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Plutonium and gamma emitters in the northeastern part of Bory Tucholskie (Poland)

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Abstract. The paper presents results of the measurements of Pu and gamma emitters of some forest litter/soil profiles collected in 1999 in the northeastern part of Bory Tucholskie forest (northern Poland). Besides the activity concentration of ^{137}Cs, ^{134}Cs, ^{40}K, ^{228}Ac, ^{238}Pu, $^{239+240}$Pu, the cumulated deposition of ^{137}Cs, ^{238}Pu and $^{239+240}$Pu were calculated. The origin of these radionuclides is discussed on the basis of the observed activity ratios. The observed average level of radioactive contamination confirms expectations for this region, however samples show a relatively large variation of cumulated deposition for both plutonium and radiocesium originated from global and Chernobyl fallout.

Key words: radioactive contamination • plutonium • ^{137}Cs • global fallout • Chernobyl • Bory Tucholskie

Introduction

The Bory Tucholskie is a large forest complex (about 20,000 ha) situated in northern Poland, west of the Vistula river. It is an area of lakes, slow streams and shadow rivers well known for their tourism and recreation values. The majority of this complex is formed by pine (*Pinus sylvestris*) trees which are more than 50 years old and, therefore, this forest complex is also important from the economical point of view. However, about 5000 ha of the most valuable part of it forms the National Park of Bory Tucholskie and this part of forest is excluded from any economical exploitation.

Radioactive contamination of this area started with global fallout which was likely to make cumulated deposition on a scale typical of Poland, with mean values equal to: 5.6 kBq/m^2 for ^{137}Cs and 58 Bq/m^2 of $^{239+240}$Pu (activities in 1963) [20]. More than a half of the global fallout radiocesium activity decayed till our measurements started, but due to the long half-life time of ^{239}Pu, ^{240}Pu isotopes their activities remained almost the same. Radiocesium from the Chernobyl accident reached this area as well, resulting together with the remains of global fallout in a total cumulated deposition at the beginning of 1990s of about 5 kBq/m^2 [19]. Plutonium from the Chernobyl fallout in Poland was related to relatively large aerosols, so-called fuel-like hot particles [2, 9, 15], accompanied in almost constant proportion by some specific gamma emitters like ^{144}Ce,

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^{154}Eu and ^{155}Eu [1, 4, 9, 16]. Accordingly to past studies based on both: ^{144}Ce measurements [10–12] and direct Pu measurements [10, 15], no significant deposition of Chernobyl plutonium was expected in the examined area. The $^{239+240}$Pu deposition of the Chernobyl fallout origin was expected to be below 3 Bq/m^2 [10, 11], what should be compared with the average from the global fallout of 58 Bq/m^2 appropriate for the (50–60° N) latitude belt [20]. The global fallout values were confirmed for lowlands of Poland by studies of soil profiles in the area of Lublin [6]. Recently, much higher global fallout Pu depositions were found in Poland, exceeding even 200 Bq/m^2 in some soil profiles from the Tatra Mountains South Poland [7, 13].

A study conducted about 150 km to the west of Bory Tucholskie on a former military test site, Borne Sulinowo [5] showed about a 5% addition of Chernobyl origin to global fallout for $^{239+240}$Pu. In more remote locations of Europe, such as Germany, the Chernobyl plutonium deposition was much lower [3]. In our earlier direct Pu measurements done for forest litter [15], the area of Bory Tucholskie was represented by a single sampling point and only traces of Chernobyl Pu were detected there in the uppermost layer of forest litter. The question of representativeness of sampling is well known to all who did any environmental studies. Therefore, to clarify the figure we decided to make some direct plutonium measurements on this area based on forest litter/soil samples collected within a distance of few kilometers. This was thought to give data on a patchy pattern of Chernobyl Pu fallout in a site rather remote from the accident. To complete the study we decided to do also gamma spectrometric analyses of all samples to obtain data on ^{137}Cs as well as on some natural gamma emitters.

Materials and methods

Forest litter and soil samples (profiles) were collected in 9 locations in the Bory Tucholskie in the area around the Radodzierz Lake (approximately 18°35' E, 53°38' N) in the summer of 1999. This lake has a water surface of 236.4 ha and is surrounded predominately by a 60–80 year old pine forest. The closest small towns are Warlubie, Osiek and Skórcz at a distance of 20–30 kilometers from each other. All sampling sites belong to the forest inspectorate Dąbrowa and are situated within the circle of a diameter of about 8 km. Exact sites were localized on the map and these localizations are presented in Fig. 1.

Litter/soil profiles were taken as monoliths. To get such a monolith, a sharp knife was used to mark a square of about 10 cm side. Then, the soil and litter around the monolith were carefully removed at a depth of 10–20 cm using a small spade and this knife. Finally, each monolith was cut off at its base, transferred to a foil bag and placed in a box. In this form, the profiles were transported to the laboratory. There, the actual sizes of monoliths were carefully determined and after drying at 105°C overnight they were sliced into about 3–4 cm thick layers accordingly to some morphological properties. Each layer sample was homogenized by grounding, weighed and placed in a measurement vessel. Characteristics of soil samples are presented in Table 1. Beside our code and geographical coordinates, the number of forest sections of the forest inspectorate Dąbrowa is provided in this table, so the sampling site is easy to be found on the official maps of Polish forestry.

Prepared samples were measured for the presence of gamma-emitters using a low background gamma ray spectrometer with HPGe detectors (10% and 15% rel.

Fig. 1. Map of sampling sites in surroundings of Radodzierz Lake.

Table 1. Characteristic of collection data of soil samples from the Bory Tucholskie

Code	Forest section no.	Depth (cm)	Description	Longitude E	Latitude N	Dry mass (d.w.) (g)	Area (m^2)
P1-1	206	0 – 3.5	Pine forest	18°35'08.8"	53°38'02.8"	88.8	0.010 ± 0.001
P1-2		3.5 – 6				225.6	0.010 ± 0.001
P1-3		6 – 10				485.3	0.010 ± 0.001
P1-4		10 – 14				389.1	0.010 ± 0.001
P2-1	180	0 – 4	Mixed	18°34'56.2"	53°37'56.8"	103.0	0.010 ± 0.001
P2-2		4 – 6	(dedicious			216.9	0.010 ± 0.001
P2-3		6 – 11	and coniferous)			365.9	0.010 ± 0.001
P3-1	361	0 – 3	Pine forest	18°39'31.5"	53°38'07.8"	34.3	0.010 ± 0.001
P3-3		3 – 7				110.1	0.010 ± 0.001
P3-3		7 – 12				450.6	0.010 ± 0.001
P3-4		12 – 17				495.0	0.010 ± 0.001
P4-1	210	0 – 4	Pine forest	18°33'47.7"	53°37'43.7"	69.3	0.010 ± 0.001
P4-2		4 – 8				275.9	0.010 ± 0.001
P4-3		8 – 12				436.8	0.010 ± 0.001
P4-4		12 – 15				634.0	0.010 ± 0.001
P5-1	62	0 – 4	Pine forest	18°33'58.7"	53°37'26.9"	27.5	0.010 ± 0.001
P5-2		4 – 7				94.9	0.010 ± 0.001
P5-3		7 – 10.5				403.2	0.010 ± 0.001
P5-4		10.5 – 15				557.3	0.010 ± 0.001
P5-5		15 – 19				413.9	0.010 ± 0.001
P6-1	126	0 – 2	Pine forest	18°34'07.2"	53°37'05.5"	13.8	0.0081 ± 0.001
P6-2		2 – 5.5				55.4	0.0081 ± 0.001
P6-3		5.5 – 8				47.2	0.0090 ± 0.001
P6-4		8 – 12				476.4	0.0100 ± 0.001
P6-5		12 – 16				526.1	0.0110 ± 0.001
P7-1	293	0 – 4	Pine forest	18°34'47.6"	53°37'13.6"	31.5	0.011 ± 0.001
P7-2		4 – 7				289.3	0.011 ± 0.001
P7-3		7 – 11				398.5	0.011 ± 0.001
P7-4		11 – 14				249.1	0.011 ± 0.001
P8-1	130	0 – 3	Coniferous	18°33'37.7"	53°38'20.3"	24.3	0.0072 ± 0.0010
P8-2		3 – 6				45.7	0.0072 ± 0.0010
P8-3		6 – 9				163.5	0.0072 ± 0.0010
P8-4		9 – 13				296.4	0.0072 ± 0.0010
P9-1	37	0 – 2	Mixed	18°31'45.7"	53°39'56.3"	20.6	0.0090 ± 0.0010
P9-2		2 – 4	(dedicious			116.3	0.0081 ± 0.0010
P9-3		4 – 7	and coniferous)			313.1	0.0081 ± 0.0010
P9-4		7 – 10				263.5	0.0081 ± 0.0010

efficiency) shielded by 10 cm of lead. The calibration method was described elsewhere [12]. Those measurements were performed in the autumn of 1999, but not published yet. The analyzed lines were 662 keV of ^{137m}Ba (in equilibrium with ^{137}Cs), 605 keV and 796 keV of ^{134}Cs, 1461 keV of ^{40}K and 911 keV of ^{228}Ac (in equilibrium with ^{228}Ra).

All the samples were then kept in the laboratory in sealed plastic bags for further radiochemical analyses. Due to some man-power shortage caused by other projects running in the laboratory, the radiochemical analyses were done not earlier than in the autumn of 2006. Unfortunately, during storage all samples from site P1 and one sample from P2 (P2-2) were accidentally lost, and therefore they were not available for Pu analysis.

Prior to any radiochemical work, all samples were ashed in a muffle oven at 600°C. Then, about 10 g of the ash sub-samples were taken for Pu determination. The applied procedure was typical of our laboratory, it follows a general idea of IAEA procedure used for the Chernobyl project [8]. Our small modifications were already described in details elsewhere [10, 14, 15]. All alpha spectrometric sources were obtained using the NdF_3 co-precipitation method [18]. The measurements were performed using a Silena AlphaQuattro alpha spectrometer equipped with Canberra PIPS detectors.

Quality assurance was achieved by measurements of the IAEA Soil-375 reference material. For $^{239+240}$Pu plutonium, we obtained 0.33 ± 0.04 Bq/kg, whereas the certified value is 0.30 Bq/kg (95% confidence level is from 0.26 Bq/kg to 0.34 Bq/kg). For ^{137}Cs, we obtained for reference data 5559 ± 70 Bq/kg and for ^{40}K 423 ± 28 Bq/kg which is not different from the certified values by more than 5%.

Table 2. Activity concentration of gamma emitters calculated for summer 1999

Code	^{137}Cs (Bq/kg d.w.)	^{134}Cs (Bq/kg d.w.)	^{40}K (Bq/kg d.w.)	^{228}Ra (Bq/m^2)	^{137}Cs (Bq/m^2)
P1-1	339 ± 14	2.6 ± 1.1	104 ± 14	< 1.7	3009 ± 325
P1-2	58.4 ± 1.5	0.4 ± 0.1	211 ± 9	4.6 ± 0.5	1318 ± 136
P1-3	32.1 ± 1.5	< 0.1	252 ± 29	3.3 ± 0.7	1558 ± 172
P1-4	10.6 ± 0.5	< 0.3	226 ± 21	5.0 ± 0.9	412 ± 46
P2-1	131.5 ± 2.7	1.4 ± 0.3	211 ± 10	2.6 ± 0.9	1354 ± 138
P2-2	10.7 ± 0.4	< 0.3	210 ± 9	3.2 ± 0.7	232 ± 25
P2-3	28.6 ± 1.1	< 0.2	247 ± 24	5.5 ± 0.8	1046 ± 112
P3-1	348 ± 7.5	3.8 ± 1.2	103 ± 16	6.6 ± 3.2	1194 ± 122
P3-2	224.9 ± 7.2	1.9 ± 0.4	151 ± 10	3.2 ± 1.1	2476 ± 260
P3-3	40.7 ± 1.9	< 0.2	267 ± 30	3.7 ± 1.1	1834 ± 203
P3-4	8 ± 0.6	< 0.2	253 ± 31	6.7 ± 1.4	396 ± 50
P4-1	108.4 ± 3.8	1.2 ± 0.4	139 ± 12	5.5 ± 1.6	751 ± 80
P4-2	89.8 ± 3.6	0.5 ± 0.2	141 ± 10	2.6 ± 0.7	2478 ± 267
P4-3	51.3 ± 2.4	< 0.2	254 ± 29	6.1 ± 1.1	2241 ± 248
P4-4	5.3 ± 0.6	< 0.1	370 ± 62	6.4 ± 1.2	336 ± 51
P5-1	83.1 ± 2.8	< 0.9	170 ± 23	< 4	229 ± 24
P5-2	168.6 ± 5.1	1.5 ± 0.3	136 ± 10	6.3 ± 1.3	1600 ± 167
P5-3	21.2 ± 1.2	< 0.2	283 ± 26	6.8 ± 0.9	855 ± 98
P5-4	5.2 ± 0.4	< 0.1	315 ± 42	6.6 ± 1.1	290 ± 37
P5-5	2.4 ± 0.3	< 0.1	260 ± 26	3.7 ± 0.9	99 ± 16
P6-1	113.9 ± 4.7	< 1.2	< 52	2.5 ± 6.7	194 ± 21
P6-2	437.0 ± 13	4.3 ± 0.8	61 ± 9	1.1 ± 1.5	2988 ± 311
P6-3	332.0 ± 9	1.6 ± 0.6	137 ± 12	7.9 ± 2.2	1741 ± 180
P6-4	46.3 ± 2.3	< 0.1	295 ± 36	5.0 ± 0.9	2206 ± 247
P6-5	7.1 ± 0.6	< 0.2	308 ± 41	5.9 ± 1.2	340 ± 45
P7-1	95.7 ± 2.8	< 0.9	124 ± 15	< 2.7	194 ± 21
P7-2	32.0 ± 1.4	< 0.2	106 ± 8	1.5 ± 0.5	842 ± 92
P7-3	14.5 ± 0.7	< 0.1	257 ± 18	6.4 ± 0.6	525 ± 58
P7-4	4.0 ± 0.3	< 0.2	242 ± 18	3.7 ± 1.0	91 ± 11
P8-1	375.5 ± 9.3	5.3 ± 1.0	< 32	< 3.7	1267 ± 131
P8-2	287.6 ± 6.5	2.1 ± 0.4	107 ± 9	5.8 ± 1.6	1825 ± 187
P8-3	37.9 ± 1	< 0.2	236 ± 10	5.5 ± 0.6	861 ± 89
P8-4	6.4 ± 0.4	< 0.2	246 ± 21	6.2 ± 1.2	263 ± 31
P9-1	69.9 ± 2.1	< 0.9	191 ± 18	5.3 ± 3.2	160 ± 17
P9-2	107.1 ± 2.2	0.6 ± 0.3	231 ± 10	4.8 ± 0.9	1538 ± 157
P9-3	22.8 ± 0.8	< 0.1	262 ± 20	5.3 ± 0.8	881 ± 93
P9-4	10.9 ± 0.7	< 0.3	264 ± 20	6.1 ± 1.2	355 ± 42

Results and discussion

Table 2 presents the results of gamma spectrometric measurements for ^{137}Cs, ^{134}Cs, ^{40}K and ^{228}Ac, decay being corrected for the date of sampling (August 1st 1999). In case of ^{137}Cs the results are presented as activity concentration (Bq/kg) and in the last column as surface deposition (Bq/m^2).

The ^{137}Cs activity in the majority of profiles shows the highest values in the uppermost layer or in a layer second from the top. The maximum activity for ^{137}Cs reached 437 ± 13 Bq/kg (d.w.) for sample P6-2. The minimum activity was 4.0 ± 0.3 Bq/kg (d.w.) for P7-4. About a half of the samples still showed (in 1999) traces of ^{134}Cs at a level of few Bq/kg. The decay correction factor for ^{134}Cs/^{137}Cs activity ratio within 13.3 year which elapsed since May 1st 1986, to reference day for activity (August 1st 1999) is equal to 56. Within relatively large uncertainties this suggests a strong domination of the Chernobyl origin cesium in those layers (usually two or three from the top).

As one can predict, the activity concentration of natural isotopes ^{40}K and ^{228}Ac (^{228}Ra) typically increases with the depth. The values were between < 32 Bq/kg (d.w.) to 370 ± 62 Bq/kg (d.w.) and < 1.7 to 7.9 ± 2.2 Bq/kg (d.w.) for ^{40}K and ^{228}Ac, respectively. These values seems to be typical of forest litter/soil samples of Poland lowlands.

The surface deposition for radiocesium (^{137}Cs) or plutonium seems to be more informative, than the activity concentration (see also Fig. 2). For the majority of profiles, the highest values for surface activity of ^{137}Cs in a single layer sample was found in layers 2 or 3, however the maximum of 3009 ± 325 Bq/m^2 was found for P1-1 sample. The deepest (4 or 5) layers show much lower surface activity from below 100 Bq/m^2 (P7-4 and P5-5) to 412 ± 46 Bq/m^2 (P1-4).

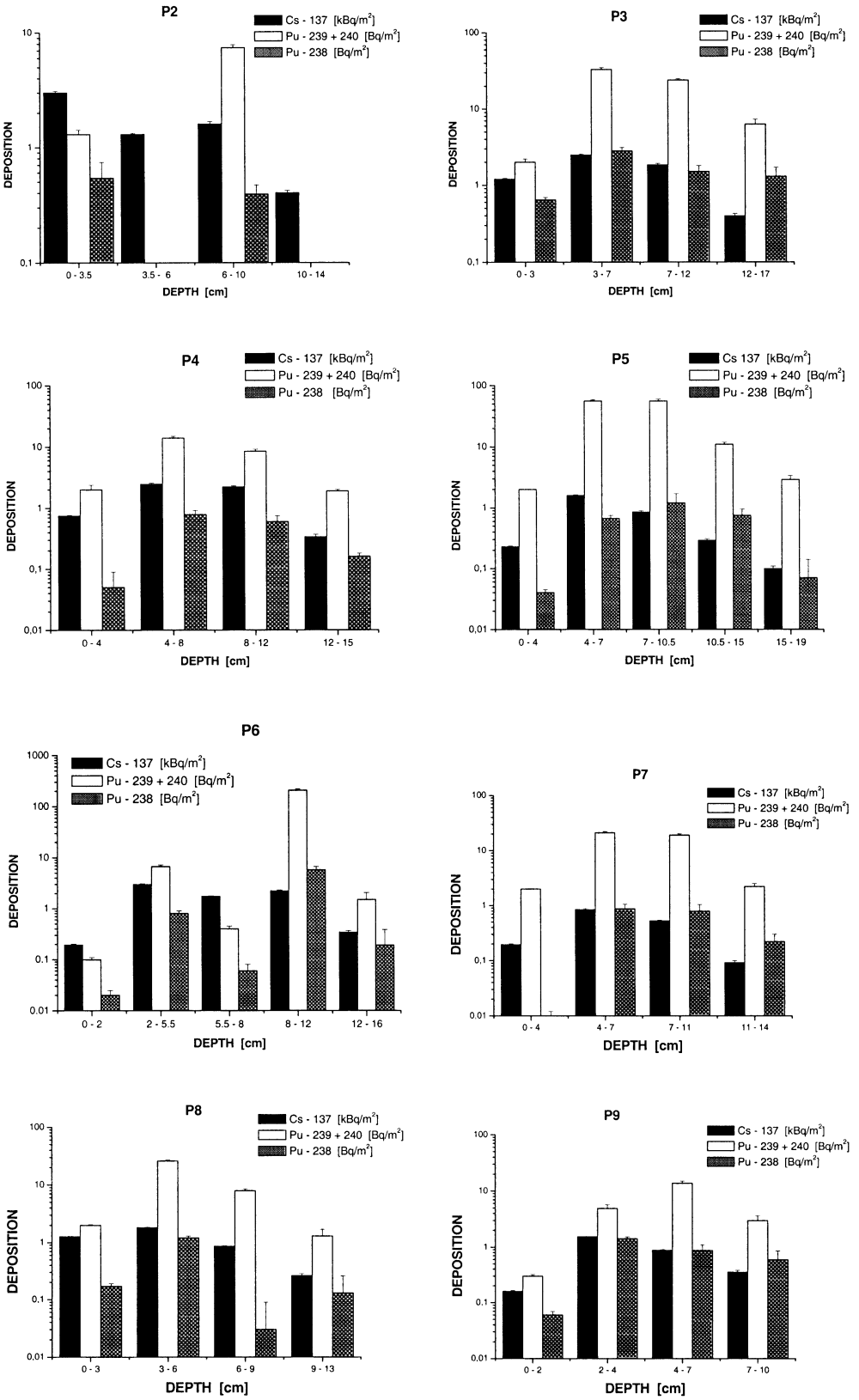

Fig. 2. Inventories of ^{137}Cs and plutonium isotopes in analyzed profiles from sites P2-P9.

Results for plutonium activity concentration and surface activity in the analyzed samples are presented in Table 3. The maximum activity concentration for a single layer were 5.9 ± 0.3 Bq/kg (d.w.) and 0.27 ± 0.03 Bq/kg (d.w.) for $^{239+240}$Pu and ^{238}Pu, respectively.

Surface activity of plutonium in separate layers varies in a wide range (see Fig. 2). The maximum was found for sample P6-4 amounting to 206 ± 11 Bq/m^2 for $^{239+240}$Pu only. Typically, the maximum values appeared for layers 2 and 3, what indicates the depth between 3 and 12 cm.

Plutonium activity isotopic ratio (^{238}Pu/$^{239+240}$Pu, for example) is useful for distinguishing the Chernobyl fraction from global fallout using values typical of the global fallout equal to 0.03 and in the Chernobyl fallout equal

Table 3. Results for $^{239+240}$Pu and ^{238}Pu analyses: recoveries, activity concentration, inventories in layer, activity ratio for ^{238}Pu/$^{239+240}$Pu and calculated percentage fraction (F) of $^{239+240}$Pu of Chernobyl

Code	Chem. yield Y (%)	$^{239+240}$Pu (Bq/kg d.w.)	$^{239+240}$Pu (Bq/m^2)	^{238}Pu (Bq/kg d.w.)	^{238}Pu (Bq/m^2)	^{238}Pu/$^{239+240}$Pu	F (%)
P2-1	70 ± 3	0.13 ± 0.01	1.3 ± 0.2	0.054 ± 0.007	0.54 ± 0.09	0.41 ± 0.07	80 ± 14
P2-3	89 ± 4	0.43 ± 0.03	7.4 ± 0.9	0.023 ± 0.005	0.39 ± 0.09	0.05 ± 0.01	5 ± 4
P3-1	97 ± 4	0.95 ± 0.05	2.9 ± 0.4	0.210 ± 0.018	0.64 ± 0.08	0.22 ± 0.02	40 ± 8
P3-2	78 ± 4	3.30 ± 0.20	33.0 ± 3.9	0.270 ± 0.030	2.80 ± 0.41	0.08 ± 0.01	12 ± 5
P3-3	82 ± 3	0.53 ± 0.03	24.0 ± 2.6	0.019 ± 0.005	1.50 ± 0.34	0.04 ± 0.01	< 4
P3-4	56 ± 3	0.13 ± 0.02	6.3 ± 1.2	0.026 ± 0.009	1.30 ± 0.42	0.20 ± 0.07	36 ± 13
P4-1	97 ± 4	0.33 ± 0.02	1.9 ± 0.2	0.027 ± 0.004	0.16 ± 0.03	0.08 ± 0.01	11 ± 5
P4-2	83 ± 3	0.56 ± 0.03	14 ± 1.7	0.032 ± 0.005	0.79 ± 0.15	0.06 ± 0.01	6 ± 4
P4-3	60 ± 2	0.21 ± 0.02	8.5 ± 1.1	0.015 ± 0.004	0.60 ± 0.16	0.07 ± 0.02	9 ± 5
P4-4	80 ± 3	0.06 ± 0.01	3.7 ± 0.5	0.001 ± 0.001	0.05 ± 0.04	0.01 ± 0.01	< 3
P5-1	90 ± 3	0.17 ± 0.01	0.40 ± 0.04	0.014 ± 0.002	0.04 ± 0.01	0.08 ± 0.01	11 ± 5
P5-2	36 ± 2	5.90 ± 0.30	56 ± 6.4	0.211 ± 0.025	0.67 ± 0.10	0.04 ± 0.005	< 4
P5-3	37 ± 2	1.90 ± 0.10	56 ± 6.9	0.029 ± 0.012	1.20 ± 0.51	0.02 ± 0.01	< 3
P5-4	88 ± 3	0.20 ± 0.02	11 ± 1.4	0.013 ± 0.004	0.75 ± 0.21	0.07 ± 0.02	9 ± 5
P5-5	31 ± 1	0.07 ± 0.01	2.9 ± 0.6	0.002 ± 0.002	0.07 ± 0.07	0.02 ± 0.02	< 3
P6-1	88 ± 3	0.08 ± 0.01	0.1 ± 0.014	0.012 ± 0.003	0.02 ± 0.01	0.16 ± 0.04	28 ± 9
P6-2	67 ± 4	1.10 ± 0.10	6.7 ± 0.8	0.131 ± 0.016	0.80 ± 0.13	0.12 ± 0.02	19 ± 6
P6-3	87 ± 3	0.09 ± 0.01	0.4 ± 0.1	0.012 ± 0.004	0.06 ± 0.02	0.14 ± 0.04	23 ± 9
P6-4	93 ± 4	4.36 ± 0.23	206 ± 23	0.120 ± 0.018	5.70 ± 1.10	0.03 ± 0.001	< 3
P6-5	62 ± 3	0.03 ± 0.01	1.5 ± 0.5	0.004 ± 0.004	0.19 ± 0.19	0.13 ± 0.13	< 36
P7-1	77 ± 3	0.22 ± 0.01	0.4 ± 0.1	0.006 ± 0.002	0.01 ± 0.005	0.03 ± 0.01	< 3
P7-2	94 ± 4	0.83 ± 0.05	21 ± 2.3	0.034 ± 0.007	0.87 ± 0.20	0.04 ± 0.01	< 5
P7-3	63 ± 3	0.54 ± 0.04	19 ± 2.1	0.022 ± 0.007	0.79 ± 0.26	0.04 ± 0.01	< 6
P7-4	50 ± 2	0.10 ± 0.01	2.2 ± 0.4	0.010 ± 0.004	0.22 ± 0.08	0.10 ± 0.04	15 ± 8
P8-1	92 ± 4	0.26 ± 0.02	0.9 ± 0.1	0.051 ± 0.006	0.17 ± 0.03	0.19 ± 0.03	35 ± 8
P8-2	99 ± 5	4.70 ± 0.20	26 ± 2.8	0.210 ± 0.018	1.20 ± 0.16	0.04 ± 0.00	3 ± 3
P8-3	86 ± 4	0.36 ± 0.03	7.9 ± 1.0	0.001 ± 0.001	0.03 ± 0.06	0.003 ± 0.001	< 1
P8-4	82 ± 4	0.03 ± 0.01	1.3 ± 0.4	0.003 ± 0.003	0.13 ± 0.13	0.10 ± 0.11	15 ± 13
P9-1	53 ± 2	0.12 ± 0.01	0.3 ± 0.04	0.030 ± 0.004	0.06 ± 0.01	0.25 ± 0.04	47 ± 10
P9-2	84 ± 3	0.34 ± 0.02	4.9 ± 0.9	0.100 ± 0.010	1.42 ± 0.18	0.29 ± 0.03	56 ± 9
P9-3	68 ± 3	0.36 ± 0.03	13.8 ± 1.8	0.023 ± 0.006	0.87 ± 0.25	0.06 ± 0.02	7 ± 5
P9-4	70 ± 4	0.09 ± 0.02	3.0 ± 0.7	0.019 ± 0.008	0.59 ± 0.28	0.20 ± 0.10	36 ± 15

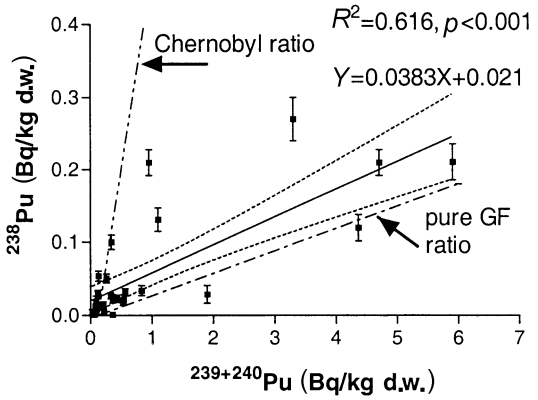

Fig. 3. Correlation plot for activity inventories in separate layers between ^{238}Pu and $^{239+240}$Pu (GF = global fallout).

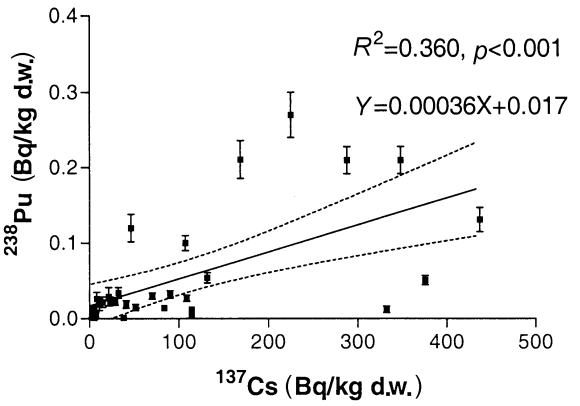

Fig. 5. Correlation plot for activity inventories in separate layers between ^{137}Cs and ^{238}Pu.

(for Poland) to 0.50 [10]. The last columns in Table 3 present the values of such ratio and the resulting percentage of Chernobyl fraction (F) of $^{239+240}$Pu activity calculated using a simple formula discussed in papers [10, 15].

For the majority of samples, the plutonium activity ratio ranged from 0.02 ± 0.01 to 0.08 ± 0.01 indicating global fallout as the main but not the only one source of plutonium, however in some samples of layers 1 or 2, for a given site, the ratio was significantly higher, reaching even a value of 0.41 ± 0.07 what points out to the Chernobyl fallout. In the deepest analyzed layers (below 10 cm) the activities were generally low and, therefore, less precisely determined which makes the ratio values to bear large uncertainties, especially when the alpha spectra showed traces of the ^{228}Th peak at 5.43 MeV. The correction for this interference (based on the ^{224}Ra peak) increased the uncertainties of determination of ^{238}Pu activity.

Significant correlation was found between the activity concentrations of $^{239+240}$Pu and ^{238}Pu. The squared Pearson correlation factor is equal to 0.616 with $p < 0.001$ (see Fig. 3). The mixed origin of Pu observed in samples with the domination of global fallout for $^{239+240}$Pu is clearly depicted in this figure. The position of experimental points is above the straight line defined by a slope equal to global fallout value of this ratio in the direction to the line with a slope of Chernobyl fallout ratio.

Activity concentration between $^{239+240}$Pu and ^{137}Cs are not correlated (see Fig. 4). However, the correlation between ^{238}Pu and ^{137}Cs is weakly correlated (see Fig. 5). The Pearson squared correlation factor R^2 is equal to 0.078 (significance level $p = 0.123$) or 0.360 ($p < 0.001$) for the first and second pair of the above isotopes, respectively. The lack of correlation was expected since both plutonium and radiocesium have mixed, the Chernobyl and global fallout, origins and in both the proportions between them were different. Different environmental properties and a long time which elapsed from the events of deposition (13 and about 36 years) also disturbs any relationship between those radionuclides. However, the observed weak correlation between ^{238}Pu and ^{137}Cs suggests that Chernobyl fallout dominates over the total measured activity of radiocesium. Moreover, it suggests that the fractionation between radiocesium and plutonium during the time which elapsed since Chernobyl deposition is not very high.

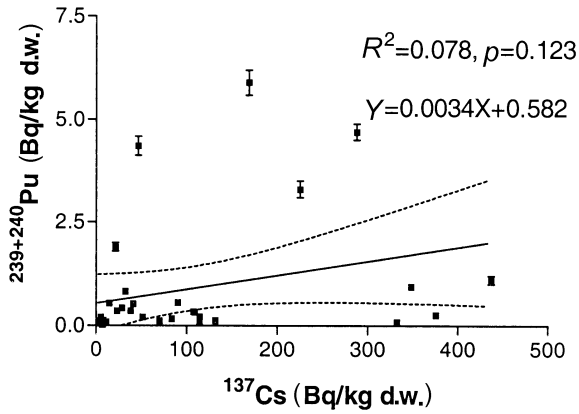

Fig. 4. Correlation plot for activity inventories in separate layers between ^{137}Cs and $^{239+240}$Pu.

The cesium found in given layer samples might come not only from direct deposition, but partially also from the vertical bio-circulation. For example, the root uptake to the plant and then fall on the litter surface of death needles or leaves is perhaps one of the important such vertical circulation mechanisms. Another mechanism is the incorporation into mycelium and transport within it, bioturbance (physical transport upward of humus or litter particles done by small animals) etc. Also the plutonium found in the top layer samples might come not only from direct deposition, but from a similar bio-circulation [10, 14].

The sum of surface deposition for a given place is the deposition inventory. The values for all analyzed profiles are presented in Table 4. Mean (arithmetic) deposition inventory of ^{137}Cs for all the examined profiles was equal to 4.4 ± 2.05 kBq/m^2, which is generally consistent with known estimations of deposition for this area, which gives values between 3 and 8 kBq/m^2 [19]. However, the variation of mean is surprisingly high. Great variation of the deposition inventory was also found for plutonium, the mean for total $^{239+240}$Pu being 76.4 ± 70.6 Bq/m^2 and for total ^{238}Pu 3.4 ± 2.2 Bq/m^2.

The total plutonium alpha-emitters ($^{238+239+240}$Pu) deposition of exclusively Chernobyl fallout origin can thus be calculated for each sampling site (profile). The results for such sum are presented in Table 5. The mean value sum of alpha-emitting Pu isotopes is 3.13 ± 2.4 Bq/m^2. Thus, the deposition of plutonium alpha

Table 4. Total inventory of ^{137}Cs and plutonium isotopes in examined sites

Site	^{137}Cs (Bq/m^2)	$^{239+240}$Pu (Bq/m^2)	^{238}Pu (Bq/m^2)
P1	6300 ± 395		
P2	2630 ± 180	8.7 ± 0.9*	0.9 ± 0.2*
P3	5900 ± 355	66.4 ± 4.8	6.2 ± 0.7
P4	5810 ± 379	27.8 ± 2.4	1.6 ± 0.5
P5	3070 ± 199	126 ± 9.5	2.7 ± 0.6
P6	7470 ± 438	215 ± 23	6.7 ± 1.1
P7	1650 ± 111	43.4 ± 3.2	1.9 ± 0.3
P8	4220 ± 247	34.6 ± 3.0	1.5 ± 0.3
P9	2930 ± 188	21.9 ± 2.2	2.9 ± 0.4

* for Pu data underestimated due to lost of sample P2-2.

Table 5. Chernobyl plutonium deposition (sum of ^{238}Pu and $^{239+240}$Pu) in the Bory Tucholskie from 1999 compared with those in the Augustów Primeval Forest from the same year published previously [10]

Chernobyl fraction $^{238+239+240}$Pu	Range (Bq/m^2)	Mean (Bq/m^2)	Standard deviation (Bq/m^2)
Bory Tucholskie	1.3 ± 0.3 to 7.7 ± 3.0	3.1	2.4
Augustow Primeval Forest	3.6 ± 0.4 to 24.2 ± 3.0	10.5	7.1

emitters of exclusively Chernobyl origin in the Bory Tucholskie is comparable with the values found for southern Finland [17] and are lower by a factor of about three than found previously in a similar study for more easterly situated another large forest complex of Poland: the Augustów Primeval Forest [10]. For comparison, the calculated exclusive global fallout component shows an order of magnitude higher mean value of 62.3 Bq/m^2. This is very close to the UNSCEAR mean for the appropriate latitude belt, which is 58 Bq/m^2 for $^{239+240}$Pu and 2.5 Bq/m^2 for ^{238}Pu [20], resulting in 60.5 Bq/m^2 for all plutonium alpha-emitters. However, the standard deviation is extremely high: 60 Bq/m^2. This is for very high deposition value observed for one location, site P6, where the total cumulated deposition is about 220 Bq/m^2. Excluding this site, the mean drops to 36 Bq/m^2 and the standard deviation turns to 5.9 Bq/m^2. Such low global fallout values suggest that some of Pu activity was perhaps not taken into consideration to deposition inventory balance for the majority of samples, may be Pu went deeper to soil layers or to other elements of the ecosystem. On the other hand, high spatial (horizontal) variation of deposition inventory of both global and Chernobyl fallouts may suggest the existance of some local mechanisms which can transport radionuclides horizontally. It could be abiotic – like erosion or biological transportation by roots or within mycelium. Another explanation could be similar for both fallouts deposition variation which can be explained, for example, by some aerodynamical properties of forest canopy, which has not much changed with time. Another, more simple explanation can be by assuming a local increase or decrease of fallen needles (on which the fallout was deposited initially) governed by dimensions of tree and distance from it. At present, we have not identified the reason for the observed variation, although we are aware of it.

Conclusions

The mean deposition of the Chernobyl or global fallout of plutonium and radiocesium in the analyzed samples from the Bory Tucholskie was found at a level close to our expectations.

The mean deposition of all the Pu alpha-emitters in the studied area was equal to 3.13 Bq/m^2 which forms a little less than 5% of similar deposition from the global fallout. The mean radiocesium deposition was found to be equal to 4.4 kBq/m^2.

The deposition of both Pu and radiocesium within relatively small area shows a significant variation, ranging up and down from the mean value by a factor of at least 2, what results in a standard deviation of the mean comparable with the mean value itself. Moreover, the global and the Chernobyl fallout show similar variations. In the case of the global fallout, the main variation is caused mostly by one profile which has an enormous inventory, being three times higher than other Pu depositions. The reason of such a large local scale variation of the deposition remains as yet unknown.

References

1. Broda R (1987) Gamma spectroscopy analysis of hot particles from the Chernobyl fallout. Acta Phys Pol 10:935–950
2. Broda R, Kubica B, Szeglowski Z, Zuber K (1989) Alpha emitters in Chernobyl hot particles. Radiochim Acta 48:89–96
3. Bunzl K, Kracke W (1990) Simultaneous determination of ^{238}Pu, $^{239+240}$Pu, ^{241}Pu, ^{241}Am, ^{241}Cm, ^{244}Cm, ^{89}Sr, ^{90}Sr in vegetation samples, and aplication to the Chernobyl-fallout contaminated grass. J Radioanal Nucl Chem, Articles 138;1:83–91
4. Jaracz P, Mirowski S, Trzcińska A *et al*. (1995) Calculation and measurements of ^{154}Eu and ^{155}Eu in "fuel-like" hot particles from Chernobyl fallout. J Environ Radioact 26:83–97
5. Komosa A (1998) Studies of contamination of Borne-Sulinowo Region (Poland) with plutonium and heavy metals. Polish J Environ Studies 7:89–93
6. Komosa A (1999) Migration of plutonium isotopes in forest soil profiles in Lublin region (Eastern Poland). J Radioanal Nucl Chem 240:19–24
7. Kubica B, Mietelski JW, Gołaś J *et al*. (2002) Concentration of ^{137}Cs, ^{40}K, ^{238}Pu, $^{239+240}$Pu radionuclides and some heavy metals in soil samples from two main valleys from Tatra National Park. Polish J Environ Studies 11;5:537–545
8. LaRosa JJ, Cooper E, Ghods-Esphahani A, Jansta V, Makarewicz M, Shawky S, Vajda N (1992) Radiochemical methods used by the IAEA's Laboratories at Seibersdorf for the determination of ^{90}Sr, ^{144}Ce and Pu radionuclides in the environment samples collected for the International Chernobyl Project. J Environ Radioact 17:183–209
9. Mietelski JW (1998) Transuranic elements and Sr-90 in samples from forests of Poland, a review. Nukleonika 43;4:449–458
10. Mietelski JW (2003) Application of nuclear spectrometry to bio-availability study of radionuclides from "fuel-like

hot particles" in forest environment. Report IFJ 1921/B. Instytut Fizyki Jądrowej, Kraków (in Polish)
11. Mietelski JW, Baeza AS, Guillen J *et al*. (2002) Plutonium and other alpha emitters in mushrooms from Poland, Spain and Ukraine. Appl Radiat Isot 56:717–729
12. Mietelski JW, Jasińska M, Kozak K, Ochab E (1996) The method of measurements used in the investigation of radioactive contamination of forests in Poland. Appl Radiat Isot 47:1089–1095
13. Mietelski JW, Kubica B, Gaca P *et al*. (2008) ^{238}Pu, $^{239+240}$Pu, ^{241}Am, ^{90}Sr and ^{137}Cs in mountain soil samples from the Tatra National Park (Poland). J Radioanal Nucl Chem 280 (in print) (DOI 10.2007/s10967-007-7026-1)
14. Mietelski JW, Szwałko P, Tomankiewicz E *et al*. (2004) ^{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 cases study. J Radioanal Nucl Chem 262;3:645–660
15. Mietelski JW, Wąs B (1995) Plutonium from Chernobyl in Poland. Appl Radiat Isot 46;11:1203–1211
16. Myasoedov BF, Pavlotskaya FI (1989) Measurements of radioactive nuclides in the environment. Analyst 114:255–263
17. Paatero J, Jaakkola T, Reponen A (1994) Determination of the ^{241}Pu deposition in Finland after the Chernobyl Accident. Radiochim Acta 64:139–144
18. Sill CW (1987) Precipitation of actinides as fluorides or hydroxides for high resolution alpha spectrometry. Nucl Chem Work Management 7:201–215
19. Strzelecki R, Wołkowicz S, Szewczyk J, Lewandowski P (1993) The map of cesium contamination of Poland. Radiological maps of Poland. Part. I. Państwowy Instytut Geologiczny, Warszawa (in Polish)
20. UNSCEAR (1982) Ionizing radiation sources and biological effects. Report to the General Assembly with annexes. United Nations Scientific Committee on Effects of Atomic Radiation, New York