

## MS 14: Accelerator Mass Spectrometry and Applications 4

Time: Friday 11:00–13:00

Location: PH/HS2

## Invited Talk

MS 14.1 Fri 11:00 PH/HS2

**AMS detection of actinides at high mass separation** — ●PETER STEIER<sup>1</sup>, ROSMARIE EIGL<sup>2</sup>, JOHANNES LACHNER<sup>1</sup>, ALFRED PRILLER<sup>1</sup>, FRANCESCA QUINTO<sup>3</sup>, AYA SAKAGUCHI<sup>4</sup>, STEPHAN WINKLER<sup>1</sup>, and ROBIN GOLSER<sup>1</sup> — <sup>1</sup>University of Vienna, Faculty of Physics, Vienna, Austria — <sup>2</sup>Hiroshima University, Earth and Planetary Systems Science, Hiroshima, Japan — <sup>3</sup>Institut für Nukleare Entsorgung, KIT, Eggenstein-Leopoldshafen, Germany — <sup>4</sup>University of Tsukuba, Center for Research in Isotopes and Environmental Dynamics, Tsukuba, Japan

AMS is the mass spectrometric method with the highest abundance sensitivity, which is a prerequisite for measurement of the long-lived radioisotope <sup>236</sup>U ( $t_{1/2}=23.4$  million years). The most successful application so far is oceanography, since anthropogenic <sup>236</sup>U is present in the world oceans at <sup>236</sup>U:<sup>238</sup>U from  $10^{-11}$  to  $10^{-8}$ . We have explored methods to increase the sensitivity and thus to reduce the water volume required to 1 L or less, which significantly reduces the sampling effort. High sensitivity is also necessary to address the expected typical natural isotopic ratios on the order <sup>236</sup>U:<sup>238</sup>U =  $10^{-13}$ , with potential applications in geology.

With a second 90° analyzer magnet and a new Time-of-Flight beam line, VERA is robust against chemical impurities in the background, which e.g. allows measuring Pu isotopes directly in a uranium matrix. This simplifies chemical sample preparation for actinide detection, and may illustrate why AMS reaches lower detection limits than other mass spectrometric methods with nominally higher detection efficiency.

MS 14.2 Fri 11:30 PH/HS2

**Determination of Actinides in Pacific Ocean Water by AMS with Respect to the Fukushima Accident** — ●KARIN HAIN<sup>1</sup>, BOYANA DENEVA<sup>1</sup>, THOMAS FAESTERMANN<sup>1</sup>, NICOLAI FAMULOK<sup>1</sup>, LETICIA FIMIANI<sup>1</sup>, JOSÉ M. GÓMEZ-GUZMÁN<sup>1</sup>, GUNTHER KORSCHINEK<sup>1</sup>, FLORIAN KORTMANN<sup>2</sup>, CHRISTOPH LIERSE V. GOSTOMSKI<sup>2</sup>, and PETER LUDWIG<sup>1</sup> — <sup>1</sup>Physics Department, Technische Universität München, James-Frank-Str.1, 85748 Garching — <sup>2</sup>Radiochemie München, Technische Universität München, Walther-Meißner-Str.3, 85748 Garching

A chemical separation procedure for plutonium (Pu) and neptunium (Np) was developed using extraction chromatography to determine their amounts and isotopic ratios in sea water. <sup>241</sup>Am, which causes isobaric background on <sup>241</sup>Pu in mass-spectrometric measurements, was successfully separated from the Pu fraction by this method. Water samples which were spiked with <sup>242</sup>Pu and <sup>237</sup>Np or <sup>239</sup>Np, respectively, were used for chemical yield determination. Chemical yields of Pu and Np, which were determined by alpha and gamma spectrometry at the Radiochemie München, of more than 85% were obtained. The developed method was applied to analyze the amount of Pu and Np in the certified reference material, IAEA-443, by accelerator mass spectrometry (AMS) at the Maier-Leibnitz-Laboratory to check the applicability of the method. The amounts of <sup>240</sup>Pu, <sup>241</sup>Pu and <sup>237</sup>Np obtained in this study are in agreement with the literature values within the uncertainties. This method can be used for the analysis of Pu and Np in Pacific Ocean water samples collected after the Fukushima accident.

MS 14.3 Fri 11:45 PH/HS2

**Determination of plutonium and uranium in environmental samples from Fukushima** — ●STEPHANIE SCHNEIDER<sup>1</sup>, MARCUS CHRISTL<sup>2</sup>, GEORG STEINHAUSER<sup>3</sup>, and CLEMENS WALTHER<sup>1</sup> — <sup>1</sup>Institut für Radioökologie und Strahlenschutz, Leibniz Universität Hannover, Hannover — <sup>2</sup>Laboratory of Ion Beam Physics, ETH Zürich, Switzerland — <sup>3</sup>Department of Environmental and Radiological Health Sciences, Colorado State University, United States of America

During the Fukushima accident significant amounts of radioactive nuclides were released into the atmosphere, including small amounts of actinides. Previous investigations showed a strong localization of plutonium, which indicates a particulate release of this element from the NPP. As plutonium is of a high radiological relevance if incorporated, further investigations on vegetation, litter and soil samples taken at 8 different sampling sites in the vicinity of the damaged reactor were conducted. Accelerator mass spectrometry is a powerful tool to determine

even lowest concentrations. For these measurements the samples were first leached and then uranium and plutonium were chemically separated using extraction chromatography. Here we use the <sup>236</sup>U/<sup>238</sup>U and the <sup>240</sup>Pu/<sup>239</sup>Pu isotopic ratios as indicators to distinguish between global fallout and releases from a nuclear reactor.

MS 14.4 Fri 12:00 PH/HS2

**I-129 in der Atmosphäre auf der Zugspitze** — ●ABDELOUAHED DARAOUI<sup>1</sup>, CLEMENS WALTHER<sup>1</sup>, CHRISTOF VOCKENHUBER<sup>2</sup> and HANS-ARNO SYNAL<sup>2</sup> — <sup>1</sup>Institut für Radioökologie und Strahlenschutz (Uni Hannover), Hannover, Deutschland — <sup>2</sup>Labor für Ionenstrahlphysik (ETH), Zürich, Schweiz

Der Transport von Radionukliden über die Luft und das Meer kennt keine Grenzen. So kann das Iod-129 aus den Wiederaufbereitungsanlagen Sellafield in England und La Hague in Frankreich in der Atmosphäre bei der Zugspitze bestimmt werden. In der Umweltforschungstation Schneefernerhaus (2650 m ü. NN) und auf der Zugspitze Gipfel (2962 m ü. NN) werden Iod-127 und Iod-129 in der Atmosphäre gemessen. Es wurden die Schnee Proben zwischen 2012 und 2014 und eine wöchentliche Aerosolproben zwischen August und Oktober 2014 gesammelt. Nach der Iod-Extraktion durch eine Anionenaustausch-Methode für Schnee Proben und Lösungsmittel-extraktion für Aerosolfilter wurde I-127 durch ICP-MS und I-129 durch Beschleuniger-massenspektrometrie (AMS) untersucht. Die ersten Ergebnisse der Einträge von I-129 im Vergleich zu I-127 zeigen eine erhebliche Variationsbreite. Der Vergleich der Messwerte von Iod-129 mit den meteorologischen Daten zeigt die Möglichkeit auf, dass die höchste Konzentration von I-129 mit Luftmassen aus Nordwesteuropa kommt, also aus Richtung der Wiederaufbereitungsanlagen. Hier wird das aktuelle Ergebnis zu überraschend hohen I-129-, und I-127-Gehalten sowie zu I-129/I-127-Isotopenverhältnissen bewertet und diskutiert.

MS 14.5 Fri 12:15 PH/HS2

**Die Verteilung von <sup>236</sup>U/<sup>238</sup>U im Nordatlantik und den angrenzenden Ozeanen** — ●MARCUS CHRISTL<sup>1</sup>, NURIA CASACUBERTA<sup>1</sup>, JOHANNES LACHNER<sup>1</sup>, SASCHA MAXEINER<sup>1</sup>, CHRISTOF VOCKENHUBER<sup>1</sup>, JÜRGEN HERRMANN<sup>2</sup>, MAXI CASTRILLEJO<sup>3</sup>, PERE MASQUE<sup>3</sup>, MICHIEL RUTGERS VAN DER LOEFF<sup>4</sup> und HANS-ARNO SYNAL<sup>1</sup> — <sup>1</sup>Labor für Ionenstrahlphysik, ETH Zürich, Schweiz — <sup>2</sup>Bundesamt für Seeschifffahrt und Hydrographie, Hamburg, Deutschland — <sup>3</sup>Universitat Autònoma de Barcelona, Spanien — <sup>4</sup>Alfred-Wegener-Institut, Bremerhaven, Deutschland

Die technischen Entwicklungen der letzten Jahre, wie zum Beispiel die Einführung von Helium als stripper Gas, machen kompakte AMS Systeme wahrscheinlich zu den sensitivsten verfügbaren Analysegeräten für Ultra-Spurenstoffanalysen von Aktiniden in der Umwelt. Solche Systeme eignen sich im Besonderen zur Spurenanalyse von <sup>236</sup>U, da konventionelle Massenspektrometrie hier nicht die erforderliche Unterdrückung des durch Moleküle oder Nachbarmassen verursachten Untergrundes bietet.

Am AMS System *Tandy* der ETH Zürich wurden während der letzten Jahre einzigartige Datensätze von <sup>236</sup>U/<sup>238</sup>U im Atlantischen und Arktischen Ozean, sowie im Mittelmeer und der Nordsee produziert.

In diesem Vortrag soll zunächst ein kurzer Überblick über die neuesten technischen Entwicklungen im Bezug auf Schwerionen Analysen an kompakten AMS Anlagen der ETH Zürich gegeben werden. Im zweiten Teil werden dann die produzierten Datensätze zusammengefügt und das gegenwärtige Verständnis dieses neuen Ozean-Tracers diskutiert.

MS 14.6 Fri 12:30 PH/HS2

**AMS with <sup>93</sup>Zr** — ●BOYANA DENEVA, THOMAS FAESTERMANN, LETICIA FIMIANI, JOSÉ GÓMEZ GUZMÁN, KARIN HAIN, PETER LUDWIG, and GUNTHER KORSCHINEK — Technische Universität München, Physik Department, Garching

The isotope <sup>93</sup>Zr with its long half life of  $1.6 \times 10^6$  years is produced in a nuclear reactor as a fission product or by neutron capture from the stable <sup>92</sup>Zr in the reactor cladding. It is thus the most important zirconium isotope considering spent nuclear fuel storage and disposal assessment on long terms. The quantitative detection of <sup>93</sup>Zr allows also a measurement of <sup>92</sup>Zr ( $n, \gamma$ ) <sup>93</sup>Zr reaction cross section relevant for the nucleosynthesis s-process, and to search for traces of <sup>93</sup>Zr from interstellar material in exclusive deposits or for hydrological and ra-

radioactive waste tracing. One of the difficulties in identifying  $^{93}\text{Zr}$  by AMS is the isobaric interference of the stable  $^{93}\text{Nb}$ . The adequate separation of these isobars is our goal in the ongoing measurements. In order to investigate the possibility of using the energy loss in a passive absorber we carried out stopping power measurements of  $^{92}\text{Zr}$  and  $^{94}\text{Zr}$  to estimate that of  $^{93}\text{Zr}$ . An experimental determination of the stopping power of  $^{93}\text{Nb}$  is performed by using silicon nitride passive absorbers of different thicknesses. The results are compared with SRIM simulations and the estimations for  $^{93}\text{Zr}$  to understand the behavior of  $^{93}\text{Zr}$  ions in a  $\text{Si}_3\text{N}_4$  passive absorber. This is crucial for the developments and optimizations of an AMS set up for the detection of  $^{93}\text{Zr}$ .

MS 14.7 Fri 12:45 PH/HS2

**AMS of actinides in ground- and seawater: a new procedure for simultaneous analysis of U, Np, Pu, Am and Cm isotopes below ppq levels** — ●FRANCESCA QUINTO<sup>1</sup>, MARKUS LAGOS<sup>1</sup>, MARKUS PLASCHKE<sup>1</sup>, THORSTEN SCHÄFER<sup>1</sup>, PETER STEIER<sup>2</sup>, and HORST GECKEIS<sup>1</sup> — <sup>1</sup>Institut für Nukleare Entsorgung, KIT, Eggenstein-Leopoldshafen, Germany — <sup>2</sup>VERA Laboratory, Univer-

sity of Vienna, Vienna, Austria

U-236, Np-237, Pu isotopes and Am-243 were determined in ground- and seawater samples at levels below ppq with a maximum sample size of 0.250 l. Such high sensitivity measurement was possible by using accelerator mass spectrometry (AMS) with an improved gas stripping and an additional high resolving magnet. The use of non-isotopic tracers was investigated in order to allow the determination of those nuclides, namely Np-237 and Am-243, for which isotopic tracers for mass spectrometry are rarely available. The actinides were concentrated from the sample matrix via iron hydroxide co-precipitation and measured sequentially without previous chemical separation from each other. The analytical method was validated with the analysis of IAEA 443 seawater Reference Material and applied to background samples from the Colloid Formation and Migration project at the Grimsel Test Site and to sea- and freshwater samples affected solely by global fallout. The sensitivity of the presented analytical method provides the capability to study the long-term release of actinide tracers in field experiments as well as the transport of actinides in a variety of environmental systems.