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Plutonium and other alpha emitters in mushrooms from Poland, Spain and Ukraine

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Abstract

The paper presents results on Pu, U and Th isotope activity concentration measurements in some mushroom samples collected in Poland, Spain and Ukraine. The sampling sites differ a lot with regard to observed levels of Pu, its origin and isotope ratios as well as the environmental properties. Some of the Polish samples were collected in the north-eastern part of the country with up to 30 Bq/m^2 of Chernobyl Pu deposition. Other Polish and the Spanish samples are from areas with almost exclusively global fallout Pu present. Ukrainian samples were collected in a highly contaminated area with a deposition of about 3.7 kBq/m^2 of Chernobyl $^{239+240}Pu$. The maximum $^{239+240}Pu$ activity concentration was found equal to (54 ± 4) Bq/kg (dw—dry weight) for a Ukrainian *Cantharellus cibarius* sample. Ukrainian samples have an extremely high radiocesium level, with maximum of (51 ± 4) MBq/kg (dw). The maximum $^{239+240}Pu$ activity concentration for Polish samples was (81 ± 5) mBq/kg (dw) for *Xerocomus badius*. From the isotopic ratio in this sample it can be concluded that Chernobyl fallout is the origin of Pu. More than twice as large was the Spanish maximum for *Hebeloma cylindrosporum* but with only global fallout Pu. Some aspects of the transfer of nuclides to fruit bodies is discussed and in some cases the transfer factors or aggregation coefficients were calculated. Especially high transfer factors were found for *Hebeloma cylindrosporum* from Spain.

Keywords: Plutonium; Radionuclides in mushrooms; Alpha emitters; Chernobyl; Global fallout; Uranium; Thorium

1. Introduction

Plutonium is considered rather insoluble in the environment. Therefore it is commonly not expected, that it might go to the fruit bodies of mushrooms or to the plants. However, some traces of plutonium were found in such samples analysed in the past even for the global fallout contamination levels (Mietelski et al., 1993; Barnett et al., 1997). This problem seemed to be interesting for a more careful examination. Till now the data on this topic is rather scarce as can be seen from a recent review of the work on mushroom contaminations (Jacquiot and Daillant, 2000). A laboratory experiment under controlled conditions on the transfer of plutonium to fruit bodies of mushrooms is now being conducted in a collaboration of Spanish laboratories, continuing previous experiments on radiocesium and radiostrontium uptake by mushrooms. (Baeza et al., 2000). Complementary to the laboratory experiments we made some field studies to get more data on the contamination levels and to get a general information on the order of magnitude of transfer factors or aggregation coefficient (AC) values observed in different environments. The transfer factor (TF) is defined as the ratio of activity

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concentration in dry mass of mushroom sample to the activity concentration in soil or forest litter, whereas the AC is defined as the ratio of activity concentration in dry mass of mushroom sample to the surface activity in soil or forest litter. We hoped, that the data obtained now would be helpful for the discussion and interpretation of the experiment conducted under controlled conditions. The data on Pu concentration for some mushroom samples from highly contaminated area of Ukraine were obtained and included already after finishing Pu measurements of Polish or Spanish samples. Those results were needed to verify and clarify some predictions suggested by Polish or Spanish sample results.

With the aim to compare the Pu behaviour with that of some natural nuclides we were also looking for U and Th activities in the case of Polish and Spanish samples. To make calculations of transfer factors or aggregation coefficients possible some soil or forest litter samples were analysed as well. However, the majority of samples were originating from other investigation projects conducted by our laboratories in the past, therefore data on radionuclide concentrations in soil or forest litter were in some cases already available.

In Poland the plutonium fallout from Chernobyl occurred predominantly in the form of small hot particles (Mietelski, 1998) in the farthest north-eastern Poland. The maximum deposition for all Pu alpha emitters from Chernobyl there was about $30\,Bq/m^2$, approximately two-third of it was $^{239+240}Pu$ (Mietelski and Wąs, 1995). In the other Polish sites, the plutonium deposition is almost solely due to global fallout (Fig. 1). Chernobyl Pu was never found in any Spanish sample.

The percentage F of Chernobyl origin $^{239+240}Pu$ in a given sample can be calculated (Mietelski and Wąs, 1995) from a simple formula:

$$F = 100\%\frac{A_{ch}}{A_{239}} = 100\%\left(\frac{A_{238}/A_{239} - 0.03}{0.55 - 0.03}\right), \quad (1)$$

where A_{ch} is the activity concentration of $^{239+240}Pu$ originating from Chernobyl, A_{239} and A_{238} are activity concentrations observed in samples for $^{239+240}Pu$ and ^{238}Pu, respectively, 0.03 is the ^{238}Pu to $^{239+240}Pu$ activity ratio in global fallout (UNSCEAR, 1982), 0.55 is the same for Chernobyl fallout in Poland (Mietelski and Wąs, 1995).

2. Materials and methods

2.1. Polish samples

More than 20 samples of fruit bodies of mushrooms from all over Poland belonging to the species: *Boletus edulis, Xerocomus badius, Macrolepiota procera, Lactarious vellereus* and *Armillariella mellea* collected in 1991 within a project for investigations in forests of Poland (Mietelski et al., 1996) were selected for analyses. All samples were measured in the past by gamma-ray spectrometry to determine the ^{134}Cs, ^{137}Cs and ^{40}K activity concentrations. These measurements were done during 1992–94 and the results were already published (Mietelski et al., 1994; Mietelski and Jasińska, 1996). The alpha activity concentrations of Pu isotopes in forest litter or humus layers for a lot of sampling sites were also known from previous measurements (Mietelski and Wąs, 1995). However, for the present work we did some additional measurements of Pu alpha activity

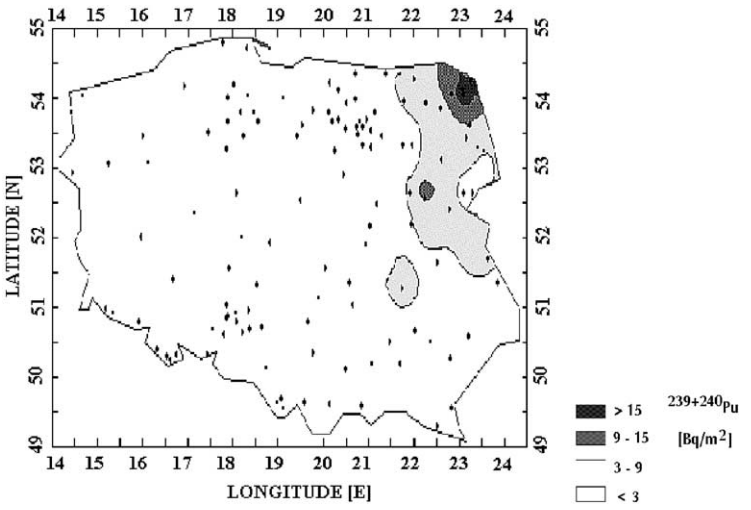

Fig. 1. Approximate deposition pattern of $^{239+240}Pu$ from Chernobyl in Poland. The map is prepared on the basis of ^{144}Ce measurements in forest litter and the $^{239+240}Pu$ to ^{144}Ce activity ratio (Mietelski and Wąs, 1995). Dots are the sampling sites.

concentrations for some further locations, as well as some Th and U determinations.

Initial results (Mietelski et al., 1993) suggested that to achieve reasonable precision for plutonium measurements in mushrooms we should use samples of masses of about 100 g (dw—dry weight). Therefore in several cases, we had to combine smaller samples to bigger sets of samples. The combined samples originated from approximately the same geographical area with approximately the same contamination observed, and were of course of the same species. The distance between sampling sites did not exceed 150 km for the areas in which mostly global fallout for Pu was observed. It was only 50 km for the areas affected with a significant amount of Chernobyl plutonium fallout. The sampling sites are displayed on the radiocesium contamination map in *Xerocomus badius* samples of Poland in Fig. 2.

Mushroom samples were cleaned only to such a degree that they could be taken as foodstuff. One sample of mushroom *Xerocomus badius* was treated in a special way. Each dried fruit body of this sample was carefully brushed using distilled water with a cleaning agent (a commercial detergent) added. Both the solution and the brushed sample were taken for analyses, in both cases relating the measured activity concentration to the total DW of sample. This sample, coded here X–X, was collected in 1993 in north-west Poland, close to locations 5, 273 and 109.

Besides plutonium, most of the samples were analysed for uranium and thorium presence also. *Boletus edulis* samples were not analysed for uranium, since the concept of uranium analyses was introduced later, when those samples were already destroyed. All nuclides were determined during one combined procedure. The part of the method applied for Pu determination has already been described in detail elsewhere (Mietelski and Wąs, 1995) and is used routinely in the Environmental Radioactivity Laboratory at Henryk Niewodniczański Institute of Nuclear Physics (INP), Kraków. This method is based on the method used in the IAEA Laboratories in Seibersdorf (LaRosa et al., 1992) for the Chernobyl Project samples. We used ^{236}Pu as a spike solution (about 10 mBq per sample). Because of the relatively short half-life of this isotope (2.85 years), for each sample the actual spike activity was calculated for the measurement day. As a tracer for uranium the ^{232}U from the ^{236}Pu decay present anyway in the Pu spike was used (about 5.2 mBq per sample). For thorium we have not used any tracer, therefore those results are just the lower limits of activity concentrations. Thorium is always separated during our Pu analyses procedure. After deposition of Pu and Th from 8 M HNO_3 on Dowex-1 anion exchange column conditioned to a nitric form, thorium is stripped off by elution using 50 ml 10 M HCl. Then, Pu is eluted using 25 ml of 0.1 M HCl-0.1 M HF solution. From both fractions alpha-spectrometric sources were prepared by means of the NdF_3 co-precipitation method (Sill, 1987).

The uranium procedure was a special modification for this project of the procedure used in Cáceres for water samples (Dregge and Boden, 1984) and it was applied almost without any changes at both Kraków and Cáceres Laboratories (for Polish and Spanish samples, respectively). The original 8 M HNO_3 effluent from the Dowex-1 column, which was supposed to contain the majority of uranium, was gently evaporated from the initial volume of about 200 ml to about 20–30 ml, and then again diluted to 200 ml with deionised water. The

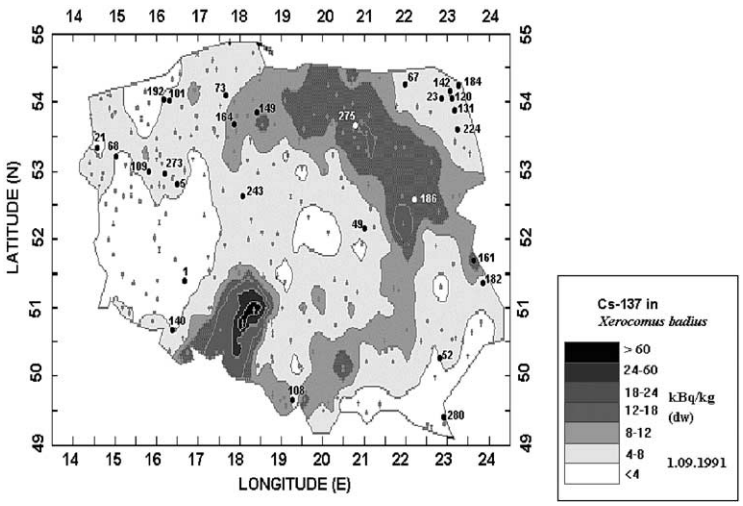

Fig. 2. Approximate map of radiocesium activity concentration in Polish *Xerocomus badius* samples collected in 1991 (Mietelski et al., 1994). The small dots mark 280 sampling sites, some of them are numbered. The numbers are the codes used also in present paper.

co-precipitation of U with hydroxides was done after adjusting the pH to 9 using ammonia. The precipitate was separated from the supernatant by centrifuging, and then it was dissolved using at least 50 ml of hot 10 M HCl. After boiling for 1 h and significant evaporation of this solution, a fresh portion of 80 ml of 9 M HCl was added and the sample was again heated for 5 min. After cooling down the solution was passed through the anion-exchange Dowex-1 column, 10 cm high, 6 mm of inner diameter, conditioned with 9 M HCl. Uranium and iron were expected to fix on the column. Iron was stripped off using 25 ml of 8 M HNO_3. Uranium was then eluted using 50 ml of 8 M HNO_3 and then 100 ml of water (collected into one beaker). The U fraction was evaporated to almost dryness and then diluted to 0.5 M HNO_3. The iron fraction was also evaporated, converted to 0.5 M HNO_3 and the NdF_3 co-precipitation was done to obtain a source. Sources prepared from this fraction did not contain any measurable amount of U. During evaporation in hot nitric acid the majority of uranium turns to U^{+6} ions, which do not co-precipitate with NdF_3. Therefore, even if some uranium would be present in the Fe fraction it will remain in solution. The filtered solution was combined with the 0.5 HNO_3 solution, which contained the U fraction. To this combined solution 0.8 g of Mohr salt was added as a reducing agent for uranium (Sill, 1987). This salt converts U ions to a lower oxidation step (+3 or +4), making uranium co-precipitation with NdF_3 possible. Sources were then prepared using this method.

For each 10 samples one blank sample spiked with tracer was analysed to determine the actual true background of the spectrometer. To achieve quality control of analyses we applied the procedure to Reference Materials (RM): IAEA 300—Baltic Sea sediment (IAEA, 1997) and IAEA Soil 6—Pre-Chernobyl soil from Austria (IAEA, 1994). To our best knowledge, there is no RM for mushrooms with certified Pu activity value available as yet.

Measurements of alpha activities were performed using a Silena AlphaQuattro alpha-spectrometer equipped with two INP (Kraków) manufactured SBSi detectors and two Canberra PIPS ion-implanted detectors. Example of alpha spectra of Pu, U and Th fractions are presented in Figs. 3, 4 and 5A, respectively.

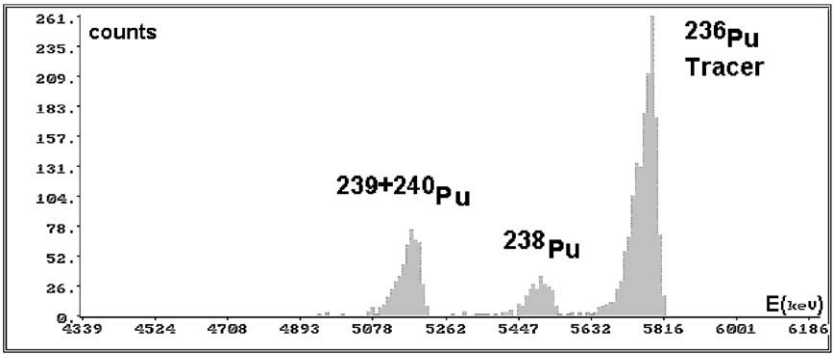

Fig. 3. Alpha spectrum of the Pu fraction of *Xerocomus badius* sample No. 142 from north-eastern Poland; the isotopic composition is typical for Chernobyl fallout. Measurement time 415832 s, detector Canberra PIPS 450 mm^2, spectrometer Silena Alpha Quattro.

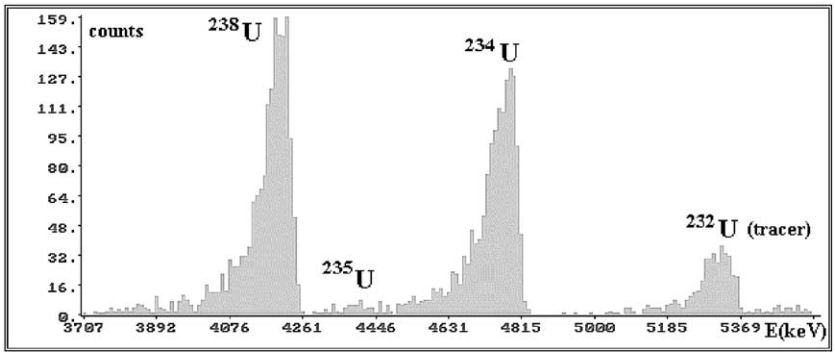

Fig. 4. Alpha spectrum of the U fraction of *Xerocomus badius* combined samples No. 120 + 131 from north-eastern Poland. Measurement time 356866 s, detector Canberra PIPS 450 mm^2, spectrometer Silena Alpha Quattro.

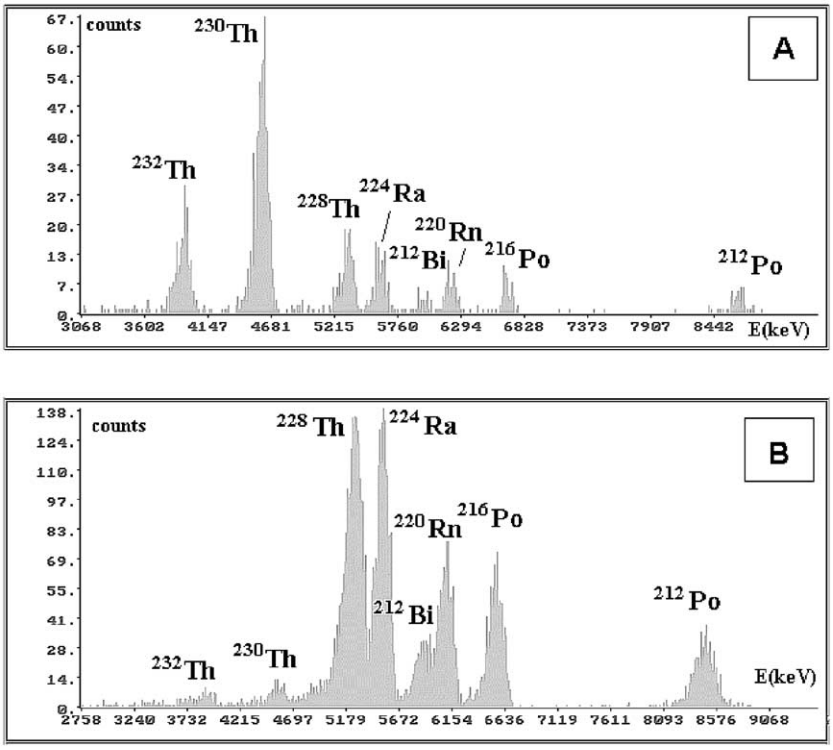

Fig. 5. Comparison of thorium fraction alpha spectra (site 49) for (A) fruit bodies of mushroom *Xerocomus badius* and (B) for the leaves of plant—bilberry *Vaccinium myrtillus.* Measurement time 76521 and 160942 s, for first and second spectrum, respectively, SBSi detectors of 300 mm^2 area each, Silena AlphaQuattro spectrometer.

2.2. Spanish samples

Plutonium in Spanish samples was expected to be present only due to global fallout. Neither traces of the Chernobyl accident nor of the Palomares accident were supposed to be found. For naturally occurring radionuclides like uranium and thorium isotopes we were interested in the differences between Spanish and more northern European sites. Both the environmental conditions and type of forests are different from those in central or Eastern Europe.

Samples of fruit bodies of the species *Boletus sp., Hebeloma cylindrosporum, Amanita muscaria, Omphalotus olearis, Agaricus campestris* were collected in central Spain. We were not able to find the same species as in Poland. The samples were washed prior to the analyses. Forest soil was collected only in one location (Muñoveros, near Segovia), about 50 km west from the foothill of Sierra de Guadarrama mountains, in a pine wood forest where samples of *Heboloma cylindrosporum* were collected. From previous, as yet unpublished results obtained at the University of Extremadura this particular species was expected to have an unusual high transfer factor at least for thorium. Samples were analysed for all gamma emitters using a typical low-background gamma-ray spectrometer with a germanium detector. Then, they were analysed for the presence of Th and Pu alpha emitters using about the same procedure as described above for the analyses of the Polish samples. The U isotope analysis procedure was also practically identical as described above for the analyses of Polish samples. The main differences in the procedures compared to those described above and used in Kraków were the following: the spikes were ^{242}Pu and separately ^{232}U; larger and not pressurised columns were used for Pu and Th separation; Pu was stripped off the Dowex-1 column using a solution of 0.1 M HCl in which a few crystals of hydroxylamine were dissolved (Iranzo et al., 1992). There was also a small difference in one detail of the U elution from the DOWEX-1 column. Here only 8 M HNO_3 (no water) was used for the elution. Sources were also prepared using the NdF_3 co-precipitation method. Measurements of alpha activities were performed applying an alpha-spectrometer with ion-implanted silicon detectors.

2.3. Ukrainian samples

Five samples of fruit bodies of mushrooms of the species *Xerocomus badius, Suillus luteus, Boletus edulis, Paxillus involutus* and *Cantharellus cibarius* were collected at a highly contaminated area within the 30 km exclusion zone of Chernobyl NPP. The whole area was a subject of intense investigation within the last years. Forest litter and soil was collected to the depth of 25 cm, and analysed for Pu. Prior to the Pu determination, all mushroom samples were analysed for the presence of gamma emitters using a gamma-ray spectrometer with germanium detector and then for plutonium following the routine procedure (Bondarenko et al., 2000a; Bondarenko et al., 2000b) used at the Laboratory of Radiochemistry of the Research Center for Radiation Medicine, Academy of Medical Sciences of the Ukraine. Sources were prepared using an electroplating and measured using an alpha-spectrometer with ion-implanted detectors. ^{242}Pu was used as a spike.

3. Results and discussion

The results of plutonium alpha activity concentrations in RM-s, obtained in our laboratories are presented in Table 1. The obtained consistency is satisfactory. Quoted uncertainties, here and in further Tables, are standard deviations due to counting statistics.

3.1. Polish samples

The results for Pu activity concentrations in mushrooms from Poland are presented in Table 2. The obtained arithmetic mean for the recovery of plutonium (including parallel measurements for additional forest litter samples) was 72%, with a standard deviation equal to 17%.

The maximum activity concentration was equal to (81 ± 5) mBq/kg (dw) and (31 ± 2) mBq/kg (dw) for $^{239+240}$Pu and ^{238}Pu, respectively. It was found in a *Xerocomus badius* from Augustów Primeval Forest (north-eastern Poland). This were combined samples from sites coded 120 and 131 (codes of areas as in other papers by Mietelski et al., using samples collected during the 1991 sample collection campaign). The relatively high ^{238}Pu content suggests the Chernobyl accident as a dominant origin of the Pu. The estimated percentage of Chernobyl origin $^{239+240}$Pu, F, is given in last column of Table 2. Other samples of this species, even collected nearby this area (for example coded 23, 142, 224) showed lower activity concentrations by a factor of five or even ten, still having a high ^{238}Pu to $^{239+240}$Pu activity ratio and a high value of F. The highest $^{239+240}$Pu activity concentration of *Boletus edulis* samples was found for the sample from mountains in the South of Poland (coded 108), equal to (28 ± 4) mBq/kg, with ^{238}Pu below the detection limit of 0.6 mBq/kg. This means that global fallout is the origin of Pu. Samples of other species had Pu activity concentrations of the order of a few mBq/kg (dw).

Thorium results are presented in Table 3. The activity concentrations were calculated under the assumption of complete, equal to 100%, recovery of this element. The actual recovery was for sure lower. Data from reference material analyses suggest a recovery of about 50%. This means that activities were at least as high as those presented in Table 3 and probably equal to the double of those values. There is no doubt, that thorium is present in traces in the examined fruit bodies. Since there is no reason for systematic differences in recovery between the species, one can notice that on the average samples of *Boletus edulis* seem to have lower Th activities than the other species. The determined activity concentrations were on the level from a few mBq/kg (dw) up to (228 ± 15) mBq/kg and (616 ± 24) mBq/kg for ^{232}Th and ^{230}Th, respectively. The maximum activity concentration was found in a *Xerocomus badius* sample from site 49 (central Poland). The ratio of ^{230}Th to ^{232}Th varies from

Table 1
Obtained activity concentrations for plutonium isotope concentrations in analysed reference materials compared with certified values and confidence limits (IAEA, 1994,1997), all results in mBq/kg dry mass

Description	Name	$^{239+240}$Pu (mBq/kg dw)		^{238}Pu (mBq/kg dw)		Laboratory
		Measured	Certified value (Confidence limits)	Measured	Certified value (Confidence limits)	
Baltic Sea sediment	IAEA 300	I:3419±186 II: 3622±214	3550 (3440–3650)	I:171±15 II: 177±20	150[a]	Kraków (2 analyses)
Soil from Austria	IAEA Soil 6	1029±75	1040 (962–1110)	23±6	[b]	Kraków
Soil from Austria	IAEA Soil 6	1070±60	1040 (962–1110)	40±12	[b]	Cáceres

[a] No data for confidence limits for ^{238}Pu, only "Information value" is available.
[b] No data for ^{238}Pu; as it is global fallout Pu it should be about 30 mBq/kg.

Table 2
Results of plutonium measurements for mushroom samples from Poland, the percentage of Chernobyl origin plutonium, F, is defined by Eq. (1)

Species	Site code	Dry mass (g)	Recovery (%)	$^{239+240}$Pu (mBq/kg dw)	^{238}Pu (mBq/kg dw)	F (%)
Boletus edulis	108	76.9	39.1±1.6	27.8±3.8	<0.6	<30
	120+131	201.5	70.1±2.5	3.0±0.4	1.0±0.3	58±19
	182+186	157.8	52.5±1.6	4.1±0.5	0.2±0.3	4±5
	67+184	225.3	58.1±1.5	2.1±0.3	0.2±0.2	13±13
	1+140	224.9	70.2±1.8	3.0±0.3	0.5±0.2	26±11
	52+280	190.8	54.5±1.7	3.2±0.4	0.3±0.2	12±8
	68+101+192	98.8	85.5±2.2	2.5±0.5	0.9±0.5	64±38
	5+21+109+273	222.2	56.3±1.5	4.3±0.6	0.6±0.4	21±14
	73+149+164+243	197.8	61.7±1.9	4.1±0.5	1.0±0.2	41±10
	275	90.4	80.3±2.0	2.0±0.4	0.8±1.2	⩽100
Xerocomus badius	23	38.8	69.1±1.8	5.2±1.0	0±1.1	<23
	49	73.3	69.5±1.9	3.1±0.6	0.1±0.6	<3
	67+184	143.6	58.1±1.6	4.6±0.5	2.4±0.4	95±19
	120+131	142.2	83.4±2.4	81.0±5.0	31.0±2.0	68±6
	142	189.8	81.3±2.2	16.7±1.1	7.9±0.7	85±9
	161	158.4	85.6±2.2	1.3±0.3	0.8±0.3	113±50
	224	42.1	89.0±2.1	2.9±0.7	1.9±1.0	120±70
	X–X	114	96.7±2.2	1.7±0.3	0.8±0.2	85±26
The wash of X–X sample		114	95.2±2.1	1.2±0.3	1.2±0.3	>100
Macrolepiota procera	23	16.8	94.4±2.1	5.4±1.7	3.3±2.5	112±92
	142	68.3	72.8±1.9	9.0±1.1	3.6±0.8	71±18
Lactarious vellereus	49 (1st half)	75.1	36.5±1.4	6.3±1.1	2.6±0.7	74±24
	49 (2nd half)	64.5	58.2±2.4	6.1±1.1	1.5±0.7	42±21
	161	162.3	40.5±1.6	4.9±0.7	0.2±0.3	2±3
Armillariella mellea	224	57.5	59.5±1.7	3.5±0.8	0.4±0.7	16±29

0.65±0.15 to 2.70±0.20. In almost all cases samples of different species originating from the same location have similar values of this ratio.

Although no radium isotope measurements were done some information on radium activity could be deduced. The interesting features are small values of ^{228}Th to ^{230}Th or to ^{232}Th activity ratios for mushrooms. For instance in animal bone samples (Mietelski et al., 2000) the ^{228}Th is far more active than ^{230}Th or ^{232}Th. This is due to the incorporation of ^{228}Ra by the organism, and subsequent decay of this nuclide through ^{228}Ac to ^{228}Th. Similar disequilibrium between ^{228}Th and ^{232}Th can be observed for plant samples, whereas nothing like that happens in the case of all examined species of mushrooms. It can be easily illustrated qualitatively just by a comparison of spectra for plant and mushroom sample (Fig. 5A and B). Although the difference seems to be obvious, we were not able to describe this feature quantitatively as yet. The ^{228}Th to ^{232}Th activity ratio in the sample was disturbed due to some ^{228}Th added together with the ^{236}Pu+^{232}U spike, but since the thorium recovery was unknown, the proper correction factor was undefined. The observed difference is apparently caused by a difference in radium accumulation properties between plant and mushroom. This suggests, that the ^{228}Ra activity concentrations for all examined samples do not exceed significantly those for ^{232}Th.

Results for uranium analyses are presented in Table 4. The recovery for the ^{232}U tracer varied from (16±2)% to (106±10)%, with the arithmetic mean equal to 53% and a standard deviation of 27%. If one would assume a 50% recovery for thorium, uranium activity concentrations seem to be higher than those for thorium. The average for *Xerocomus badius* is not too much different from activities determined for samples of other species (although no results for *Boletus* were available). The maximum ^{238}U activity was again observed for a sample of *Xerocomus badius* from site 49, which was (240±70) mBq/kg (dw). The uranium isotopic ratio, ^{234}U to ^{238}U

Table 3
Results for thorium isotope activity concentrations in mushroom samples calculated under the assumption, that the recovery of thorium was 100%

Species	Site code	^{232}Th (mBq/kg dw)	^{230}Th (mBq/kg dw)	^{230}Th:^{232}Th
B. edulis	120 + 131	2.5 ± 0.4	2.5 ± 0.4	1.03 ± 0.25
	1 + 5 + 21 + 52 + 68 + 73 + 101 + 108 + 140 + 149 + 164 + 192 + 243 + 273 + 280	4.0 ± 0.2	3.9 ± 0.2	0.97 ± 0.07
	275 + 182 + 186	4.0 ± 0.5	4.9 ± 0.6	1.21 ± 0.22
X. badius	23	41 ± 7	48 ± 8	1.19 ± 0.29
	49	228 ± 15	616 ± 24	2.70 ± 0.20
	67	5.8 ± 0.6	4.8 ± 0.6	0.83 ± 0.13
	120	8.0 ± 0.7	8.3 ± 0.7	1.04 ± 0.14
	142	1.9 ± 0.3	1.5 ± 0.2	0.78 ± 0.16
	161	3.8 ± 0.5	3.4 ± 0.4	0.91 ± 0.15
	224	31 ± 3	28 ± 2	0.90 ± 0.11
	X–X	3.2 ± 0.5	2.3 ± 0.4	0.74 ± 0.16
Wash X–X		6.0 ± 0.7	8.8 ± 0.8	1.47 ± 0.21
M. procera	23	33 ± 5	21 ± 4	0.65 ± 0.15
	142	3.1 ± 0.6	3.0 ± 0.7	0.96 ± 0.30
L. vellereus	49 (two halves)	17 ± 2	26 ± 3	1.51 ± 0.23
	161	13 ± 2	14 ± 2	1.08 ± 0.18
A. mellea	224	9.4 ± 1.1	13.6 ± 1.4	1.44 ± 0.22

The real activities should be considered at least as high as presented here, but the ratio presented in the last column is not affected by this possible systematic error.

Table 4
Results for uranium determination in mushrooms from Poland

Species	Site code	Recovery (%)	^{238}U (mBq/kg dw)	^{234}U (mBq/kg dw)	^{235}U (mBq/kg dw)	^{234}U:^{238}U
Xerocomus badius	23	16 ± 2	70 ± 21	62 ± 13		0.95 ± 0.22
	49	51 ± 5	240 ± 70	240 ± 20	9 ± 5	0.99 ± 0.05
	67	19 ± 4	50 ± 20	60 ± 10		1.15 ± 0.19
	120	41 ± 5	160 ± 50	140 ± 10	6 ± 4	0.86 ± 0.05
	142	34 ± 3	130 ± 30	89 ± 7		0.69 ± 0.06
	161	106 ± 10	15 ± 4	14 ± 2		0.95 ± 0.14
	224	97 ± 10	70 ± 20	66 ± 9		1.00 ± 0.16
	X–X	46 ± 5	22 ± 6	23 ± 3		1.05 ± 0.16
Wash of X–X		69 ± 6	18 ± 4	17 ± 2		0.97 ± 0.12
L. vellereus	49 (two halves)	79 ± 7	120 ± 30	140 ± 20		1.17 ± 0.16
	161	40 ± 3	170 ± 30	170 ± 10	10 ± 5	0.98 ± 0.07
M. procera	23	42 ± 5	270 ± 90	120 ± 20		0.46 ± 0.08
	142	50 ± 5	100 ± 30	62 ± 6		0.65 ± 0.06
A. mellea	224	45 ± 5	100 ± 30	50 ± 7		0.50 ± 0.07

was about unity or a little higher than one for *Xerocomus badius* and *Lactarius vellereus* samples (as it was also for forest litter samples), whereas for samples of *Macrolepiota procera* and *Armillariella mellea* this ratio was surprisingly low, varying from 0.46 ± 0.08 to 0.65 ± 0.06. Although both uranium isotopes belong to one uranium series, in environmental samples this ratio is often not equal to unity. It is > 1 for water samples

and <1 for mineral material. In biological samples this ratio might be expected to be >1, since the water transports ions to organisms. For some of our samples we obtained an opposite result. This surprising feature needs perhaps wider and more careful studies in future.

3.2. Spanish samples

Results for the alpha emitter measurements in samples collected in Spain are presented in Tables 5–7. A relatively high $^{239+240}$Pu concentration equal to (164±21) mBq/kg (dw) was found for the sample of *Hebeloma cylindrosporum* mushroom. A second relatively high result, equal to (51.0±0.5) mBq/kg (dw), was observed for *Agaricus campestris* mushroom. For other species plutonium was present on the level of a few mBq/kg (dw). The presented thorium results are calculated under the assumption of 100% recovery, since no tracer was available. Therefore the presented results are surely underestimated. Surprisingly high thorium activity concentrations were found again for the *Hebeloma cylindrosporum* species. It was (4.25±0.14) Bq/kg (dw) for ^{232}Th and (2.43±0.10) Bq/kg (dw) for ^{230}Th. Similarly high activity concentrations of uranium were found for this species also: (3.38±0.16) Bq/kg (dw) and (3.41±0.16) Bq/kg (dw) for ^{238}U and ^{234}U, respectively. The ^{234}U to ^{238}U activity ratio (see Table 7) was close to unity for *Hebeloma cylindrosporum*, whereas it was about 1.3 for the *Boletus* sample.

3.3. Ukrainian samples

The results for plutonium and, for comparison, for radiocesium, are presented in Table 8. The results for radiocesium are given only because of the striking extremely high activity concentration levels (up to

Table 5
Results for plutonium isotope concentrations obtained for Spanish samples

Sample	Dry mass (g)	Recovery (%)	$^{239+240}$Pu (mBq/kg dw)	^{238}Pu (mBq/kg dw)
Forest soil—Muñoveros	9.82	81.5±2.4	246±23	<7.6
Hebeloma cylindrosporum	33.09	32.6±2.0	164±21	<2.0
Boletus sp.	104.81	63.2±1.6	5.3±0.9	<0.9
Amanita muscaria	1159.1	64.8±3.2	0.9±0.2	<0.03
Omphalotus olearius	66.46	91.7±1.2	8.3±0.6	<0.9
Agaricus campestris	50.83	95.9±1.0	51±0.5	<1.9

Table 6
Results for thorium isotope concentrations obtained for Spanish samples, calculated under the assumption, that the recovery of thorium was 100%

Sample	^{232}Th (Bq/kg dw)	^{230}Th (Bq/kg dw)	^{232}Th:^{230Th}
Forest soil—Muñoveros	11.1±0.3	6.9±0.2	1.62±0.06
Hebeloma cylindrosporum	4.25±0.14	2.43±0.10	1.75±0.09
Boletus sp.	0.365±0.017	0.336±0.016	1.09±0.07
Amanita muscaria	0.0611±0.0026	0.0535±0.0024	1.14±0.07
Omphalotus olearius	1.1±0.03	0.75±0.022	1.46±0.05
Agaricus campestris	0.305±0.017	0.249±0.015	1.23±0.1

The real activities should be considered at least as high as presented here, but the ratio presented in the last column is not affected by this possible systematic error.

Table 7
Results for uranium isotope concentration obtained for Spanish samples

Sample	Recovery (%)	^{238}U (Bq/kg dw)	^{235}U (Bq/kg dw)	^{234}U (Bq/kg dw)
Forest soil—Muñoveros	20.7±1.2	7.30±0.50	0.42±0.09	7.00±0.50
Hebeloma cylindrosporum	44.7±1.9	3.38±0.16	0.16±0.02	3.41±0.16
Boletus sp.	51.7±2.3	0.74±0.04	0.040±0.006	0.99±0.05
Amanita muscaria	35.6±1.5	0.120±0.006	0.007±0.001	0.150±0.007

Table 8
Results for plutonium and radiocesium for Ukrainian samples

Species	Dry mass (g)	^{137}Cs (MBq/kg dw)	$^{239+240}$Pu (Bq/kg dw)	^{238}Pu (Bq/kg dw)
Xerocomus badius	20.3	21.4±1.5	2.96±0.24	1.13±0.09
Suillus luteus	21.3	14.4±1.0	3.38±0.27	2.04±0.16
Paxillus involutus	20.3	50.7±3.6	2.78±0.22	1.7±0.14
Boletus edulis	13.1	0.517±0.036	<LD	<LD
Cantharellus cibarius	21.7	2.12±0.15	53.78±4.30	24.1±1.93

Table 9
Statistical parameters on plutonium transfer factors from the leaf layer and humus layer to different species of mushrooms from Poland

Species	Number of samples	Layer	TF Mean	Median	Std. Dev.	Min.	Max
Boletus edulis	4	Leaf	1.91×10^{-2}	8.38×10^{-3}	2.72×10^{-2}	1.00×10^{-3}	5.86×10^{-2}
		Humus	1.86×10^{-3}	1.39×10^{-3}	1.64×10^{-3}	9.77×10^{-4}	4.17×10^{-3}
Xerocomus badius	6	Leaf	1.03×10^{-2}	3.18×10^{-3}	1.41×10^{-3}	0.08×10^{-3}	37.8×10^{-3}
		Humus	3.95×10^{-3}	3.19×10^{-3}	4.11×10^{-3}	5.50×10^{-4}	1.30×10^{-2}
Lactarius vellereus	3	Leaf	4.55×10^{-2}	6.16×10^{-3}	2.97×10^{-2}	1.11×10^{-2}	6.36×10^{-2}
		Humus	8.51×10^{-3}	6.47×10^{-3}	3.71×10^{-3}	6.27×10^{-3}	1.28×10^{-2}
M. procera	2	Leaf	2.17×10^{-3}	2.17×10^{-3}	—	1.67×10^{-3}	2.67×10^{-3}
		Humus	1.94×10^{-3}	1.94×10^{-3}	—	1.88×10^{-3}	2.00×10^{-3}
A. mellea	1	Leaf	1.03×10^{-3}	—	—	—	—
		Humus	6.64×10^{-4}	—	—	—	—

The data on Pu concentration in forest litter was available only for 4 sites of *Boletus edulis* sampling.

Table 10
Aggregation coefficients (AC) (m^2/kg) for $^{239+240}$Pu, calculated for samples from Poland. Only for 4 sites of *Boletus edulis* sampling data on Pu concentration in forest litter was available

Species	Number of samples	AC Mean	Median	Std. Dev.	Min.	Max.
Boletus edulis	4	2.54×10^{-4}	2.11×10^{-4}	1.37×10^{-4}	1.40×10^{-4}	4.53×10^{-4}
Xerocomus badius	6	1.39×10^{-3}	1.05×10^{-3}	1.41×10^{-3}	0.08×10^{-3}	37.8×10^{-3}
Lactarius vellereus	3	3.14×10^{-3}	2.77×10^{-3}	0.72×10^{-3}	2.68×10^{-3}	3.96×10^{-3}
Macrolepiota procera	2	4.43×10^{-4}	4.43×10^{-4}	—	3.22×10^{-4}	5.65×10^{-4}
Armillariella mellea	1	9.18×10^{-5}	—	—	—	—

50.7 MBq/kg dw!). Despite this fact, plutonium was present on the level of a few Bq/kg (dw), except for a *Cantherellus cibarius* sample, for which a $^{239+240}$Pu activity concentration of 53.8 Bq/kg (dw) was found while that of ^{238}Pu was equal to 24.1 Bq/kg (dw). The ^{238}Pu to $^{239+240}$Pu activity ratio which varies from 0.38 to 0.61 left no doubts, that the Chernobyl accident is the origin of Pu in the samples.

The comparison of activity concentrations found for Polish mushroom samples with previously (Mietelski and Wąs, 1995) or currently determined forest litter activity concentrations at the same sites, resulted in data needed for the estimation of the transfer factors (TF) or aggregation coefficients (AC). The statistical parameters for plutonium transfer factors for Polish samples are presented in Table 9. The aggregation coefficients calculated on the basis of the Pu activity concentrations observed in mushroom and in leaf and humus layers of forest litter are presented in Table 10. The relatively high values (on the level of 10^{-2}) for the transfer factor from leaf layer are most likely artefacts caused by the low leaf layer Pu activity observed in these cases. The aggrega-

Table 11
Data on transfer factors for Th isotopes for sample 67 and 49 of *Xerocomus badius* and 49 of *Lactarius vellereus*

Sample	TF(^{232}Th, leaf)	TF (^{230}Th, leaf)	TF(^{232}Th, humus)	TF(^{230}Th, humus)
X.badius, 49	0.26 ± 0.02	0.57 ± 0.04	$(7.8\pm0.7)\cdot10^{-2}$	$(1.9\pm0.1)\cdot10^{-1}$
X.badius, 67	$(1.8\pm0.2)\cdot10^{-3}$	$(1.8\pm0.2)\cdot10^{-3}$	$(3.9\pm0.5)\cdot10^{-3}$	$(2.4\pm0.3)\cdot10^{-3}$
L.vellerreus, 49	$(1.9\pm0.3)\cdot10^{-2}$	$(2.4\pm0.3)\cdot10^{-2}$	$(5.8\pm0.8)\cdot10^{-3}$	$(8.1\pm1.0)\cdot10^{-3}$

Only for a few samples the forest litter samples were still available for analyses and only then calculations were possible.

Table 12
Uranium, thorium and plutonium transfer factors for *Hebeloma cylindrosporum* mushroom collected in Spain

Radionuclide	TF
^{232}Th	0.383 ± 0.016
^{230}Th	0.352 ± 0.018
^{238}U	0.46 ± 0.04
^{235}U	0.38 ± 0.10
^{234}U	0.49 ± 0.04
$^{239+240}$Pu	0.67 ± 0.11

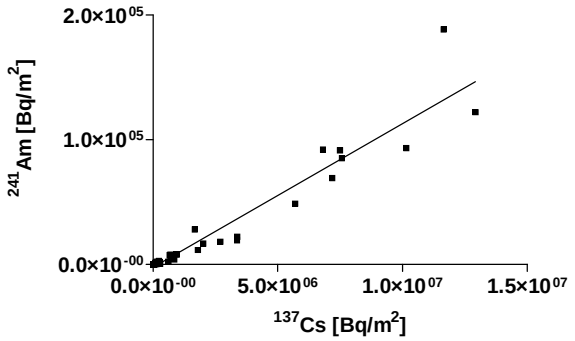

Fig. 6. Correlation plot between the ^{241}Am and ^{137}Cs ground deposition (Buzinny, not published data) in areas close to the site in which Ukrainian samples were collected (both surface activities determined by gamma-ray spectrometry).

tion coefficients are likely a little overestimated, since plutonium situated below examined forest litter layers was not taken into consideration. Few results obtained for TF for thorium isotopes are presented in Table 11.

Calculated TF values for all examined nuclides in a Spanish mushroom *Hebeloma cylindrosporum* are presented in Table 12. All calculated TFs for *Hebeloma cylindrosporum* are much bigger than, for example, those calculated for other species in Polish or Ukrainian samples. This species is a specific Mediterranean one, and does not grow in the more northern countries. Obtained TF values suggest a possible important bioindicator property of this particular species for U, Th and Pu isotopes, as was found for radiocesium only in the case of a Japanese species of *Hebeloma* earlier (Yoshida and Muramatsu, 1994). However, even for similar species as measured in Poland and Spain, for instance for *Boletus sp.*, the U and Th concentrations, but not that of Pu, seem to be higher for the Spanish than for the Polish samples. This might perhaps be a result of a mycelium localisation within mineral soil rather than only organic forest litter and a more intense decomposition process as the result of generally higher biomass turnover rate in the warmer Spanish climate, compared to other countries mentioned above.

For Ukrainian samples the Pu activity concentration and its deposition in ground was estimated from depositions of ^{137}Cs and ^{241}Am, since the environmental behaviour of plutonium and americium might be similar and the ^{241}Am and ^{137}Cs depositions were locally well correlated (Fig. 6). This was realised during other investigations conducted at this area (not published data, analyses of forest litter and soil samples were performed for sites, corresponding to those of Table 1 in the paper Buzinny et al., 1998). The slope of the correlation line (0.0115 ± 0.0006) in Fig. 6 might be considered roughly as the average ^{241}Am:^{137}Cs ratio. The deposition of $^{239+240}$Pu was proportional to the ^{241}Am deposition with a Am/Pu ratio of about 1.3 (Ageev et al., 2000). The activity concentration of ^{137}Cs in forest litter was equal to 344 kBq/kg (dw). This suggests about 4 kBq/kg (dw) for ^{241}Am and about 3 kBq/kg (dw) for $^{239+240}$Pu. The total deposition of ^{137}Cs at the mushroom sampling area calculated from the measured concentrations of this isotope not only in forest litter but also in the soil profile down to 25 cm is equal to 420 kBq/m^2. This gives approximately 4.8 kBq/m^2 for the ^{241}Am deposition and about 3.7 kBq/m^2 for $^{239+240}$Pu. This value was taken for the estimation of transfer factors (TF_{Pu}) or aggregation coefficients (AC_{Pu}) presented in Table 13 along with the TF_{Cs} or AC_{Cs} values for radiocesium. The value obtained for AC_{Cs} for *Xerocomus badius* was slightly higher than the maximum value ($=44$ m^2/kg) for Polish samples of this species obtained previously (Mietelski and Jasińska, 1996), and it is eleven times higher than the mean value for the Polish *Xerocomus* samples. A comparison of aggregation coefficients for radiocesium and plutonium shows, that plutonium from Chernobyl fallout close to the accident site is 2 up to 5 orders of magnitude less

Table 13
Data on the transfer factors (TF) and aggregation coefficients (AC) for samples collected in Ukraine

Species	TF_{Cs}	TF_{Pu}	AC_{Cs} (m^2/kg)	AC_{Pu} (m^2/kg)
Xerocomus badius	62.3	0.98×10^{-3}	51	0.80×10^{-3}
Suillus luteus	41.8	1.1×10^{-3}	34	0.91×10^{-3}
Paxillus involutus	147.	0.91×10^{-3}	121	0.75×10^{-3}
Boletus edulis	1.50	n.d.	1.23	n.d.
Cantharellus cibarius	6.15	18.0×10^{-3}	5.05	14.6×10^{-3}

intensively transferred to fruit bodies of mushrooms than radiocesium. *Cantherellus cibarius* seems to be a species, which might accumulate Pu a little more than other examined species.

The values obtained for plutonium aggregation coefficients for Polish and Ukrainian samples seem to be roughly similar. The majority of mean results for various species are on the level of $10^{-3}\,m^2$/kg.

For a comparison, the value of AC, which can be calculated from single results on Pu activity concentration in a *Boletus edulis* sample from Great Britain, equal to 282 mBq/kg (dw) (Barnett et al., 1997) would be about $5 \times 10^{-3}\,m^2$/kg. It was calculated assuming the plutonium deposition from global fallout equal to $58\,Bq/m^2$ (UNSCEAR, 1982), on the average, for a latitude belt from 50°N to 40°N. However, the data from the Chernobyl area published recently (Ageev, 2000) showed AC results lower by about two orders of magnitude for most of the examined species, including such ones as: *Xerocomus badius, Suillus luteus* and *Boletus edulis*. Such low values were now observed also for one sample of *Xerocomus badius* from Poland.

An interesting question is: what is the dominant mechanism of transfer of alpha emitters to the fruit body growing period? There are at least three different explanations:

(1) external contamination of fruit bodies with traces of soil (litter) particles;
(2) soak of the fruit bodies with the ions from soil or litter solutions during the fruit body growing period;
(3) incorporation of ions by the mycelium, similar to the root uptake by the plants.

There are several suggestions, that the majority of Pu is somehow incorporated into the fruit bodies. The most important argument for this assumption are results obtained for the Polish sample of the *Lactarius* mushroom from site 49, which was subdivided into two samples which had the same Pu activity concentration. It is rather unlikely, that an external contamination would split so evenly. The results obtained for the specially washed sample (X–X) show, that even intense brushing and soaking with water, which perhaps removes some of the fungi tissues (the water taken for analyses was of a intense dark-brown colour), remove less Pu or U than Th. The difference in the mechanical cleaning ratio seems to be against the supposition of an external contamination, at least as the exclusive mechanism. On the basis of the present data, we cannot distinguish between the two other possibilities quoted above.

The dosimetric interpretation of the observed activities depends mostly on the speciation of U, Th and Pu within the fruit bodies. If their chemical forms are refractory, the small value of the f_1 coefficient, which describes the probability of the intake of a given radionuclide through the ingestion pathways, prevents transfer of those radionuclides to humans. However, it is not sure any longer, whether the f_1 value is still so small in this case.

4. Conclusions

Plutonium, thorium and uranium were observed in fruit bodies of mushrooms in measurable amounts. The highest activity concentrations were observed, as expected, on the highly contaminated area close to the Chernobyl NPP. Surprisingly, elevated levels of all these radionuclides were found in a sample of *Hebeloma cylindrosporum* collected in Spain. This species seems to be particularly interesting for further investigations. For other species we can expect in laboratory experiments under controlled conditions transfer factors at the level of 10^{-3}, when nuclides are applied not directly to fruit bodies but to the mycelium. Such values were obtained for samples collected at very different locations. Alpha spectra obtained for the thorium fraction suggest no radium accumulation properties of examined mushroom species.

The speciation of alpha-emitting radionuclides observed in mushrooms is not known as yet. This is a very important question for proper dosimetric interpretation of observed activities. The details on the mechanism of radionuclide incorporation into fruit bodies remains unclear, however the incorporation itself seems to be proved.

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References

Ageev, V.A., Odinstov, A.A., Satsyuk, V.A., Lashko, A.P., 2000. About the transuranium elements and fission products content in the biological samples taken in the 30-km ChNPP zone. Scientific Papers of the Institute for Nuclear Research, No. 2, pp. 161–167 (in Ukrainian).

Baeza, A.S., Guillen, J., Paniagua, J.M., Hernandez, S., Martin, J.L., Diez, J., Manjon, J.L., Moreno, G., 2000. Radiocesium and radiostrontium uptake by fruit bodies of *pleurotus eryngii* via mycelium and aerial absorption. Appl. Radiat. Isotopes 53, 455–462.

Barnett, C.L., Dodd, B.A., Boresford, N.A., Self, P.L., Frankland, J.C., Howard, B.J., 1997. Radionuclide contamination of fruit bodies of macrofungi in England and Wales: a survey of contamination levels and dietary intake habits. Final Report of Project No. T07051W1, MAFF contract No. RP0425, Institute of Terrestrial Ecology, Grange-over-Sands.

Bondarenko, O.A., Aryasov, B.B, Tsygankov, N.Ya., 2000a. Evaluation of the plutonium content in the human body due to global and Chernobyl fallout. J. Radioanal. Nucl. Chem. 243, 473–478.

Bondarenko, O.A., Onischuk, Yu., Melnichuk, D., Tsygankov, N.Ya., 2000b. An approach to retrospective decomposition of plutonium intake in result of the global and the Chernobyl Accident based on isotope analysis. NRC5. Fifth International Conference on Nuclear and Radiochemistry. Pontresina, Switzerland, September 3–8, 2000. Book of Extended Abstracts, Vol. 2, pp. 440–443.

Buzinny, M.G., Likhtarjov, I., Los', I., Talerko, N., Nikolay Tsigankov, N.Ya., 1998. Radiocarbon analysis of annual tree rings from the vicinity of the chernobyl NPP. In: W.G., Mook, Van der Plicht, J. (Eds.), Proceedings of the 16th International Radiocarbon Conference RADIOCARBON, Vol. 40, pp. 373–380.

Dregge, P., Boden, R., 1984. Review of chemical separation techniques applicable to alpha spectrometric measurements. Nucl. Instru. Methods 223, 181–187.

IAEA, 1994. AQCS 1994–95 Catalogue, IAEA Press, Vienna, p. 27.

IAEA, 1997. AQCS, Alternation to the 1994–95 Catalogue, IAEA Press, Vienna, p. 6.

Iranzo, E., Espinosa, A., Bellido, A., Gascó, C., Aragón, A., Gutierrez, J., 1992. Summary Report on the Palomares Surveillance Programme. CIEMAT/IMA/UGIA/M5A01/03/92. CIEMAT, Madrid, Spain.

Jacquiot, L., Daillant, O., 2000. Bio-accumulation of Trace Elements and Radiocesium by Macrofungi, Review, Part II. Bull. Observatoire Mycologique 17, 2.

LaRosa, J.J., Cooper, E., Ghods-Esphahani, A., Jansta, V., Makarewicz, M., Shawky, S., Vajda, N., 1992. Radiochemicals methods used by the IAEA's laboratories at Seibersdorf for the determination of ^{90}Sr, ^{144}Ce, and Pu radionuclides in environmental samples collected for the International Chernobyl Project. J. Environ. Radioactivity 17, 183–209.

Mietelski, J.W., 1998. Transuranic elements and Sr-90 in samples from forests of Poland, a review. Nukleonika 43, 449–458.

Mietelski, J.W., Jasińska, M., 1996. Radiocesium in bilberries from Poland: comparison with data for mushroom samples. J. Radioecol. 4, 15–25.

Mietelski, J.W., Wąs, B., 1995. Plutonium from Chernobyl in Poland. Appl. Radiat. Isot. 46, 1203–1211.

Mietelski, J.W., LaRosa, J.J, Ghods, A., 1993. Results of ^{90}Sr, and ^{238}Pu, $^{239+240}$Pu, ^{241}Am, measurements in some samples of mushrooms and forest soil from Poland. J. Radioanal. Nucl. Chem. Articles 170, 243–258.

Mietelski, J.W., Jasińska, M., Kubica, B., Kozak, K., Macharski, P., 1994. Radioactive contamination of Polish mushrooms. Sci. Total Environ. 157, 217–226.

Mietelski, J.W., Jasińska, M., Kozak, K., Ochab, E., 1996. The method of measurements used in the investigation of radioactive contamination of forests in Poland. Appl. Radiat. Isotopes 47, 1089–1095.

Mietelski, J.W., Gaca, P., Jasińska, M., 2000. Plutonium and other alpha-emitters in bones of wild, herbivorous animals from north-eastern Poland. Appl. Radiat. Isotopes 53, 251–257.

Sill, C.W., 1987. Precipitation of actinides as fluorides or hydroxides for high resolution alpha spectrometry. Nucl. Chem. Waste Manage. 7, 201–215.

UNSCEAR Report to the General Assembly with Annexes, 1982. New York, p. 228 & 238.

Yoshida, S., Muramatsu, Y., 1994. Accumulation of radiocesium in basidiomycetes collected from Japanese forests. Sci. Total Environ. 157, 197–205.