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### Multi-Ion-Counting-ICP-MS technique for precise determination of Uranium and Thorium series isotopes

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Most likely multi-ion-counting is the most suitable technique for the precise determination of low intensity U- and Th-series isotopes.

We developed a method for the measurement of  $^{234}\text{U}/^{235}\text{U}$  ratios on two, respectively three, multipliers using a ThermoFinnigan "AXIOM" MC-ICP-MS. The instrument was tuned to one amu spacing between multipliers. A  $^{233}\text{U}/^{236}\text{U}$  double spike was applied for online internal standardisation, mass bias correction and multiplier cross-calibration making isotope ratio determination independent on changing mass fractionation and signal fluctuations due to plasma source instabilities. The  $2\sigma$  internal precisions of better than 1 ‰ are reached in less than 30 minutes consuming about 5 ng of total U. Repeated measurements of uranium CRM 112A (NBL960) were performed to test the reproducibility and accuracy of the method. An average  $^{234}\text{U}/^{238}\text{U}$  value of  $(5.2858 \pm 0.0009) \cdot 10^{-5}$  was determined in accordance with a previously published value of  $(5.2860 \pm 0.0095) \cdot 10^{-5}$  [1].

In combination with the high sensitivity (20 GHz/ppm) and low detection limits (approx. 0.01 fg) for  $^{230}\text{Th}$  measurements on a MC-ICP-MS this method allows precise U/Th age determination.

Furthermore, the multi-ion-counting technique allows precise determination of  $^{231}\text{Pa}/^{233}\text{Pa}$ - and  $^{226}\text{Ra}/^{228}\text{Ra}$ -ratios. Both isotope pairs being important geochronological tracers. On seawater samples from the SE Atlantic  $^{231}\text{Pa}$  concentrations were determined from 4 to 10 l with an external precision in between 2 to 5 % ( $2\sigma$ ). Ra-isotope measurements via MC-ICP-MS are shown to be slightly less precise than performed by TIMS. However, MC-ICP-MS data acquisition is faster (10 min) and more robust.

It can also be shown that the low concentrations of  $^{227}\text{Ac}$  (half-life: 21.77 years), being a promising proxy for ocean upwelling and ocean circulation, can be determined within a statistical uncertainty of about 5% in 10 to 20 l of seawater. This is four times more precise than with the usual alpha counting technique.

#### Reference

[1] Cheng et al. (2000) *ChemGeol* **169**, 17-33.

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### Monitoring Depleted Uranium contamination in the biosphere of Iraq using MC-ICP-MS

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Contamination of the biosphere with artificial long-lived radionuclides has become an important environmental issue. A potential health hazard of still unknown dimension represents the use of Depleted Uranium (DU) munitions in the military theatre in the Gulf War I and II. The release of significant quantities of uranium dust to the biosphere has a different quality as natural occurring uranium. Inhalation of small amounts of these oxidized fine particles may give rise to internal exposure to low-level, high-energy ionising radiation over many years.

We studied the uranium concentration and isotope composition of surface water and topsoil from different sites of Baghdad, Basra, and the Suweirah farming area. The samples, including also obviously contaminated surface soils, were collected from the Uranium Medical Research Centre field team after coalition operation Iraqi Freedom. Total soil samples, separated soil fine-fractions (< 150 micrometers) and evaporated water samples were leached in hot aqua regia. Uranium fractions, spiked with an enriched  $^{233}\text{U}$  tracer, were purified by ion-exchange chromatography and analysed by double-focusing multicollector ICP-MS Neptune using an Aridus desolvation introduction system. Analytical blanks were below 1 picogram. Signal sensitivity of 0.3-0.4 V ng<sup>-1</sup>ml and low  $^{235}\text{U}/^{238}\text{U}$  formation and  $^{238}\text{U}$  tailing enables detection of 1-2 femtogram  $^{236}\text{U}$  and  $^{236}\text{U}/^{238}\text{U}$  below  $1 \times 10^{-7}$ .

The uranium concentrations in the soil fine-fraction varies from about 1 to 2,600 mg/kg and is positively correlated with the  $^{238}\text{U}:^{235}\text{U}$  ratio, which ranges from 139.3 to 542.1. The  $^{234}\text{U}:^{238}\text{U}$  ratio ranges from  $7.42 \times 10^{-6}$  to  $7.05 \times 10^{-5}$  and is negatively correlated with  $^{238}\text{U}:^{235}\text{U}$ .  $^{236}\text{U}$  is present in all samples, ranging from 0.23 to 30,724 ng/kg with a  $^{236}\text{U}:^{238}\text{U}$  ratio of  $1.4 \times 10^{-7}$  to  $1.2 \times 10^{-5}$ . The latter generally correlates positively with the  $^{238}\text{U}:^{235}\text{U}$ . The data, however, also suggest the use of depleted uranium with different amounts of  $^{236}\text{U}$ . A DU signature could be found in two of six water samples (5-42 µg/l uranium,  $^{238}\text{U}:^{235}\text{U}$  of 194 to 427,  $^{236}\text{U}:^{238}\text{U}$  of  $1$  to  $2.4 \times 10^{-5}$ ). The four water samples contain only natural uranium (below 2 µg/l,  $^{238}\text{U}:^{235}\text{U}$  around 137.8) and no detectable traces of  $^{236}\text{U}$ .

Our results demonstrate a general presence of depleted uranium in superficial soils and also in two water samples from the three regions. This warrants further objective, interdisciplinary evaluation of the environmental and health impact of the uranium contamination of the biosphere in Iraq.