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Radionuclide Isotopic Ratio Analysis at Ultra-Trace Concentration Levels for CTBT Applications

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CENTER FOR PHYSICAL SCIENCES AND TECHNOLOGY

INTRODUCTION AND MAIN RESULTS

Actinide isotopic composition forensics technology at ultra-trace concentration level.

$^{137}Cs/^{239+240}Pu$, $^{240}Pu/^{239}Pu$ ratios reveal nuclear contamination at sub ppt concentration level.

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CTBT: SCIENCE AND TECHNOLOGY CONFERENCE
8 SEPTEMBER ONLINE DAY
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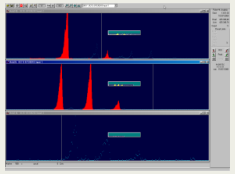
Measurement Techniques and Protocols

Three different techniques for measurements of soil samples:

- alpha spectrometry - for plutonium-238 and plutonium-239,240 determination



- inductively coupled plasma mass spectrometry (ICP-MS) paired with Apex IR module - for plutonium-239 and plutonium-240

- gamma-spectrometric system – for caesium-137

Isotopic Measurements

Three sample set was taken. Time used to collect samples and to start samples preparation and measurement – 24 h.

Caesium-137: soil samples were prepared in plastic containers of the standard geometry for gamma spectrometry measurement. The reference standards with radionuclides (^{57}Co, ^{139}Ce, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{54}Mn and ^{65}Zn) free of the coincidence-summing effects were used for the efficiency calibration. The gamma spectrometer was equipped with the HPGe coaxial gamma-X-ray detector (GMX-series with a 0.5 mm thickness Be window) made by Ortec (USA). The relative efficiency of the detector was 33 %, the energy resolution at 1332.5 keV was 1.8 keV. All samples were counted for the fixed time of 50,000 s. The detection limit for ^{137}Cs is 0.185 Bq. Additionally, activity concentration of ^{37}Cs was measured in the IAEA-TEL-2014-03 reference soil sample for analytical quality control. **Work total – 3 x 13 h of measurement = 36 h.**

Plutonium was extracted by acid leaching from the soil matrix and purified using the anion-exchange (Bio-Rad AG-1×8 100-200 mesh, Bio Rad Company) method followed by the extraction chromatography technique (double step separation protocol). Initially, samples were dried at 500°C for 2h, then left for 12h in 700°C to burn organic part of soil. The analytical procedure has been reported in [1]. **Work total - 14h of drying and ashing, 3 x 1.5h preparation, 25h of alpha- and 1h of ICP-MS measurements = 44.5 h.**

Results

Sampling locations in Lithuania

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Results

Sample no.	^{137}Cs, Bq/kg	$^{239+240}$Pu, Bq/kg	^{238}Pu/$^{239+240}$Pu	^{240}Pu/^{239}Pu (k=2)	^{137}Cs/$^{239+240}$Pu
1	23.0±1.8	0.45±0.04	0.02±0.04	0.195±0.012	51.1
2	12.5±1.0	0.48±0.05	0.03±0.04	0.187±0.016	26.0
3	148.8±11.8	1.40±0.03	0.18±0.03	0.243±0.002	106.3

CONCLUSIONS

1. The ICP-MS technique enables significantly faster measurement of ^{239}Pu and ^{240}Pu isotopes compared to alpha spectrometry.
2. Using the APEX IR sample introduction system for ICP-MS stabilizes plutonium isotopic signals by 67%, allowing for shorter analysis times.
3. A standard error of up to 8.5% for the ^{240}Pu/^{239}Pu isotopic ratio was achieved using single-detector ICP-MS (limit of detection for ^{239}Pu and ^{240}Pu was 5 ppq).
4. It has been demonstrated that rapid radionuclide analysis can be completed in less than 48 hours (plus 24 hours for sampling).
5. Based on the activity and atomic ratios of ^{137}Cs/$^{239+240}$Pu, ^{238}Pu/$^{239+240}$Pu, and ^{240}Pu/^{239}Pu, it was determined that the predominant sources are global fallout and the Chernobyl accident.

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