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Plutonium and ^{137}Cs in forest litter: an approximate map of plutonium from Chernobyl deposition in North-eastern and Eastern Poland

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Abstract. The present article contains information about the activities and origin of ^{137}Cs, ^{238}Pu and $^{239+240}$Pu in the North-eastern Poland. Analyzed samples were collected in 59 locations where forest litter A_0 and humus A_1 were collected in 1991. An approximate map of the Chernobyl fallout component $^{239+240}$Pu was prepared on the basis of received results. The largest Chernobyl $^{239+240}$Pu contamination occurred in north-eastern and eastern part of the investigated area, reaching 22.1 ± 1.6 Bq·m^{-2}.

Key words: plutonium • cesium • Chernobyl • hot particles • radioactive contamination • map of deposition plutonium

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Introduction

The Chernobyl fallout in remote places contains only traces of non-volatile elements like plutonium, americium or strontium. The radioisotopes of non-volatile elements are transported on larger aerosols, the tiny pieces of nuclear fuel forming fallout of the so-called fuel like hot particles [5, 6]. According to numerical simulations [25], at the most north-eastern part of Poland the deposition of aerosols with aerodynamic diameters of about 4–5 μm occurred. Many of such big radioactive aerosols were "fuel like hot particles". Such particles were found in Poland by many investigators [2–4, 8–10, 15]. In 1991, a project on radionuclide deposition in all over Poland, based on mushrooms and forest litter studies, was launched. Within this project almost each forest inspectorate in Poland collected and sent us samples of forest litter and some mushrooms following our instructions. Samples were examined initially only for gamma emitters. Resulting approximate maps of deposition of different isotopes from Chernobyl fallout were already published some years ago [14, 18]. The deposition pattern of ^{144}Ce ($T_{1/2}$ = 284 d, still present in the forest litter/humus in 1991 or 1992 when measurements took place) was then reinterpreted by us as a possible pattern of Chernobyl -origin Pu [16] since both of them were the constituents of "fuel like hot particles". Those maps are presented in Fig. 1. Although direct measurements of Pu in forest litter from this area supported this approach, the number of analyzed there samples from North-eastern Poland was too low to produce any map directly. The

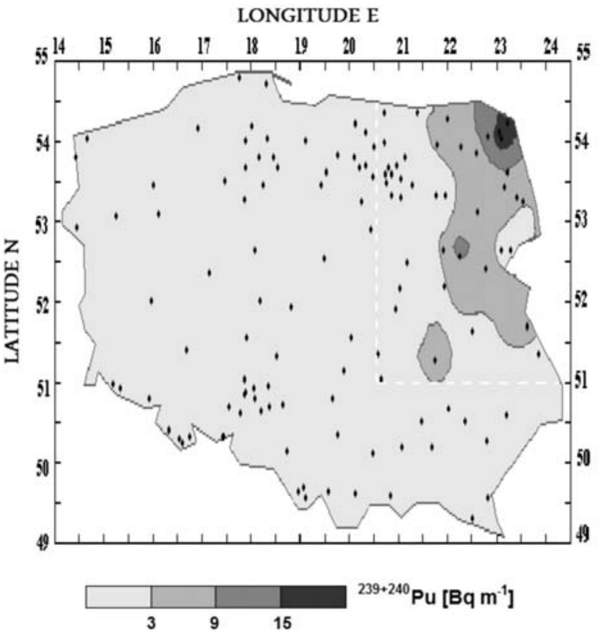

Fig. 1. Approximate map of the $^{239+240}$Pu deposition Chernobyl fallout component in Poland, based on ^{144}Ce deposition (decay corrected for 1 September 1991). White dotted line marks area of the present study [14].

aim of the work presented now was to fill this gap and to produce an approximate map of Chernobyl-origin Pu based on all available samples from North-eastern Poland left in our laboratory from the past project from 1991. This was the subject of two works for Master of Science degree done in 2007.

Material and methods

Samples

Samples of forest litter A_0 and next layer – the humus A_1 were collected in 1991 by forest inspectorate workers following the detailed instruction prepared by us. Details of the whole project were described elsewhere [17, 19]. The whole set of samples taken from the described here analyses consisted of 59 pairs: layer A_0 from an area of 30 × 30 cm^2 and A_1 from an area of 20 × 20 cm^2. The samples were kept dry in plastic bags in the laboratory. Prior to analyses for Pu, all the samples were additionally dried over night at 105°C, homogenized by means of grinding and measured for gamma emitters using a low background gamma-ray spectrometer with an HPGe detector.

Radiochemical procedure

After gamma spectrometric measurements, all the samples were ashed at 600°C, subsamples of up to 10 g of ash were taken for radiochemical analyses. At tracer of ^{242}Pu was added. The radiochemical procedure applied was rather standard one, which is in use in our laboratory since 1993. It follows the ideas of procedure applied in the IAEA Laboratories Seibersdorf for the Chernobyl project [12]. It consist of complete wet mineralization using subsequently hot HF, HNO$_3$ and HCl (with some H$_3$BO$_3$ added), Pu oxidation step adjustment in nitric acid solution using hydrazine and NaNO$_2$, separation of Pu on Dowex-1 from 8 M HNO$_3$ with a final source preparation by means of NdF$_3$ co-precipitation [22]. Sources were measured using a Silena AlphaQuattro spectrometer equipped with four Canberra silicon (PIPS) detectors with 450 mm^2 area each. Spectra were analyzed using a home made ALF software [14] and activity concentrations as well as all other calculations were done in MS Excel file.

Quality assurance

Together with the samples we analysed two IAEA reference materials: Soil 375 and Soil 6. The recommended value for $^{239+240}$Pu for Soil 6 is 1.01 Bq·kg^{-1} and 95% certified value interval is 0.96÷1.11 Bq·kg^{-1}. We obtained a little too low value: 0.90±0.08 Bq·kg^{-1}. However, in the case of Soil 375 the recommended value for $^{239+240}$Pu is 0.30 Bq·kg^{-1} and the 95% certified interval is 0.26÷0.34 Bq·kg^{-1}. For ^{238}Pu, the recommended value is 0.071 Bq·kg^{-1} and the 95% certified interval is 0.056÷0.085 Bq·kg^{-1}. Here, we obtained 0.27±0.03 Bq·kg^{-1} for $^{239+240}$Pu and 0.071±0.012 Bq·kg^{-1} for ^{238}Pu. Thus, possible systematic errors should not exceed 10% of the determined values and likely are much smaller.

Calculations of Chernobyl fraction of Pu

Differences in the Pu activity ratio (^{238}Pu/$^{239+240}$Pu) in global fallout and in Chernobyl fallout allows us to distinguish them. The discussion of details for this problem was done already previously [19]. The appropriate algorithm is the following:

$$\begin{aligned} A_{239+240} &= A_g + A_{ch} \\ A_{238} &= \zeta A_g + \xi A_{ch} \end{aligned} \tag{1}$$

where: $A_{239+240}$ – observed activity concentration of $^{239+240}$Pu [Bq·kg^{-1}]; A_{238} – observed activity of ^{238}Pu [Bq·kg^{-1}]; A_g – *a priori* unknown global fallout component of $^{239+240}$Pu [Bq·kg^{-1}]; A_{ch} – *a priori* unknown Chernobyl fallout component of $^{239+240}$Pu [Bq·kg^{-1}]; ζ – activity ratio ^{238}Pu to $^{239+240}$Pu in global fallout (assumed 0.03); ξ – activity ratio ^{238}Pu to $^{239+240}$Pu in Chernobyl fallout (assumed 0.5).

As a solution, we have:

$$A_g = \frac{\xi A_{239+240} - A_{238}}{\xi - \zeta} \tag{2}$$

$$A_{ch} = \frac{-\zeta A_{239+240} + A_{238}}{\xi - \zeta} \tag{3}$$

Thus, the percentage F of Chernobyl component in the observed activity can be described as:

$$F = \frac{A_{ch}}{A_{ch} + A_g} \cdot 100\% = \frac{A_{ch}}{A_{239+240}} \cdot 100\% = \frac{A_{238}(A_{239+240})^{-1} - \zeta}{\xi - \zeta} \cdot 100\% \tag{4}$$

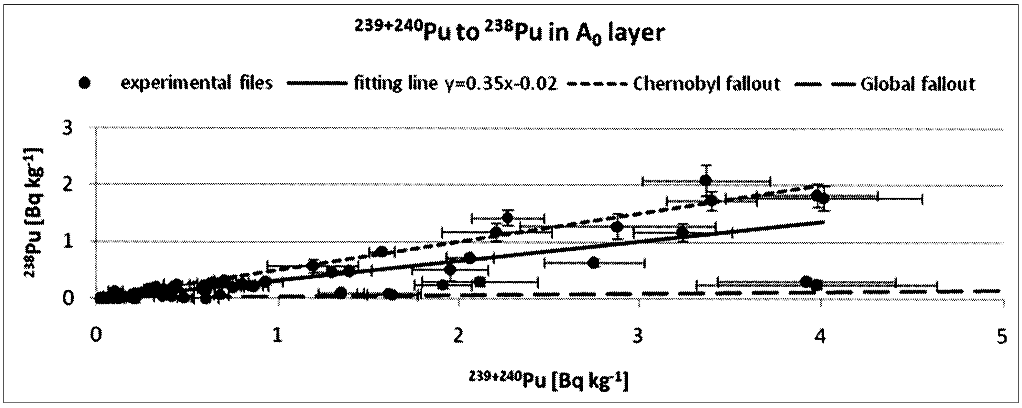

Fig. 2. Correlation between $^{239+240}$Pu and ^{238}Pu in A_0 layer. Fit (solid) is the regression line ($y = 0.35x - 0.02$). Dotted lines are the mean values for global and Chernobyl (more sloppy) fallouts.

Calculation of surface deposition

The result of measurement is the activity concentration. To turn it to deposition, an assumption must be done that the forest inspectors collecting the samples were following our instructions. Next, the following formula was applied:

$$D_{ch} = \frac{A_{ch} m}{s} \tag{5}$$

where: D_{ch} – Chernobyl component of $^{239+240}$Pu activity deposition in a given layer [Bq·m^{-2}]; A_{ch} – taken from Eq. (3); m – mass of the whole sample from a given layer [kg]; s – surface of the layer [m^2].

In the case of ^{137}Cs we use a similar formula. However, since we were not able to distinguish between the Chernobyl and global fallout componets (since ^{134}Cs was not detectable any more) in calculations of activity deposition we use the total activity concentration of ^{137}Cs.

Preparation of map

The map of Pu deposition was prepared using Golden Software Surfer 8. The raw data was in the form of a matrix of columns XYZ, were X and Y were geographical coordinates of the site and Z was calculated as deposition of $^{239+240}$Pu of Chernobyl origin. The software produced from this raw data the regular grid by kriging interpolation using default values for this method. The final map was produced as contour map, filled then with patterns.

Results and discussion

Table 1 presents the results on ^{137}Cs and plutonium activity concentrations and depositions as well as the data on $^{239+240}$Pu Chernobyl fraction.

Received values of ^{137}Cs activity on the studied area was between 6±1 Bq·kg^{-1} and 2062±51 Bq·kg^{-1} in A_0 layer or from 19±1 Bq·kg^{-1} to 2346±184 Bq·kg^{-1} in A_1 layer. In both layers the average radiocesium activity was near 700 Bq·kg^{-1}. The range of the deposition in the litter layer was from 5±1 Bq·m^{-2} to 3516±733 Bq·m^{-2} and in the humus layer was from 105±10 Bq·m^{-2} to 11,440±1100 Bq·m^{-2}. The cumulated deposition values in both the examined layers was near 4 kBq·m^{-2}, on the average. However, some radiocesium, which was surely present in the deeper layers of soil profile, was not taken into account.

Minimum activity of the $^{239+240}$Pu in A_0 layer was 0.02±0.01 Bq·kg^{-1}, whereas the maximum activity reaches 4.01±0.54 Bq·kg^{-1}. For A_1 layer, the minimum was 0.14±0.02 Bq·kg^{-1} and the maximum was 6.88±0.53 Bq·kg^{-1}. Average activity in the litter layer was 1.11 Bq·kg^{-1} and in the humus layer is larger by about two times.

Activity of the ^{238}Pu were between 0.01±0.01 Bq·kg^{-1} to 2.09±0.27 Bq·kg^{-1} in A_0 layer and 0.02±0.01 Bq·kg^{-1} to 2.20±0.31 Bq·kg^{-1} in A_1 layer.

As can be noticed from values of factor F (defined in Eq. (4), displayed in 10th column of Table 1), the clear evidence of Chernobyl fallout was found for plutonium in booth layers. In A_0 layer this is the dominant component in the majority of samples (Fig. 2), whereas in A_1 layer it is not so common (Fig. 3). One should recall here that it is not the present situation – samples were

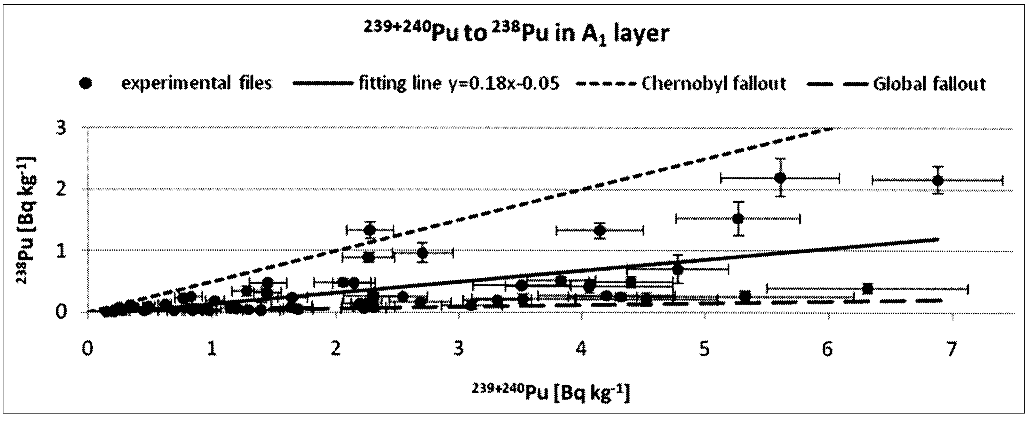

Fig. 3. Correlation between $^{239+240}$Pu and ^{238}Pu in A_1 layer. Fit (solid) is the regression line ($y = 0.18x - 0.05$) Dotted lines are the mean values for global and Chernobyl (more sloppy) fallouts.

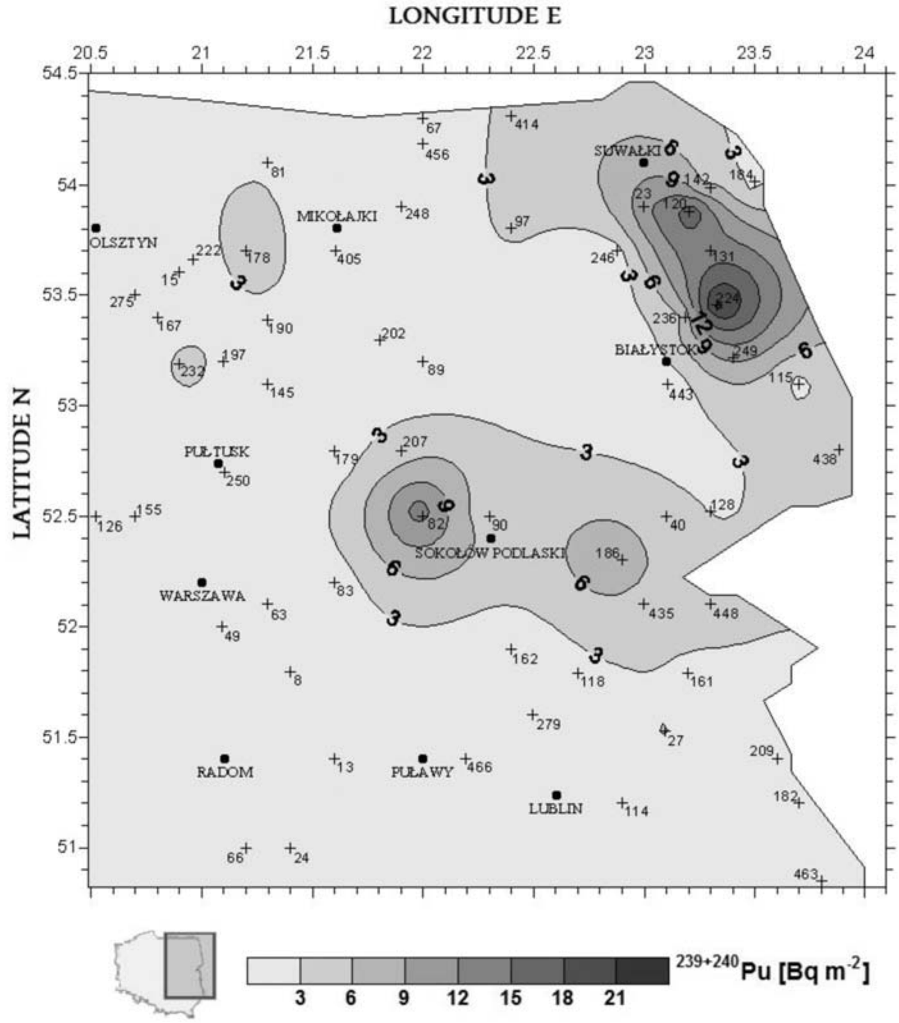

Fig. 4. Approximate map of the $^{239+240}$Pu deposition Chernobyl fallout component in North-eastern and Eastern Poland, based on samples in 59 locations (marked crosses, decay corrected for 1 September 1991).

collected only 5 years after the Chernobyl accident. In that short time plutonium cannot moved largely to deeper layers. The Chernobyl component of $^{239+240}$Pu activity deposited in both examined layers in the studied area was near 3.4 Bq·m^{-2}, on the average.

The present map of the Chernobyl fallout component $^{239+240}$Pu (Fig. 4) was made on the basis of received results displayed in the last column of Table 1. The largest Chernobyl $^{239+240}$Pu contamination occurred in north-eastern and eastern parts of the investigated area and it reaches at the maximum 22.1 ± 1.6 Bq·m^{-2}. By definition, another half of this value was deposition of ^{238}Pu from Chernobyl. Since samples were collected only 5 years after the Chernobyl accident, one can assume that the majority of Pu from Chernobyl was present in the analyzed two top forest litter/hums layers. This is not necessarily true in the case of Pu from global fallout, which can be already driven to deeper layers. Such situation is supported by deposition calculations made for the total Pu content. The majority of samples show a much lower total deposition than the average of 58 Bq/m^2 for the 50°–60° N latitude belt given in UNSCEAR reports [24]. This means that apparently an important part of global fallout plutonium is deposited deeper than the examined layers. It is very likely that it happens also in the case of ^{137}Cs, therefore, no attempt was made to show deposition map for radiocesium. Such maps are known for Poland [1, 23].

The presented map is, in general, confirming our earlier map (Fig. 1), which was quoted in Introduction. It supports the findings about the enhanced presence of non-volatile constituents of Chernobyl fallout in North-eastern Poland. Such a feature was reported even in very early studies [2, 20]. The Chernobyl plutonium deposition pattern seems to be similar to that for ^{90}Sr suggested from the studies on bilberry leaves [7]. Those findings support a general knowledge [11] of the traces of radioactive plume from Chernobyl (Fig. 5). The deposition of mostly non-volatile radioisotopes from the Chernobyl plume moving at altitude of about 500 m toward North-eastern Poland was predicted in another early paper [13]. However, the present results suggest that some deposition of non-volatile radioisotopes occurred also in Central-eastern Poland – the area of Bielsk Podlaski, Mińsk Mazowiecki or Siedlce as was already suggested by the geographical distribution of ^{90}Sr [7].

Figure 6 present the correlation between the ^{137}Cs activity and the observed activity of $^{239+240}$Pu and the Chernobyl fallout component of $^{239+240}$Pu in both layer. The lack of correlation among these elements can be noticed. This suggests that the proportions between the activity of ^{137}Cs and the $^{239+240}$Pu in Chernobyl plume over Poland changed from site to site with distance, since plutonium was transported on larger aerosols and likely only in plume from the initial explosion, not

Table 1. Results of the ^{137}Cs and plutonium isotopes activity and deposition in the examined forest litte/humus samples as well as the calculated Chernobyl fraction for $^{239+240}$Pu (F) and calculated values for the solely Chernobyl plutonium fallout. All data decay corrected for 1991

Code	Layer	^{137}Cs (Bq/m^2)	^{137}Cs (Bq/m^2)	Chemical yield (%)	$^{239+240}$Pu (Bq/kg)	^{238}Pu (Bq/kg)	$A_{238}/A_{239+240}$	$A_{ch(239+240)}$ (Bq/kg)	F (%)	$A_{ch(239+240)}$ (Bq/m^2)
8	A_0	104 ± 4	174 ± 11	90	0.05 ± 0.01	0.01 ± 0.01	0.27 ± 0.07	0.03 ± 0.01	52 ± 15	0.043 ± 0.003
	A_1	383 ± 16	2980 ± 260	75	2.19 ± 0.14	0.15 ± 0.01	0.07 ± 0.01	0.17 ± 0.05	8 ± 3	1.32 ± 0.13
13*	A_0	423 ± 12	233 ± 13	52	0.10 ± 0.04	0.14 ± 0.05	1.40 ± 0.72	0.10 ± 0.04	100**	0.055 ± 0.007
	A_1	855 ± 23	2640 ± 210	74	0.82 ± 0.09	0.26 ± 0.05	0.32 ± 0.07	0.50 ± 0.13	61 ± 17	1.55 ± 0.14
15	A_0	1637 ± 69	1480 ± 150	74	0.93 ± 0.10	0.30 ± 0.04	0.32 ± 0.06	0.58 ± 0.11	62 ± 14	0.519 ± 0.040
	A_1	967 ± 47	3380 ± 270	80	2.30 ± 0.24	0.27 ± 0.06	0.12 ± 0.03	0.42 ± 0.14	18 ± 6	1.49 ± 0.15
23*	A_0	607 ± 19	620 ± 60	89	3.24 ± 0.27	1.18 ± 0.16	0.36 ± 0.06	2.30 ± 0.42	71 ± 14	2.28 ± 0.17
	A_1	355 ± 15	1800 ± 130	71	2.70 ± 0.25	0.97 ± 0.16	0.36 ± 0.07	1.89 ± 0.40	70 ± 16	9.60 ± 0.82
24	A_0	84 ± 4	58 ± 4	84	0.48 ± 0.04	0.02 ± 0.01	0.05 ± 0.01	0.02 ± 0.01	4 ± 3	0.012 ± 0.003
	A_1	86 ± 4	611 ± 55	82	1.19 ± 0.08	0.06 ± 0.01	0.05 ± 0.01	0.06 ± 0.03	5 ± 3	0.402 ± 0.056
27	A_0	372 ± 16	460 ± 30	96	0.68 ± 0.04	0.07 ± 0.01	0.11 ± 0.01	0.11 ± 0.02	16 ± 3	0.137 ± 0.011
	A_1	518 ± 22	2360 ± 200	90	4.06 ± 0.68	0.43 ± 0.10	0.11 ± 0.03	0.65 ± 0.24	16 ± 6	2.97 ± 0.31
40	A_0	363 ± 16	338 ± 23	87	1.57 ± 0.07	0.83 ± 0.05	0.53 ± 0.04	1.57 ± 0.07	100**	1.460 ± 0.076
	A_1	1060 ± 150	5350 ± 840	87	2.54 ± 0.20	0.26 ± 0.05	0.10 ± 0.02	0.40 ± 0.12	16 ± 5	2.02 ± 0.20
49*	A_0	174 ± 6	104 ± 6	83	0.10 ± 0.01	0.01 ± 0.01	0.14 ± 0.05	0.02 ± 0.01	24 ± 10	0.014 ± 0.002
	A_1	562 ± 16	1280 ± 100	88	0.97 ± 0.05	0.03 ± 0.02	0.03 ± 0.02	0.01 ± 0.04	1 ± 4	0.018 ± 0.018
63	A_0	216 ± 9	406 ± 26	86	0.20 ± 0.02	0.04 ± 0.01	0.18 ± 0.04	0.06 ± 0.02	32 ± 9	0.121 ± 0.011
	A_1	79 ± 3	1530 ± 130	88	2.22 ± 0.18	0.07 ± 0.02	0.03 ± 0.01	< 0.06	< 3	< 0.24
66	A_0	186 ± 12	483 ± 40	89	0.60 ± 0.07	0.01 ± 0.01	0.02 ± 0.01	< 0.02	< 3	< 0.012
	A_1	290 ± 26	3770 ± 440	90	0.87 ± 0.07	0.07 ± 0.01	0.08 ± 0.02	0.09 ± 0.03	10 ± 4	1.16 ± 0.12
67	A_0	764 ± 34	550 ± 60	85	0.70 ± 0.06	0.33 ± 0.03	0.47 ± 0.06	0.66 ± 0.10	9 ± 16	0.473 ± 0.032
	A_1	174 ± 11	1130 ± 90	89	2.30 ± 0.17	0.08 ± 0.05	0.04 ± 0.02	0.03 ± 0.13	1 ± 5	0.19 ± 0.16
81	A_0	924 ± 59	1330 ± 130	71	1.19 ± 0.25	0.58 ± 0.11	0.49 ± 0.14	1.15 ± 0.27	97 ± 31	1.66 ± 0.14
	A_1	337 ± 23	2300 ± 200	72	4.52 ± 0.58	0.22 ± 0.10	0.05 ± 0.02	0.17 ± 0.23	4 ± 5	1.18 ± 0.33
82	A_0	103 ± 6	273 ± 21	80	0.13 ± 0.03	0.03 ± 0.01	0.24 ± 0.07	0.06 ± 0.01	44 ± 14	0.151 ± 0.012
	A_1	576 ± 44	11,000 ± 1200	96	3.51 ± 0.40	0.44 ± 0.04	0.13 ± 0.02	0.72 ± 0.14	21 ± 5	13.49 ± 1.13
83	A_0	545 ± 24	583 ± 39	85	0.21 ± 0.02	0.01 ± 0.01	0.05 ± 0.07	0.01 ± 0.03	5 ± 14	0.011 ± 0.010
	A_1	348 ± 14	3070 ± 260	85	0.69 ± 0.06	0.03 ± 0.01	0.04 ± 0.01	0.02 ± 0.02	3 ± 3	0.187 ± 0.035

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Table 1. Results of the ^{137}Cs and plutonium isotopes activity and deposition in the examined forest litte/humus samples as well as the calculated Chernobyl fraction for $^{239+240}$Pu (F) and calculated values for the solely Chernobyl plutonium fallout. All data decay corrected for 1991 (*cont.*)

Code	Layer	^{137}Cs (Bq/m^2)	^{137}Cs (Bq/m^2)	Chemical yield (%)	$^{239+240}$Pu (Bq/kg)	^{238}Pu (Bq/kg)	$A_{238}/A_{239+240}$	$A_{ch(239+240)}$ (Bq/kg)	F (%)	$A_{ch(239+240)}$ (Bq/m^2)
89	A_0	487 ± 15	150 ± 30	82	0.28 ± 0.03	0.16 ± 0.02	0.57 ± 0.09	0.28 ± 0.03	100**	0.087 ± 0.005
	A_1	443 ± 10	300 ± 60	60	0.34 ± 0.03	0.12 ± 0.02	0.34 ± 0.06	0.22 ± 0.05	66 ± 15	0.153 ± 0.013
90*	A_0	2062 ± 51	1072 ± 60	70	2.27 ± 0.20	1.43 ± 0.14	0.63 ± 0.08	2.27 ± 0.20	100**	1.180 ± 0.067
	A_1	1990 ± 140	8220 ± 850	81	4.40 ± 0.34	0.50 ± 0.09	0.11 ± 0.02	0.78 ± 0.22	18 ± 5	3.24 ± 0.30
97	A_{0L}	925 ± 42	1390 ± 120	68	2.75 ± 0.28	0.65 ± 0.07	0.24 ± 0.03	1.21 ± 0.20	44 ± 8	1.82 ± 0.13
	A_{0H}	683 ± 85	1940 ± 280	65	3.92 ± 0.48	0.32 ± 0.06	0.08 ± 0.02	0.42 ± 0.15	11 ± 4	1.21 ± 0.15
	A_1	93 ± 13	1670 ± 250	65	0.44 ± 0.11	0.03 ± 0.01	0.06 ± 0.02	0.03 ± 0.02	7 ± 4	0.548 ± 0.077
114	A_0	241 ± 10	468 ± 31	87	0.36 ± 0.05	0.04 ± 0.01	0.12 ± 0.03	0.07 ± 0.02	20 ± 6	0.138 ± 0.013
	A_1	19 ± 1	428 ± 46	84	0.14 ± 0.02	0.02 ± 0.01	0.12 ± 0.04	0.03 ± 0.01	19 ± 9	0.623 ± 0.070
115	A_0	33 ± 3	50 ± 10	65	0.02 ± 0.01	0.01 ± 0.00	0.31 ± 0.17	0.01 ± 0.00	59 ± 34	0.018 ± 0.002
	A_1	841 ± 39	2310 ± 190	83	2.14 ± 0.17	0.48 ± 0.04	0.22 ± 0.03	0.89 ± 0.13	41 ± 7	2.44 ± 0.20
118	A_0	520 ± 22	2070 ± 130	84	1.61 ± 0.18	0.10 ± 0.02	0.06 ± 0.02	0.11 ± 0.06	7 ± 4	0.435 ± 0.077
	A_1	43 ± 1	1450 ± 120	81	0.91 ± 0.07	0.05 ± 0.01	0.05 ± 0.01	0.05 ± 0.03	5 ± 3	1.62 ± 0.23
120*	A_0	870 ± 41	860 ± 80	73	3.98 ± 0.33	1.83 ± 0.20	0.46 ± 0.06	3.64 ± 0.58	91 ± 16	3.66 ± 0.25
	A_1	1042 ± 57	3360 ± 280	80	6.88 ± 0.53	2.17 ± 0.22	0.32 ± 0.04	4.18 ± 0.65	61 ± 11	13.48 ± 1.09
126	A_0	561 ± 27	551 ± 39	84	0.60 ± 0.05	0.18 ± 0.03	0.31 ± 0.05	0.35 ± 0.07	59 ± 12	0.347 ± 0.027
	A_1	219 ± 10	851 ± 75	78	3.31 ± 0.28	0.21 ± 0.03	0.06 ± 0.01	0.23 ± 0.10	7 ± 3	0.90 ± 0.10
128	A_0	1230 ± 250	3520 ± 730	89	0.80 ± 0.07	0.26 ± 0.03	0.32 ± 0.05	0.50 ± 0.08	62 ± 12	1.43 ± 0.10
	A_1	192 ± 27	3820 ± 610	95	1.14 ± 0.08	0.07 ± 0.01	0.06 ± 0.01	0.07 ± 0.04	6 ± 3	1.43 ± 0.18
131*	A_0	462 ± 19	470 ± 50	75	1.95 ± 0.21	0.52 ± 0.19	0.27 ± 0.10	0.98 ± 0.42	50 ± 22	1.01 ± 0.14
	A_1	787 ± 32	2150 ± 170	69	5.61 ± 0.48	2.20 ± 0.31	0.39 ± 0.06	4.32 ± 0.81	77 ± 16	11.87 ± 0.99
142*	A_0	360 ± 10	270 ± 30	37	3.37 ± 0.35	2.09 ± 0.27	0.62 ± 0.10	3.37 ± 0.35	100**	2.50 ± 0.15
	A_1	790 ± 24	2230 ± 160	60	4.78 ± 0.41	0.71 ± 0.23	0.15 ± 0.05	1.21 ± 0.51	25 ± 11	3.38 ± 0.38
145	A_0	1920 ± 130	2630 ± 270	56	1.39 ± 0.12	0.50 ± 0.05	0.36 ± 0.05	0.97 ± 0.15	70 ± 12	1.33 ± 0.09
	A_1	420 ± 20	1370 ± 110	81	5.32 ± 0.87	0.27 ± 0.10	0.05 ± 0.02	0.23 ± 0.24	4 ± 5	0.74 ± 0.17
155	A_0	87 ± 3	52 ± 3	89	0.12 ± 0.03	0.01 ± 0.01	0.09 ± 0.03	0.02 ± 0.01	14 ± 7	0.010 ± 0.001
	A_1	397 ± 25	1410 ± 140	83	0.46 ± 0.03	0.05 ± 0.01	0.10 ± 0.01	0.07 ± 0.02	15 ± 4	0.247 ± 0.022
161*	A_0	260 ± 8	208 ± 12	87	0.44 ± 0.07	0.25 ± 0.06	0.57 ± 0.17	0.44 ± 0.07	100**	0.352 ± 0.024
	A_1	1026 ± 29	2370 ± 190	89	3.83 ± 0.28	0.53 ± 0.06	0.14 ± 0.02	0.88 ± 0.18	23 ± 5	2.04 ± 0.17

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Table 1. Results of the ^{137}Cs and plutonium isotopes activity and deposition in the examined forest litte/humus samples as well as the calculated Chernobyl fraction for $^{239+240}$Pu (F) and calculated values for the solely Chernobyl plutonium fallout. All data decay corrected for 1991 (*cont.*)

Code	Layer	^{137}Cs (Bq/m^2)	^{137}Cs (Bq/m^2)	Chemical yield (%)	$^{239+240}$Pu (Bq/kg)	^{238}Pu (Bq/kg)	$A_{238}/A_{239+240}$	$A_{ch(239+240)}$ (Bq/kg)	F (%)	$A_{ch(239+240)}$ (Bq/m^2)
162	A_0	551 ± 23	836 ± 55	89	0.23 ± 0.02	0.10 ± 0.01	0.43 ± 0.05	0.20 ± 0.03	85 ± 14	0.297 ± 0.020
	A_1	513 ± 32	1320 ± 130	91	0.25 ± 0.02	0.08 ± 0.01	0.32 ± 0.04	0.16 ± 0.02	62 ± 11	0.401 ± 0.032
167	A_0	396 ± 11	340 ± 30	71	0.13 ± 0.01	0.08 ± 0.01	0.60 ± 0.10	0.13 ± 0.01	100**	0.113 ± 0.006
	A_1	1819 ± 42	2760 ± 270	68	1.44 ± 0.11	0.33 ± 0.03	0.23 ± 0.03	0.60 ± 0.10	42 ± 7	0.912 ± 0.074
178	A_0	479 ± 24	610 ± 60	59	0.26 ± 0.02	0.11 ± 0.02	0.43 ± 0.08	0.22 ± 0.04	84 ± 18	0.279 ± 0.022
	A_1	2350 ± 180	11,000 ± 1100	90	1.44 ± 0.15	0.48 ± 0.06	0.33 ± 0.05	0.93 ± 0.16	65 ± 13	4.55 ± 0.37
179*	A_0	1720 ± 140	2050 ± 230	65	1.29 ± 0.10	0.47 ± 0.07	0.36 ± 0.06	0.92 ± 0.18	71 ± 15	1.120 ± 0.039
	A_1	775 ± 56	5260 ± 550	80	1.39 ± 0.15	0.04 ± 0.12	0.03 ± 0.08	< 0.25	< 18	< 0.011
182	A_0	434 ± 13	616 ± 36	87	1.63 ± 0.14	0.07 ± 0.01	0.04 ± 0.01	0.05 ± 0.04	3 ± 2	0.072 ± 0.017
	A_1	389 ± 34	4950 ± 560	85	0.81 ± 0.06	0.06 ± 0.09	0.08 ± 0.11	0.08 ± 0.19	10 ± 24	1.01 ± 0.49
184*	A_0	606 ± 43	1020 ± 100	65	2.06 ± 0.13	0.73 ± 0.05	0.35 ± 0.03	1.42 ± 0.19	69 ± 10	2.38 ± 0.15
	A_1	402 ± 9	460 ± 60	71	1.64 ± 0.12	0.24 ± 0.03	0.15 ± 0.02	0.41 ± 0.08	25 ± 5	0.466 ± 0.040
186*	A_0	203 ± 8	201 ± 13	65	0.07 ± 0.04	0.04 ± 0.05	0.57 ± 0.72	0.07 ± 0.04	100**	0.069 ± 0.012
	A_1	1265 ± 71	3910 ± 370	80	2.27 ± 0.19	1.34 ± 0.13	0.59 ± 0.07	2.27 ± 0.19	100**	7.01 ± 0.54
190	A_0	807 ± 33	610 ± 70	55	0.42 ± 0.04	0.18 ± 0.03	0.43 ± 0.08	0.36 ± 0.07	86 ± 19	0.271 ± 0.021
	A_1	1950 ± 180	9400 ± 1000	89	6.31 ± 0.81	0.40 ± 0.08	0.06 ± 0.01	0.45 ± 0.22	7 ± 4	2.17 ± 0.26
197	A_0	1538 ± 91	2060 ± 200	74	0.58 ± 0.05	0.18 ± 0.03	0.32 ± 0.06	0.36 ± 0.07	62 ± 14	0.478 ± 0.038
	A_1	1020 ± 240	6700 ± 1600	84	4.32 ± 0.43	0.26 ± 0.06	0.06 ± 0.01	0.27 ± 0.15	6 ± 4	1.80 ± 0.24
202	A_0	111 ± 4	80 ± 10	71	0.08 ± 0.01	0.01 ± 0.01	0.19 ± 0.07	0.03 ± 0.01	34 ± 15	0.017 ± 0.002
	A_1	1282 ± 75	3800 ± 330	59	2.05 ± 0.23	0.49 ± 0.06	0.24 ± 0.04	0.91 ± 0.17	45 ± 10	2.71 ± 0.23
207	A_0	763 ± 30	633 ± 40	79	0.21 ± 0.02	0.09 ± 0.01	0.41 ± 0.05	0.17 ± 0.02	80 ± 14	0.139 ± 0.009
	A_1	1998 ± 95	5240 ± 470	95	2.26 ± 0.21	0.90 ± 0.08	0.40 ± 0.05	1.77 ± 0.26	78 ± 14	4.63 ± 0.37
209	A_0	1214 ± 50	2900 ± 190	87	0.84 ± 0.07	0.23 ± 0.03	0.28 ± 0.04	0.44 ± 0.08	53 ± 11	1.053 ± 0.080
	A_1	303 ± 13	1860 ± 160	89	3.10 ± 0.25	0.12 ± 0.06	0.04 ± 0.02	0.06 ± 0.14	2 ± 5	0.36 ± 0.18
222	A_0	83 ± 3	20 ± 10	75	0.14 ± 0.01	0.04 ± 0.01	0.32 ± 0.03	0.08 ± 0.01	61 ± 9	0.025 ± 0.002
	A_1	186 ± 12	2190 ± 180	75	1.69 ± 0.12	0.06 ± 0.01	0.04 ± 0.01	0.02 ± 0.04	1 ± 2	0.246 ± 0.099
224*	A_0	865 ± 65	940 ± 110	59	3.40 ± 0.25	1.74 ± 0.16	0.51 ± 0.06	3.40 ± 0.25	100**	3.70 ± 0.20
	A_1	374 ± 23	2320 ± 190	68	5.27 ± 0.50	1.53 ± 0.27	0.29 ± 0.06	2.92 ± 0.66	55 ± 14	18.39 ± 1.61

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Table 1. Results of the ^{137}Cs and plutonium isotopes activity and deposition in the examined forest litte/humus samples as well as the calculated Chernobyl fraction for $^{239+240}$Pu (F) and calculated values for the solely Chernobyl plutonium fallout. All data decay corrected for 1991 (*cont.*)

Code	Layer	^{137}Cs (Bq/m^2)	^{137}Cs (Bq/m^2)	Chemical yield (%)	$^{239+240}$Pu (Bq/kg)	^{238}Pu (Bq/kg)	$A_{238}/A_{239+240}$	$A_{ch(239+240)}$ (Bq/kg)	F (%)	$A_{ch(239+240)}$ (Bq/m^2)
232	A_0	800 ± 67	1300 ± 150	62	0.33 ± 0.04	0.20 ± 0.03	0.62 ± 0.11	0.33 ± 0.04	100**	0.537 ± 0.033
	A_1	674 ± 81	8100 ± 1100	56	0.61 ± 0.05	0.13 ± 0.03	0.21 ± 0.06	0.24 ± 0.07	39 ± 13	2.89 ± 0.28
236	A_0	310 ± 16	980 ± 80	77	0.75 ± 0.06	0.20 ± 0.04	0.27 ± 0.06	0.38 ± 0.09	50 ± 13	1.19 ± 0.11
	A_1	175 ± 15	2660 ± 270	61	1.02 ± 0.08	0.19 ± 0.02	0.18 ± 0.03	0.33 ± 0.07	33 ± 7	5.07 ± 0.43
246	A_0	80 ± 5	20 ± 10	81	0.04 ± 0.02	0.01 ± 0.01	0.30 ± 0.21	0.02 ± 0.01	58 ± 42	0.005 ± 0.001
	A_1	565 ± 14	860 ± 90	74	0.76 ± 0.07	0.23 ± 0.03	0.30 ± 0.05	0.45 ± 0.08	58 ± 12	0.676 ± 0.056
248*	A_{0L}	539 ± 42	1010 ± 110	67	0.64 ± 0.09	0.28 ± 0.08	0.44 ± 0.14	0.55 ± 0.18	87 ± 31	1.04 ± 0.11
	A_{0H}	1020 ± 120	2450 ± 330	83	1.91 ± 0.16	0.25 ± 0.06	0.13 ± 0.03	0.41 ± 0.14	21 ± 7	0.99 ± 0.11
	A_1	234 ± 12	2170 ± 160	86	0.84 ± 0.07	0.04 ± 0.05	0.05 ± 0.06	0.03 ± 0.11	4 ± 13	0.29 ± 0.20
249	A_0	679 ± 23	570 ± 60	74	2.21 ± 0.31	1.18 ± 0.15	0.53 ± 0.10	2.21 ± 0.31	100**	1.84 ± 0.12
	A_1	508 ± 13	1490 ± 110	82	4.15 ± 0.36	1.34 ± 0.12	0.32 ± 0.04	2.58 ± 0.38	62 ± 11	7.57 ± 0.61
250	A_0	605 ± 25	965 ± 62	87	0.41 ± 0.04	0.04 ± 0.01	0.10 ± 0.02	0.06 ± 0.01	14 ± 4	0.093 ± 0.008
	A_1	186 ± 7	3420 ± 290	79	0.35 ± 0.03	0.08 ± 0.01	0.23 ± 0.04	0.15 ± 0.03	42 ± 9	2.70 ± 0.23
275	A_0	1001 ± 25	670 ± 70	64	2.12 ± 0.32	0.31 ± 0.07	0.14 ± 0.04	0.52 ± 0.17	24 ± 9	0.348 ± 0.038
	A_1	746 ± 34	3450 ± 250	76	2.68 ± 0.25	0.18 ± 0.04	0.07 ± 0.02	0.20 ± 0.10	8 ± 4	0.95 ± 0.12
279	A_0	89 ± 4	33 ± 2	85	0.02 ± 0.01	< 0.01	0.17 ± 0.11	0.01 ± 0.01	30 ± 22	0.002 ± 0.001
	A_1	775 ± 19	860 ± 68	88	0.37 ± 0.03	0.08 ± 0.01	0.22 ± 0.03	0.15 ± 0.02	41 ± 7	0.167 ± 0.014
405	A_0	754 ± 23	1310 ± 110	77	3.98 ± 0.66	0.26 ± 0.08	0.07 ± 0.02	0.30 ± 0.20	8 ± 5	0.53 ± 0.11
	A_1	79 ± 3	1010 ± 60	66	1.28 ± 0.10	0.05 ± 0.01	0.04 ± 0.01	0.03 ± 0.04	2 ± 3	0.375 ± 0.096
414	A_0	146 ± 6	90 ± 10	50	0.21 ± 0.03	0.07 ± 0.03	0.33 ± 0.13	0.13 ± 0.06	64 ± 28	0.085 ± 0.011
	A_1	780 ± 27	4310 ± 280	56	1.27 ± 0.12	0.35 ± 0.07	0.27 ± 0.06	0.66 ± 0.17	52 ± 14	3.66 ± 0.33
435	A_0	2020 ± 82	2420 ± 160	79	2.88 ± 0.54	1.28 ± 0.22	0.45 ± 0.11	2.54 ± 0.55	88 ± 25	3.04 ± 0.25
	A_1	526 ± 23	1650 ± 140	89	4.20 ± 0.56	0.28 ± 0.05	0.07 ± 0.01	0.33 ± 0.14	8 ± 3	1.04 ± 0.12
438	A_0	249 ± 9	330 ± 30	93	4.01 ± 0.54	1.78 ± 0.22	0.44 ± 0.08	3.53 ± 0.60	88 ± 19	4.71 ± 0.34
	A_1	86 ± 4	320 ± 20	82	2.24 ± 0.12	0.16 ± 0.03	0.07 ± 0.02	0.19 ± 0.09	9 ± 4	0.720 ± 0.084
443	A_0	440 ± 14	510 ± 50	83	1.34 ± 0.12	0.10 ± 0.05	0.08 ± 0.04	0.14 ± 0.12	10 ± 9	0.159 ± 0.042
	A_1	688 ± 21	7740 ± 460	76	1.63 ± 0.16	0.10 ± 0.01	0.06 ± 0.01	0.10 ± 0.05	6 ± 3	1.11 ± 0.13
448	A_0	205 ± 9	407 ± 27	91	0.36 ± 0.05	0.09 ± 0.04	0.24 ± 0.13	0.16 ± 0.10	45 ± 27	0.324 ± 0.059
	A_1	195 ± 9	4160 ± 360	90	0.48 ± 0.05	0.10 ± 0.04	0.20 ± 0.08	0.18 ± 0.09	37 ± 18	3.79 ± 0.46

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Table 1. Results of the ^{137}Cs and plutonium isotopes activity and deposition in the examined forest litte/humus samples as well as the calculated Chernobyl fraction for $^{239+240}$Pu (*F*) and calculated values for the solely Chernobyl plutonium fallout. All data decay corrected for 1991 (*cont.*)

Code	Layer	^{137}Cs (Bq/m^2)	^{137}Cs (Bq/m^2)	Chemical yield (%)	$^{239+240}$Pu (Bq/kg)	^{238}Pu (Bq/kg)	$A_{238}/A_{239+240}$	$A_{ch(239+240)}$ (Bq/kg)	*F* (%)	$A_{ch(239+240)}$ (Bq/m^2)
456	A_0	153 ± 5	170 ± 20	71	0.86 ± 0.08	0.23 ± 0.04	0.26 ± 0.05	0.42 ± 0.09	49 ± 11	0.460 ± 0.038
	A_1	824 ± 26	4780 ± 300	84	3.52 ± 0.42	0.23 ± 0.08	0.06 ± 0.02	0.26 ± 0.18	7 ± 5	1.51 ± 0.24
463	A_0	6 ± 1	9 ± 2	83	0.19 ± 0.03	0.01 ± 0.01	0.07 ± 0.02	0.02 ± 0.01	9 ± 4	0.025 ± 0.004
	A_1	226 ± 14	105 ± 10	89	0.27 ± 0.03	0.03 ± 0.01	0.12 ± 0.02	0.05 ± 0.01	19 ± 5	0.023 ± 0.002
466	A_0	22 ± 3	5 ± 1	86	0.05 ± 0.02	0.01 ± 0.01	0.16 ± 0.08	0.01 ± 0.01	27 ± 14	0.003 ± 0.001
	A_1	76 ± 4	224 ± 21	92	0.20 ± 0.02	0.02 ± 0.01	0.12 ± 0.03	0.04 ± 0.01	19 ± 6	0.111 ± 0.011

* Result taken from earlier work [19].
** In further calculations assumed 100%.

Fig. 5. Migration of the Chernobyl radioactive cloud over Poland [11].

from the later fire. The mixing origin (Chernobyl/global fallouts) of radiocesium also acts destructively towards correlations.

Dosimetric aspect of the presence of Pu in the environment of Poland was a subject of study based on hospital diet [21]. The inhalation of Pu by Polish subjects was not investigated yet. We hope that the presented here approximate map could be helpful for future retrospective studies in this field.

Conclusion

The results on radiocesium and plutonium activity and deposition in forest litter presented now will contribute to database on the radioactive contamination in Polish forests. The presented approximate map of Pu of Chernobyl origin seems to give some general overview on the scale and deposition pattern of the Chernobyl-origin Pu in Poland. The maximum deposition of this plutonium (including ^{238}Pu) seems to be below a half of the mean global fallout values for Poland. The north-eastern part of Poland (for example, Augustów Primeval Forest) seems to be the site with the highest values of Chernobyl-origin Pu. Enhanced levels of Pu can be found also in the area of Mińsk Mazowiecki–Siedlce–Bielsk Podlaski.

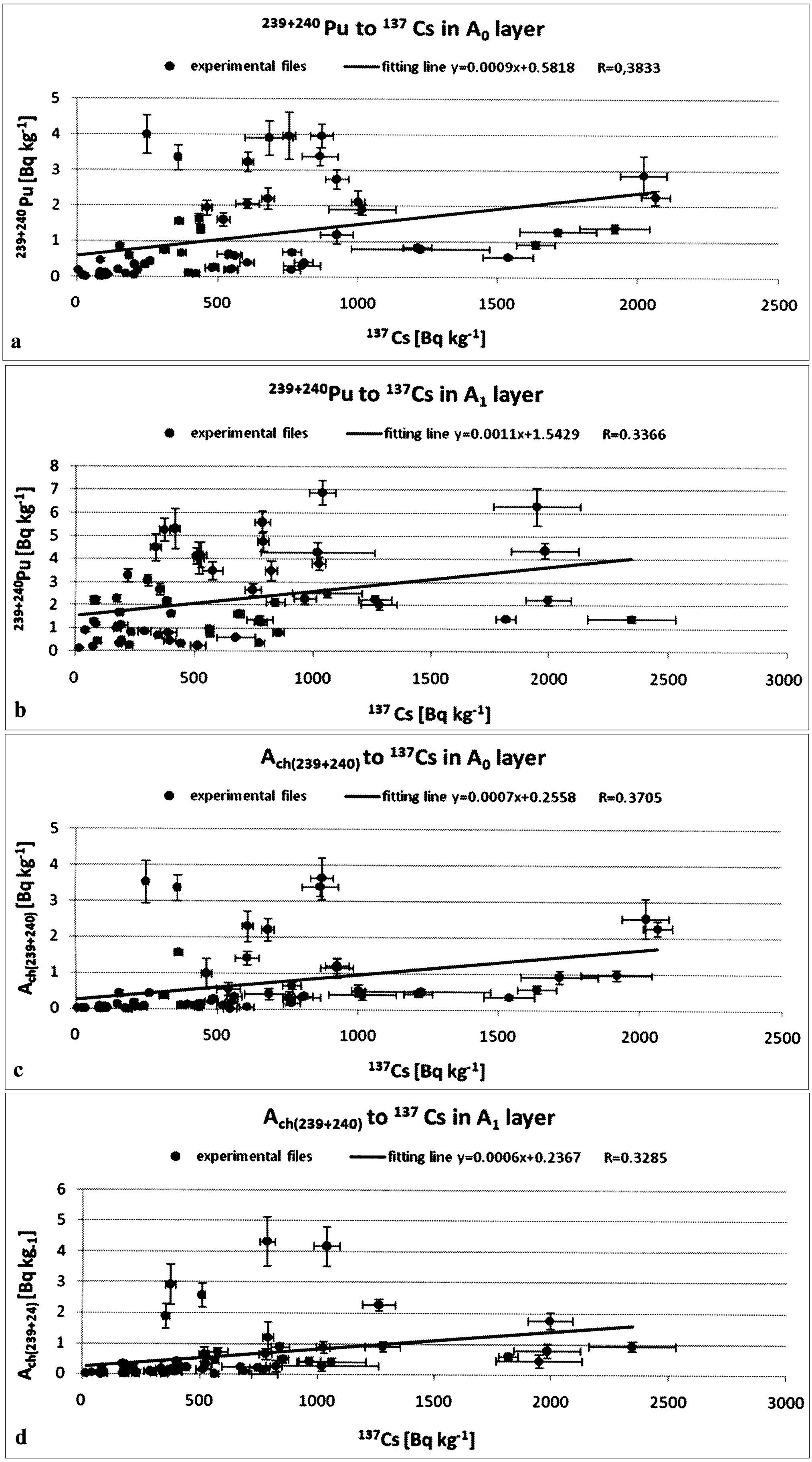

Fig. 6. Correlation between ^{137}Cs activity and (a, b) the total activity of $^{239+240}$Pu or (c, d) ^{137}Cs with the solely Chernobyl-origin activity of $^{239+240}$Pu. The top row concerns forest litter layer, the bottom concerns forest humus layer. Data for 1991.

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