

Nuclear Debris Characterization by Fission Isotope Assessment

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Introduction

Isotopic composition analysis allows the identification of an artificial source of radionuclides.



1 Fig. Fallout in Northern hemisphere



2 Fig. Chernobyl NPP accident (1986)
 $^{240}\text{Pu}/^{239}\text{Pu} = 0.180 \pm 0.007(1\sigma)$
(Kelley et al., 1999)

$^{240}\text{Pu}/^{239}\text{Pu} = 0.403 \pm 0.003(1\sigma)$
(Muramatsu et al., 2000)

$^{238}\text{Pu}/^{239+240}\text{Pu} = 0.04 \pm 0.01$
(Mietelski and Was, 1995)

$^{238}\text{Pu}/^{239+240}\text{Pu} = 0.51 \pm 0.03$
(Remeikis et al., 1995)

$^{137}\text{Cs}/^{239,240}\text{Pu}$ activity ratio for the global fallout is 28 ± 3

(for the 2021 year)

The goal is to determine radionuclide local contamination source vs. global contamination through the isotopic ratio assessment.



Fig 3. Geographical sampling situation (in dark green) and the Chernobyl NPP (the source) location

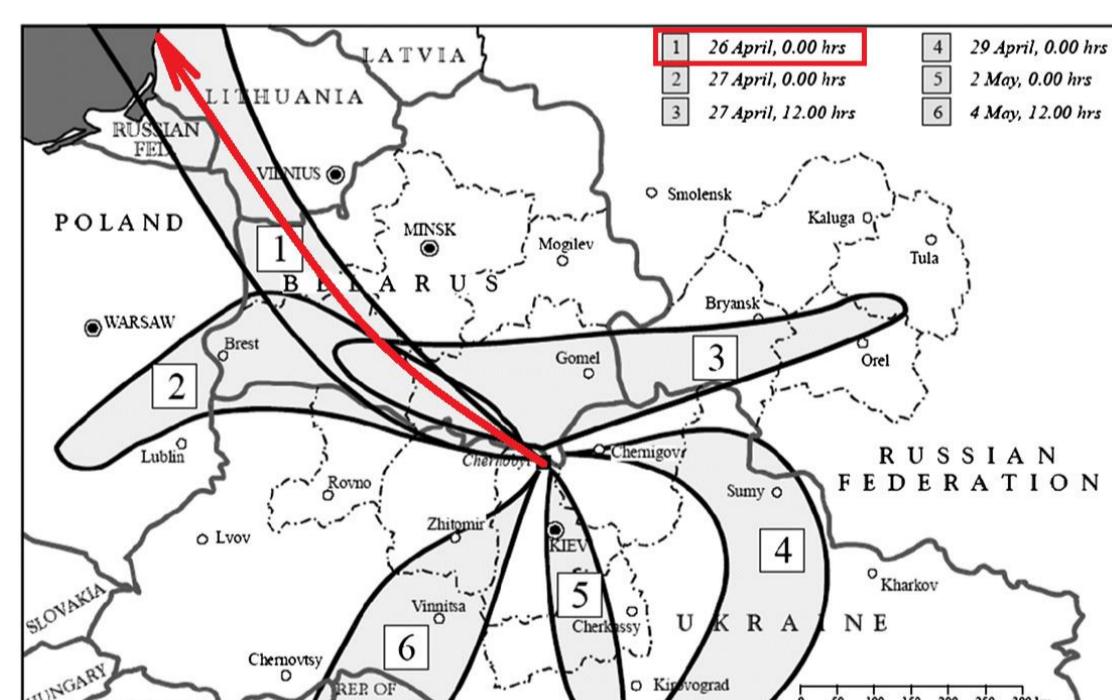


Fig 4. Modelled Chernobyl NPP accident emission dispersion
(Mozilov et al., 1993)

Sampling and measurements

Samples:

- undisturbed meadow and forest soils, 0-5 cm depth profile, with upper layer.

Measurement equipment:

- HPGe gamma spectrometry – for caesium-137,
- Inductively-coupled plasma mass spectrometry – for plutonium-239 and plutonium-240,
- alpha spectrometry – for plutonium-238 and plutonium-239,240 determination.

Emission source identification through caesium and plutonium isotopic information

Plutonium isotope activity concentrations ranged from 0.01 Bq kg⁻¹ to 0.25 Bq kg⁻¹ ^{238}Pu and from 0.05 Bq kg⁻¹ to 1.80 Bq kg⁻¹ $^{239,240}\text{Pu}$ in meadow and forest soil samples. The calculated $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratios and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios are 0.02–0.30 and 0.18–0.26, respectively. Relatively lower 0.38–0.05 Bq kg⁻¹ ^{238}Pu activity concentration and 0.07–0.53 Bq kg⁻¹ $^{239,240}\text{Pu}$ activity concentration were measured in meadow soil, while high activity concentration was found in forest soils. 0.05 - 0.09 Bq kg⁻¹ ^{238}Pu and 0.74 - 1.80 Bq kg⁻¹ $^{239,240}\text{Pu}$. $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios did not show significant difference in meadow and forest soil samples. The increased $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratio to 0.30 and the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio to 0.26 in the southern, southeastern and western parts of Lithuania indicate the source (Chernobyl accident emissions).

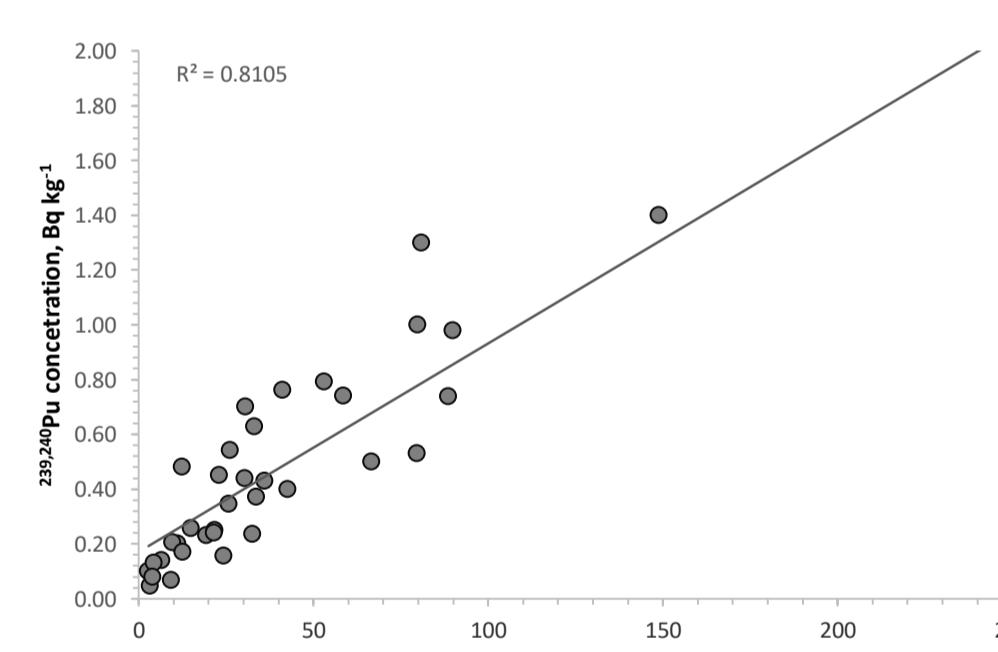


Fig 5. ^{137}Cs and $^{239,240}\text{Pu}$ correlation

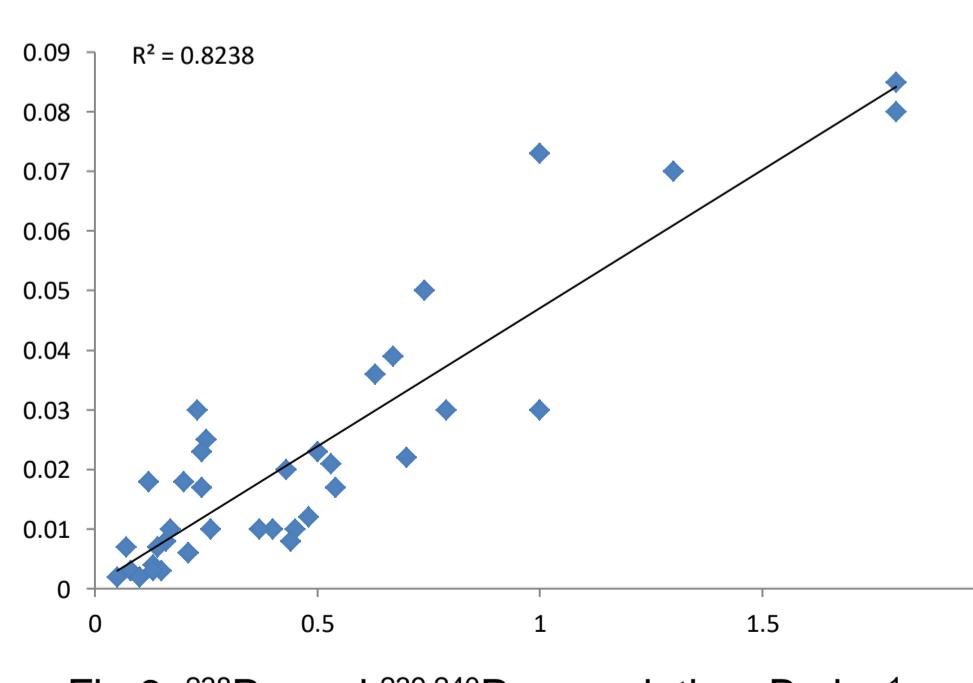


Fig 6. ^{238}Pu and $^{239,240}\text{Pu}$ correlation, Bq kg⁻¹

Sample preparation

- Undisturbed meadow and forest soil samples, were sieved (50 g dry mass + ^{242}Pu tracer added);
- Pu chemical separation and concentration: ion exchange resin DOWEX 1X8 + afterwards U/TEVA, TRU resins.

1 Table. ^{137}Cs and Pu isotopes in undisturbed meadow and forest soils

Sample no.	Location, sample type	Specific activity, Bq/kg			Activity ratio	Isotopic ratio	Activity ratio
		^{137}Cs	^{238}Pu	$^{239+240}\text{Pu}$			
-							
1	Lieponys	23.0±1.8	0.010±0.002	0.45±0.04	0.02±0.04	0.195±0.012	51.1
2	Prienai	12.5±1.0	0.012±0.002	0.48±0.05	0.03±0.04	0.187±0.016	26.0
3	Skriaudupis	148.8±11.8	0.250±0.018	1.40±0.03	0.18±0.03	0.243±0.002	106.3
4	Kapčiamiestis	58.6±3.1	0.050±0.007	0.74±0.04	0.07±0.04	0.212±0.010	79.2
5	Kapčiamiestis (co-méginiys)	89.9±5.5	0.073±0.020	1.00±0.08	0.07±0.08	0.204±0.007	89.9
6	Druskininkai	33.1±2.6	0.036±0.008	0.63±0.04	0.06±0.04	0.190±0.005	52.5
7	Latežeris	81.0±6.5	0.070±0.006	1.30±0.04	0.05±0.04	0.187±0.004	84.0
8	Latežeris (5-10 cm gylyje)	36.1±2.9	0.020±0.002	0.43±0.04	0.05±0.04	0.193±0.008	84.0
9	Latežeris (co-méginiys)	44.0±3.5	0.080±0.007	1.80±0.03	0.04±0.03	0.192±0.003	24.4
10	Latežeris (co-méginiys 2)	210.6±16.8	0.018±0.002	0.12±0.02	0.15±0.02	0.221±0.027	1755.0
11	Marcinkony	66.7±3.5	0.023±0.002	0.50±0.03	0.05±0.03	0.199±0.012	133.4
12	Varėna	79.8±6.4	0.030±0.003	1.00±0.08	0.03±0.08	0.184±0.004	79.8
13	Dieveniškės	24.4±1.2	0.008±0.005	0.16±0.02	0.05±0.02	0.204±0.030	152.5
14	Dieveniškės (co-méginiys)	250.0±20.0	0.085±0.008	1.80±0.04	0.05±0.04	0.189±0.003	138.9
ZIL-2	Ignalinos AE pusiasalyje	6.66±0.3	0.006±0.002	0.21±0.02	0.03±0.02	0.188±0.029	31.7
SIL	Šilutė	94.3±4.1	0.039±0.005	0.67±0.03	0.06±0.03	0.190±0.008	140.9
Z1	Nagliaj alėja	171.3±7.3	0.006±0.001	0.04±0.01	0.17±0.50	0.205±0.027	4564.5
Z2	Skypačiai	46.8±3.4	0.002±0.001	0.02±0.01	0.12±0.85	0.207±0.069	2476.3
Z3	Pamavys	65.7±4.2	0.005±0.003	0.03±0.01	0.15±0.49	0.230±0.030	2173.4
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15	Vilkaviškis	19.4±1.2	0.030±0.002	0.23±0.02	0.12±0.02	0.243±0.018	84.3
16	Vilkaviškis (co-méginiys)	11.1±0.9	0.018±0.002	0.20±0.03	0.09±0.03	0.236±0.021	55.5
17	Virbalis	21.8±1.0	0.025±0.004	0.25±0.02	0.10±0.02	0.242±0.014	87.2
18	Gražiškiai	9.3±0.7	0.007±0.001	0.07±0.01	0.10±0.01	0.214±0.032	132.9
19	Lazdijai	21.5±1.0	0.023±0.003	0.24±0.03	0.10±0.03	0.263±0.016	89.6
20	Veisiejai	42.7±1.0	0.010±0.002	0.40±0.04	0.03±0.04	0.196±0.012	106.8
21	Skriaudupis	6.5±0.5	0.007±0.001	0.14±0.02	0.04±0.03	0.196±0.032	46.4
22	Kapčiamiestis	14.9±1.1	0.010±0.002	0.26±0.03	0.04±0.03	0.217±0.020	57.3
23	Kapčiamiestis (co-méginiys)	32.6±2.2	0.017±0.002	0.24±0.03	0.07±0.03	0.168±0.022	135.8
24	Kapčiamiestis (co-méginiys 2)	9.7±0.9	0.006±0.001	0.21±0.02	0.03±0.02	0.205±0.018	46.2
25	Leipalingis	30.5±2.4	0.022±0.003	0.70±0.04	0.03±0.04	0.189±0.0073	43.6
26	Latežeris	30.4±2.4	0.008±0.001	0.44±0.05	0.02±0.05	0.184±0.009	69.1
27	Marcinkony	12.6±0.9	0.010±0.001	0.17±0.02	0.05±0.02	0.190±0.034	74.1
28	Varėna	26.2±1.6	0.017±0.002	0.54±0.02	0.03±0.02	0.198±0.007	48.5
29	Senoji Varėna	53.0±4.2	0.030±0.003	0.79±0.03	0.04±0.03	0.199±0.011	67.1
30	Eišiškės	3.3±0.3	0.002±0.001	0.05±0.01	0.04±0.01	0.195±0.038	66.0
31	Šalčininkai	4.4±0.3	0.004±0.001	0.13±0.02	0.03±0.03	0.199±0.021	33.8
32	Dieveniškės	4.0±0.9	0.003±0.001	0.08±0.01	0.04±0.01	0.210±0.036	50.0
33	Dieveniškės (co-méginiys)	33.6±2.7	0.010±0.001	0.37±0.03	0.03±0.03	0.191±0.011	90.8
34	Stakiai	79.7±6.4	0.021±0.002	0.53±0.03	0.04±0.03	0.187±0.010	150.4
35	Igliškėlis	2.8±0.9	0.002±0.001	0.10±0.01	0.02±0.01	0.231±0.036	28.0
ZIL-1	Ignalinos AE pusiasalyje	5.29±0.2	0.003±0.001	0.15±0.02	0.02±0.01	0.195±0.029	35.3
ZIL-6	Tilžė (pri Drūkščių ež.)	4.1±0.2	0.003±0.001	0.13±0.01	0.02±0.01	0.177±0.021	