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^{137}Cs, ^{40}K, ^{90}Sr, $^{238, 239+240}$Pu, ^{241}Am and $^{243+244}$Cm in forest litter and their transfer to some species of insects and plants in boreal forests: Three case studies

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Results for ^{137}Cs, ^{40}K, ^{90}Sr, $^{238,239+240}$Pu, ^{241}Am and $^{243+244}$Cm measurements in plant, insects and forest litter samples collected at three sites in Poland are presented. New results are compared with some existing data for locations examined during previous studies. Insect samples were introduced now for the first time. Relatively high activities of ^{90}Sr were noticed for spruce bark beetle (*Ips typographus*) and those for ^{137}Cs, plutonium and ^{241}Am for forest dung beetle (*Anoplotrupes stercorosus*). Faster than caused by physical decay decrease of radiocesium activity was noticed for the majority of plant and litter samples. The results for $^{239+240}$Pu for litter were similar to previous results, but the activities of ^{238}Pu were smaller. The activity ratio between ^{241}Am and $^{239+240}$Pu was found lower than expected for known proportions between global and Chernobyl fallout. Thus a kind of dynamic behavior of Pu and Am in the forest ecosystem is concluded. Transfer factors and aggregation coefficients were estimated and discussed for both plants and insects as well as between insects and the part of plants (or litter) they feed on. Many of them were never presented before.

Introduction

Radiocesium behavior in the soil-litter-plant system in European boreal forests was a subject of investigations carried out on relatively wide scale following the Chernobyl accident. Since the deposition of radiostrontium, plutonium and americium was much lower in sites remote from Chernobyl and since the analyses are much more complicated, the data about the presence of those radionuclides in plants or other parts of forest ecosystem in Europe are rather scarce, when compared to radiocesium data.

During our investigation we focused particularly on the dynamics of Pu isotopes, ^{241}Am, $^{243+244}$Cm and ^{90}Sr in a boreal forest ecosystem. For comparison, activities of radiocesium and natural ^{40}K were also a subject of the study. Present investigations were conducted on three locations, chosen from those, for which some results from the past studies were available.

We included some insect species to investigate part of the forest ecosystem. There are several reasons of turning the interest to the radioactivity of insects. Since terrestrial invertebrates, especially the insects, are rather excluded from the human diet within European cultural circle they usually were not a subject of intense radiological survey. However, insects are included to the diet in many other human cultures as well as they are very important part of the natural food chains. It seemed interesting to see how do the artificial radionuclides go to selected species of insects of different living and feeding habits. There is also another reason, which makes insects an interesting kind of environmental sample. Taking samples of soil or plants, one has always doubts concerning representation of sampling site. If one is collecting flying insects into a pheromone trap he/she may be sure, that he/she collects individuals, which occupy larger area, so he/she already deals with a kind of a well-averaged sample. There is also one more reason for turning the interest to insects. During last years a concept of doses to the biota started to be a matter of increasing concern.[1–4] Providing data for future dose estimation is a question of the day. The doses from man-made radionuclides should be compared with the doses from natural isotopes. The majority of inner dose is expected to come from ^{40}K. Therefore, in our study about insects besides man-made radionuclides like: ^{137}Cs, ^{90}Sr, $^{238,239+240}$Pu, ^{241}Am and $^{243+244}$Cm, the activities of ^{40}K were determined as well.

Experimental

Samples

Samples of forest litter and top layer soil, plants and insects were collected mostly during the summer of 2001 in three forests of Poland (Fig. 1). One location was in the North-Eastern Poland, near the village of Płaska in Augustów Primeval Forest (ca. 54°04'N, 23°05'E). At this location an additional set of plant samples was collected during next summer (year 2002) also. That area was of our particular interest, since it was an example of the site, where the Chernobyl fallout containing relatively intense hot particle fraction

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occurred, what was established and intensively examined during previous studies.[5–11] The deposition of Chernobyl-origin Pu alpha emitters (sum of $^{238,239+240}$Pu), reconstructed for 1986, was found in Augustów Primeval Forest equal[5,6] up to 30 Bq/m^2 that for ^{241}Pu was[10] up to 1 kBq/m^2 and for ^{90}Sr was[8] up to 2 kBq/m^2, whereas the ^{137}Cs was present on the level not exceeding 5 kBq/m^2 (this site was coded "120" in cited past projects). Two other locations were control areas at South of Poland. Namely: one site nearby Węgierska Górka in Beskid Mountains (ca. 49°41'N, 19°03'E) and another one near Tułowice in Silesia (ca. 50°40'N, 17°32'E). The site at Węgierska Górka was very close to the site (No 39A) from our previous studies in 1987 and 1993.[12–14] It was in the mountains, at 700 m above sea level. The third site – the Tułowice was close to site coded "203" in the past project.[5]

One additional insect sample was collected in 1999 in the Gorce Mountains, also in South of Poland (ca. 49°36'N, 20°07'E). All the southern locations were expected to have a typical isotopic composition of Chernobyl fallout for the remote sites, but with ^{137}Cs deposition[15] ranging from 20 to 50 kBq/m^2 and with dominating global fallout origin of ^{90}Sr[8,13] or Pu and Am isotopes.[5–7,13] More details on sampling sites are presented in Table 1.

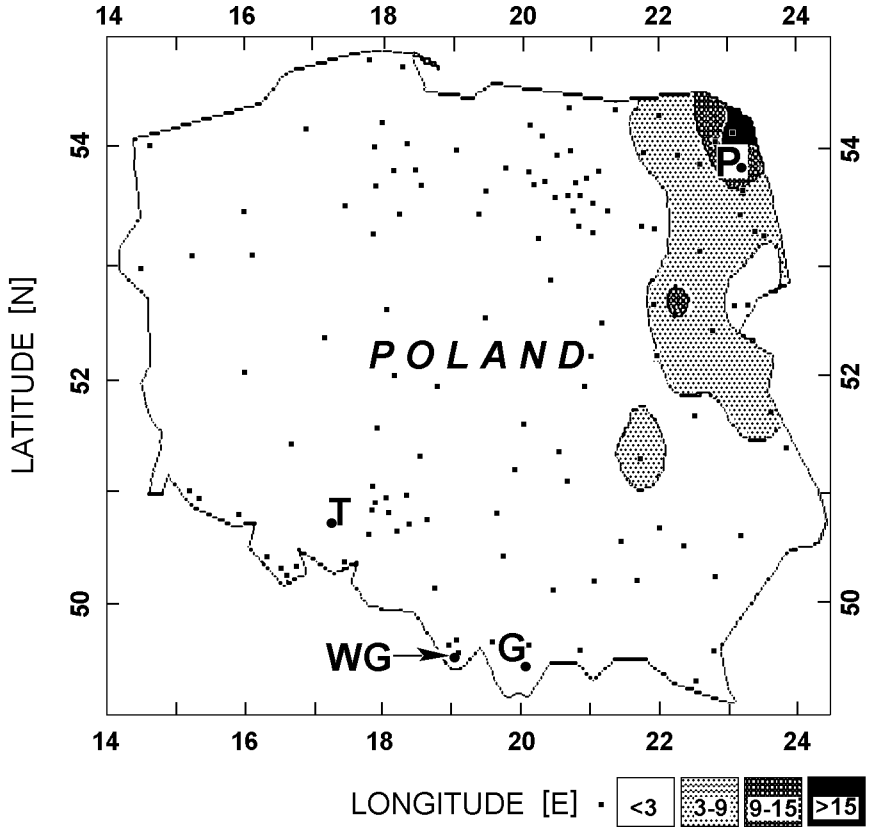

Fig. 1. Sampling sites on the approximate map $^{239+240}$Pu deposition (expressed in Bq/m^2) from Chernobyl fallout[37] (no global fallout included) based on the ^{144}Ce approximate deposition map,[5,38] small rectangles are sampling sites in that project. Present study sites marked with dark full circles: P – Płaska, T – Tułowice, W – Węgierska Górka, G – Gorce

Table 1. Details on sampling sites

Name of site of the forest district	Name of region	Forest compartment/ forest services code	Altitude, m asl	Soil	Forest type	Main tree species	Age, year	Density, %	Mean trunk diameter $D_{1..3}$, cm	Mean tree height, m	Wood biomass, m^3/ha	Collected plant or insect samples
Tułowice	Niemodlin Forest	Przechód/ 124d	180	PZha	CMf	90% pine 10% spruce	100 (pine) 70 (spruce)	65	34 40	27 27	401 45	AS, pine and spruce needles, bilberry, fern, spruce bark, litter
		Przechód/ 144f	180	PZha	CMf	Oak and pine	5	–	–	–	–	pine rots, pine stem of 3 years old tree
		Przechód/ 180a	180	PZha	CMw	Oak and pine	5	–	–	–	–	HA
Węgierska Górka	Beskid Mts.	Żabnica/ 32c	700	CMd	CH	Spruce	78	65	27	26	250	All kind
Płaska	Augustów Primeval Forest	Jazy/ 269	110	PZha	Cf	Pine	125	60	35	26	240	LM, pine needles, heather
		Jazy/ 310d	110	PZha	Cf	Pine	5	–	–	–	–	HA, pine roots, pine stem of 3 years old tree
		Jazy/ 312d	110	PZha	CMf	70% spruce, 30% pine Single alder and oaks.	87 87	65	33 31	26 26	270 80	IT(1), AS (1&2), spruce bark and needles, litter, moss, alder
		Królowa Woda										IT(2)

Forest type abbreviations: C – coniferous, D – deciduous, M – mixed, f – fresh, w – wet, H- mountain forest, PZha-haplic podzols, CMd = distric cambisoils.

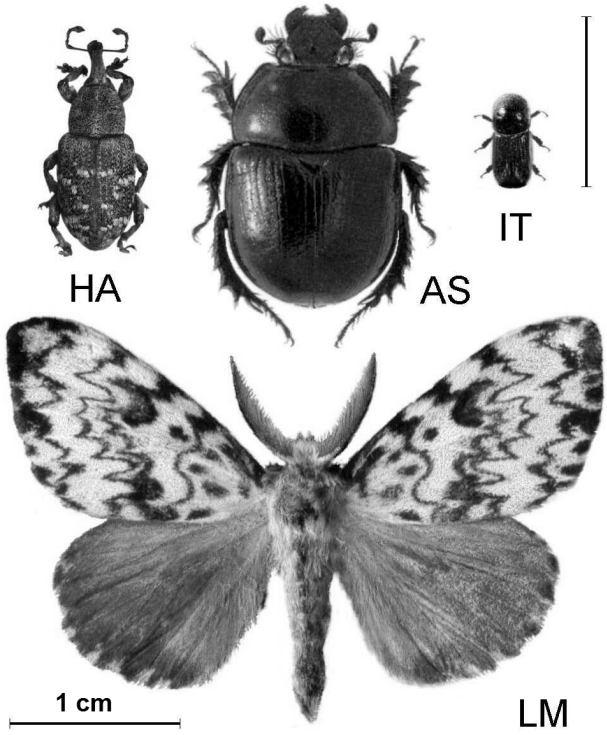

Fig. 2. Investigated species of insects: AS – forest dung beetle (*Anoplotrupes stercorosus*), HA – large pine weevil (*Hylobius abietis*), IT – spruce bark beetle (*Ips typographus*), LM – male of the nun moth (*Lymantria monacha*)

The following species of insects (Fig. 2) were subject of study:

(1) Forest dung beetle – *Anoplotrupes stercorosus* (Hartm.), (Coleoptera, Geotrupidae) one of most common and relatively big European forest beetle, species tested by us recently for biomonitoring of radionuclides[11] collected into traps as a grown form (imago), being of interest since the young form (larva) develops in the soil taking nutrients from humus clusters, formed there by its parents (abbreviation – AS).

(2) Large pine weevil – *Hylobius abietis* (L.), (Coleoptera, Curculionidae) another common species of a beetle, larva lives in the soil feeding on the roots of pine, easy to collect into pheromone traps (abbreviation – HA).

(3) Spruce bark beetle – *Ips typographus* (L.), (Coleoptera, Scolytidae) another common species of a small beetle, lives and feeds in the cambium of spruce, collected by pheromone traps (abbreviation – IT).

(4) Nun moth – *Lymantria monacha* (L.), (Lepidoptera, Lymantriidae) a moth which lives and feeds as caterpillar mostly on the pine or spruce needles, easy to collect flying males into pheromone traps (abbreviation – LM).

A large collection of AS beetle from Płaska was subdivided into two samples. Two separate samples of beetle IT from Płaska were collected in the forest with a distance of a few kilometers between them.

Samples of the most important insect food components were also collected. Namely the forest litter (leaf and humus layers), pine (*Pinus sylvestris*) roots, needles (from current year – C, and two previous years coded C+1 or C+2), stem of three year old trees, spruce (*Picea excelsa*) bark and needles (current year C or previous years C+1). Some additional plant samples like leaf of bilberry (*Vaccinium myrtilus*), fern (*Athyrium* sp.), leaf of alder (*Alnus glutinosa*) and heather (*Calluna vulgaris*) were also collected, mainly for better recognition of the bio-availability of selected radionuclides in given environment according to the recommendation of ICP Forest Manual.[16] Sampling sites were also characterized regarding typical factors used in forestry (Table 1).

Additional archive samples from 1999 collected in the Gorce Mountains were a rather huge (310 g of dry mass) set of IT trapped into pheromone traps. No additional data about the location was available in this case.

Gamma-spectrometric measurements

The analysis of gamma-emitter activities, mainly of ^{137}Cs was performed by means of low background gamma-ray spectrometry with a HPGe detector. The number of individuals in the samples was estimated in a statistical way by determining the mass of a known quantity of individuals and comparing the result to the mass of the whole sample. The normalization masses used for presentation of the activity concentration in insect samples were determined after incineration in 400 °C. Gamma-activities of other than insect samples were determined for grounded material, after drying them overnight at 105 °C in an oven. Activity concentrations were here given for dry samples.

Radiochemical procedure for Pu, Sr, Am, Cm

All samples were incinerated at 600 °C. We include Am and Cm to our investigation when it was already running, therefore, for some samples activities for those radionuclides are not available. Chemical yield tracers (^{85}Sr, ^{236}Pu, ^{243}Am) were added and the samples were wet mineralized using hot mineral acids.[17] Radionuclides were separated sequentially, during one combined procedure. Pu was separated[6,17] on an anion exchange column filled with Dowex-1 from 8M HNO_3, Sr on a Sr-Resin™ (EiChrom) also from 8M HNO_3,[18] and Am together with Cm was separated using Dowex-1 and methanol-acids solutions.[7,19] Alpha-spectrometric sources (Pu, Am+Cm) were prepared using NdF_3 co-precipitation.[20] The measurements were performed using a Silena AlphaQuattro spectrometer with Canberra PIPS detectors. The activity of ^{90}Sr was determined using a liquid scintillation spectrometer Wallac 1414-003 Guardian. The ^{85}Sr recovery was determined by means of gamma-spectrometry.

The activities for insects were normalized to the mass of ashes, all others were normalized to dry mass at 105 °C. Because all measurements were performed within one year no decay correction was needed due to the long half-lives of the determined isotopes. The conversion factors from activity concentrations in Bq/kg to activity inventory in Bq/m^2 were determined for each soil or litter layer. It was just the dry mass of the whole given layer sample divided by the area of sampling frame equal to 19.5 × 19.5 cm^2.

The $^{239+240}$Pu peaks were considered free of any interference (Fig. 3). However, in some cases, alpha-spectra contained some tiny traces of ^{228}Th and ^{224}Ra, unfortunately large enough to cause some interference resulting in increased detection limit for ^{241}Am (Fig. 4) or even ^{238}Pu. Peak areas were calculated by a fitting procedure provided by our ALF software described elsewhere.[5,6]

Quality assurance

Analyses of blank samples and reference material[21] Soil-IAEA 375 were performed to achieve quality assurance. The results for reference material measurements are presented in Table 2.

Origin of radionuclides

The analysis of plutonium activity concentration ratios (^{238}Pu to $^{239+240}$Pu) allowed us to distinguish the Chernobyl and global fallout components of plutonium activities.[5] This ratio in global fallout was[22] 0.03±0.01, and for Chernobyl fallout in Poland it was in 1986 about 0.55.[5,6] Due to the radioactive decay which affected mostly the activity of ^{238}Pu, now this ratio could be assumed to 0.50±0.03. Thus, the percentage F_{Pu} of Chernobyl $^{239+240}$Pu can be calculated as:

$$F_{Pu} = \left(\frac{A_{238}/A_{239} - 0.03}{0.50 - 0.03} \right) \cdot 100\% \qquad (1)$$

where A_{238} is the activity of ^{238}Pu, A_{239} is the activity of $^{239+240}$Pu. The percentage of global fallout component is $100\% - F_{Pu}$.

Table 2. Activities (in Bq/kg) determined for analyzed reference material (IAEA-375) compared with certified values[21]

Nuclide	Mass, g	Determined activity (corrected for reference date) D	Certified value, C	95% Confidence interval	Difference (D–C)/C, %
^{137}Cs	75.4	5559 ± 66	5280	5200÷5360	+5.3
^{40}K	75.4	423 ± 28	424	417÷432	–0.2
^{90}Sr	5.2	104 ± 10	108	101÷114	–0.4
$^{239+240}$Pu	5.2	0.267 ± 0.025	0.30	0.26÷0.34	–11
$^{239+240}$Pu	12.8	0.259 ± 0.026	0.30	0.26÷0.34	–13.7
^{238}Pu	5.2	0.076 ± 0.010	0.071	0.056÷0.085	+7
^{238}Pu	12.8	0.052 ± 0.009	0.071	0.056÷0.085	–27
^{241}Am	12.8	0.126 ± 0.033	0.13	0.11÷0.15	–3.1

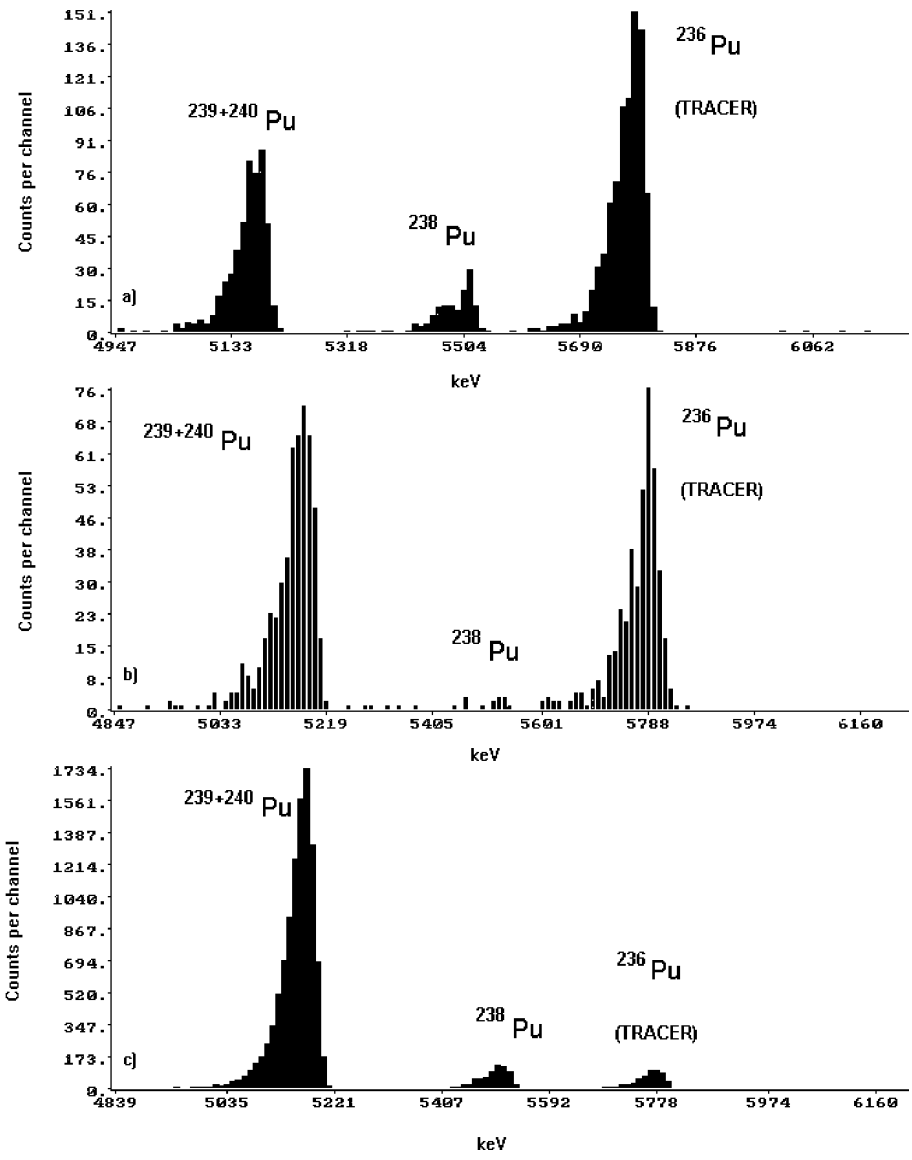

Fig. 3. Alpha-particle spectra of plutonium separated from samples of: beetle "AS" (a), plant – needles of spruce C+1 (b) or forest litter O_H (c) from Płaska

The activity of Chernobyl-origin ^{241}Am can be estimated on the basis of $^{243+244}$Cm activity. The present activity ratio of $^{243+244}$Cm to ^{241}Am in Chernobyl NPP vicinity is about 5%,[39] and for global fallout can be assumed to be very close to zero.[22] The value of this ratio is continuously decreasing predominately as the result of the ingrown of ^{241}Am activity from the ^{241}Pu decay. The percentage F_{Am} of Chernobyl ^{241}Am can be calculated as:

$$F_{Am} = \left(\frac{A_{Cm} / A_{Am}}{0.05} \right) \cdot 100\% \qquad (2)$$

where A_{Cm} is the activity of $^{243+244}$Cm, A_{Am} is the activity of ^{241}Am. The rest is the percentage of global fallout component equal to $100\% - F_{Am}$.

^{134}Cs was not measurable in any sample, therefore, no data on the proportion between Chernobyl and global fallout in the present samples were available. However, the general pattern in Płaska and Węgierska Górka was recognized during our past studies on the investigated sites.[5,12,14] In all cases the very top layer of forest litter contained only Chernobyl-origin ^{137}Cs, the global fallout fraction was small in investigated layers but it was increasing with depth. For example in the O_H layer the global fallout fraction was about 10%.[5,14]

Transfer factors and aggregation coefficients

Usually the transfer factor *TF* is defined as the ratio of the activity A_i of a given radionuclide in the insect or plant to its activity A_0 in the given layer of forest soil,

litter (here layer O_H was chosen) or in the plant considered the insect fodder:

$$TF = \frac{A_i}{A_0} \tag{3}$$

In some cases the activity ratio of ^{238}Pu to $^{239+240}$Pu pointed out the layer from which plutonium is most likely incorporated to plant or insect. In those cases the activity A_0 *TF* was calculated as the weighted sum of activities in layers O_L and O_H, in a way that the activity ratio ^{238}Pu to $^{239+240}$Pu was equal to that observed in the insect or plant:

$$TF = \frac{A_i}{aA_{0_F} + bA_{0_H}} \tag{4}$$

where *a, b* mean weights.

Estimations of the aggregation coefficients *AC*, for cumulated deposition of given radionuclide were also possible. *AC* can be defined as the ratio of observed activity concentration to total deposition A_S of a given radionuclide on the investigated area:

$$AC = \frac{A_i}{A_S} \tag{5}$$

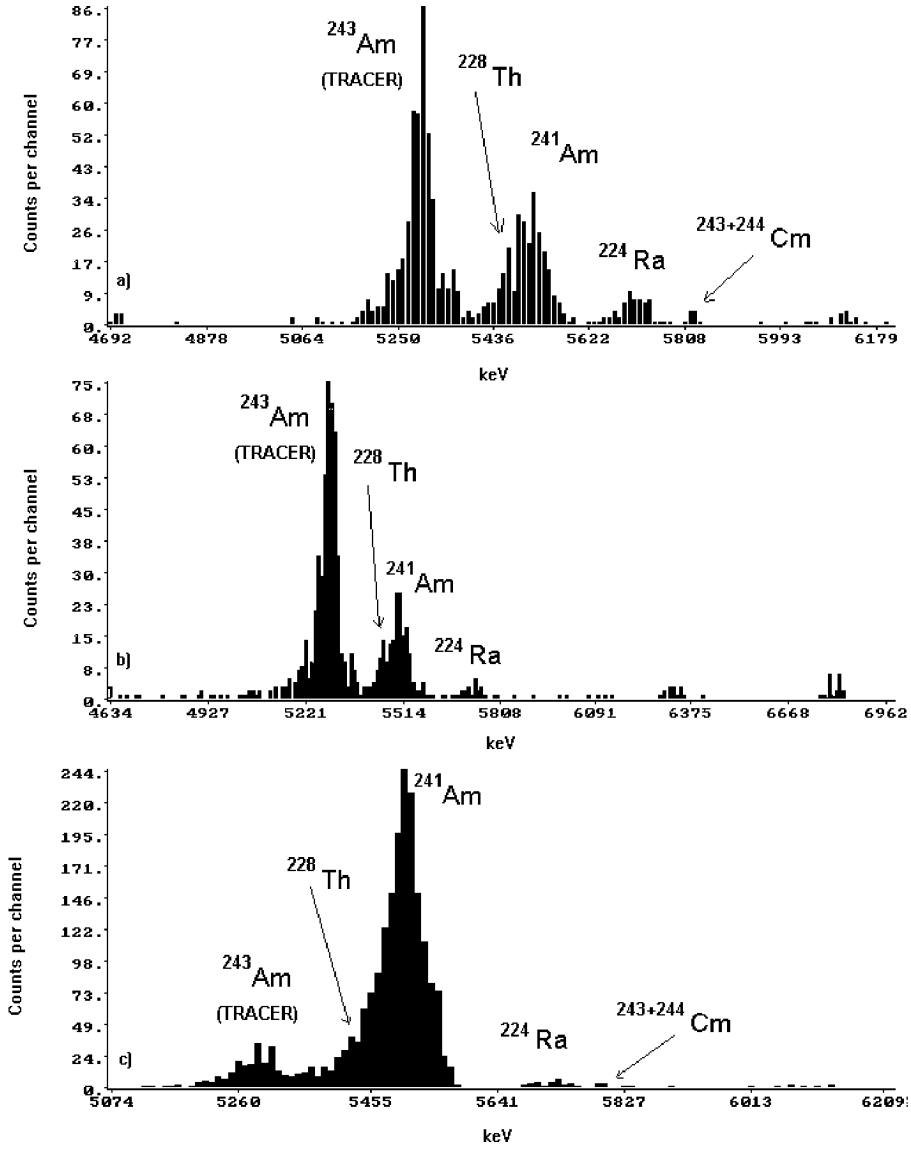

Fig. 4. Alpha-particle spectra of americium (and curium) separated from samples of: beetle "AS" (a), plant – needles of spruce C+1 (b) or forest litter O_H (c) from Płaska. Interfering effect of 5.4 MeV. ^{228}Th peak on 5.5 MeV ^{241}Am peak is visible

Results and discussion

For all the samples the mean recoveries of tracers and their standard deviations were 65±12%, 74±29% and 42±12%, for plutonium, strontium and americium (with curium), respectively.

Soil and litter samples

Results for activities of forest moss/litter/soil system samples are presented in Table 3. In addition, the second column of this table contains conversion factors from activity concentration in Bq/kg to activity inventory in Bq/m^2.

It seems, that the majority of ^{137}Cs was present in the O_H or O_F layers. Usually, ^{40}K activity increased whereas ^{90}Sr decreased with the depth. The majority of Pu activity was present in the O_H or A (in case of Tułowice) layers.

A relatively low level of ^{238}Pu indicates the global fallout as the Pu source in the case of Tułowice and Węgierska Górka sites, whereas in O_F layer of Płaska the Chernobyl origin can be clearly recognized. In general, this confirms our past knowledge on those sites but some new observation will be discussed later in this text.

The $^{239+240}$Pu activity 10.86±0.61 Bq/kg found now for the O_H level at Węgierska Górka was exactly the same as the one found for the A_1 layer in 1987,[13] where it was 10.8±0.4 Bq/kg. A similar effect was found for present O_F samples and past A_0 samples, with $^{239+240}$Pu activities of 2.25±0.19 Bq/kg and 2.19±0.12 Bq/kg, for present and 1987 samples, respectively. However, ^{238}Pu activities now were lower by a factor from three to five while compared to the results from 1987. This might be explained by a kind of dynamic equilibrium between litter compartments for global fallout Pu: what goes out from litter down to soil (including slow turning the upper litter to humus, than soil, etc.) comes back with fallen needles and leaves. ^{238}Pu was deposited mainly from Chernobyl, its fluxes downward and upward are apparently not equilibrated yet. Cesium activities were now lower by more than a factor of two than those found in 1987 or 1993,[14] strontium in O_H and in former A_h samples were similar, but in upper layers now we found activities lower by a factor of four than in the past.[13]

Radiocesium found in soil or litter samples from Tułowice was higher by a factor of two lower compared to those found at this area in 1991.[5] Activities of neither Pu and Am nor radiostrontium isotopes were determined there previously. The previous analyses for Płaska site were performed previously for samples collected in 1991. For forest litter the data for gamma-emitters[5] and plutonium[5,6] are available. Results for ^{40}K were similar, about 100 Bq/kg. Present results for ^{137}Cs were about three and four times lower than those for samples from 1991, when they were 1042±57 Bq/kg for A_0 (leaf layer) and 870±41 Bq/kg for A_1 (humus layer) samples. Similar to the results from Węgierska Górka the results for plutonium activities remained almost unchanged for $^{239+240}$Pu and were lower for ^{238}Pu. For samples from 1991 $^{239+240}$Pu activities were 3.98±0.33 Bq/kg and 6.88±0.53 Bq/kg, for A_0 and A_1 layers, respectively, whereas now they were 4.315±0.289 Bq/kg and 8.379±0.478 Bq/kg for O_F (comparable with former A_0) and O_H (comparable with former A_1) layers, respectively. For ^{238}Pu in 1991 they were 1.8±0.2 Bq/kg and 2.17±0.22 Bq/kg for A_0 and A_1 layers, respectively. Now they were 1.529±0.123 Bq/kg and 0.637±0.045 for O_F and O_H layers, respectively.

^{90}Sr activity at Płaska was significantly higher than at two remaining locations as expected already from previous studies.[8,23]

Table 3. Results on the radionuclides activity for forest litter, moss and soil samples

Site (layer)	Mass of layer, kg/m^2	^{137}Cs, Bq/kg$_{dry}$	^{40}K, Bq/kg$_{dry}$	^{90}Sr, Bq/kg$_{dry}$	$^{239+240}$Pu, mBq/kg$_{dry}$	^{238}Pu, mBq/kg$_{dry}$	^{241}Am, mBq/kg$_{dry}$	$^{243+244}$Cm, mBq/kg$_{dry}$
Tułowice								
O_L (0–2 cm)	2.02	82 ± 2	42 ± 9	10 ± 1	48 ± 5	5 ± 1		
O_F (2–4 cm)	2.15	163 ± 5	32 ± 8	11 ± 2	107 ± 8	6 ± 2	31 ± 16	<2
O_H (4–6 cm)	3.35	786 ± 17	36 ± 8	20 ± 3	2430 ± 181	169 ± 18		
A (6–8 cm)	11.03	358 ± 9	107 ± 9	5 ± 1	3250 ± 233	136 ± 24	1260 ± 128	<10
AE_{es} (8–11 cm)	19.75	101 ± 2	106 ± 10	<1	415 ± 41	9 ± 8	220 ± 58	<6
Węgierska Górka								
O_L (0–1 cm)	0.92	41 ± 2	30 ± 16	11 ± 1	670 ± 47	23 ± 9	233 ± 32	<3
O_F (1–3 cm)	2.07	185 ± 6	68 ± 16	10 ± 1	2250 ± 187	89 ± 16		
O_H (3–7 cm)	2.56	394 ± 11	233 ± 26	8 ± 1	10860 ± 614	293 ± 29		
A (7–12 cm)	3.27	189 ± 4	313 ± 29	2 ± 0	5080 ± 335	148 ± 21		
Płaska								
O_L+Moss* (0–3 cm)	0.69	132 ± 5	71 ± 25	18 ± 2	322 ± 27	32 ± 12	172 ± 18	5 ± 2
O_F (3–7 cm)	2.65	346 ± 10	105 ± 21	64 ± 7	4315 ± 289	1529 ± 123	2980 ± 228	66 ± 9
O_H (7–9 cm)	3.68	197 ± 6	169 ± 23	60 ± 8	8379 ± 478	637 ± 45	2380 ± 198	21 ± 6
A (9–13 cm)	10.75	28 ± 1	293 ± 27	8 ± 1	581 ± 55	53 ± 32	184 ± 35	18 ± 10

* *Climacium dendroides.*

Table 4 presents sums of surface activity observed in analyzed soil or litter layers. The results for Cs, Pu, Am and Cm isotopes activity, presented in Tables 3 and 4, suggest that significant amounts of total deposition of those radionuclides was found in analyzed layers. For example for $^{239+240}$Pu the sum of activity was about 80% of average Pu deposition from the global fallout for appropriate latitude belt being[22] 58 Bq/m^3. On the contrary, the majority of ^{90}Sr seems to be absent in the examined litter/soil layers. Since Sr is mobile in forest environment, and even if it was deposited as small hot particles at Płaska site, it was expected that the 15 years elapsed since the deposition occurred was apparently long enough to incorporate radiostrontium into environmental processes.[24] The content found in the investigated layers seems to be equal to about 10% of total deposition, estimated from the former studies on ^{90}Sr content in animal bones[23] or bilberry leaves[8] and including data on global fallout.[22]

Presence of Cm isotopes in Płaska confirmed our earlier finding for Augustów Primeval Forest.[7] Hot particles from Chernobyl fallout seems to be responsible for increasing ^{241}Am to $^{239+240}$Pu activity ratio for Płaska samples. It clearly exceeds the value of 0.3 typical now for global fallout.[25]

Table 4. Total amount of activity (in Bq/m^2) found in investigated part of soil profile

Site (layer)	^{137}Cs	^{40}K	^{90}Sr	$^{239+240}$Pu	^{238}Pu	^{241}Am	$^{243+244}$Cm
Tułowice (0–11 cm)	9100 ± 211	3550 ± 360	166 ± 47	52.5 ± 4.0	2.2 ± 0.5		
Węgierska Górka (0–12 cm)	2050 ± 56	1790 ± 209	58 ± 6	49.7 ± 3.1	1.4 ± 0.2		
Płaska (0–13 cm)	2030 ± 63	4100 ± 450	490 ± 60	48.7 ± 3.1	7.0 ± 8.4	18.7 ± 1.7	0.45 ± 0.16

Table 5. Results on activities found for plant samples

Site/Species	Dry mass, g	^{137}Cs, Bq/kg$_{dry}$	^{40}K, Bq/kg$_{dry}$	^{90}Sr, Bq/kg$_{dry}$	$^{239+240}$Pu, mBq/kg$_{dry}$	^{238}Pu, mBq/kg$_{dry}$	^{241}Am, mBq/kg$_{dry}$	$^{243+244}$Cm, mBq/kg$_{dry}$
Tułowice								
Bilberry leaf	17.5	570 ± 26	370 ± 52	6 ± 1	11 ± 5	<40	12 ± 5	<1
Pine needles C	26.8	504 ± 17	270 ± 45	2 ± 1	2 ± 1	<32	<2	<1
Pine needles C+1	48.3	95 ± 4	156 ± 28	9 ± 2	4 ± 1	3 ± 1		
Pine stem of 3 years old tree	23.8	54 ± 2	127 ± 13	6 ± 1	153 ± 16	<22		
Root of pine	105.1	81 ± 3	27 ± 15	8 ± 1	29 ± 5	<2	16 ± 5	<1
Spruce needles C	25.4	139 ± 4	146 ± 15	10 ± 1	0 ± 1	<24	<2	<1
Spruce needles C+1	28.1	134 ± 7	177 ± 34	18 ± 3	12 ± 2	<1	6 ± 2	<1
Spruce bark 1	337.8	107 ± 3	35 ± 11	127 ± 22	25 ± 2	<2		
Spruce bark 2	354.1	88 ± 3	40 ± 15	49 ± 11	21 ± 2	3 ± 1		
Fern	87.2	1220 ± 39	660 ± 38	12 ± 2	10 ± 3	8 ± 2		
Węgierska Górka								
Bilberry leaf	23.8	13 ± 1	247 ± 27	2 ± 1	42 ± 4	<21	22 ± 4	<1
Spruce bark	54.7	8 ± 1	117 ± 14	62 ± 9	11 ± 2	<2	<1	<1
Spruce needles C+1	38.3	7 ± 1	113 ± 19	13 ± 2	15 ± 2	<2	8 ± 2	<1
Fern	32.3	8 ± 1	1000 ± 120	10 ± 1	138 ± 11	3 ± 2	370 ± 43	<1
Płaska-2001								
Bilberry leaf	63.6	435 ± 13	257 ± 30	56 ± 42	11 ± 1	3 ± 1	11 ± 8	<1
Pine needles C	99.9	163 ± 6	140 ± 29	6 ± 1	308 ± 23	<9	383 ± 28	<1
Pine needles C+1	63.7	30 ± 2	85 ± 21	8 ± 1	87 ± 9	<4	27 ± 5	<1
Pine needles C+2	22.1	21 ± 2	94 ± 24	<4	141 ± 14	<1	13 ± 7	<1
Pine stem of 3 years old tree	16.0	11 ± 2	266 ± 34	8 ± 2	152 ± 14	14 ± 10	<58	<1
Root of pine	115.8	19 ± 1	24 ± 14	43 ± 10	232 ± 17	53 ± 7	164 ± 12	7 ± 2
Spruce needles C	114.0	237 ± 7	143 ± 23	5 ± 1	107 ± 10	<1	176 ± 14	<1
Spruce needles C+1	117.0	74 ± 3	86 ± 19	22 ± 3	130 ± 10	<3	24 ± 3	<2
Spruce bark	275.3	183 ± 5	38 ± 18	10 ± 3	208 ± 13	31 ± 3	93 ± 13	<1
Fern	96.8	1270 ± 49	420 ± 54	32 ± 5	244 ± 22	8 ± 2	186 ± 24	5 ± 3
Alder	59.1	4 ± 1	214 ± 28	<2	17 ± 3	13 ± 3	21 ± 4	<1
Płaska-2002								
Pine needles C	60.2	160 ± 5	201 ± 12	6.0 ± 0.6	<20	<20	4.9 ± 3.0	<1
Pine needles C+1	59.8	104 ± 3	219 ± 12	2.9 ± 0.3	2.5 ± 0.6	<2	9.6 ± 5.0	<1
Spruce needles C	66.5	161 ± 6	236 ± 15	19.7 ± 2.0	103 ± 2	1.6 ± 0.9	6.9 ± 3.5	<1
Spruce needles C+1	59.1	25 ± 1	160 ± 12	25.6 ± 3.1	3.1 ± 1.8	<2	<13	<1
Heather	46.4	301 ± 10	216 ± 14	16.1 ± 1.6	43 ± 7	<2	39.7 ± 8.9	<1

Plant samples

Table 5 contains results for plant samples from the investigated areas.

As for litter samples obtained now results for ^{137}Cs in bilberry can be compared with results from 1991.[5] In the case of Tułowice, present activities of ^{137}Cs were 63±6% of that from 1991. On the contrary, the samples from Płaska showed 153±4% activity from 1991. This could be explained in a speculative way by the increasing bio-availability of radiocesium from hot particle fallout, but it can as well be just a fluctuation.

Ferns from Węgierska Górka were the subject of previous analyses for ^{137}Cs in samples from 1987 and 1993[12,14] and for ^{90}Sr and Pu in samples collected in 1987.[13] There was a hypothesis of a strong decreasing trend of radiocesium activity in ferns. Sample from 1987 showed ^{137}Cs activity of 15900±234 Bq/kg[12] but it was only 870±32 Bq/kg for samples from 1993[14] at exactly the same site. The trend was confirmed – now the sample from 2001 showed only 8±1 Bq/kg. There was no such strong trend of radiocesium activity removal found in litter, so it must be a trend of bio-availability. For ^{90}Sr, the activity between 1987 and 2001 decreased by a factor of four. In 1987 $^{239+240}$Pu was not found in fern with the detection limit of 30 mBq/kg,[13] but now its activity was 138±11 mBq/kg. For two other locations ^{137}Cs activities in fern were still relatively high. Ferns are known for their usually high AC for radiocesium.[26]

Using an AC value of 0.20 m^2/kg for bilberry discussed above and observed ^{90}Sr activity of 56±42 Bq/kg the deposition could be estimated as 280±210 Bq/m^2. This is only 20% of previously estimated values, far below global fallout deposition for this radionuclide. This means, that ^{90}Sr is effectively removed from shallow root areas of bilberries not only in Płaska, but in the two other locations, where the activities of ^{90}Sr in bilberries are much lower.

Of other plant samples, only spruce needles (C+1) from Węgierska Górka were analyzed for ^{137}Cs in samples from 1987.[12] The activity found for that sample was 300±12 Bq/kg, and now it is only 7±1 Bq/kg. In the present study the majority of radiocesium was always found for "C" needles, the youngest ones, for both pine and spruce samples. Such feature was observed by many other authors before[26–30] and was interpreted as translocation of ^{137}Cs or as a result of lower content of lignin.[31] In our work a similar effect can be seen for potassium, but not for all samples. It was clearly noticeable for pine samples from 2001, for instance. Nothing like that was found for radiostrontium. The results suggest, that the translocation effect cannot be excluded for Pu and Am. It was suggested only for needles of pine collected in Płaska during 2001, but it was not confirmed by the samples from 2002, or by the samples from other locations. Nevertheless, the non-zero values of Pu and Am activities in plant samples seem to be an interesting observation. The highest activities for Pu and Am in plant samples were found for some pine and spruce samples.

During the Chernobyl accident there was a discrepancy noticed[10] between ^{238}Pu to $^{239+240}$Pu or ^{241}Pu to $^{239+240}$Pu activity ratios reported for grass or other plant samples and direct measurement of those ratios in fallout.[32,33] Present results show for all three sites, that the root uptake of global fallout Pu is the main source of measurable amounts of plutonium in plants. It seems, that plant samples used for Pu activity ratio determination in 1986 in cited above papers, contained besides external contamination with "fresh" Chernobyl fallout some overlooked global fallout Pu as well. This was shifting the activity ratios of both ^{238}Pu to $^{239+240}$Pu and ^{241}Pu to $^{239+240}$Pu to the lower values. Therefore, it should be remembered for any future projects using plant samples for reconstruction of actinides fallout in trace amounts.

Large differences between activities of Pu and Am in needles from Płaska between samples from years 2002 and 2001 suggest, that there is an important difference between Pu and Am content in needles within the tree canopy. Samples from 2001 with relatively high Pu and Am activities were collected from the almost top of the tree (at the seventh branch from the top, freshly cut down tree) whereas samples from 2002 were collected from the bottom of the canopy.

Radiocesium found for single heather sample was not high, although this species was recognized in the past[34] as one characterized by a high transfer factor for radiocesium. The activity of Pu was moderate, equal to 43±7 mBq/kg.

Insects

Table 6 presents the results for ashes of insect samples. Some intuitive expectations were confirmed, but some other not. For example the analyzed moth species (LM) as caterpillar feeds on pine needles. Therefore, it was expected for them to contain relatively high concentrations of ^{137}Cs or ^{90}Sr. It appeared, that it was not exactly so. Traces (up to 0.65±0.13 Bq/kg for ash) of $^{239+240}$Pu and ^{90}Sr (up to 97±15 Bq/kg ash), but not ^{137}Cs were found, but only for samples of this species from North-East Poland. The beetle IT showed mostly elevated levels of ^{90}Sr. Median activity for this nuclide was 806 Bq/kg for ash weight (what approximately respect to 77 Bq/kg dry weight), whereas the maximum value exceeds 1400 Bq/kg$_{ash}$ (about 210 Bq/kg$_{dry}$). The activity of ^{90}Sr found in samples from North-Eastern Poland was about two or even three times higher than for the rest of the country.

Table 6. Activities found in ashed samples of insects (in Bq/kg$_{ash}$)

Site/Species	Individuals	^{137}Cs	^{40}K	^{90}Sr	$^{239+240}$Pu	^{238}Pu	^{241}Am	$^{243+244}$Cm
Tułowice								
LM	~310	<50	7010 ± 890	<11	0.074 ± 0.060	0.173 ± 129	0.27 ± 0.10	
AS	600	5030 ± 96	3000 ± 570	26 ± 4	0.99 ± 0.12	0.294 ± 67	0.33 ± 0.09	
HA	~2367	750 ± 21	4650 ± 272	290 ± 31	0.52 ± 0.06	<0.34	0.14 ± 0.10	
IT	~6350	410 ± 23	1730 ± 205	557 ± 31	0.023 ± 0.011	0.011 ± 0.009	<0.02	
Węgierska Górka								
IT	~24750	14 ± 3	1900 ± 138	765 ± 96	0.011 ± 0.003	0.013 ± 0.003	<0.005	
Gorce								
IT	~37500	200 ± 12	2960 ± 106	806 ± 110	0.024 ± 0.018	<0.007	<0.11	
Płaska								
LM	~550	<73	5170 ± 1380	97 ± 15	0.65 ± 0.13	<0.11	<0.7	
AS (1)	376	6720 ± 83	6910 ± 470	142 ± 18	5.49 ± 0.39	1.49 ± 0.17	3.01 ± 0.50	0.19 ± 0.06
AS (2)	342	6710 ± 64	8480 ± 360	161 ± 21	5.69 ± 0.41	1.20 ± 0.13	2.00 ± 0.22	0.10 ± 0.04
HA	448	423 ± 47	2130 ± 419	177 ± 19	2.07 ± 0.17	0.10 ± 0.04	0.72 ± 0.22	
IT (1)	~12250	60 ± 5	1000 ± 99	1270 ± 320	0.024 ± 0.008	<0.01	<0.07	
IT (2)	~4970	290 ± 21	2100 ± 236	1430 ± 190	<0.10	<0.02	<0.09	

Abbreviations explained in Experimental section. Symbol "~" before the number of individuals taken for analyses means only statistical estimation of this number based on the average mass of single imago.

Table 7. Results on ^{238}Pu to $^{239+240}$Pu and $^{243+244}$Cm to ^{241}Am activity ratios, percentages of Chernobyl origin $^{239+240}$Pu (F_{Pu}) or ^{241}Am (F_{Am}). Last column contains results of calculated activity ratio of ^{241}Am to $^{239+240}$Pu only for Chernobyl-origin activities. Data for chosen samples

Site/Layer	^{238}Pu/$^{239+240}$Pu	F_{Pu}	$^{243+244}$Cm/^{241}Am	F_{Am}	^{241}Am/$^{239+240}$Pu (Chernobyl-origin)
Tułowice					
O_L	0.10 ± 0.02	15.8 ± 5.6			
O_F	0.06 ± 0.02	5.5 ± 4.8			
O_H	0.07 ± 0.01	8.4 ± 3.2			
A	0.04 ± 0.01	2.5 ± 3.1			
A (deeper)	0.02 ± 0.02	0.0 ± 4.8			
Węgierska Górka					
O_L	0.03 ± 0.01	0.9 ± 3.9			
O_F	0.04 ± 0.01	2.0 ± 3.0			
O_H	0.0027 ± 0.003	0.0 ± 2.6			
A	0.06 ± 0.03	7.0 ± 6.4			
Płaska					
O_L+Moss	0.10 ± 0.04	14.8 ± 8.5	0.029 ± 0.012	58 ± 24	2.1 ± 0.5
O_F	0.35 ± 0.04	69.0 ± 8.3	0.022 ± 0.003	44 ± 7	0.44 ± 0.10
O_H	0.08 ± 0.01	9.8 ± 2.9	0.009 ± 0.003	18 ± 5	0.51 ± 0.22
A	0.09 ± 0.06	13 ± 12	0.098 ± 0.057	196 ± 115	5 ± 5
Płaska					
Bilberry leaf	0.27 ± 0.09	52 ± 20			
Pine stem of 3 years old tree	0.09 ± 0.07	13 ± 14			
Root of pine	0.23 ± 0.03	42 ± 8	0.043 ± 0.013	85 ± 25	1.4 ± 0.5
Spruce bark	0.15 ± 0.02	25 ± 4			
Fern	0.03 ± 0.01	<3	0.027 ± 0.016	54 ± 33	<300
Alder	0.8 ± 0.2	156 ± 47			
Płaska					
AS (1)	0.27 ± 0.04	51.4 ± 8.2	0.06 ± 0.02	126 ± 45	1.4 ± 0.6
AS (2)	0.21 ± 0.03	38.5 ± 6.4	0.05 ± 0.02	100 ± 41	0.9 ± 0.4
HA	0.05 ± 0.02	<8			

This confirmed that suggested by the results of past investigations on the Chernobyl ^{90}Sr deposition pattern in Poland conducted for bilberry leaf samples[8] or for bones of wild herbivorous animals.[23] Beetle AS have relatively high ^{137}Cs, $^{238,239+240}$Pu, ^{241}Am and $^{243+244}$Cm activities as was expected due to their feeding habits. The influence of Chernobyl hot particles deposition was visible in increased activities of transuranic radionuclides. Activities of $^{239+240}$Pu, ^{238}Pu, ^{241}Am and especially $^{243+244}$Cm were higher for samples from Płaska than for those of Tułowice. More details about usage of this beetle species for bio-monitoring can be found in another article.[11] Results on activities for another beetle species (HA) shows lower values, but also no doubts about presence of Pu or Am.

Origin of plutonium and americium

Table 7 presents results of ^{238}Pu/$^{239+240}$Pu and $^{243+244}$Cm/^{241}Am activities ratio, calculated values of F_{Pu} [Eq. (1)] and F_{Am} [Eq. (2)] as well as the calculated activity ratio of ^{241}Am/$^{239+240}$Pu of Chernobyl-origin exclusively. Data is provided only for chosen samples.

Chernobyl-origin Pu seems to be present only in Płaska. The input of hot particle plutonium fallout is mostly seen in the O_F layer from Płaska, where Chernobyl-origin $^{239+240}$Pu consist 69.0±8.3% of total $^{239+240}$Pu activity. Despite this, for samples from Płaska Chernobyl-origin Pu was almost not present in plants (except for samples of roots of young pine, pines stem of 3 years old tree and for spruce bark), although the global fallout Pu was present there. Roots and bark of pine contained some Pu with elevated ^{238}Pu to $^{239+240}$Pu activity ratio, being a "fingerprint" for Chernobyl. Bark was perhaps keeping Pu from direct deposition in 1986.

Chernobyl-origin ^{241}Am/$^{239+240}$Pu activity ratios in both O_F or O_H samples from Płaska, which have relatively low uncertainties, were close to 0.5. From known ^{241}Pu to $^{239+240}$Pu activity ratios in Chernobyl fallout, of to 53 [10] one can calculate the present value of ^{241}Am/$^{239+240}$Pu activity ratio. After 16 years of decay it should be at least 0.78 ("at least" since some initial activity of ^{241}Am was present as well). This discrepancy suggests that Am is a somewhat more mobile or bio-available than Pu and, therefore, removed a little from the soil. This can be supported by the data about this ratio obtained for insects (AS), root of pine and O_L+moss samples, which showed higher values of this ratio, although with relatively larger uncertainties. A comparison of data for almost all plant samples, affected or not by Chernobyl fallout, presented in Table 5, suggest also that the ^{241}Am to $^{239+240}$Pu activity ratio was higher than it was expected for a given site fallout. The exceptions are old pine or spruce needles (C+2 or C+1), which can be explained by more efficient translocation of Am than Pu within the plant.

Transfer factors and aggregation coefficients

Estimated *TF*'s from forest litter to plant and insects, defined following the formulae 3 or 4 are presented in Table 8. Values of *TF* for ^{137}Cs to bilberry were comparable with data given by other authors. For example in Sweden[34] it was found 1.04±0.84 (mean and standard deviation, 22 samples). In Austria[31] it was about 0.8 (readout from a figure). Our previous study[35] gave 1.3±1.4 (mean and standard deviation, 118 samples), other Polish result was 0.74±0.11.[36] The *TF* for ^{137}Cs to heather from Płaska is about 4 times lower than reported for Sweden samples,[34] but is almost exactly equal to 0.99±0.18 or 1.23±0.16 reported for Central Poland samples.[36]

As expected *TF*'s for ^{90}Sr seems to be similar to those for radiocesium in the majority of cases, but those values are a little misleading, since only a small fraction of radiostrontium is kept in O_H litter layer. *TF*'s for ^{241}Am between litter and majority of plants seems to be a little higher than that for Pu, which is two order of magnitude lower than that for radiocesium. The variation of *TF*'s for actinides seems to be not a wide one.

TF's between litter and insects were different for different species. High *TF*'s were observed for all species for ^{40}K. Relatively high *TF* were noticed for AS beetle for ^{137}Cs, and order of magnitude lower not only for ^{90}Sr but also for Pu, Am and Cm isotopes. High *TF*s were found for IT beetle in the case of ^{90}Sr.

The estimated value for *TF*'s from insect food to insect, defined as ratio of activity concentration in ashes of insect samples to activity concentration in dry mass of main food component, are presented in Table 9. A high transfer factor for ^{90}Sr in the case of IT beetle seems to be the most interesting finding. It suggests that birds which feed on this species can be exposed to elevated doses on the area with high ^{90}Sr levels.

AC's for chosen samples of plants and insects and radionuclides are presented in Table 10. Most likely, the AC values for radiostrontium are overestimated by an order of magnitude, since only about 10% of total deposition of ^{90}Sr was present in the litter layers. In the case of other radionuclides authors believe, that it is not more than a factor of two. However, results for radiocesium AC to plants seems to be consistent with the values obtained in other studies. For bilberry the AC found in Sweden[34] was 0.05±0.02 m^2/kg and in our case 0.006±0.003 to 0.214±0.009 m^2/kg. In our previous study it was in average 0.14±0.13 m^2/kg. In Germany[26] it was about 0.1 m^2/kg. For ferns in Germany it was about 0.45 m^2/kg, whereas our results are from 0.0039±0.0005 m^2/kg to 0.63±0.03 m^2/kg.

Table 8. Transfer factors (Bq/kg_{ash})/(Bq/kg_{dry}) relative to activity deposited in O_H layer of humus

Species	Site	^{137}Cs	^{40}K	^{90}Sr	$^{239+240}Pu$	^{238}Pu	^{241}Am	$^{243+244}Cm$
Plant								
Bilberry	T	0.73 ± 0.04	10.3 ± 2.7	0.30 ± 0.07	0.0045 ± 0.0021			
	WG	0.033 ± 0.003	1.1 ± 0.2	0.25 ± 0.13	0.0039 ± 0.0004			
	P*	1.45 ± 0.05	2.1 ± 0.4	0.89 ± 0.67	0.0020 ± 0.0002	0.0024 ± 0.0008		
Fern	T	1.55 ± 0.06	18.3 ± 4.2	0.60 ± 0.13	0.004 ± 0.001	0.047 ± 0.013		
	WG	0.020 ± 0.003	4.3 ± 0.7	1.25 ± 0.20	0.013 ± 0.001	0.010 ± 0.007		
	P	6.45 ± 0.32	2.5 ± 0.5	0.53 ± 0.11	0.029 ± 0.003	0.013 ± 0.003	0.08 ± 0.01	0.24 ± 0.16
Pine	T, C	0.641 ± 0.026	7.5 ± 2.1	0.10 ± 0.05	0.0008 ± 0.0004			
	T, C+1	0.121 ± 0.006	4.3 ± 1.2	0.45 ± 0.12	0.0016 ± 0.0004	0.018 ± 0.006		
	T, stem of 3 years old tree	0.069 ± 0.003	3.5 ± 0.9	0.30 ± 0.07	0.063 ± 0.008			
	T, root	0.103 ± 0.004	0.8 ± 0.4	0.40 ± 0.08	0.012 ± 0.002			
	P, C	0.827 ± 0.040	0.83 ± 0.21	0.10 ± 0.02	0.037 ± 0.004		0.161 ± 0.018	
	P, C+1	0.152 ± 0.011	0.50 ± 0.14	0.13 ± 0.02	0.010 ± 0.001		0.011 ± 0.002	
	P, C+2	0.107 ± 0.011	0.56 ± 0.16		0.017 ± 0.002		0.005 ± 0.003	
	P, stem of 3 years old tree	0.056 ± 0.010	1.57 ± 0.29	0.13 ± 0.04	0.018 ± 0.002	0.022 ± 0.016		
	P, root*	0.070 ± 0.004	0.17 ± 0.10	0.69 ± 0.17	0.036 ± 0.003	0.049 ± 0.007	0.061 ± 0.006	0.16 ± 0.05
Spruce	T, C	0.177 ± 0.006	4.1 ± 1.0	0.5 ± 0.1				
	T, C+1	0.170 ± 0.010	4.9 ± 1.4	0.9 ± 0.2	0.0049 ± 0.0009			
	T, bark 1	0.136 ± 0.005	1.0 ± 0.4	6.4 ± 1.5	0.010 ± 0.001			
	T, bark 2	0.112 ± 0.005	1.1 ± 0.5	2.5 ± 0.7	0.009 ± 0.001	0.018 ± 0.006		
	W.G. C+1	0.018 ± 0.003	0.5 ± 0.1	1.6 ± 0.3	0.0014 ± 0.0002			
	W.G. bark	0.020 ± 0.003	0.50 ± 0.08	7.8 ± 1.5	0.0010 ± 0.0002			
	P C	1.20 ± 0.05	0.85 ± 0.18	0.08 ± 0.02	0.013 ± 0.001		0.074 ± 0.009	
	P C+1	0.38 ± 0.02	0.51 ± 0.13	0.37 ± 0.07	0.016 ± 0.002		0.010 ± 0.002	
	P, bark	0.93 ± 0.04	0.22 ± 0.11	0.17 ± 0.05	0.025 ± 0.002	0.049 ± 0.006	0.039 ± 0.006	
Alder	P	0.020 ± 0.005	1.27 ± 0.24		0.0020 ± 0.0004	0.020 ± 0.005	0.009 ± 0.002	
Heather	P	1.53 ± 0.07	1.3 ± 0.2	0.27 ± 0.04	0.0051 ± 0.0009		0.017 ± 0.004	
Insect, (Bq/kg_{ash})/(Bq/kg_{dry})								
LM	T		195 ± 50		0.03 ± 0.02			
	P		31 ± 9	1.62 ± 0.33	0.08 ± 0.02			
AS	T	6.4 ± 0.2	83 ± 24	1.3 ± 0.3	0.41 ± 0.06			
	P*	22.4 ± 0.5	55 ± 8	2.3 ± 0.3	0.99 ± 0.08	1.19 ± 0.15	1.08 ± 0.19	3.7 ± 1.2
	P*	24.9 ± 0.6	62 ± 8	2.6 ± 0.4	0.89 ± 0.08	1.12 ± 0.14	0.75 ± 0.09	2.3 ± 0.9
HA	T	0.95 ± 0.03	129 ± 30	14.5 ± 2.7	0.21 ± 0.03			
	P	2.15 ± 0.25	13 ± 3	3.0 ± 0.5	0.25 ± 0.03	0.16 ± 0.06	0.30 ± 0.10	
IT	T	0.52 ± 0.03	48 ± 12	27.9 ± 4.5	0.09 ± 0.05	0.07 ± 0.05		
	WG	0.04 ± 0.01	8.2 ± 1.1	96 ± 17	0.0010 ± 0.0003	0.044 ± 0.011		
	P	0.30 ± 0.03	6 ± 1	21.2 ± 6.0	0.003 ± 0.001			
	P	1.47 ± 0.12	12 ± 2	23.8 ± 4.5				

* Calculated including contribution from O_F layer using Eq. (4).
Site codes: T – Tułowice, P – Płaska, WG – Węgierska Górka.

Table 9. Ranges of estimated transfer factor from specific plant to given insect species [in (Bq/kg$_{ash}$)/Bq/kg$_{dry}$)]

Species	Related sample	^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am
IT	Spruce bark	0.51–4.2	12–560	0.11–1.1	<2
LM	Pine needle	<0.32	<0.17	<31	<200
HA	Pine root	9–39	38–46	14–18	<28

Table 10. Aggregation coefficients [Eq. (5)] for chosen samples and nuclides

Species	Sample site	^{137}Cs m^2/kg$_{dry}$	^{90}Sr m^2/kg$_{dry}$	$^{239+240}$Pu (mBq/kg$_{dry}$)/Bq/m^2	^{241}Am, (mBq/kg$_{dry}$)/Bq/m^2
Plant					
Bilberry	T	0.063 ± 0.003	0.036 ± 0.012	0.21 ± 0.10	
	WG	0.006 ± 0.001	0.03 ± 0.02	0.85 ± 0.10	
	P	0.214 ± 0.009	0.11 ± 0.09	0.23 ± 0.03	0.6 ± 0.4
Fern	T	0.134 ± 0.005	0.072 ± 0.024	0.19 ± 0.06	
	WG	0.0039 ± 0.0005	0.17 ± 0.02	2.8 ± 0.3	
	P	0.63 ± 0.03	0.065 ± 0.013	5.0 ± 0.6	9.9 ± 1.6
Pine	C, T	0.0554 ± 0.0023	0.012 ± 0.007	0.04 ± 0.02	
	C+1,T	0.0104 ± 0.0005	0.054 ± 0.020	0.08 ± 0.02	
	Stem of 3 years old tree, T	0.0059 ± 0.0003	0.036 ± 0.012	2.9 ± 0.4	
	Root , T	0.0089 ± 0.0004	0.048 ± 0.015	0.55 ± 0.10	
	C, P	0.080 ± 0.004	0.012 ± 0.003	6.3 ± 0.6	20.5 ± 2.4
	C+1, P	0.015 ± 0.001	0.016 ± 0.003	1.8 ± 0.2	1.4 ± 0.3
	C+2, P	0.010 ± 0.001	2.9 ± 0.3	0.7 ± 0.4	
	Stem of 3 years old tree, P	0.005 ± 0.001	0.016 ± 0.005	3.1 ± 0.3	
	Root, P	0.009 ± 0.001	0.09 ± 0.02	4.8 ± 0.5	8.8 ± 1.0
Spruce	C, T	0.0153 ± 0.0006	0.060 ± 0.018		
	C+1, T	0.0147 ± 0.0008	0.108 ± 0.036	0.23 ± 0.04	
	Bark 1, T	0.0118 ± 0.0004	0.77 ± 0.25	0.48 ± 0.05	
	Bark 2, T	0.0097 ± 0.0004	0.30 ± 0.11	0.40 ± 0.05	
	Bark, WG	0.0039 ± 0.0005	1.07 ± 0.19	0.22 ± 0.04	
	C+1, WG	0.0034 ± 0.0005	0.22 ± 0.04	0.30 ± 0.04	
	C, P	0.117 ± 0.005	0.010 ± 0.002	2.2 ± 0.2	9.4 ± 1.1
	C+1, P	0.036 ± 0.002	0.045 ± 0.008	2.7 ± 0.3	1.3 ± 0.2
	Bark, P	0.090 ± 0.004	0.020 ± 0.007	4.3 ± 0.4	5.0 ± 0.8
Alder	P	0.0020 ± 0.0005		0.3 ± 0.1	1.1 ± 0.2
Heather	P	0.148 ± 0.007	0.033 ± 0.005	0.9 ± 0.2	2.1 ± 0.5
Insect, m^2/kg$_{ash}$					
LM	T			0.0014 ± 0.0011	
	P		0.20 ± 0.04	0.013 ± 0.003	
AS	T	0.553 ± 0.017	0.16 ± 0.05	0.019 ± 0.003	
	P	3.31 ± 0.11	0.29 ± 0.05	0.113 ± 0.011	0.16 ± 0.03
	P	3.31 ± 0.11	0.33 ± 0.06	0.117 ± 0.011	0.11 ± 0.02
HA	T	0.082 ± 0.003	1.75 ± 0.53	0.0099 ± 0.0014	
	P	0.21 ± 0.02	0.36 ± 0.06	0.043 ± 0.004	0.04 ± 0.01
IT	T	0.045 ± 0.003	3.36 ± 0.97	0.0004 ± 0.0002	
	WG	0.0068 ± 0.0015	13.2 ± 2.1	0.0002 ± 0.0001	
	P	0.030 ± 0.003	2.59 ± 0.73	0.0005 ± 0.0002	
	P	0.14 ± 0.01	2.92 ± 0.53		

Site codes: T – Tułowice, P – Płaska, WG – Węgierska Górka.

For heather in Sweden[34] the AC for ^{137}Cs was 0.50±0.06 m^2/kg and our value is 0.148±0.007 m^2/kg. AC for Pu and Am was found of approximately three orders of magnitude lower than those for ^{137}Cs. Values for AC seem to be higher for ^{241}Am than for $^{239+240}$Pu.

Relatively high values of ^{90}Sr AC for IT beetle were found. A little lower values were those of ^{137}Cs for AS beetle.

Conclusions

A clear decrease with time of radiocesium activity for all previously examined samples was noticed. However, it was not so in the case of plutonium isotopes. The activities of $^{239+240}$Pu in litter were almost constant, but activities of ^{238}Pu were lower, suggesting the existence of some kind of dynamic equilibrium between Pu fluxes down the soil profile and backward, with dead plants or fallen leaves. This mechanism averages ^{238}Pu to $^{239+240}$Pu activity ratio. A similar mechanism works perhaps also in the case of americium, resulting in decreasing ^{241}Am to $^{239+240}$Pu ratio in litter layers below the values expected for remains of Chernobyl fallout. Those results need to be confirmed since this was apparently overlooked so far. Usually forest litter was considered to be rather a passive reservoir of transuranics.

Transfer factors and aggregation coefficients for ^{137}Cs in case of plants were generally consistent with data obtained by other authors.

Small, but well measurable amounts of transuranic radionuclides were found for plant samples. This clears-up the discrepancy in isotopic ratio of Pu in Chernobyl fallout between measurements in air and in plant samples and gives additional support to a more dynamic behavior of transuranics in boreal forest than it was usually considered. Results suggest that Pu and Am are distributed unevenly in trees and translocated within plant, but this question should be studied in a more systematical way.

Activities found in insects confirmed the expectations only partially. Surprisingly, no radiocesium was found in nun-moth (*Lymantria monacha*). High concentration of ^{90}Sr caused by high transfer factors was noticed for spruce bark beetle (*Ips typographus*). The species forest dung beetle (*Anoplotrupes stercorosus*) showed the highest transfer factors and aggregation coefficients for ^{137}Cs and transuranic nuclides. The values for transuranics were an order of magnitude lower than for radiocesium.

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