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Method development for the determination of uranium isotope ratios by MC-ICP-MS

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Accurately determined uranium isotope ratios are essential for geochronological dating and tracing of different environmental processes. In addition, they are of paramount importance for nuclear safeguards and nuclear forensics.

Multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) has become an important tool for determination of uranium isotope ratios in both liquid and solid samples. Its features are better accuracy and precision compared to alpha spectrometry, which is comparable to thermal ionization mass spectrometry while maintaining higher sample throughput. This makes it an ideal tool for many applications.

The objective of this study was to develop, validate and verify accurate analytical method for the determination of uranium isotope ratios ($^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$) with correctly estimated measurement uncertainty. Measurements were carried out with Nu plasma II, (Nu instruments Ltd, UK) MC-ICP-MS with the high-efficiency sample introduction system Aridus II TM (Cetac Technologies, NE, USA). At first, routine optimization and calibration of the MC-ICP-MS was performed. Then the appropriate aliquot concentration of purified uranium fractions was selected from several different concentrations of a uranium standard of known isotopic composition. Instrument mass bias was corrected with external standard –sample –standard bracketing technique.

The developed method was applied for analysis of uranium isotope ratios in uranium fractionations obtained from water samples of karstic aquifer (Ljubljanica catchment). After pre-concentration, uranium precipitate was further purified on UTEVA column and then the organic composition of resin was destroyed by using concentrate HNO_3 and H_2O_2 . Before measuring the proper aliquot of uranium isotope ratio on MC-ICP-MS, the exact concentration of uranium in sample was determined by two different methods, by radiochemical neutron activation analysis (RNAA) and by inductively coupled plasma mass spectrometry (ICP MS). The comparison of this two different methods was also performed.

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