The activity and atomic ratios of long-lived radionuclides are useful parameters for identifying nuclear source origin because their values depend on their generation processes. Among them $^{129}$I and $^{239,240}$Pu are the key radionuclides for prediction of the sources of radioactive contamination.

The activity ratio of Pu isotopes enables one to distinguish the global fallout from nuclear weapons testing, the Chernobyl fallout or other sources. Iodine-129 is a naturally occurring long-lived radionuclide of iodine (15.7 Myr) naturally formed by the cosmic ray reaction with Xe and fission of $^{238}$U and due to the releases from nuclear fuel reprocessing plants, and is a fresh ‘fingerprint’ of nuclear weapons testing.

The aim of this work is to distinguish the possible sources of radioactive contamination by measuring the activity concentrations of iodine-129 and plutonium radionuclides in the soil samples collected.

### Measurements

Three different techniques for measurements of soil samples were used:

- accelerator mass spectrometry - for iodine-129,
- inductively coupled plasma mass spectrometry - for iodine-129, plutonium-239 and plutonium-240,
- alpha spectrometry - for plutonium-238 and plutonium-239,240 determination.

### Result

The concentration of $^{129}$I in the soil samples was (2.01 - 0.21) × 10$^7$ atoms g$^{-1}$, whereas the concentration of $^{131}$I in the forest soil varied (4.72 ± 0.377 - 26.16 ± 0.56) × 10$^6$ atoms g$^{-1}$. An extremely high concentration of 34.16 ± 5.65 × 10$^6$ atoms g$^{-1}$ for $^{129}$I was obtained in sample S1L taken at a location 30 km from the Baltic Sea in depositional forest.

The two European reprocessing plants were transported along the European coast in the North Sea and in this work, the Kattegat and Danish Strait, a high $^{129}$I level in the Baltic Sea water has been reported.

### A source defined by iodine-129 isotope

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

$^{238}$Pu/$^{239}$Pu activity ratios and $^{238}$Pu/$^{239}$Pu atomic ratios were calculated to be 0.02 ± 0.18 and 0.18 - 0.24, respectively. In meadow soil, a relative lower $^{238}$Pu activity concentration of 0.01 ± 0.05 Bq kg$^{-1}$ and $^{238}$Pu/$^{239}$Pu concentration of 0.07 - 0.53 Bq kg$^{-1}$ were measured, whereas a higher activity concentrations were determined in forest soils, varying in 0.05 - 0.09 Bq kg$^{-1}$ of $^{239}$Pu and 0.74 - 1.80 Bq kg$^{-1}$ of $^{238}$Pu, $^{239}$Pu/$^{238}$Pu activity and $^{239}$Pu/$^{238}$Pu atomic ratios did not show any significant difference in undisturbed meadow and forest soil samples. Elevated $^{239}$Pu/$^{238}$Pu activity and $^{239}$Pu/$^{238}$Pu atomic ratios up to 0.18 and up to 0.24, respectively, in AMS4, AMS6 (southern part of Lithuania), and in AMS10 (southeastern part) areal points might be attributed to the deposition of the Chernobyl accident.

### No correlation between $^{129}$I and plutonium isotopes!

In addition, no correlation between $^{129}$I and plutonium isotopes was observed (Fig. 3).

The measured $^{129/131}$I ratio in most of the samples showed that more than 95% of $^{129}$I originated from the global fallout which included nuclear weapon testing, reprocessing facilities, etc. Nevertheless, there were a few exceptions, e.g. the sampling point at 265 km, where 10% of $^{129}$I originated from the global fallout and the rest — of the Chernobyl fallout.

### Conclusion

1. It was revealed that $^{129/131}$I atomic ratio decreased significantly distancing away from the sea deeper into the country, drawing an atomic ratio baseline for assessment of possible nuclear contamination in future.

2. The relative higher $^{131}$I concentration in meadow and forest soil might be attributed to their better vegetation coverage of soil surface, atmospheric deposition iodine (dry deposited and washout of rainfall) can be better retained in the soil without significant loss by blowing out by wind or wash out by water.

3. Plutonium isotopes are mainly attributed to the global fallout from the nuclear weapons testing, with small contribution from the Chernobyl accident, causing a relative homogeneous distribution.

4. While $^{239}$Pu/$^{238}$Pu activity and $^{239}$Pu/$^{238}$Pu atomic ratios did not show any significant difference in undisturbed meadow and forest soil samples, although higher plutonium concentrations were observed in forest soil samples.

5. The reconstructed $^{129/131}$I atomic ratios in most of the soil samples shows that more than 95% of $^{129}$I originated from the fallout, while in the southern part of Lithuania there are some locations indicating only 10% of $^{129}$I originated from the global fallout.

### References