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## ABSTRACTS

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- Stable and Radioisotope Analysis



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### Abstracts

KN 15

PROGRESS ON ISOTOPE ANALYSIS OF STABLE ISOTOPES AND RADIONUCLIDES BY ICP-MS AND LA-ICP-MS

**J. Sabine Becker**

Central Division of Analytical Chemistry, Research Centre Jülich, D-52425 Jülich, Germany; [s.becker@fz-juelich.de](mailto:s.becker@fz-juelich.de)

The significance of isotope ratio measurements by mass spectrometry has grown in the last few years due to improvement of mass spectrometers with respect to sensitivity, detection limits, precision and accuracy. [1] Because the isotope abundances of several elements are not constant and vary as a result of nuclear, biological, chemical, geochemical and physical processes isotope analysis is necessary for quite different fields of research and applications. Especially the determination of very small abundances of stable isotopes and radionuclides at the ultratrace concentration level is of increasing importance. In addition to stable isotope ratio measurements to study isotope variation in nature the analysis of long-lived radionuclides is of growing interest in quite different areas ranging from radiobioassay, environmental monitoring, decontamination and environmental remediation, health safety, nuclear waste characterization (radioactive waste control) and management of radioactive waste of high radiological toxicity for storage and disposal. [2]

Inductively coupled plasma mass spectrometry (ICP-MS) and laser ablation ICP-MS provide besides a very sensitive multielemental determination at trace and ultratrace concentration also precise analysis of stable isotopes and long-lived radionuclides, e.g., to study natural isotope variation, for characterization of environmental materials such as waters, geological, biological and medical samples, nuclear and radioactive waste materials or can be applied for geochronology. Geochronology (age dating) of geological samples is based on the decay of  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  to stable  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{206}\text{Pb}$  isotopes, respectively. In comparison to radioanalytical techniques ICP-MS and LA-ICP-MS yield an improvement with respect to smaller sample preparation steps together with improved precision by measuring the amount of accumulated decay products in small samples sizes (e.g., on single zircon grains). In addition, tracer experiments and the isotope dilution technique using enriched stable isotopes e.g., in life sciences or for speciation studies would not be thinkable without both sensitive mass spectrometric techniques.

In this lecture the state of the art, methodological and instrumental developments of ICP-MS and laser ablation ICP-MS in comparison to other mass spectrometric techniques (e.g., secondary ion mass spectrometry - SIMS, accelerator mass spectrometry - AMS, thermal ionization mass spectrometry - TIMS and glow discharge mass spectrometry - GDMS [3]) for isotope ratio measurements of stable and long-lived radionuclides in different sample types, especially in the main application fields of environmental research, life sciences and health controls or for characterization of nuclear samples, will be discussed.

[1] J.S. Becker, *J. Anal. At. Spectrom.* **20** (2005) 1173.

[2] J.S. Becker, *Intern. J. Mass Spectrom.* **242**, 185 (2005).

[3] M. Betti, *Intern. J. Mass Spectrom.* **242**, 169 (2005).

Fr01

ICP-MS AS PERFECT TOOL FOR THE CHARACTERISATION OF NUCLEAR FUEL SAMPLES?

**I. Günther-Leopold**, N. Kivel, J. Kobler Waldis, B. Wernli Paul Scherrer Institut, Department of Nuclear Energy and Safety Research, Isotope and Elemental Analysis, Villigen PSI, Switzerland, 5232 [ines.guenther@psi.ch](mailto:ines.guenther@psi.ch)

The characterisation of nuclear fuel cycle materials for long-lived radionuclides is of major importance for fuel licensing, safety studies, increased fuel enrichment for power reactors and for the storage of nuclear waste in geological repositories.

Radioactive isotopes are traditionally quantified by means of radioactivity counting techniques ( $\alpha$ ,  $\beta$ ,  $\gamma$ ). However, these methods often require extensive matrix separation and sample purification. In contrast, the low detection limits for a large number of elements combined with the ability to analyse the isotopic composition of the elements helped to establish inductively coupled plasma mass spectrometry (ICP-MS) as a powerful multi-element technique for the characterisation of nuclear materials. Especially the technique of multicollector inductively coupled plasma mass spectrometry (MC-ICP-MS) is attracting much attention in the last years because it highly increases the precision of the isotopic measurements for a wide range of elements and starts to complement the classical technique of thermal ionisation mass spectrometry.

Within the hot laboratory of the Paul Scherrer Institut ICP-MS (quadrupol and multicollector) is used since many years for the characterization of nuclear fuel samples (burnup analysis, characterisation of fresh fuel samples, determination of the isotopic vectors of fission products and actinides etc.). Hyphenations with HPLC and laser ablation were installed as complementary systems and are used extensively depending on the sample type and application. A comparison of the analytical performance of both ICP-MS techniques for the determination of the isotopic composition and the elemental concentration of different nuclides in nuclear fuel samples, the advantages and limitations, the accuracy and precision of the results and typical applications will be discussed in the presentation.

## Fr 02

### ISOTOPIC CHARACTERIZATION OF SOME ALPHA EMITTERS IN CUBAN SOIL: A PRELIMINARY EVALUATION BY Q-ICP-MS

**M.L. Cozzella**<sup>1</sup>, N. Cogliati<sup>2</sup>, M. Mariani<sup>3</sup>, R. Sibello Hernandez<sup>4</sup>, S. Spezia<sup>5</sup>, E. Trivellone<sup>3</sup>

<sup>1</sup> ENEA, Radiation Protection Institute, via Anguillarese 301, 00123 Santa Maria di Galeria (Roma), Italy.

<sup>2</sup> CESI S.p.A., via Nino Bixio 39, 29100 Piacenza, Italy.

<sup>3</sup> Politecnico di Milano, Dipartimento di Ingegneria Nucleare, CeSNEF, via Ponzio 34/3, 20133 Milano, Italy

<sup>4</sup> Centro de Estudios Ambientales de Cienfuegos, Carretera Castillo del Jagua Km 1 ½, Cienfuegos, Cuba

<sup>5</sup> Thermo Fisher Scientific, Strada Rivoltana, 20090 Rodano (MI), Italy

[letizia.cozzella@casaccia.enea.it](mailto:letizia.cozzella@casaccia.enea.it)

The determination of alpha emitters, namely of uranium, thorium and plutonium isotopes, in the soils, is a useful way in order to evaluate the radioactive fallout and to better understand the isotopic distribution profiles in different soil layers.

As the Cuba Island has never been contaminated by artificial human activities employing alpha emitters, these soils could be specifically considered as a reference soil samples for the Central American zone.

The analytical results of the radionuclidic soil contents, in particular for some isotopes of actinides, are strongly influenced by the characteristics of the samples investigated; some problems related to inhomogeneity, wide dispersion in the particles size and effects of extra low-level concentrations, were considered.

The samples of soil were treated, for a standardized time, with HNO<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> solutions following the 3050b EPA Method (“*Acid digestion of sediments, sludges and soils*”), in order

to extract by leaching the plutonium and the other radionuclides eventually absorbed on the soil surface. Plutonium analyses were performed after an anionic-chromatography purification step, in view of its very low concentration; the analyses of thorium and uranium isotopes were directly carried out on the leached solutions.

The results obtained didn't show any particular increasing in the plutonium concentration accomplished by significantly results in thorium and uranium analyses. In addition, a comparison between actinides and other transition metal ions concentrations, was evaluated in order to correlate the concentration distribution profiles for the different tracers in the examined soil layers.

All the analyses were performed separately in two labs in order to compare and to validate the obtained results. Furthermore, with the aim of checking the repeatability, reproducibility and uncertainty related to the data, the two labs performed the same analytical procedures on the N° 375 IAEA reference soil.

### Fr 03

#### MEASUREMENT OF ISOTOPE RATIOS BY ION CHROMATOGRAPHY COUPLED TO MULTICollector ICP-MS

**M. Moldovan**, S. García-Ruiz, J.A. Rodríguez-Castrillón, J.I. García-Alonso, University of Oviedo, Department of Physical and Analytical Chemistry, Julián Clavería 8, Oviedo, Spain, 33006. E-mail: [moldovanmariella@uniovi.es](mailto:moldovanmariella@uniovi.es)

In recent years, the development of multicollector ICP-MS (MC-ICP-MS) instruments has resulted in an important tool for highly precise isotope ratio measurements opening new horizons in research. Recent instrumental developments made MC-ICP-MS competitive to thermal ionization mass spectrometry (TIMS) as the reference in the field. As the ICP is perfectly suited for hyphenation to chromatographic separation techniques like gas chromatography (GC) or liquid chromatography (LC), MC-ICP-MS opens the possibility to precisely determine element isotope ratios on transient signals.

Isotope ratios of Rb, Sr, Nd, Sm and Pb could be useful tools for mineral dating and/or source identification, but in order to obtain meaningful results the chromatographic separation of Rb and Sr ( $^{87}\text{Rb}$  from  $^{87}\text{Sr}$ ), Nd and Sm ( $^{144}\text{Nd}$  from  $^{144}\text{Sm}$ ), and Pb from Cu (matrix interferences on mass bias) must be achieved first.

In the present work, the on-line coupling between a ion chromatographic system and a MC-ICP-MS instrument has been evaluated for the separation of Rb-Sr, Nd-Sm and Cu-Pb. By injecting large volumes of sample (from 1 to 5 ml) "flat-topped" chromatographic peaks can be obtained but still maintaining adequate chromatographic resolution. The measurement of isotope ratios in those flat-topped peaks by MC-ICP-MS could become a single step alternative to the traditionally long off-line measurements.

Three different application areas were studied: food authenticity (Sr isotope ratios in cider); mineral dating (Rb-Sr and Nd-Sm geochronology) and archaeology (Pb isotope ratios in Bronze Age artifacts).

### Fr 04

#### EVALUATION OF TRUENESS FOR ISOTOPIC MEASUREMENTS IN NUCLEAR PROBLEMATICS : APPLICATION TO CESIUM ISOTOPIC COMPOSITION IN NUCLEAR FUEL SAMPLES

**H. Isnard**, M. Aubert, F. Chartier, E. Ducarme, B. Tran  
CEA Saclay, DEN/DPC/SECR/LANIE, 91191 Gif sur Yvette, France.  
[helene.isnard@cea.fr](mailto:helene.isnard@cea.fr)

In the nuclear domain, precise and accurate isotopic composition determination of elements in spent nuclear fuels is mandatory to validate neutron calculation codes and for nuclear waste disposal. The trueness of the isotopic ratios is evaluated using standard reference isotopic

materials to correct for mass fractionation effects in thermal ionisation mass spectrometry (TIMS) and for mass bias effects in inductively coupled plasma mass spectrometry with multicollection (MC-ICPMS). For evaluation of trueness for Cs isotopic measurements in nuclear applications no standard reference material is available. Natural cesium is monoisotopic ( $^{133}\text{Cs}$ ) whereas artificial cesium has 4 isotopes ( $^{133}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{135}\text{Cs}$ ,  $^{137}\text{Cs}$ ). Moreover its analysis by total evaporation in TIMS is not possible due to its low evaporation temperature.

A comparison on cesium isotopic composition in spent nuclear fuels has been performed using TIMS and by MC-ICPMS techniques. By TIMS, isotopic fractionation has been evaluated studying the behaviour of the cesium isotope ratios ( $^{133}\text{Cs}/^{137}\text{Cs}$  and  $^{135}\text{Cs}/^{137}\text{Cs}$ ) against time. By MC-ICPMS technique, the mass bias effects have been corrected using external standard bracketing approach using elements (Eu, Sb) close to cesium masses. The results obtained by the two techniques show very good agreement. Relative difference on  $^{133}\text{Cs}/^{137}\text{Cs}$  and  $^{135}\text{Cs}/^{137}\text{Cs}$  ratios for two nuclear samples, analysed after radiochemical separation, is lower than 0.25%.

#### Fr 05

#### INVESTIGATION OF URANIUM OXIDE SAMPLES USING ICP-SFMS: DETERMINATION OF IMPURITIES AND ISOTOPE RATIOS

**Zs. Stefánka**, Zs. Varga and T. Bíró, Institute of Isotopes of the Hungarian Academy of Sciences, Department of Radiation Safety, Konkoly-Thege M. út 29-33. Budapest, Hungary, H-1121 e-mail: [stefanka@iki.kfki.hu](mailto:stefanka@iki.kfki.hu)

During the last decade the risk of terrorism has been increased. The smuggling of nuclear materials is a new challenge for the national forensic and/or radioanalytical laboratories. The task of the categorization and characterisation of nuclear material of unknown origin has been delegated to the Institute of Isotopes of the Hungarian Academy of Sciences in 1996 by governmental decree. One important part of the new task is the analysis of seized nuclear material, including the determination of isotopic enrichment, geometric dimension and impurities. Since the level of trace elements and the isotopic enrichment ( $^{235}\text{U}/^{238}\text{U}$ ) is depending on fabrication methods and intended use of the material. Therefore, these differences could be used for characterisation and identification the origin of unknown material.

Inductively coupled plasma mass spectrometry (ICP-MS) is ideal for the above mentioned analytical task, because it combines the possibilities of multielemental analysis and isotopic determination from a small amount of samples with high sensitivity over a wide dynamic range. Since uranium as a matrix component was found to suppress the analyte signals in ICP-MS, its removal is essential. Therefore, a matrix separation step based on off-line extraction chromatography using uranium selective resin (UTEVA, Eichrom) was used in order to improve detection limits and prevent the instrument from uranium contamination. The method is available for analysis of 19 elements (Ag, Al, As, Ba, Bi, Cd, Co, Cr, Cu, Fe, Ga, In, Li, Mn, Ni, Pb, Sr, Tl, Zn). The accuracy of the method was systematically investigated by spiking procedure using different concentrations of trace elements. The average recovery was found to be 88-115%.

Since the investigated materials are forensic samples the reduction of sample amount required for analysis has a great importance. Therefore, a direct solid sampling methods based on laser ablation assisted sample introduction has been developed and applied for  $^{235}\text{U}/^{238}\text{U}$  ratio determination. The results were compared with conventional solution nebulization ICP-MS and gamma spectrometry. Future work will concentrate on the development a laser ablation method for direct multielemental determination in uranium oxide samples.

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## FROM MARIE CURIE TO GLEN SEABORG AND BEYOND: HOW SF-ICPMS IS (IN)VALUABLE IN ENVIRONMENTAL RADIOCHEMISTRY

**Michael E. Ketterer**, Department of Chemistry and Biochemistry, Northern Arizona University, Box 5698, Flagstaff, AZ 86011-5698 USA [Michael.Ketterer@nau.edu](mailto:Michael.Ketterer@nau.edu)

Radium and plutonium are two vastly different elements. The former is naturally occurring and was discovered in uranium ore extracts by Madame Curie; the latter is almost entirely synthetic and was discovered by Glen Seaborg and co-workers in 1941. However, both share the characteristics of having at least one long-lived, alpha-emitting isotope. As such, the classical determination of the alpha-emitting Ra and Pu isotopes is by alpha spectrometry. Prior to the development of ICPMS, thermal ionization MS was used in determination of Ra, Pu, and other long-lived radioisotopes in certain specialty applications. However, ICPMS has greatly expanded the possibilities for *routine* determination of these and other long-lived radioactive elements at environmental levels.

This presentation will focus on studies of Pu in the environment, both in terms of forensic and geochemical tracing applications. Pu is globally distributed in the Earth's surface environment as a result of atmospheric weapons tests, nuclear accidents, and nuclear fuel reprocessing; this global dispersion creates new tracer opportunities for studying earth processes. Sector ICPMS offers capabilities for facile, routine, high-throughput determinations of Pu activities and atom ratios, and is pragmatic for small-mass samples containing Pu of stratospheric fallout origin. Determination of Pu by ICPMS involves dissolution, pre-concentration and purification of Pu by ion exchange or extraction chromatography, and MS determination; detection limits are 0.1-10 fg for each isotope. We have used  $^{239+240}\text{Pu}$ , along with  $^{137}\text{Cs}$ , in studies of erosion and soil redistribution, and chronostratigraphic dating of recent sediments. A new application is the use of soil  $^{239+240}\text{Pu}$  inventories and depth profiles to understand human and natural disturbances of soils containing atmospherically deposited pollutants, especially near point sources such as smelters. Pu atom ratios, and the mass spectrometric analysis of individual "hot" particles will be discussed as a method for distinguishing Pu of specific sources vs. the omnipresent stratospheric fallout "background". One current topic under development is the use of sector ICPMS to measure the  $^{244}\text{Pu}$  signature of stratospheric fallout, and its global inventories in surface soils. Preliminary ICPMS results from Northern Hemisphere soils indicate that the stratospheric fallout atom ratio  $^{244}\text{Pu}/^{239}\text{Pu}$  is about 0.0001.

In contrast to Pu, Ra is naturally occurring and the determination of long-lived  $^{226}\text{Ra}$  is of interest in environmental monitoring of "naturally occurring radioactive material", and in certain chronology applications. Despite the importance of  $^{226}\text{Ra}$  as an analyte, ICPMS is still greatly underutilized in its determination. This talk will discuss high-throughput procedures for determination of  $^{226}\text{Ra}$  in soils and sediments by total dissolution, cation exchange separation, and isotope dilution with a  $^{228}\text{Ra}$  spike. Several ongoing and potential applications of ICPMS in environmental studies of  $^{226}\text{Ra}$  will also be discussed.

The overall theme of this research is that mass spectrometry enables data to be generated with a throughput and scope to allow large-scale studies of these fascinating ultra-trace environmental constituents.

Fr06

## INVESTIGATION OF URANIUM POLLUTION IN A FORMER URANIUM MINING AREA IN HUNGARY

**Gy. Záray**<sup>1</sup>, Zs. Varga<sup>2</sup>, V. G. Mihucz<sup>3</sup>, E. Tatár<sup>1</sup>, I. Virág<sup>1</sup>, R. van Grieken<sup>4</sup>, M. Kázmér<sup>5</sup>

<sup>1</sup>Eötvös Loránd University, Department of Analytical Chemistry, Pázmány Péter sétány 1/A. H-1117 Budapest, Hungary

<sup>2</sup>Hungarian Academy of Sciences, Institute of Isotopes, Department of Radiation Safety, Konkoly-Thege Miklós 29-33. H-1121 Budapest, Hungary

<sup>3</sup>Hungarian Satellite Centre of Trace Elements Institute to UNESCO, Pázmány Péter sétány 1/A. H-1117 Budapest, Hungary

<sup>4</sup>University of Antwerp, Department of Chemistry, Micro- and Trace Analysis Centre (MiTAC) Campus Drie Eiken, Universiteitplein 1, BE-2610 Antwerp-Wilrijk, Belgium

<sup>5</sup>Eötvös Loránd University, Department of Palaeontology, Pázmány Péter sétány 1/C. H-1117 Budapest, Hungary [zaray@ludens.elte.hu](mailto:zaray@ludens.elte.hu)

The search for natural sources of nuclear raw materials in Hungary started in 1953 under Soviet supervision. One year later, a significant accumulation of uranium ore was discovered in the southwestern part of Hungary, in the village of Kővágószőlős. The uranium mine, established in 1954, was closed in 1990 and the recultivation process started in 1995. During this process, the tailings were covered with loess soil layer of 1.0 – 1.5 m thickness and various tree species were planted. In order to evaluate the extent of uranium contamination in the territory of the abandoned mine, soil (sampling depth 0-0.3 m) and leaf samples of *Robinia pseudoacacia*, *Eleagnus angustifolia*, *Quercus pubescens*, *Populus alba* and *Populus × canescens*, planted in 2002, were taken from the recultivated tailings and analysed by inductively coupled plasma sector field mass spectrometry (ICP-SFMS) after microwave-assisted digestion. Moreover, leaf samples and tree ring of a *Populus × canescens*, grown spontaneously on one of the tailings, which was not covered by the time of the sampling, were also analysed by ICP-SFMS with conventional nebulisation and laser ablation, respectively. Thus, comparison of the U concentration in *Populus × canescens* trees grown in the loess-covered and uncovered tailings could be performed. The results may be helpful for mapping of the uranium contamination in the affected region and redistribution of U due to its uptake and translocation in the trees.

### Fr07

#### U-TH-PU ISOTOPE ANALYSES OF SOIL BY ID-MC-ICP-MS AND LA-ICP-MS ANALYSES OF SPHERICAL PARTICLES FROM THE ELBMARSCH, NORTHERN GERMANY.

**A. Gerdes**, J.W. Goethe-University, Institute of Geosciences, Senckenberganlage 28, Frankfurt am Main, Germany, 60054  
[gerdes@em.uni-frankfurt.de](mailto:gerdes@em.uni-frankfurt.de)

Sensitive single and multi-collector SF-ICP-MS have been applied to characterize micrometer sized spherical particles and different grain size fractions of three soil samples from northern Germany. Samples were collected in close vicinity to a nuclear power plant and the GKSS research centre in the Elbmarsch, c.30 km SE of Hamburg. In 1990 a leukaemia cluster has been discovered among the children living in the small Elbmarsch community. Despite of various investigations, which could not find any link between this cluster and the nuclear facilities, scepticism and accusations persist and is demanding for more detailed studies.

Various kinds of spherical particles, 10-1000µm in size, have been identified, imaged by optical and backscatter electron microscopy and analysed for 60 major and trace elements by LA-ICP-MS using UP213 laser system coupled to a Element II. Th and U contents usually vary from 0.004 to 90 and 0.005 to 20 ppm, respectively. Elevated concentrations have been identified only in zircon (e.g., 100-600 ppm U). The mean <sup>238</sup>U/<sup>235</sup>U of LA-ICP-MS analyses on spherical particles (n=90) gave 138.1 ±0.5 and is thus identical with the natural ratio

(137.88). Instead the particles often characterised by elevated contents of heavy elements, such as Pb, Cr, Ni, Cu, As, Cd and/or Zn relative to the average continental crust. Spherical particles can be grouped in biological material, fly ashes of different composition and natural minerals.

Uranium, thorium and plutonium isotopes were analysed by isotope dilution MC-ICP-MS in 20 different soil grain size fractions and different types of spherical particles. Dissolution with hot acid mixtures was performed in three subsequent steps to study the effect on the overall results, e.g. behaviour of the different isotopes and the parent daughter ratios.

## Fr08

### ULTRATRACE-LEVEL ENVIRONMENTAL TRANSURANIC ANALYSIS BY ICP-SFMS FOR ORIGIN ASSESSMENT

**Z. Varga**, Z. Stefánka, Hungarian Academy of Sciences, Institute of Isotopes, Department of Radiation Safety, Konkoly-Thege Miklós 29-33. H-1121 Budapest, Hungary

[varga@iki.kfki.hu](mailto:varga@iki.kfki.hu)

Long-lived transuranic radionuclides (e.g.,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  or  $^{241}\text{Am}$ ) have been dispersed in the environment as a consequence of atmospheric weapons tests, nuclear power plant or satellite accidents. Measurement of these radionuclides due to their long half-lives and high dose contributions are of high importance. Moreover, the isotopic composition is characteristic to its source, which can be exploited in environmental monitoring, safeguards and for the analysis of illicit (confiscated) nuclear materials<sup>1-3</sup>.

The aim of our study was to develop novel and simple methods for ultratrace-level transuranic ( $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{241}\text{Am}$ ) analysis by inductively coupled plasma sector field mass spectrometry (ICP-SFMS). Samples from various locations contaminated with global fallout, nuclear power plant accidents (Chernobyl), reprocessing plant release (Sellafield) and nuclear weapons tests (Pacific test site) were analysed. The sample preparation developed and applied for the analysis is presented together with their figures of merit. The results were used to assess the origin of contamination based on isotope ratio measurements and the time of release was estimated using  $^{241}\text{Pu}/^{241}\text{Am}$  ratio.

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<sup>3</sup> Betti, M., Tamborini, G., Koch, L.: Use of secondary ion mass spectrometry in nuclear forensic analysis for the characterization of plutonium and highly enriched uranium particles. Analytical Chemistry 1999, 71, 2616