MS 11: SIMS / Accelerator Mass Spectrometry and Applications 4

Time: Friday 10:30–12:30 Location: DO24 1.205

Invited Talk MS 11.1 Fri 10:30 DO24 1.205

Environmental sample analysis by SIMS in the search for undeclared nuclear activities — •Magnus Hedberg — European Commission, Joint Research Centre (JRC), Institute for Transuranium Elements, P.O. Box 2340, D-76125 Karlsruhe, Germany

When uranium is processed in industrial quantities, it is very difficult to avoid the release of micron to submicron-sized aerosol particles containing the isotopic signature of the handled materials to the immediate environment. This allows nuclear safeguards authorities to monitor the used nuclear materials by analysing dust samples from the facilities. Until recently, particle analyses have predominantly been performed by SIMS using the small geometry CAMECA IMS 3F-7F instruments. The performance of these instruments is however limited for theses samples by the occurrence of isobaric interferences that cannot be resolved without compromising the transmission. A recent breakthrough to solve this problem has been the implementation of Large Geometry (LG)-SIMS, mainly by the CAMECA IMS 1280HR. These instruments are like the small geometry SIMS, based on a double focusing mass spectrometer, but with the implementation of a large-radius magnetic sector and improved secondary ion optics. In short, the LG-SIMS today provides isotopic data on particle analysis in safeguards applications that are at a state of the art level in a timely way. The latter is important for the safeguards application, where timely analysis can be critical. A short introduction to nuclear safeguards and forensics analysis are given together with a presentation of how LG-SIMS is used for safeguards analysis.

MS 11.2 Fri 11:00 DO24 1.205

AMS measurements of global fallout U-236 and Pu in an ombrotrophic peat profile: evidence for their post depositional migration — •Francesca Quinto[1], Erich Hrnecek[1], Michael Krachler[1], William Shotyk[2], Peter Steier[3], Stephan Winkler[3], and Robin Golser[3] — [1]European Commission Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany — [2]Department of Renewable Resources, University of Alberta, 839 General Services Building, Edmonton, AB, Canada T6G 2H1 — [3]VERA Laboratory, Faculty of Physics, University of Vienna, Währinger Straße 17, A-1090 Vienna, Austria

U-236, Pu-239, Pu-240, Pu-241 and Pu-242 were analysed in an ombrotrophic peat core representing the last 80 years of atmospheric deposition. The determination of these isotopes at femtogram and attogram levels was possible by using ultra-clean laboratory procedures and accelerator mass spectrometry. Since the Pu isotopic composition characteristic for global fallout, as well as anthropogenic U-236, were identified in peat samples pre-dating the period of atmospheric atom bomb testing, migration of Pu and U within the peat profile is clearly indicated. The vertical profile of the U-236/U-238 isotopic ratio represents the first observation of the U-236 bomb peak in a terrestrial environment. Comparing the abundances of the global fallout derived U-236 and Pu-239 along the peat core, the post depositional migration of plutonium exceeds that of uranium. These results highlight, for the first time, the mobility of Pu and U in a peat bog with implications for their migration in other acidic, organic rich environments.

MS 11.3 Fri 11:15 DO24 1.205

AMS of I-129: cross contamination and its correction — •Christof Vockenhuber — ETH Zurich, Labor für Ionenstrahlphysik, Zürich, Schweiz

Low-energy AMS is well suited for measurements of the long-lived nuclide ^{129}I because the interfering stable isobar ^{129}Xe does not form negative ions, thus high ion energies are not required for discrimination in the final detector. Furthermore, low-energy AMS has the advantage that in combination with helium stripping the most probable charge state can be selected; in our case at the TANDY running at 300 kV we select charge state 2+ with a transmission of $> 50\%$. With a proper spectrometer at the high-energy side interferences of the stable isotope ^{127}I can be completely eliminated.

Contrary to many AMS nuclides ^{129}I readily forms negative ions and the overall efficiency is high. The challenges lie more in the ion source where cross contamination can be quite severe due to the volatile nature of iodine. This is particularly of importance when analyzing samples that are influenced from anthropogenic sources because the isotopic ratios can span several orders of magnitude. On the other hand special care must be taken when analyzing samples with low isotopic ratios (^{129}I/^{127}I $< 10^{-13}$) or samples with very low iodine content (carrier free samples) due to the very same reason.

This talk will discuss the advantages and challenges of low-energy AMS of I-129 with the focus on the issues with cross contamination and its correction.

MS 11.4 Fri 11:30 DO24 1.205

Anwendung von AMS zur Bestimmung von Iod-129 in Bodenprofilen in der Nord- und Südhalbkugel — •Abdelouahed Daraoui[1], Mareike Schwinger[1], Beate Riebe[1], Christof Vockenhuber[2], Hans-Arno Synal[2] und Clemens Walther[1] — [1]Institut für Radioökologie und Stahlenschutz, Uni Hannover, Deutschland — [2]Labor für Ionenstrahlphysik, Zürich, Schweiz

Die Ausbreitung von I-129 in der Umwelt ist in den letzten Jahrzehnten verstärkt in den Fokus der Wissenschaft gerückt. Das Gleichgewicht zwischen dem stabilen I-127 und dem I-129 ist aufgrund der kontinuierlichen Freisetzung von I-129 durch die Wiederaufarbeitungsanlagen (Sellafield und La Hague), aber auch durch in der Vergangenheit durchgeführte Atomwaffentests sowie durch nukleare Unfälle (Chernobyl und Fukushima) stark verändert. Der Iod-Gehalt im Boden ist das Ergebnis des Eintrags von Iod aus der Atmosphäre über nasse und trockene Deposition. Das Migrationsverhalten von I-129 im Boden wird unter anderem durch seine Wechselwirkung mit Metalloxiden sowie organischen Bestandteilen bestimmt. AMS ist eine sensitive Methode für die Bestimmung von I-129. Die Nachweisgrenze für das I-129/I-127 liegt im Bereich von 1E-14. Damit ist die AMS für die Untersuchung von I-129 sowohl in pränuklearen wie auch in kontaminierten Umweltproben geeignet. In dieser Arbeit berichten wir über die Untersuchung der Deposition und des Migrationsverhaltens von I-127 und I-129 in Böden der Nordhalbkugel (Deutschland, Ukraine) und der Südhalbkugel (Japan, Chile). Auf diese Weise ist es möglich, Rückschlüsse bezüglich der jeweiligen Kontaminationsquellen zu ziehen.

MS 11.5 Fri 11:45 DO24 1.205

^{236}U and ^{129}I as tracers of water masses in the Arctic Ocean — •Núria Casacuberta[1], Marcus Christl[1], Christof Vockenhuber[1], Clemens Walther[2], Michiel Van-der-Loeff[3], Pere Masqué[4], and Hans-Arno Synal[1] — [1]Laboratory of Ion Beam Physics, ETH-Zurich, Switzerland — [2]Institut für Radioökologie und Strahlenschutz, Leibniz Universität Hannover, Germany — [3]AWI-Geochemistry, Alfred Wegener Institut Für Polar und Meeresforshung, Bremerhaven, Germany. — [4]Institut de Ciència i Tecnologia Ambientals, Universitat Autònoma de Barcelona, Bellaterra, Spain

Recently ^{236}U attested to be a new transient oceanographic tracer: it is conservative in seawater and far from having reached steady state in the oceans. Its main sources in the North Atlantic are global fallout and European reprocessing plants. In this study, concentrations of ^{236}U and ^{129}I of eight deep profiles in the Arctic Ocean collected in 2011-2012 were determined with a compact ETH Zurich AMS system (TANDY). Results on ^{236}U/^{238}U show a steep gradient, from the lowest ever-reported ^{236}U/^{238}U atomic ratio in open ocean water (5±5) x 10^{-12} up to (3700±80) x 10^{-12}. Whereas the very low ratios are indicative for deep old waters, high ratios in shallow and surface waters show a clear signature of Atlantic Waters (AW) penetrating to the Arctic Ocean. The combination of ^{236}U with ^{129}I, both being released by the nuclear reprocessing plants of Sellafield and La Hague, with a distinct temporal input function, is used to estimate transit time of AW distributions in the Arctic Ocean.

MS 11.6 Fri 12:00 DO24 1.205

Analysis of Primordial Nuclides in High Purity Copper with Accelerator Mass Spectrometry — Nicolai Famulok, •Karin Hain, Thomas Faestermann, Leticia Fimiani, José Gomez Guzman, Peter Ludwig, Gunther Korschinek, and Stefan Schönert — Technische Universität München, Physik Department, Garching

The sensitivity of experiments in rare event physics like neutrino or direct dark matter detection crucially depends on the background level. Therefore, all material surrounding the detectors requires low contamination of radionuclides to not create additional background. A significant contribution originates from the primordial actinides thorium

and uranium and the progenies of their decay chains.

At the Maier Leibnitz Laboratorium in Munich the applicability of ultra-sensitive Accelerator Mass Spectrometry (AMS) for the direct detection of thorium and uranium impurities in a copper matrix was tested for the first time. For this special purpose, Th and U were extracted from the ion source as a copper compound. Two different samples of copper and one sample of a copper alloy were investigated. The lowest concentrations achieved with these first AMS measurements were $(1.4\pm0.6)\cdot10^{-11}$ g/g for thorium and $(7\pm4)\cdot10^{-14}$ g/g for uranium which correspond to (56 ± 16) μBq/kg and (0.9 ± 0.5) μBq/kg, respectively.

The particular requirements on the AMS technique and the developed measurement procedure will be presented, followed by a discussion of the results of the first measurements.

MS 11.7 Fri 12:15 DO24 1.205

The upgrade of VERA for natural ^{236}U - first results on detection efficiency and background — •Peter Steier, Joannes Lachner, Alfred Priller, Stephan Winkler, and Robin Golser — Universität Wien, Fakultät für Physik, VERA Labor, Währingerstraße 17, 1090 Wien, Österreich

Interest in the long-lived radioisotope ^{236}U ($t_{1/2}$=23.4 million years) has significantly increased recently, due to the emergence of environmental and earth science applications. Of the AMS instruments suited, none could achieve the sensitivity to address the expected typical natural isotopic ratios on the order ^{236}U:^{238}U = 10^{-13}. One major limitation is the relatively low total detection efficiency. Stripping with helium was shown to be advantageous at lower energies [1]. We have implemented this capability at VERA, which improved the yield by a factor of four. The second instrumental limitation is background caused by ^{235}U hydrides. With our recently installed additional 90° magnet, we can suppress this background by several orders of magnitude. Our first measurements indicate a new instrumental limit below ^{236}U:^{238}U = 10^{-14}, however, improved chemical procedures are still under development to extract such pure uranium from geological samples. Measurements of anthropogenic ^{236}U and other actinides profit from the higher sensitivity allowing for smaller samples.

[1] Vockenhuber, C., et al., The potential of He stripping in heavy ion AMS. Nucl. Instr. and Meth. in Phys. Res. B 294 (2013) 382-386.