

# Nuclear Event Fallout Inhomogeneity on Small- and Medium-Scale in Meadows and Forests

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## Introduction

The isotopic composition analysis leads to the determination of the artificial radionuclide source.



Fig 1. Fall-out in the Northern hemisphere

$$^{240}\text{Pu}/^{239}\text{Pu} = 0.180 \pm 0.007(1\sigma)$$

(Kelley et al., 1999)

$$^{238}\text{Pu}/^{239+240}\text{Pu} = 0.04 \pm 0.01$$

(Mietelski and Was, 1995)

$^{137}\text{Cs}/^{239,240}\text{Pu}$  activity ratio for the global fallout is  $32 \pm 3$  (reference year 2016)

**The objective is to assess a radionuclide emission source vs. global nuclear fall-out by isotopic information.**



Fig 2. Chernobyl NPP accident (in 1986)

$$^{240}\text{Pu}/^{239}\text{Pu} = 0.403 \pm 0.003(1\sigma)$$

(Muramatsu et al., 2000)

$$^{238}\text{Pu}/^{239+240}\text{Pu} = 0.51 \pm 0.03$$

(Remeikis et al., 1995)



Figure 3. Geographical location of sampling place (coloured in dark green) and the Chernobyl NPP (the source) situation

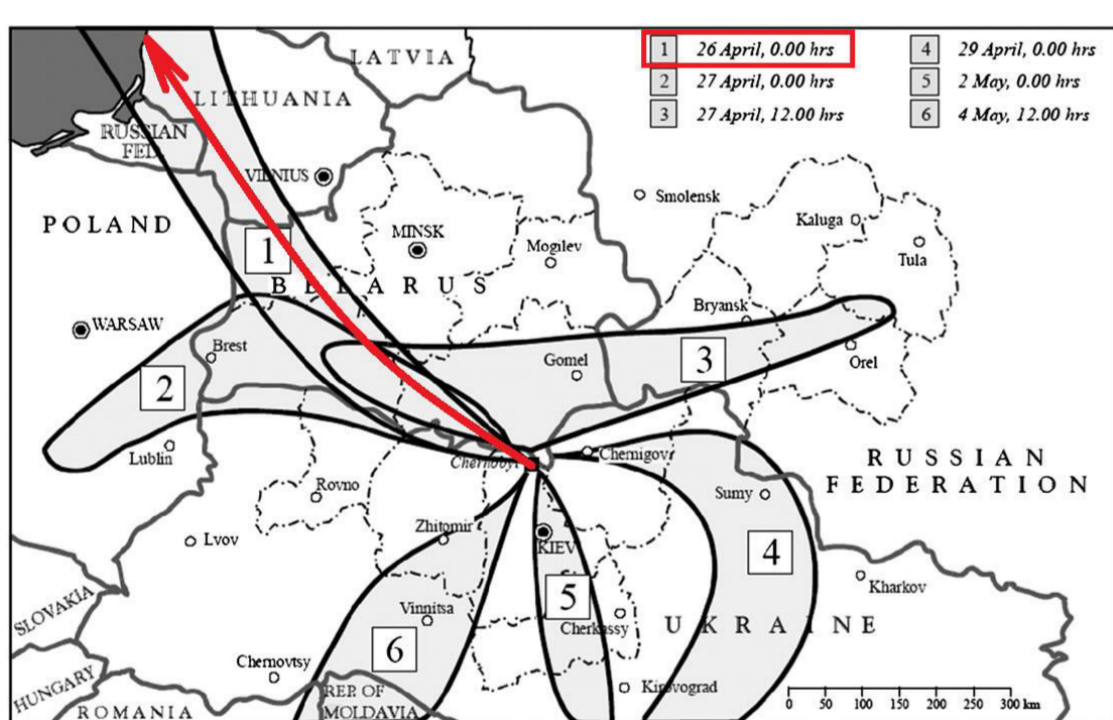


Figure 4. Chernobyl NPP plume paths modeled (Mozilov et al., 1993)

## Sampling and techniques

### Samples:

- undisturbed grassland meadows and forest soils, 0-5 cm profile, with litter.

### Techniques:

- HPGe gamma spectrometry – for cesium-137,
- inductively coupled plasma mass spectrometry – for iodine-127, plutonium-239 and plutonium-240,
- alpha spectrometry - for plutonium-238 and plutonium-239,240 determination.

## A source defined by cesium and plutonium isotopes

The activity concentrations of plutonium isotopes ranged from  $0.01 \text{ Bq kg}^{-1}$  to  $0.25 \text{ Bq kg}^{-1}$  for  $^{238}\text{Pu}$  and from  $0.05 \text{ Bq kg}^{-1}$  to  $1.80 \text{ Bq kg}^{-1}$  for  $^{239,240}\text{Pu}$  in meadow and forest soil samples.

$^{238}\text{Pu}/^{239,240}\text{Pu}$  activity ratios and  $^{240}\text{Pu}/^{239}\text{Pu}$  atomic ratios were calculated to be 0.02-0.18 and 0.18-0.24, respectively. In meadow soil, a relative lower  $^{238}\text{Pu}$  activity concentration of  $0.01\text{--}0.05 \text{ Bq kg}^{-1}$ , and  $^{239+240}\text{Pu}$  concentration of  $0.07\text{--}0.53 \text{ Bq kg}^{-1}$  were measured, whereas a high activity concentrations were determined in forest soils, varying in  $0.05\text{--}0.09 \text{ Bq kg}^{-1}$  of  $^{238}\text{Pu}$  and  $0.74\text{--}1.80 \text{ Bq kg}^{-1}$  of  $^{239,240}\text{Pu}$ .  $^{238}\text{Pu}/^{239,240}\text{Pu}$  activity and  $^{240}\text{Pu}/^{239}\text{Pu}$  atomic ratios did not show any significant difference in undisturbed meadow and forest soil samples. Elevated  $^{238}\text{Pu}/^{239,240}\text{Pu}$  activity and  $^{240}\text{Pu}/^{239}\text{Pu}$  atomic ratios up to 0.18 and up to 0.26, respectively, in southern part of Lithuania attribute to the deposition of the Chernobyl accident (a source).

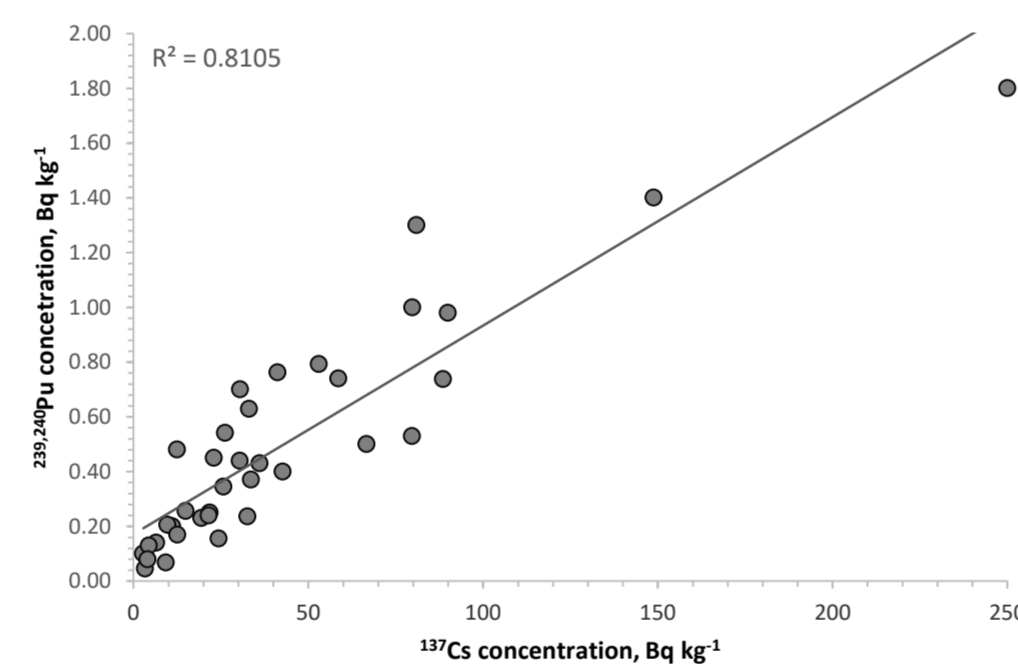


Figure 5. Correlation between  $^{239,240}\text{Pu}$  and  $^{238}\text{Pu}$

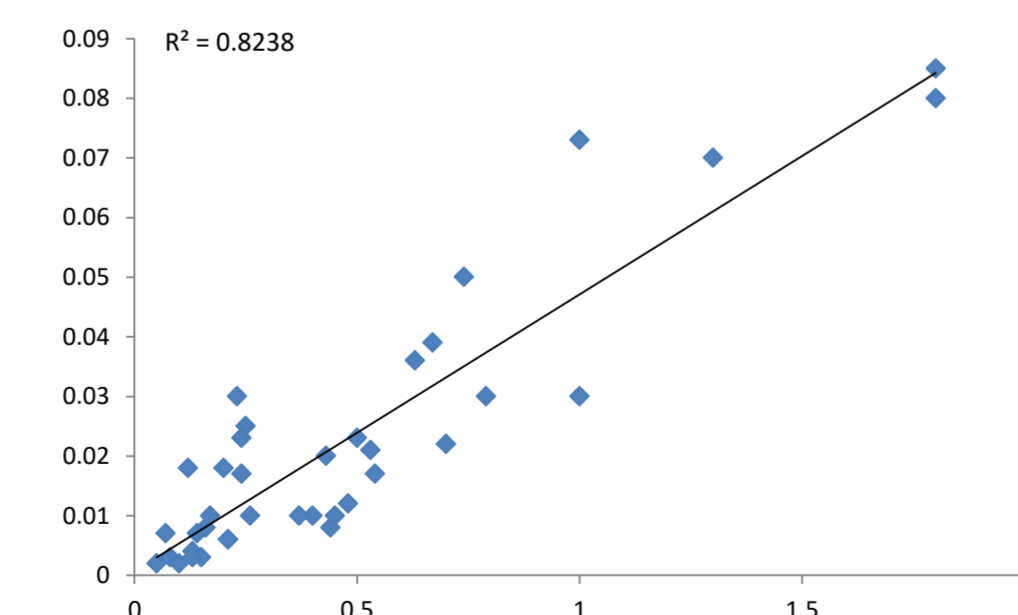


Figure 6. Correlation between  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$ , in  $\text{Bq kg}^{-1}$

## Sample preparation

- undisturbed grassland and forest soils (50 g of dry weight +  $^{242}\text{Pu}$  tracer added);
- Pu isotopes separation and purification step:* strong acid anion exchange resin DOWEX 1X8 + U/TEVA, TRU columns afterwards.

Table 2.  $^{137}\text{Cs}$  and Pu isotopes in meadow and forest soils

Sample code	Location, sample type	Specific activity, Bq/kg			Isotopic ratio		Activity ratio
		$^{137}\text{Cs}$	$^{238}\text{Pu}$	$^{239+240}\text{Pu}$	$^{238}\text{Pu}/^{239+240}\text{Pu}$	$^{240}\text{Pu}/^{239}\text{Pu}$	
<b>Forest soil</b>							
AN1	Lieponys	23.0±1.8	0.010±0.002	0.45±0.04	0.02±0.04	0.195±0.012	51.1
AN2	Prienai	12.5±1.0	0.012±0.002	0.48±0.05	0.03±0.04	0.187±0.016	26.0
AN3	Skriaudupis AMS4	148.8±11.8	0.250±0.018	1.40±0.03	0.18±0.03	0.243±0.002	106.3
AN4	Kapčiamiestis	58.6±3.1	0.050±0.007	0.74±0.04	0.07±0.04	0.212±0.010	79.2
AN5	Kapčiamiestis (co-sample) AMS11	89.9±5.5	0.073±0.020	1.00±0.08	0.07±0.08	0.204±0.007	89.9
AN6	Druskininkai	33.1±2.6	0.036±0.008	0.63±0.04	0.06±0.04	0.190±0.005	52.5
AN7	Latežeris	81.0±6.5	0.070±0.006	1.30±0.04	0.05±0.04	0.187±0.004	84.0
AN8	Latežeris (5-10 cm depth)	36.1±2.9	0.020±0.002	0.43±0.04	0.05±0.04	0.193±0.008	84.0
AN9	Latežeris (co-sample) AMS8	44.0±3.5	0.080±0.007	1.80±0.03	0.04±0.03	0.192±0.003	24.4
AN10	Latežeris (co-sample 2) AMS8	210.6±16.8	0.018±0.002	0.12±0.02	0.15±0.02	0.221±0.027	1755.0
AN11	Marcinkony	66.7±3.5	0.023±0.002	0.50±0.03	0.05±0.03	0.190±0.012	133.4
AN12	Varėna	79.8±6.4	0.030±0.003	1.00±0.08	0.03±0.08	0.184±0.004	79.8
AN13	Dieveniškės	24.4±1.2	0.008±0.005	0.16±0.02	0.05±0.02	0.204±0.030	152.5
AN14	Dieveniškės (co-sample) AMS10	250.0±20.0	0.085±0.008	1.80±0.04	0.05±0.04	0.189±0.003	138.9
ZIL-2	Ignalina NPP peninsula	6.66±0.3	0.006±0.002	0.21±0.02	0.03±0.02	0.188±0.029	31.7
SIL	Šilutė	94.37±4.1	0.039±0.005	0.67±0.03	0.06±0.03	0.190±0.008	140.9
<b>Undisturbed meadow soil</b>							
AN15	Vilkaviškis AMS7	19.4±1.2	0.030±0.002	0.23±0.02	0.12±0.02	0.243±0.018	84.3
AN16	Vilkaviškis (co-sample) AMS6	11.1±0.9	0.018±0.002	0.20±0.03	0.09±0.03	0.236±0.021	55.5
AN17	Virbalis	21.8±1.0	0.025±0.004	0.25±0.02	0.10±0.02	0.242±0.014	87.2
AN18	Gražiškiai	9.3±0.7	0.007±0.001	0.07±0.01	0.10±0.01	0.214±0.032	132.9
AN19	Lazdijai	21.5±1.0	0.023±0.003	0.24±0.03	0.10±0.03	0.263±0.016	89.6
AN20	Veisiejai	42.7±1.0	0.010±0.002	0.40±0.04	0.03±0.04	0.196±0.012	106.8
AN21	Skriaudupis AMS5	6.5±0.5	0.007±0.001	0.14±0.02	0.04±0.03	0.196±0.032	46.4
AN22	Kapčiamiestis	14.9±1.1	0.010±0.002	0.26±0.03	0.04±0.03	0.217±0.020	57.3
AN23	Kapčiamiestis (co-sample)	32.6±2.2	0.017±0.002	0.24±0.03	0.07±0.03	0.168±0.022	135.8
AN24	Kapčiamiestis (co-sample 2)	9.7±0.9	0.006±0.001	0.21±0.02	0.03±0.02	0.205±0.018	46.2
AN25	Leipalingis	30.5±2.4	0.022±0.003	0.70±0.04	0.03±0.04	0.189±0.0073	43.6
AN26	Latežeris	30.4±2.4	0.008±0.001	0.44±0.05	0.02±0.05	0.184±0.009	69.1
AN27	Marcinkony	12.6±0.9	0.010±0.001	0.17±0.02	0.05±0.02	0.190±0.034	74.1
AN28	Varėna	26.2±1.6	0.017±0.002	0.54±0.02	0.03±0.02	0.198±0.007	48.5
AN29	Senoji Varėna AMS13	53.0±4.2	0.030±0.003	0.79±0.03	0.04±0.03	0.199±0.011	67.1
AN30	Eišiškės	3.3±0.3	0.002±0.001	0.05±0.01	0.04±0.01	0.195±0.038	66.0
AN31	Šalčininkai	4.4±0.3	0.004±0.001	0.13±0.02	0.03±0.03	0.199±0.021	33.8
AN32	Dieveniškės	4.0±0.9	0.003±0.001	0.08±0.01	0.04±0.01	0.210±0.036	50.0
AN33	Dieveniškės (co-sample)	33.6±2.7	0.010±0.001	0.37±0.03	0.03±0.03	0.191±0.011	90.8
AN34	Stakiai	79.7±6.4	0.021±0.002	0.53±0.03	0.04±0.03	0.187±0.010	150.4
AN35	Iglisškės	2.8±0.9	0.002±0.001	0.10±0.01	0.02±0.01	0.231±0.036	28.0
ZIL-1	Ignalina NPP peninsula	5.29±0.2	0.003±0.001	0.15±0.02	0.02±0.02	0.195±0.029	35.3
ZIL-6	Tilžė (near lake Druškščiai)	4.1±0.2	0.003±0.001	0.13±0.01	0.02±0.01	0.177±0.021	31.5

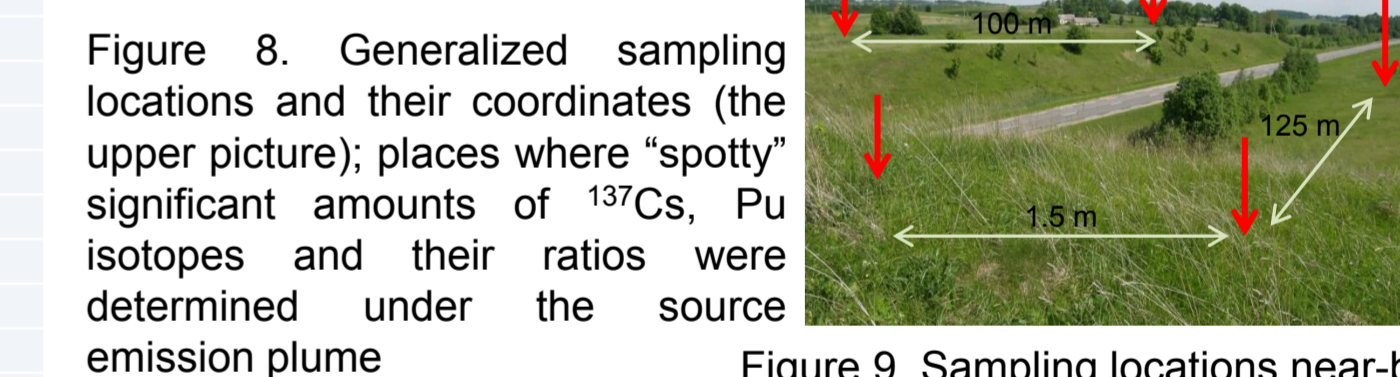
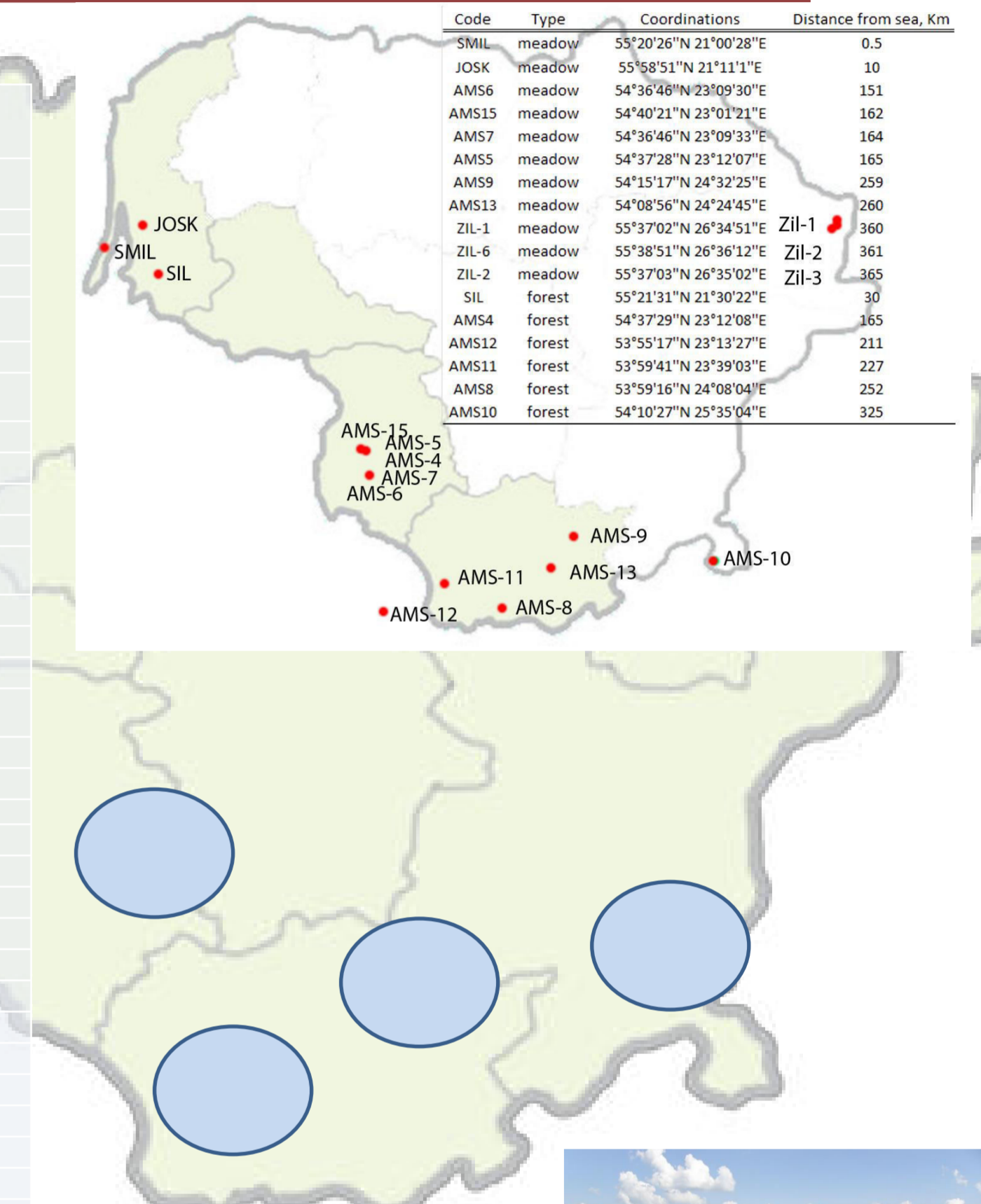


Figure 9. Sampling locations near-by

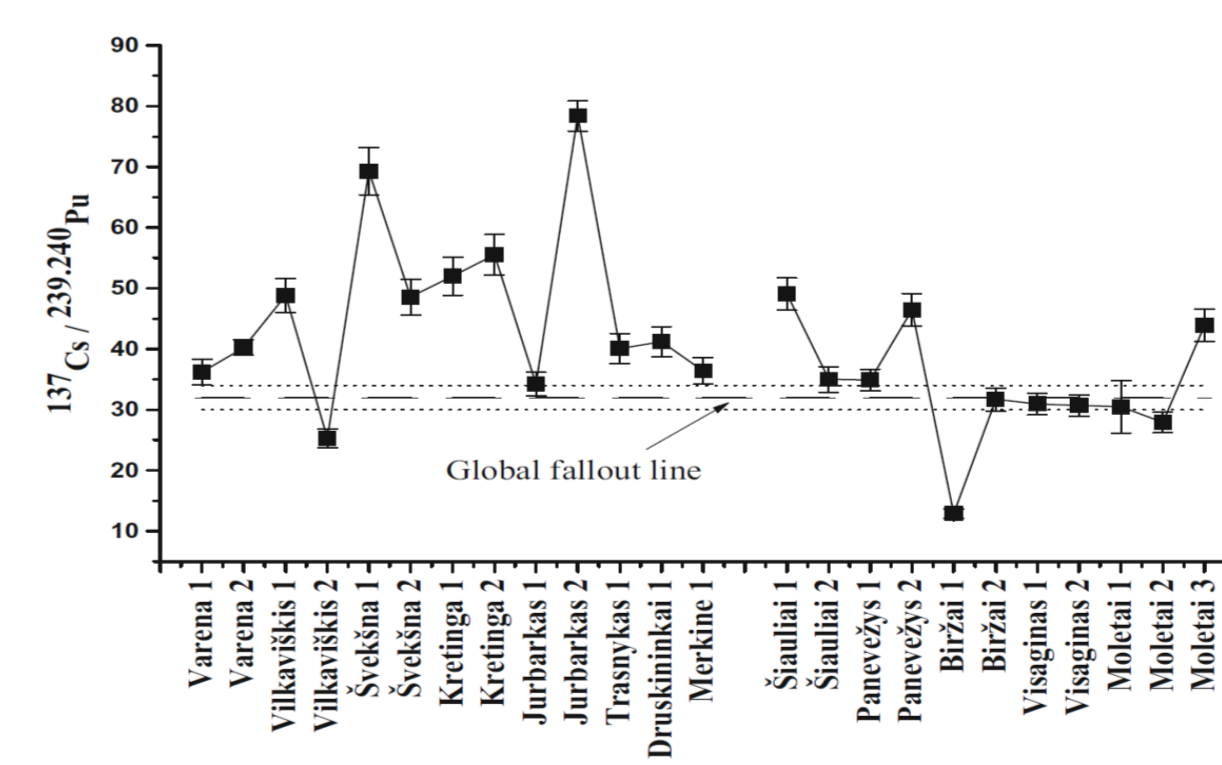


Figure 7. Activity concentration ratio  $^{137}\text{Cs}/^{239,240}\text{Pu}$  values in comparison with the global fallout line ( $32 \pm 2$  value, Luksiene et al. 2015)

## Emission source: local vs. alobal

$$A_{\%} = \frac{A_{\text{sample}} - A_{\text{global}}}{A_{\text{local}} - A_{\text{global}}} \times 100\%$$

where  $A_{\%}$  are total abundances of local emission contribution to the total amount in %,  $A_{\text{sample}}$  is a measured experimental value and  $A_{\text{local}}, A_{\text{global}}$  are typical activity and atom ratios.

## Conclusion

- $^{137}\text{Cs}$  and plutonium isotopes were mainly attributed to the global fallout from the nuclear weapons testing, with small contribution from the Chernobyl accident in southern part (up to 43%).
- In general,  $^{238}\text{Pu}/^{239,240}\text{Pu}$  activity and  $^{240}\text{Pu}/^{239}\text{Pu}$  atomic ratios did not differ significantly in undisturbed meadow and forest soil samples, although higher plutonium concentrations were observed in forest soil samples.
- Significant inhomogeneous plutonium isotopic and concentration distributions were observed during the experimental data analysis in certain sampling points, although local precipitation was absent during these days because of the anticyclone conditions, possibly due to different surface patterns.