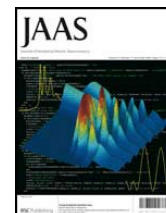


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**Optimisation and application of ICP-MS and alpha-spectrometry for determination of isotopic ratios of depleted uranium and plutonium in samples collected in Kosovo****Sergei F. Boulyga, Corrado Testa, Donatella Desideri and J. Sabine Becker**

The determination of environmental contamination with natural and artificial actinide isotopes and evaluation of their source requires precise isotopic determination of actinides, above all uranium and plutonium. This can be achieved by alpha spectrometry or by inductively coupled plasma mass spectrometry (ICP-MS) after chemical separation of actinides. The performance of a sector-field ICP-MS (ICP-SFMS) coupled to a low-flow micronebulizer with a membrane desolvation unit, "Aridus", was studied with respect to precise isotopic measurements of uranium and plutonium at the ultratrace level. The UH<sup>+</sup>/U<sup>+</sup> formation rate was about  $3 \times 10^{-5}$  and a sensitivity for <sup>238</sup>U of up to  $4 \times 10^9$  counts s<sup>-1</sup> ppm<sup>-1</sup> was achieved. The limit of quantification (LOQ, 10s) for <sup>236</sup>U and <sup>239</sup>Pu using the experimental arrangement described above was 0.6 pg l<sup>-1</sup> in aqueous solution and 0.13 pg g<sup>-1</sup> in soil, respectively. ICP-SFMS was used in comparison to alpha spectrometry to measure the U and Pu concentrations and isotopic compositions in two soil samples and in one penetrator collected in Kosovo. ICP-MS permitted the determination of U and Pu isotope ratios including the <sup>236</sup>U abundance and <sup>240</sup>Pu/<sup>239</sup>Pu ratio at ultratrace levels in soil samples of up to 0.1 g. Depleted uranium (<sup>235</sup>U/<sup>238</sup>U = 0.002 02 ± 0.000 01) was determined in one penetrator and one soil sample. Pu concentrations of  $(5.5 \pm 1.1) \times 10^{-13}$  g g<sup>-1</sup> and  $(4.4 \pm 0.5) \times 10^{-13}$  g g<sup>-1</sup> (<sup>240</sup>Pu/<sup>239</sup>Pu = 0.35 ± 0.10 and 0.27 ± 0.07, respectively) were found in both soil samples from Kosovo. Besides plutonium, <sup>236</sup>U ( $3.1 \times 10^{-5}$  g g<sup>-1</sup>) and <sup>241</sup>Am ( $1.7 \times 10^{-12}$  g g<sup>-1</sup>) were also detected in the penetrator sample, which indicates the previous existence of neutron-related processes and points to a possible presence of spent reactor uranium in munitions. However, the most probable plutonium contamination sources in analyzed soil samples from Kosovo are mixed fallout including spent reactor fuel due to the Chernobyl nuclear power plant accident in 1986 and plutonium due to nuclear weapon tests. Additional plutonium contamination could not be determined in the Kosovo soil sample containing depleted uranium with a detection limit of about 10<sup>-13</sup> g g<sup>-1</sup>.

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