^{137}Cs and ^{90}Sr in multilayered soils: results from batch, column, and fallout investigations. Nucl. Technol. **59**, 291 (1982).
16. Forsberg, S., Strandmark, M.: Migration and chemical availability of ^{137}Cs and ^{90}Sr in Swedish long-term experimental pastures. Water Air Soil Pollut. **127**, 157 (2001).
17. Fuhrmann, M., Lasat, M. M., Ebbs, St. D., Kochian, L. V., Cornish, J.: Uptake of cesium-137 and strontium-90 from contaminated soil by three plant species; application to phytoremidiation. J. Env. Qual. **31**, 904 (2002).
18. Raes, F., Graziani, G., Stanners, D., Girardi, F.: Radioactivity measurements in air over Europe after the Chernobyl accident. Atmosph. Env. **24A**(4), 909 (1990).
19. Tatzber, F., Irlweck, K.: Accumulation of fallout nuclides and natural radioactivity in Austrian snails. Health Phys. **46**(2), 448 (1984).
20. Schönhofer, F., Tataruch, F., Friedrich, M.: Strontium-90 in antlers of red deer: an indicator of environmental contamination by strontium-90. Sci. Total Env. **157**, 323 (1994).
21. Landstetter, C., Wallner, G.: Determination of strontium-90 in deer bones by liquid scintillation spectrometry after separation on Sr-specific ion exchange columns. J. Env. Radioact. **87**, 315 (2006).
22. Moreno, J., Vajda, N., Danesi, P. R., Larosa, J. J., Zeiller, E., Sinojmeri, M.: Combined procedure for the determination of ^{90}Sr, ^{241}Am and Pu radionuclides in soil samples. J. Radioanal. Nucl. Chem. **226**(1–2), 279 (1997).
23. Hrnecek, E., Feichtinger, J.: Assessment of soil contamination in the vicinity of a research reactor during the decommissioning process. J. Radioanal. Nucl. Chem. **264**, 377 (2005).
24. Vockenhuber, Ch., Ahmad, I., Golser, R., Kutschera, W., Liechtenstein, V., Priller, A., Steier, P., Winkler, S.: Accelerator mass spectrometry of heavy long-lived radionuclides. Int. J. Mass Spectrom. **223**–**224**, 713 (2003).
25. Mietelski, J. W., Dorda, J., Was, B.: Pu-241 in samples of forest soil from Poland. Appl. Radiat. Isotopes **51**, 435 (1999).
26. De Bortoli, M., Ganglione, P., Malvicini, A., Van der Sticht, E.: Five years experience of ^{90}Sr and ^{137}Cs herbage to milk transfer under field conditions. In: *Radiation Protection*. Vol. 2 (Snyder, W. S., ed.) Pergamon Press, Oxford (1968).