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Technical Meeting on Developments in Non-Radiocarbon Accelerator Mass Spectrometry Techniques and Relevant Applications

Virtual Event

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Booklet of Abstracts¹

¹ Listed in alphabetical order of the represented IAEA Member State/Surname of the main author

ABOUT THE MEETING

Introduction

Accelerator Mass Spectrometry (AMS) initially developed at low-energy nuclear physics laboratories in the late 70s - beginning of 80s, has been proved to be not only an ultra-sensitive technique for counting individual atoms but also an accelerator-based method with a vast potential for analytical applications related to problems of modern society. Nowadays, AMS is used in archaeology, biomedicine applications, climate change studies, environmental monitoring, hydrology and oceanography, nuclear safeguards, forensics, and many other fields of increased societal and economical interest.

AMS is used to detect radionuclides and stable nuclides with very low concentrations, between 10^{-12} and 10^{-16} . In contrast to other methods, AMS requires much shorter measuring times, typically less than one hour, as well as much smaller amounts of samples, often at sub-milligram range. The most widely employed isotope is radiocarbon (^{14}C), which used by at least 85% of the analyses performed in AMS laboratories. Besides radiocarbon, the use of other isotopes, such as ^{10}Be , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{129}I , or U and Pu isotopes is continuously increasing, due to their unique analytical capabilities for a wide spectrum of applications.

The present Technical Meeting on Developments in Non-Radiocarbon Accelerator Mass Spectrometry Techniques and Relevant Applications is driven by the need to promote the unique analytical capabilities of non-radiocarbon AMS, disseminate the relevant knowledge and expertise and advancing technologies to IAEA Member States together with AMS's large potential for contributing to their socio-economic development.

Topics

A non-exhaustive list of topics to be covered by the Technical Meeting is given below:

- Advanced accelerator systems for AMS with radioisotopes besides radiocarbon
- New and advanced AMS techniques
- Advances in sample preparation methods
- Current and emerging non-radiocarbon AMS applications
- Analytical requirements in terms of sensitivity and measurement precision
- New and Future Facilities
- Networking

Objectives

The purpose of the event is to:

1. Review progress made on the technologies associated with the sample preparation and subsequent use in AMS of radioisotopes besides radiocarbon.
2. Review technological developments and achievements at AMS laboratories using radioisotopes besides radiocarbon.

3. Discuss current and emerging applications based on anthropogenic and cosmogenic radioisotopes apart from radiocarbon.
4. Record training needs in the field addressed by the Technical Meeting, especially for scientists from developing countries
5. Collect and evaluate scientific information for the preparation of an IAEA TECDOC, if required.
6. Identify special research and development topics of common interest for the participants to assess the need of initiating an IAEA Coordinated Research Project (CRP)

Expected Outputs

1. Meeting report including recommendations on the meeting objectives.
2. Layout of an IAEA TECDOC tentatively entitled “Developments in Non-Radiocarbon Accelerator Mass Spectrometry Techniques and Relevant Applications”

Participants will be asked to contribute on voluntary basis to the publication of an IAEA TECDOC. For the preparation of the TECDOC, an editing committee to be coordinated by the Scientific Secretary of the meeting will be assigned by the participants. If agreed by the participants, a summary report of the meeting will be jointly prepared for submission to a scientific journal.

Event Web Page: <https://www.iaea.org/events/evt1904256>

Tracing biological uptake of plutonium for radioecology applications

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Abstract: Development of capabilities and methods of analysis using AMS has provided a tool for extremely high sensitivity analysis of a wide range of heavy isotopes including fission products, such as I-129 and Tc-99, and activation/decay products such as plutonium isotopes (239, 240, 241, 242 and 244), Pa-231, Np-237, Am-241 and the uranium isotopes U-236 and U-233.

The 1MV VEGA AMS system at ANSTO's Centre for Accelerator Science (CAS) has one of the highest sensitivities in the world for rare heavy isotopes, which offers a significant advantage in investigation of trace signatures of nuclear contamination in the environment [1]. Achievement of sub-femtogram detection limits for a wide range of isotopes is possible this system. The ability to interrogate a wide range of anthropogenic nuclear isotopes allows the potential to distinguish nuclear material signatures of local contamination sources from global fallout background and identify the relative contributions.

Significantly, with this improved tool for investigation it has become possible to not only characterise and quantify environmental deposition of plutonium contamination, but also the interaction of such contamination with biota and food webs by examining uptake in organisms living and feeding within contaminated zones. This information can then be used to inform radiological dose models for humans and wildlife and provide a basis for managing contaminated zones. Additionally, organ specific uptake can be determined to understand the interaction between contaminant form and biological uptake. Here a couple of recent examples of the application of the VEGA AMS Actinides capability are presented.

Following the 2011 Fukushima accident, Accelerator Mass Spectrometry (AMS) was used to quantify and compare Pu uptake by local biota (earthworms and wild boar) near the accident site to uptake at a site near Nagasaki, Japan, where Pu had been dispersed by the 1945 nuclear weapon detonation. Results revealed that the uptake of Pu was greater in biota near Nagasaki where >90% was sourced from the weapon detonation (vs from global fallout), whereas near Fukushima, >90% of Pu in biota was from global fallout and <10% from the accident [2]. In both cases, the local sources were able to be distinguished from global fallout using their isotopic signatures (Figure 1).

Nuclear tests were conducted by the British government in Australia at the Montebello islands, Western Australia from 1952-56. Much of this area is now a protected Marine Environment however ongoing uptake of Pu occurs in marine biota at the Montebello Islands. AMS was utilised to study the residual environmental distribution of nuclear material as well as the biological burden of a wide range of the endemic biota. The high sensitivity afforded by the VEGA Actinide analysis capability at CAS, ANSTO allowed for the inclusion of protected marine species in the study, including flatback sea turtles, using small, non-lethal skin punches [3]. Pu released during the local tests was found to be the dominant source in the tissues of local fish, reptiles and crustaceans and sea cucumbers. Dose rates to local biota, including the sea turtles that nest on contaminated beaches, were determined to be low today compared with doses in the decade following the tests. Additionally, each test was found to have produced a

distinct Pu isotopic signature, which was used to determine their respective Pu dose impacts (Figure 2).

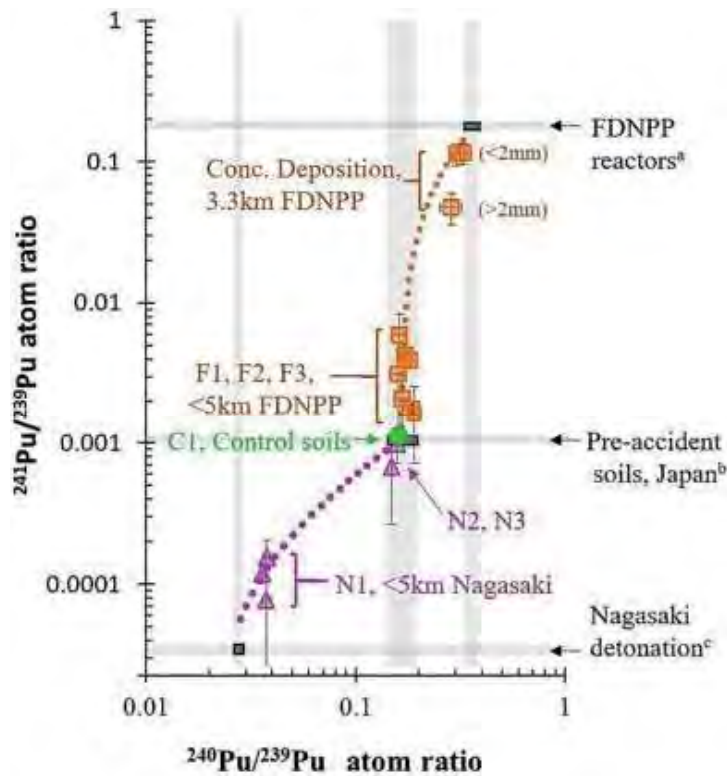


Figure 1: Pu atom ratios from study samples near the FDNPP (\square), near Nagasaki (Δ) and control site (\circ). Dark grey boxes are Pu-source reference ranges for (a) FDNPP reactors, (b) pre-accident soils of Japan, and (c) the Nagasaki detonation (see S4 for references used). Dashed lines are atom ratio mixing lines.

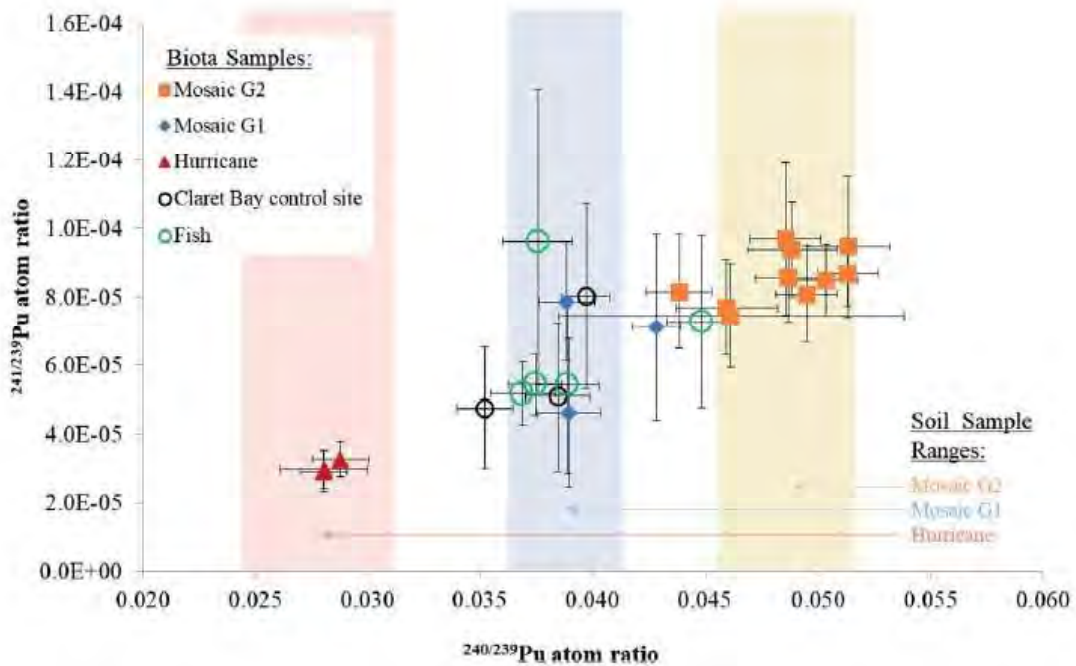


Figure 2: $^{241}/^{239}\text{Pu}$ vs $^{240}/^{239}\text{Pu}$ atom ratios from biota samples identified by their respective collection sites, except for the fish samples which were gathered from seawater areas between detonation sites.

In addition to results from these studies, several technical challenges for application of this technique to emergency response and environmental monitoring scenarios will be discussed, from sample preparation logistics, contamination concerns, and availability of appropriate calibration materials, isotope dilution tracers and reference standards.

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Advances in the use of multiple cosmogenic nuclide pairs driving emerging applications in Earth sciences

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Abstract: Cosmogenic nuclides, both *meteoric* – produced in the Earth's atmosphere, and *in-situ* – produced in minerals within the Earth's surface, have been routinely used in the Earth Sciences for more than three decades. Here we summarise the principles behind the main Earth science applications of the in-situ produced radionuclides ^{26}Al ($T_{1/2}=0.7\text{Myr}$), ^{10}Be ($T_{1/2}=1.4\text{Myr}$), ^{14}C ($T_{1/2}=5\text{kyr}$), and ^{36}Cl ($T_{1/2}=0.3\text{Myr}$) and review the current state-of-the-art with respect to analytical procedures and novel applications, providing examples of recent work and development involving these nuclides completed at ANSTO's Centre for Accelerator Science.

Although sample preparation and chemistry procedures for ^{26}Al , ^{10}Be , and ^{36}Cl have changed little since their initial publication, more recently there have been efforts to optimise [1] and even automate [2] some of the steps that are involved in the chemistry process. Sample extraction also continues to be a major limitation for in situ ^{14}C applications, several labs attempting automation to increase sample throughput and minimise opportunities for human error [3]. Due to limitations related to usable target minerals as well as the complexity of sample preparation steps, the workhorse for Earth science applications has been in-situ-produced ^{10}Be followed by ^{26}Al , the latter almost exclusively analysed in conjunction with ^{10}Be . Cosmogenic nuclides such as ^{36}Cl and in-situ-produced ^{14}C , have seen far less applications to date, however, this is likely to change going forward. First, applications of ^{36}Cl in magnetite [4] will extend the applicability of basin-wide denudation rate studies to non-quartz bearing lithologies. Second, in terms of in-situ-produced ^{14}C , major optimisations will result from the new generation of the simpler and more robust metal-based extraction systems (or even laser) that have already achieved lower blank levels, shorter extraction times, and substantially improved reproducibility of standard materials [5,6].

Surface exposure dating is the simplest and most straightforward application of cosmogenic nuclide analysis. Surface exposure dating has been extensively applied to the dating of glacially derived sediment (e.g., moraines, erratic boulders) and glacially polished bedrock. The application assumes that the datable surface or sediment was depleted in cosmogenic nuclides prior to the exposure event, an assumption that does not always hold and so has confounded to some extent the reliable application of the technique to glacial deposits. More recently, however, the paired use of ^{10}Be and in-situ ^{14}C , due to the latter's short half-life, has allowed for the inherited nuclide concentration to be explicitly constrained [7], and so paired nuclide surface exposure dating applications will likely become the norm in the future.

In surfaces and landscapes that are undergoing continuous and steady erosion, cosmogenic nuclides such as ^{10}Be and ^{26}Al have been utilised to quantify the rate of surface lowering. The technique has been successfully applied to both outcrops or soils, to calculate at-a-site surface erosion rates, and to river sediment, to calculate basin-wide average landscape lowering rates (or denudation rates). Cosmogenic ^{10}Be and ^{26}Al have now been applied in river basins from a range of climatic and tectonic settings, ranging from a few square-km to basins as large as the Amazon. Cosmogenic nuclide triplets, such as ^{26}Al - ^{10}Be - ^{14}C in modern river sediment have also been used recently to quantify the timescales of sediment transport, storage and reworking as the sediment moves from source areas to depositional sinks [8].

Burial dating employs two (or more) cosmogenic nuclides and is used to quantify the time elapsed since a sample was shielded from cosmic radiation by burial. The method is based on the principle that the ratio of two cosmogenic radionuclides will decrease from a set value in proportion to the duration of burial. The method assumes that prior to burial, the parcel of sediment that is being dated experienced a simple and continuous exposure history and burial completely shielded the material from cosmic radiation. The above have limited the use of burial dating to sediment found in caves or as part of thick alluvial or colluvial deposits. A new approach to burial dating – called isochron burial dating – uses the variation in measured ^{26}Al and ^{10}Be (or any other nuclide pair) in a group of individual sediment clasts collected from the same stratigraphic layer to solve explicitly for the post-burial component that will be common among the samples [9]. The new technique is not limited by the constraints of conventional burial dating and has been successfully applied to sediments where reworking might have occurred and to deposits partially exposed to cosmic radiation. A particularly salient application has been the dating of hominid and hominin fossil bearing deposits [10].

In summary, although single-nuclide-based conventional applications of cosmogenic nuclides still dominate the field by volume, a number of new applications have emerged in the last few years that make use of multiple nuclide pairs and leverage analytical advances, especially regarding ^{26}Al , ^{36}Cl , and in-situ ^{14}C . Given the array of capabilities encompassing sample preparation and analysis of all four radionuclides, ANSTO's Centre for Accelerator Science is well positioned to capitalise on and contribute to advances in these novel applications.

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Technical and operational strategies to bolster cosmogenic isotope AMS

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Abstract: ANSTO Centre for Accelerator Science routinely applies cosmogenic isotope AMS measurements of ^{10}Be and ^{26}Al for earth science studies, such as quantitative geomorphology and reconstruction of glacier chronologies and past climate changes. The isotope of choice for most studies is ^{10}Be , but ^{26}Al as a part of $^{26}\text{Al}/^{10}\text{Be}$ nuclide pair is also widely used in studies of burial dating and settings with complex exposure histories. The majority of routine samples yield $^{10}\text{Be}/^9\text{Be}$ or $^{26}\text{Al}/^{27}\text{Al}$ ratios of 10^{-13} or above. However, more recent applications often push the limits of the technique. For example, dating of young glacial deposits [1], using onshore and offshore bedrock and sediment cores to reconstruct the glaciation histories of polar regions [2-4], or to reconstruct paleo-erosion rate records [5] and estimating coastal cliff retreat rates [6].

Here we present technical method development that reduces measurement losses thus increasing sensitivity, and an operational strategy to increase accessibility and broaden the cosmogenic isotope user base. The former enables new applications with low isotope concentrations and expedites the measurement and sample preparation, whereas the latter seeks to make the method more accessible to user communities that do not readily have access to high performance, ultra-sensitive AMS infrastructure.

The limiting factor is the low overall detection efficiency for cosmogenic isotopes (Al, Be) affecting precision and sensitivity for applications where the amount of available sample material is small and/or rare isotope concentration is low. Measurement efficiency is characterized by losses in AMS measurement that occur in the ion source, ion transport, charge exchange, and ion detection. Due to these losses, it is not uncommon that more than 99% of the rare isotopes in the sample do not contribute to the statistical precision of the measurement [7-10].

To compensate for the measurement inefficiency, one has to process more quartz to ensure the quantity of rare isotope in the sample is sufficiently high to yield desired statistical precision. This, in turn, leads to labour-intensive and time-consuming sample processing with increased analytical costs. The result often is that the sample preparation forms a bottleneck and limits the throughput.

Often the approach in many AMS user facilities, including ANSTO, is to establish relationships with satellite sample preparation facilities that reduce the pressure on the local sample preparation laboratory. However, this might only move the bottleneck elsewhere, for example personnel time or available instrument time at the AMS laboratory might become the limiting factor.

We benchmark our current measurement capability for ^{10}Be and ^{26}Al and introduce the ongoing method development work with regards to the ion source, ion transport, and ion detection. Optimizing the performance and operation of the negative ion Cs-sputter sources has significant scope for improvement but is challenging. One often needs to compromise between competing requirements, for example, maintaining a high sputtering rate to allow expedient consumption of the sample material but at the same time keeping the source insulators clean for longevity. On the other hand, as a user facility, we work with different sample preparation laboratories that process samples from varying lithological settings, and any increases in method sensitivity can't compromise our selectivity against interferences.

Operationally we aim to expand the standard satellite laboratory concept further by making our facilities remotely accessible. This may include monitoring the measurement progress, having remote control of the sample queue, and/or full remote operation of the accelerator. Early review of the protocols, interlocks, infrastructure, and policies required to do this safely and to ensure data quality will be discussed. This could be a possible avenue to increase the availability of the technique to a broader user base especially in cases where significant investments, that are required to acquire, operate, and maintain an AMS facility, are limiting the use of the method.

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The EU funded project RADIATE for providing easy, flexible and efficient access for Non-Radiocarbon AMS research

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Abstract: For 25 years the research group Isotope Physics operates the Vienna Environmental Research Accelerator (VERA) based on a 3 MV tandem accelerator (Fig. 1) [1]. VERA is one of the first accelerators dedicated and designed for universal accelerator mass spectrometry (AMS) across the nuclear chart and has been continuously extended ever since (Fig. 2).

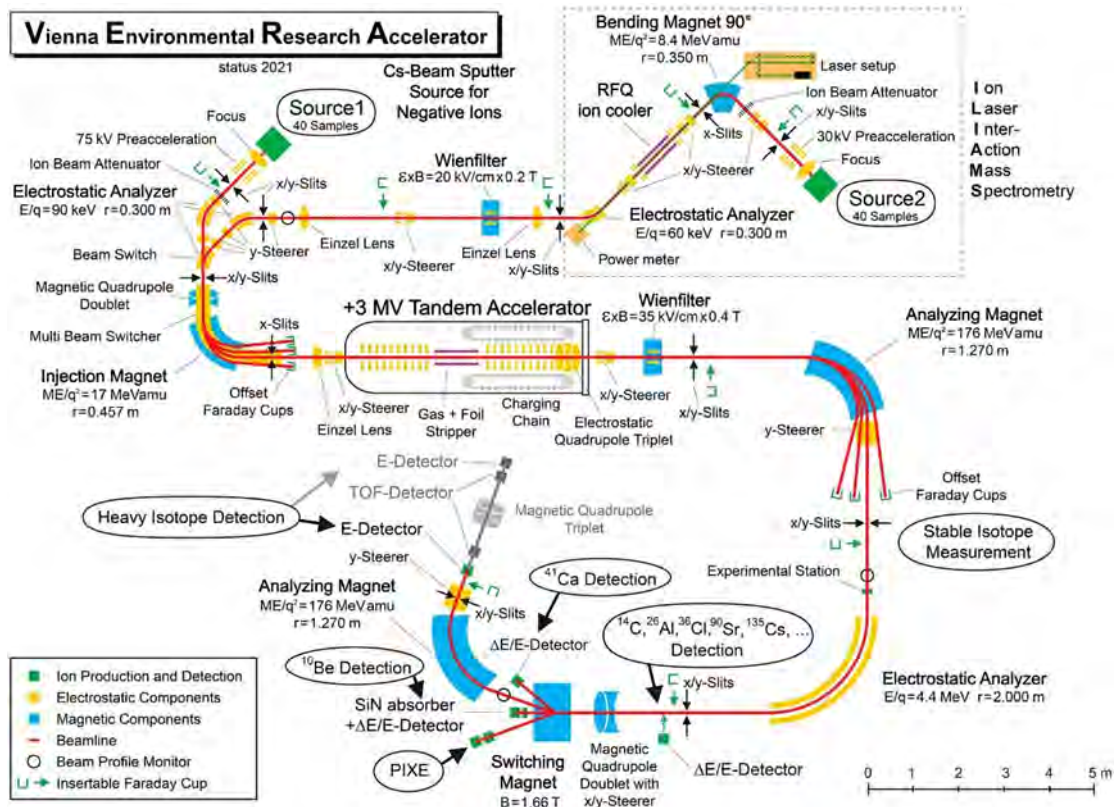


Figure 1: Overview of the VERA facility (<https://isotopenphysik.univie.ac.at/en/vera/>).

VERA can measure all standard AMS isotopes like the long-lived radionuclides ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , and ^{129}I (Fig. 2) and is especially well-suited for the determination of actinide isotopes (U, Np, Pu, Am). AMS measurements on the complete set of established isotopes are performed for (i.e., hands-off) and in cooperation with many national and international partners.

VERA is also well-known for hosting international guests for education and training of both, AMS sample preparation and running an AMS facility, often yielding into performing common (i.e., hands-on) AMS measurements. VERA researchers have been invited for knowledge exchange especially contributing in setting-up new AMS facilities e.g., in Czech Republic and South Africa, and “satellite” labs for sample preparation e.g., in Austria, Denmark, France, Germany, Hungary and UK.

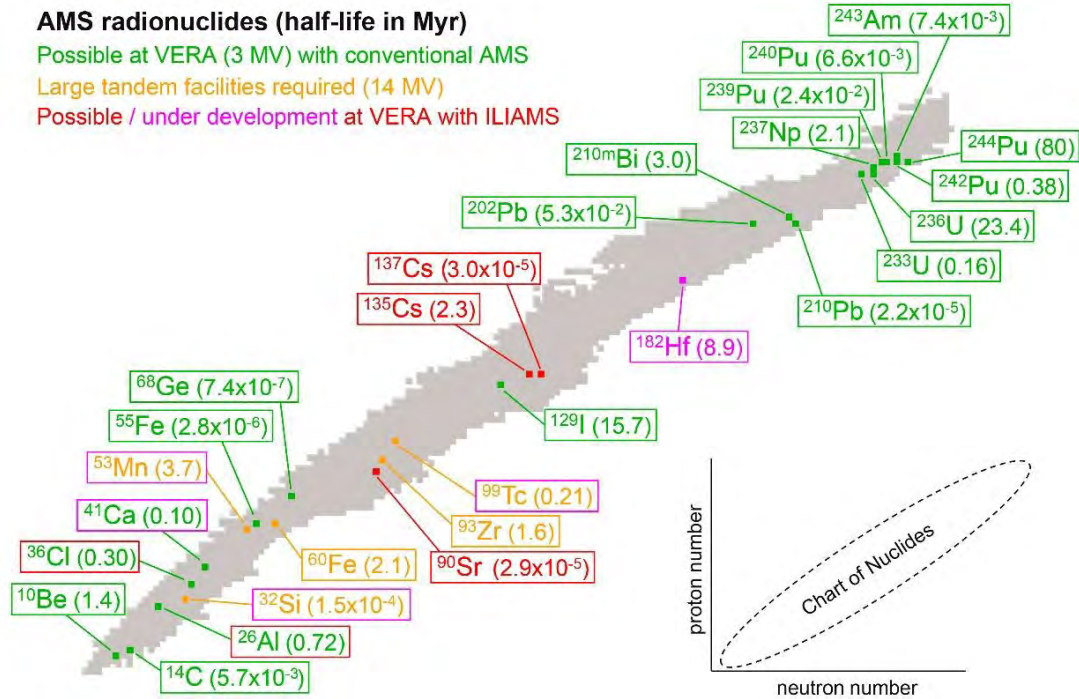


Figure 2: Long-lived radionuclides measurable by AMS at VERA and other large Tandem facilities (<https://isotopenphysik.univie.ac.at/en/vera/>).

Additionally, VERA has an outstanding reputation for the development and improvement of AMS methods and methodologies such as the yet world-wide unique ILIAMS (Ion-Laser Interaction Mass Spectrometry) system for isobar suppression [2]. Altogether, this makes VERA one of the most successful European ion beam infrastructure partners in the Horizon 2020-funded RADIATE (Research And Development with Ion Beams – Advancing Technology in Europe [3]) project, which started in 2019.

Within RADIATE, VERA contributes to several joint research activities and is the work package leader of “Beams and Sources”. Most prominently, VERA is committed to provide easy, flexible and efficient trans-national access for researchers from academia and industry. Over 1,500 hours of Non-Radiocarbon AMS at VERA is offered free of charge to users who successfully undergo a proposal procedure [4]. The focus is the attraction of new users who are not yet acquainted with AMS by providing them extensive support and training for interdisciplinary research. Besides pure AMS technology and sample preparation knowledge, VERA researchers also supply users with special expertise in the fields of Earth and environmental research, cosmochemistry, nuclear and astrophysics applications of AMS.

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Separation of Isobars via Ion-Molecule Reactions

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Abstract: The development of Accelerator Mass Spectrometry (AMS) has led to remarkable advances in the ability to detect at very low isotopic ratio several radionuclides of great interest in many scientific fields. However, expanding the application range of AMS to new radionuclides may require the use of isobaric separation techniques different from those commonly used in conventional AMS. Separation techniques based on DE/dx separation in foils or gases, gas ionization detectors or gas-filled magnets in many cases require larger accelerators. These currently limit the extension of AMS to a wider variety of radionuclides, despite the quality and imagination of AMS scientists who routinely apply these techniques to separate isobars from “classical” radionuclides. To be successful, any new isobaric separation technique will need to be integrable into existing AMS systems without major modification of installed equipment and will also need to provide efficient transmission of the radionuclide of interest to preserve the current ability of AMS to measure, in reasonable time and on milligram samples, isotopic ratios inaccessible by other techniques. Other important features to consider will be the cost and reliability of the additional equipment to be installed, the relative lengthening of the AMS line that may be required and the flexibility of these new techniques to be quickly programmed by the user to handle a wide range of radionuclides, ideally without requiring modification to hardware.

These new separation techniques will also need to be compatible with the latest compact AMS systems that industry, in collaboration with the research community, has developed over the past two decades. One promising approach consists in thermalizing ions in a section of the low energy line of the AMS system to promote highly specific ion-molecule reactions between the isobar and a gaseous reactant, using Radio Frequency Quadrupole (RFQ) ion guides commonly used in Liquid Chromatography Mass Spectrometry (LC-MS). This new method of isobar separation is facilitated by the fact that the isobar to be removed is most often member of a group in the periodic chart of elements that is adjacent to that of the radionuclide of interest. We will discuss the relevance of coupling this method to the in-source isobar suppression method realized by adding PbF₂ to the sample and mass-selecting the superhalogen molecular anion of the radionuclide being measured. The main difficulty in the application of the gas-filled ion guide technique seems to reside in the effective integration in the low energy (20-40 keV) line of the AMS system, of an RFQ-based system operating optimally in the energy range between 0.1 and 10 eV. This obstacle is being overcome through a collaboration between AMS scientists and LC-MS industry specialists, underway at the A. E. Lalonde Laboratory at the University of Ottawa. We will review briefly preliminary results of isobar suppression involving atomic and molecular species recently obtained at this laboratory.

Actinides & Fission Products AEL-AMS AMS Research Capabilities

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Abstract: The Actinides and Fission Products Unit is a facility at the A. E. Lalonde AMS Laboratory located at the University of Ottawa, where mainly samples are prepared for analysis on a 3MV accelerator mass spectrometer (AMS). In this facility we have the capability to prepare samples over a wide range of levels: from very low concentrations for environmental analysis to higher concentrations such as uranium ores. Also, two AMS approaches have been used: a conventional one in which samples are extracted and processed chemically and a “Direct approach”, in which sample material is placed in the accelerator ion source without chemical preparation.

Using the direct-AMS approach in the case of uranium ore concentrates (UOCs), several related isotope ratios ($^{231}\text{Pa}/^{238}\text{U}$, $^{230}\text{Th}/^{238}\text{U}$, $^{226}\text{Ra}/^{238}\text{U}$) were also assessed within the AMS data acquisition sequence used for measuring the $^{236}\text{U}/^{238}\text{U}$ ratios, and ^{185}Re , ^{187}Re and ^{187}Os , ^{188}Os , ^{191}Ir and ^{193}Ir in the sequence for the $^{187}\text{Os}/^{188}\text{Os}$ ratios. The sum of these results can be used to provide a “fingerprint” pattern to strengthen the capability for UOC source identification. Unexpectedly large $^{236}\text{U}/^{238}\text{U}$ ratios (up to $\geq 1 \times 10^{-7}$) have been found in several UOC samples. The $^{187}\text{Os}/^{188}\text{Os}$ ratio has also been shown for the first time to be a viable supplementary signature of UOCs. This work shows that the direct-AMS method has the potential to become an effective tool for nuclear forensics provenance assessment applications with UOCs. The implications of the results, and the need for further refinement of the sputter target preparation, the availability of standard reference materials, as well as the Cs^+ sputter ion source itself, will also be discussed.

Charcoal air filter cartridges were exposed at a reactors site for seven days to investigate the long-term accumulated emissions within a nuclear facility. For this purpose, two methodologies were applied: an extraction method and a direct analysis. The former method was proposed, using an in-house system and the high I_2 species volatility. Crushed charcoal sample with concentrated HNO_3 , and a yield tracer labelled with ^{125}I were loaded in Reactor A (Fig 1), This was heated and the vapor formed is trapped into AgNO_3 solution (Collector B). The ^{125}I was used to monitor the satisfactory efficiency of the process. In addition, a direct method was proposed because charcoal is already a material suitable for direct measurement in a Cs sputter ion source. In this direct analysis, charcoal samples were first spiked with a known quantity of stable iodine (^{127}I), in an excess over the quantity naturally present in the filter. In both methods, copper targets were prepared and ^{129}I analysed by AMS. The extraction method showed to be more reproducible than the direct measurement due to the size of the sample and results confirmed that these cartridges are very useful for monitoring the long-term emission of iodine from a nuclear facility.

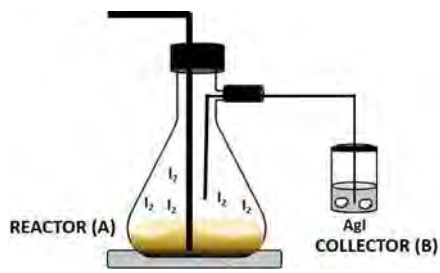


Figure 1. Diagram of extraction method to collect ^{129}I for further measurements by AMS. (A) Reactor with sample, ^{125}I and $\text{HNO}_3(\text{conc})$ (B) Collector which contained AgNO_3 1M solution. The I_2 formed in (A) and transferred to solution (B) (I^-), reacts with Ag^+ and forming AgI , which is collected for copper targets to be analysed by AMS.

This presentation provides two examples of the capabilities of the Actinides & Fission Products unit. Others can be developed in response to user requirements.

Regulatory applications of accelerator mass spectrometry

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Abstract: The Canadian Nuclear Safety Commission (CNSC) regulates the use of nuclear energy and materials to protect health, safety, security and the environment; to implement Canada's international commitments on the peaceful use of nuclear energy; and to disseminate objective scientific, technical and regulatory information to the public. Accelerator mass spectrometry (AMS) is a valuable analytical technique that can be widely employed to support regulatory-related matters such as safety case development, environmental monitoring programs and regulatory oversight of waste management.

One application of AMS to nuclear regulation is the tracing of both operational and accidental releases of isotopes including ^{99}Tc , ^{36}Cl , and ^{129}I in environmental samples. AMS has been used both in Canada and internationally to trace these isotopes as they are mobile dose contributors and therefore difficult to contain in radioactive waste management facilities. Environmental monitoring using AMS may also help establish baseline levels of these isotopes during site characterization or monitor for accidental releases during operations, decommissioning and post-closure.

AMS can contribute to regulatory oversight of waste management facilities and facilities undergoing decommissioning through the characterization of fission and neutron activation products in waste, particularly those that are difficult to measure (DTM). The CNSC requires that waste characterization include radionuclides relevant to safety, which for disposal facilities often include long-lived radionuclides such as ^{99}Tc , ^{129}I or ^{36}Cl . Furthermore, in developing scaling factors - a common method used to estimate the activity of DTM radionuclides - it is required that they be shown to be valid, which may involve some direct measurement using AMS.

The CNSC also engages in regulatory research using AMS. The purpose of regulatory research is to keep staff abreast of new scientific information, develop internal knowledge and share its research findings with stakeholders and scientists in Canada and abroad. One example of such a project is the characterization of ^{129}I production and transport in the Cigar Lake deposit, which is widely regarded as one of the best natural analogue sites for a deep geological repository.

The ability to trace accidentally released isotopes, such as those released by a leak or in a nuclear emergency, contributes to emergency preparedness and response through the analysis of several isotopes including ^{14}C , ^{129}I , and potentially ^{90}Sr . AMS has been used to assess the long-term fate of these radioisotopes in the environment as well as calculate the fallout of short lived isotopes such as ^{131}I released from Fukushima. The development of gas and laser ion sources will increase the speed and flexibility of analyses enhancing the applications of AMS for emergency preparedness and technical capability.

Applications of Fluoride Matrices in Target Materials for Heavy Element AMS Analysis

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Abstract: The necessity to use negative ions for injection into AMS systems which use tandem accelerators has, from its beginning, led to the use of molecular anions (negative ions) to inject the analyte atom. This strategy is required for those elements, typically metals, which have weak binding of an electron to the neutral atom and so produce very low (nA or less) beams from a sputter source. Early work used oxide beams, for example BeO⁻ [1], and oxides continue to be commonly used. While oxides provide stable target materials and beams, they are electrically insulating, so require mixing with a conducting binder. They also require powerful ion sources to generate usable currents and the presence of 3 oxygen isotopes can, in some cases, cause ambiguities in the beam that is mass-selected for injections. In 2009, Zhao *et al.* [2] published the results of an extensive investigation of molecular anions using fluorine, covering much of the periodic table. Not only is fluorine a mono-isotopic element, but the binding properties of molecular fluorine anions show a stoichiometric relationship which can provide useful isobar discrimination.

Noting the high currents from a lead fluoride target reported by Vockenhuber [3], the fluoride anion survey used PbF₂ as a binder which, as soon became clear, was a source of fluorine atoms. This could be explained by a property of PbF₂ discovered by Faraday [4] in which the solid poorly conducting crystalline material at room temperature became electrically conducting at temperatures ~250°C. Given the energy that is typically deposited in a target by the sputtering beam and the probability observed for formation of anions with a range of numbers of fluorine atoms, it appears that in the region near the entry of the sputtering ion, not only are electrons mobile but also the fluorine atoms.

Subsequent work has focussed on the preparation of molecular fluoride anions for use with the Isobar Separator for Anions (ISA) [5], including samples of calcium, strontium and caesium isotopes and fluoride matrix sample preparation methods. In addition, a search is on-going for other fluoride containing materials which could minimize some of the difficulties associated with certain combinations of elements with PbF₂, such as deliquescence during target storage, and volatility during initial phases of sputtering in the ion source. Methods including co-precipitation with either Fe₂O₃ or NdF₃ or simply fluxing the analyte with milliliter sized quantities of concentrated HF will be discussed, as well as analytical strategies to mitigate target volatility in the ion source.

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MILEA: Data from the new compact 300 kV multi-isotope AMS system

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Abstract: Multi-isotope AMS systems enable measurements of different long-lived radioisotopes on a single machine. Significant improvements in stripping techniques, molecule- and isobar suppression methods and advances in detector design over the last decade allowed lowering the ion energies while enhancing the efficiency at competitive background levels. A proof-of-principle experiment conducted at ETH Zurich showed that Tandem acceleration voltages of only 300 kV are sufficient for measuring of most radioisotopes measured by AMS today [1].

Based on the experiences gained with the proof-of-principle experiment, a new compact multi-isotope AMS system was designed and taken into operation in 2018 at ETH Zurich as a collaboration between Ionplus and ETH. The MILEA (Multi-Isotope Low Energy Accelerator) system, based on a 300 kV vacuum insulated Tandem accelerator, is dedicated to the measurement of ^{10}Be , ^{14}C , ^{26}Al , ^{41}Ca (biomedical applications), ^{129}I and actinides (Pu, U, Th and others). With new models and simulation tools [2], the stripper geometry and acceleration were optimized for He stripping at low energies. New approaches and developments were implemented in order to optimize the performance for all mentioned nuclides, resulting in a very small system footprint of $3.5 \times 7 \text{ m}^2$ (Fig. 1).

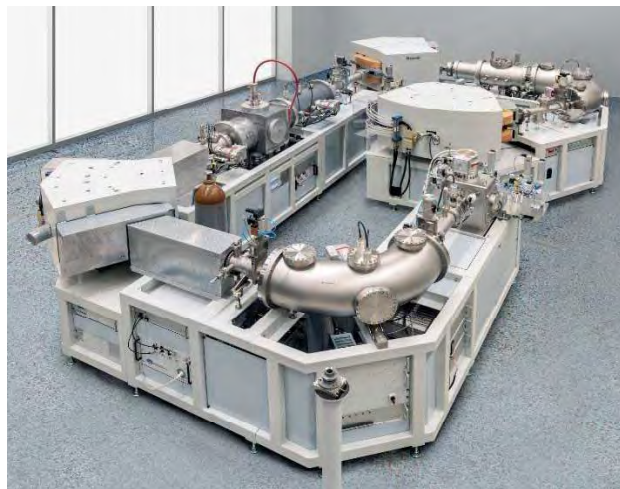


Figure 1. Perspective picture of the prototype version of the new, compact 300 kV multi-isotope AMS (MILEA) instrument. Footprint: $3.5 \times 7 \text{ m}^2$.

During 2018, extensive tests and experiments were performed on the prototype instrument to determine its measurement performance regarding transmission, background, and stability for the above-mentioned radionuclides. Since then, further experiments performed on the first three commercial installations have confirmed and, in some cases, surpassed the previously found performance parameters. An overview of the most important system features of MILEA will be given and the latest results for the different isotopes will be presented and discussed.

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AMS in the Czech Republic - building a new laboratory

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Applications of accelerator mass spectrometry (AMS) for determination of long-lived naturally occurring and man-made radionuclides at ultra-trace levels are continuously increasing in mutual synergy with research and development of technology and related fields. New, smaller and multipurpose AMS systems are being designed and produced following intensive scientific progress, bringing significant economic advantages. To acquire a system in line with the above trend, a consortium of Nuclear Physics Institute (NPI) of the Czech Academy of Sciences (CAS), Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague (CTU in Prague – FNSPE) and the Institute of Archaeology of the Czech Academy of Sciences, Prague has been established. The consortium is actually finalizing the first installation of a new Multi-Isotope Low-Energy AMS (MILEA) system outside of the country of its developers – Ionplus AG and ETH Zurich, Switzerland. MILEA system combines the established ion source technology and the vacuum insulated accelerator of the well-known device MICADAS (upgraded to 300 kV terminal voltage) with the well-proven concept of the high energy mass spectrometer layout of the ETH “TANDY” instrument. In contrast to the mentioned devices, MILEA consists of 90° low energy ESA and LE injector, and uses two analysing magnets and ESA at the high energy side, all in achromatic arrangement. Helium is used as a stripper gas. To compensate focal plane positions in multi-nuclide arrangement it uses e.g. a set of quadrupole lenses just behind the accelerator, or a set of positionable Faraday cups with various combinations of integrators after the first analysing magnet. At the back-end, an improved low-noise ΔE - E_{res} gas ionization detector provides outstanding separation and identification of interfering particles.

The consortium has been established to deal with a project “Ultra-trace isotope research in social and environmental studies using accelerator mass spectrometry”, with the acronym RAMSES. The presented MILEA system is designed for but not limited to the determination of ^{10}Be , ^{14}C , ^{26}Al , ^{41}Ca , ^{129}I , U, Pu and other actinoids. Apart from radiocarbon determination, the project scientific activities are focused on two key activities – determination of actinoids and fission products, and determination of cosmogenic radionuclides. At the early stages of the laboratory start up, current research deals with sample preparation procedures for the determination of ^{236}U , Pu and Am isotopes, ^{129}I , ^{10}Be , and ^{26}Al , respecting ultra-trace analysis requirements. Additionally, research focused on AMS target matrices and improvement of their ionization efficiency in the caesium sputter source is conducted. This is supported by the related infrastructure – new dedicated laboratories for the above-mentioned groups of radionuclides and a clean ISO 7 class laboratory at the NPI, and upgrades of (radiochemical) laboratories at FNSP (Department of Nuclear Chemistry).

The project ends in February 2023, the laboratory should be fully operational in early 2022.

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Isobar separation limits of Accelerator Mass Spectrometry and comprehensive approach by Ion Laser Photodetachment and Electron Transfer Reactions

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Abstract: Accelerator Mass Spectrometry has demonstrated its strength in the detection of rare radionuclides in the attogram (10^{-18} g) range as a result of combining efficient ion beam production and filtering. Such low levels of detection (LOD) are a result of dedicated sample preparation, anion selection, molecule dissociation and isobar suppression. The overall limiting factor for the detection of single nuclei by direct atom counting is the background of ubiquitous stable isobars. The isobar suppression is done generally by discriminating isotope of interest and isobars via their respective energy loss, since they have a difference in their proton number. In particular, the differentiation can be done by passive energy loss (stopping power) measurement, like gas-filled magnets or absorber foils, or by active methods, for example by a gas ionization detector. Furthermore, the suppression with a gas-filled magnet is enhanced due to the difference in the mean charge states. In any case, a high kinetic energy is needed if the relative proton number differences are small. Only then, the deviation of the stopping powers is big enough to overcome statistical variances and to allow a reliable particle identification. In the case of highly abundant stable isobars, like $^{60}\text{Fe}/^{60}\text{Ni}$ and $^{53}\text{Mn}/^{53}\text{Cr}$, this kinetic energy can only be reached with big tandem accelerators with terminal voltages in the range of 10 MV. For this reason, the medium mass range (e.g. ^{93}Zr , ^{99}Tc) represents the limit for conventional isobar separation techniques.

For heavier radionuclides with lower relative proton number difference to the stable isobar it is difficult to reach a sufficient LOD for radionuclides in natural concentrations, which are of great importance for societal questions like ^{99}Tc and ^{135}Cs or for astrophysical research like ^{182}Hf . However, radionuclides whose isobars do not form negative ions (e.g. ^{55}Fe , ^{129}I), or which have no stable isobar (e.g. ^{236}U , ^{244}Pu) are nevertheless precisely measurable with AMS and the highest sensitivities are reached for such radionuclides.

An advanced approach for the suppression of highly abundant stable isobars is the selective laser photodetachment of isobar anions. The difference in the electron affinity of the isobars allows to neutralize anions of a specific element by laser irradiation with a photon energy above their EA, while leaving the ion of interest with higher EA unaffected. This technique of Ion Laser InterAction Mass Spectrometry (ILIAMS) has demonstrated an extraordinarily high isobar suppression capability for a variety of radionuclides, like ^{26}Al , ^{36}Cl , ^{90}Sr , and ^{135}Cs [1].

For sufficient isobar suppression, long interaction times on the order of ms are needed, and the ion beam must be decelerated after the extraction from the ion source in a gas-filled radio-frequency quadrupole (RFQ) ion cooler. The admixture of reactive O_2 gas to the helium buffer gas has revealed an even higher isobar suppression for ^{90}Sr , as a consequence of electron

transfer reactions and ion-molecule transfer reactions.

Gas-filled RFQs are not commercially available, and their development is in the focus of ongoing research with exotic nuclei. A crucial point is the efficient deceleration and trapping of ion beams with high beam emittance like heavy molecular anions, e.g. $^{90}\text{SrF}_3^-$ or $^{182}\text{HfF}_5^-$. Sophisticated ion optical injection structures, like an elliptical injection electrode, comparable to ISOLTRAP [2], allow to slow down the ions far away from the central region where the buffer gas is leaking out of the RFQ through the central aperture. Despite of modern machining techniques for complex mechanical structures, technical challenges of aligning had to be solved for a reliable operation. To guide the thermalized anions through the gas-filled RFQ, a gradually changing potential in longitudinal direction is of particular importance and can be created by diagonally split cylindrical electrodes, which are capacitively coupled to a core rod that is carrying the RF signal. This advanced design simplifies the complex structure of diagonally split hyperbolic RF electrodes of the BECOLA setup [3]. The expected benefits of such an arrangement are the low required DC voltages and additionally the low multipole expansion coefficients.

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Radiological characterization for nuclear decommissioning with accelerator mass spectrometry

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Abstract: In the field of nuclear waste management, the quantification of a variety of nuclides is important for the radiological characterization of a material. Commonly used techniques for detection rely on the gamma decay of those nuclides. Since there are several nuclides like e.g., ^3H , ^{36}Cl , ^{41}Ca or ^{90}Sr , which undergo beta decay and emit no or only a weak gamma particle, other techniques have to be used. Common ones rely on the beta decay and become time and cost intensive with increasing half-life of the measured nuclide. Hence, accelerator mass spectrometry (AMS) can be a promising alternative in detecting those nuclides since it does not rely on the decay but measures and counts the atoms directly.

Commonly used reference nuclides in nuclear waste management are, e.g., ^{60}Co or ^{152}Eu because they can be easily quantified by gamma ray spectrometry. In case of ^{60}Co the relatively short half-life of 5.3 a might be a major disadvantage as decommissioning works might take place on a longer timescale. Thus, it is not very well suited if longer storage and control is needed. Another disadvantage in the case of reactor concrete is that ^{60}Co and ^{152}Eu are generated by neutron irradiation of ^{59}Co or ^{151}Eu , respectively, which both are contained only as trace elements with an unknown level of homogeneity. A research project therefore investigated the suitability of ^{41}Ca as a reference isotope for the characterization of reactor concrete, e.g., originating from the bio-shield of a nuclear power plant. For this purpose, several samples of concrete material were irradiated with thermal neutrons with different doses at the Mainz TRIGA reactor. The irradiated samples were then measured by gamma spectrometry and ^{41}Ca was chemically extracted as CaF_2 to produce a sputter target for the AMS measurement. Subsequently, the ^{41}Ca concentration was successfully determined at the 6MV tandetron AMS set-up CologneAMS at the University of Cologne.

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Activated graphite, e.g., from graphite moderated reactors, contains besides ^{14}C several radioactive nuclides with pure beta decay like ^{36}Cl or ^3H . The institute for nuclear physics in Cologne is aiming for a system which enables automated measurements using the AMS technique with gaseous samples, for ^{14}C , ^{36}Cl , and ^3H . The planned system will provide a high sample throughput as well as the possibility of sample dilution in cases of high activity. Therefore, a new gas-interface was built, which uses two syringes for the transport of the

sample gas into the ion source and a separate reservoir for blank gas which can be used for the dilution.

This project is supported by the Federal Ministry for Education and Research (BMBF) under contract number 15S9410B. Samples were partly supplied by the KTE (Kerntechnische Entsorgung Karlsruhe GmbH)

^{90}Sr is produced by nuclear fission and is a prominent nuclide in nuclear waste and fallout. Currently AMS is capable of measuring $^{90}\text{Sr}/\text{Sr}$ ratios below 10^{-12} , which corresponds to an activity of less than 4 mBq. Therefore, ^{90}Sr is extracted as SrF_3 in the ion source. The limiting factors for lower ratios are the background of the isobar ^{90}Zr and the weak ion source output. Since the suppression of the isobar ^{90}Zr demands a high accelerator energy, CologneAMS is developing the measurement of ^{90}Sr at the 10-MV tandem accelerator at the institute for nuclear physics. The target preparation is performed at the department for nuclear chemistry, Cologne. The recent status and development will be shown.

Linking ^{236}U -Uranium global fall-out to ^{90}Sr -Strontium: first results from a Pacific coral core

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Abstract: Uranium-236 (^{236}U) has been used applied as an ocean current tracer for almost a decade now, thanks to massive improvements in the sensitivity of Accelerator Mass Spectrometry for this isotope. Besides the discharges of the Sellafield reprocessing plant and the dispersion of ^{236}U in the aftermath of the Fukushima disaster, a significant amount of ^{236}U in the oceans is due to the global fall-out from nuclear testing – most of which occurred over a period of just a few years, with 80% of this fall-out deposited by 1964. An important improvement in the understanding of the evolution of this input would be more data on the total input and its distribution at the time. However, during this period of nuclear testing, monitoring ^{236}U fall-out was not possible for lack of a suitable sensitive measurement method. Strontium-90 (^{90}Sr) is another anthropogenic isotope whose global fall-out history has been intensively monitored in the past. Strontium as an element has a long oceanic lifetime, just like uranium, and both elements are readily built into the aragonite structure of coral skeletons. By extracting both elements from a coral core which covers the period after the maximum fall-out, it is possible to link the total ^{236}U to the total ^{90}Sr fall-out via the ratios of $^{90}\text{Sr}/\text{Sr}$, $^{236}\text{U}/\text{U}$ and the known seawater concentration of these elements.

We have measured the $^{90}\text{Sr}/\text{Sr}$ and $^{236}\text{U}/\text{U}$ ratios in a coral core from Kiribati (Tarawa). While initially we expected the selected coral core to cover the period of nuclear testing from its onset, based on the number of annual growth-bands in the core, our dataset does not include a rising edge of the ^{236}U concentration over the yearly slices, indicating that nuclear testing started before the oldest portion of this coral core. However, ^{236}U and ^{90}Sr levels for the analysed samples are well in line with post-peak oceanic levels, and so these results should still be useful to determine the global stratospheric fall-out $^{236}\text{U}/^{90}\text{Sr}$ ratio. Based on the results from this core, this ratio is 10.7 and thus the corresponding ^{236}U from the period of atmospheric testing before the comprehensive nuclear test ban treaty can be estimated as $1020\pm 80\text{kg}$, based on applying this $^{236}\text{U}/^{90}\text{Sr}$ ratio to the well-monitored global fall-out of ^{90}Sr .

[#] now at: *Naval Information Warfare Center, Pacific, San Diego, CA, United States of America*

Identifying water masses in the western Arctic Ocean using ^{233}U and ^{236}U

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Abstract: ^{233}U ($T_{1/2}=0.159$ My) has recently entered the AMS scene with very promising applications. The reason is the potential of the $^{233}\text{U}/^{236}\text{U}$ ratio to identify anthropogenic U sources [1]. Both ^{233}U and ^{236}U ($T_{1/2}=23.4$ My) were released worldwide during the atmospheric testing of nuclear weapons (i.e. global fallout). Unlike the case of ^{233}U , the operation of conventional nuclear reactors has led to the massive production of ^{236}U (i.e. due to the thermal neutron activation of ^{235}U). Consequently, those regions either affected by nuclear reactor accidents or nuclear reprocessing activities show an enhanced presence of ^{236}U which translates into lower $^{233}\text{U}/^{236}\text{U}$ ratios. This feature opens the gate to a wide variety of applications. To date, the most reasonable estimation of the global fallout $^{233}\text{U}/^{236}\text{U}$ ratio is $(1.44 \pm 0.15) \times 10^{-2}$ [1]. Considering the estimated ^{236}U worldwide global fallout budget of 1000 kg [2], about 14 kg of ^{233}U would be circulating in the general environment. This extremely low ^{233}U abundance makes its AMS determination very demanding. For instance, $^{233}\text{U}/^{238}\text{U}$ ratios of 10^{-11} in surface oceanic waters, or ^{233}U concentrations at the 10^5 at/kg level, have been reported [3]. The accurate analytical determination of ^{233}U can only be accomplished with optimized radiochemical procedures, aimed at minimizing ^{233}U background sources such as ^{232}Th and maximizing U recovery accompanied by a thorough setup of the AMS instrument. Indeed, only a few AMS facilities, with a long experience on actinides measurements, have demonstrate the potential to measure ^{233}U at environmental levels.

In this study, we present and discuss results on the ^{233}U and ^{236}U composition in seawater samples from the GN01 GEOTRACES expedition to the western Arctic Ocean in 2015. This is the first time that both radionuclides are studied in combination in the Arctic Ocean. The determinations were possible thanks to a collaborative effort between the Lamont-Doherty Earth Observatory (LDEO, New York, USA), the ETH Zürich (Switzerland) and the CNA (Seville, Spain). Iron precipitates from a set of 140 5L seawater samples were produced at LDEO and subsequently sent to the CNA, where U was purified, and the cathodes processed. Those samples with the lowest expected ratios were analysed on the 600 kV Tandy AMS facility at ETH, where sensitivities can reach 10^{-13} level or below [4]. The remaining samples were measured either on the Tandy facility or on the 1 MV AMS system at the CAN, whose ion beam design imposes serious restrictions on the abundance sensitivity that can be achieved (i.e. at the level of 10^{-11} for $^{233}\text{U}/^{238}\text{U}$).

The aim of the study was to explore for the first time the possibilities that the $^{233}\text{U}/^{236}\text{U}$ atom ratio offers to distinguish water masses in the Arctic Ocean. Recent studies mostly focus on the

analysis of ^{129}I and ^{236}U in a dual tracer approach [5]. Atlantic waters entering the Arctic Ocean often carry the isotopic signature of European reprocessing facilities (i.e. Sellafield and La Hague) and that of global fallout from lower latitudes in the Atlantic Ocean, whereas Pacific waters are labelled with the North Pacific Ocean global fallout nuclides only. Thus, in the Arctic Ocean, ^{233}U originates from global fallout while ^{236}U carries both, the bomb pulse and a prominent nuclear reprocessing signal. On the other hand, the relatively isolated Deep and Bottom Waters are expected to contain mostly the natural isotopic imprint, with ^{233}U and ^{236}U produced by natural process in the lithosphere.

We will show that: i) Pacific and Atlantic derived waters show enhanced signals of both radionuclides, which can be unravelled based on their $^{233}\text{U}/^{236}\text{U}$ signature; and ii) Deep and Bottom Waters show extremely low ^{233}U and ^{236}U concentrations close to or below detection limits with isotopic ratios distinct from known anthropogenic U sources. The comparably high $^{233}\text{U}/^{236}\text{U}$ ratios are interpreted as a relative increase of naturally occurring ^{233}U and ^{236}U and thus for gradually reaching natural $^{233}\text{U}/^{236}\text{U}$ levels in the deep Arctic Ocean. Our results set the basis for future studies using the $^{233}\text{U}/^{236}\text{U}$ ratio to distinguish anthropogenic and pre-anthropogenic U in the Arctic Ocean and beyond [6].

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Application of Accelerator Mass Spectrometry to the characterization of nuclear residues

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Abstract: Nuclear energy continues being commonly used nowadays in spite of the general tendency to substitute it by cleaner ways of energy production. In any case, one of the disadvantages of this method of energy production is the necessity of environmental control of the radioactivity especially close to the vicinity of nuclear facilities. Another important drawback is the large amount of residues that it produces. These are originated either in the normal activity of the nuclear power plants and/or on the decommissioning of these plants.

A big part of these residues will need to be stored in special facilities especially designed for this purpose. In order to optimise this process, it is very important to characterize them very well so that only the ones that strictly need this storage treatment are sent to these special stores. This would reduce strongly the amount of material that must be treated as radioactive and, consequently, the loading rhythm of the stores and their economic and social impact.

The long-lived radionuclides fulfil two important characteristics: they remain for a very long time in the nuclear residues and, in many cases, they are very difficult to detect by radiometric methods, as these do not have always enough sensitivity. In spite of this, the knowledge of their activities is essential for their appropriate evaluation. In these cases, a maximum level is fixed although the real level of the radionuclide in the residue will be much lower. The lack of sensitivity is clearly also an inconvenient for environmental samples.

To overpass this drawback, it has been shown that the use of high sensitivity radiometric methods and mass spectrometry methods can reduce strongly the detection limits for several long-lived radionuclides and can be used as an alternative for a series of nuclides whose emissions are difficult to evaluate by traditional counting techniques. Few years ago, the AMS group at CNA carried out a project dedicated to start developing the methodology of the application of Accelerator Mass Spectrometry (AMS) to the characterization low and intermediate level nuclear residues. In this project, we proposed to optimise the originally developed methodology for environmental samples and to extend the application of the technique to new radionuclides of interest for their management that have not been studied before. These studies will be mostly focused on residues generated in the decommissioning of nuclear power plants for its implications in the ongoing activities carried out in Spain, as it is performed in collaboration with ENRESA, the company in charge of nuclear residues in Spain.

Some of the radionuclides present in this kind of residues are ¹²⁹I, ¹⁴C, ³⁶Cl, ⁴¹Ca, ²³⁹Pu, ²⁴⁰Pu, ²³⁶U and ²³⁷Np. Our project tries to evaluate the limits of AMS for the determination of these radionuclides in nuclear residues. For this, our efforts are put both on the optimization of the AMS measurement and the radiochemistry. Another important point is that low and

intermediate nuclear residues can include a variety of materials that can be liquid or solid, coming from the daily processes carried out in normal operation or from decommissioning. Examples can include paper smears, resins, sludge, concrete, etc.

For many of the previously enumerated radionuclides, the expected levels will be high enough to be measured by AMS in similar conditions as environmental samples. More emphasis has to be put on the radiochemistry, as the isotopic ratios can be, in some cases, high. Sample preparation will then need to include carrier addition or isotopic dilution to reduce these ratios. Apart from this, radiochemical methods will need to be adapted to every kind of matrix.

The AMS facility at CNA is based on a 1 MV Tandetron and was installed in 2005. Its relatively low maximum terminal voltage makes it difficult to reach very low levels for some of the long-lived radionuclides that are traditionally detected by AMS, for example ^{36}Cl and ^{41}Ca . For them, we expect to evaluate the detection limits that the combination of radiochemical processes and machine set up can offer to reduce the detection limits that other techniques can offer.

In this talk we will present some of the results obtained up to now as well as the strategies and the experiments that are being currently performed in order to fulfil the described objectives.

Accelerator mass spectrometry of noble gases and medium-heavy nuclides

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Abstract: Ultra-sensitive isotopic analysis of noble gases is excluded from the capabilities of accelerator mass spectrometry (AMS) facilities based on electrostatic tandem accelerators using negative-ion injection, due to the instability of elemental negative ions of noble gases. Detection of rare medium and heavy radionuclides which require isobaric discrimination is also constrained for tandem-accelerator AMS facilities by their achievable ion energy. Our group has been developing and using a method of accelerator mass spectrometry based on an electron cyclotron resonance (ECR) ion source producing highly-charged positive ions and the superconducting heavy-ion linear accelerator ATLAS at Argonne National Laboratory (Illinois, USA).

The positive ions are accelerated, normally with no further stripping, to energies 2-5 MeV/*u* with an overall transmission from ion source throughout the accelerator of the order of 10%. A gas-filled magnetic spectrograph equipped with a position-sensitive focal-plane ionization-chamber detector is used for isobaric separation and ion identification [1]. Noble-gas accelerator mass spectrometry (NOGAMS) was used for detection of cosmogenic ^{39}Ar ($t_{1/2} = 268$ y) at isotopic abundances down to the 10^{-16} [2] and for the determination of the $^{36,38}\text{Ar}(n,\gamma)^{37,39}\text{Ar}$ reaction cross section at thermal ($kT = 25$ meV) and stellar neutron energy ($kT = 30$ keV) [3]. Owing to the ion high energy achievable at ATLAS, the method is also used for the analysis of medium-heavy rare nuclides which requires isobaric discrimination, so far up to ^{146}Sm (6.8×10^7 y) [4]. The technical aspects of the positive-ion AMS used in these studies and the applications in different fields of research will be discussed. An outlook to ongoing work will be presented.

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Geochemistry of Anthropocene: Novel application field of AMS

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Abstract

1. MALT facility overview, Japanese AMS facilities

MALT (Micro Analysis Laboratory, Tandem accelerator), The University of Tokyo, is an ion beam analysis facility equipped with a 5MV tandem accelerator, a Cs-sputter negative ion source, mass analysis system, multi-Faraday cups for the stable isotope measurements, and final detectors designed for each beam analysis techniques. Among various beam analysis methods including NRA (Nuclear Reaction Analysis), ERDA (Elastic Recoil Detection Analysis), and PIXE (Particle Induced X-ray Emission), AMS (Accelerator Mass Spectrometry) is the most promising at MALT. Actually 70% of whole machine time is used for AMS. Since the completion of MALT facility in 1993, AMS for various nuclides had been developed, ^{10}Be , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{129}I , and ^{236}U as well as ^{14}C . MALT is a pioneer of non-radiocarbon AMS in Japan. In Japan, there are 14 running accelerators for AMS, of which 8 accelerators are dedicated for ^{14}C (Table 1).

Table 1. Summary of Japanese AMS facility

No.	Facility	Location	Installation	Accelerator	Vmax	AMS
1	JAEA-Aomori	Aomori	1997	HVE	3.0	^{14}C , ^{129}I
2	Yamagata Univ.	Yamagata	2010	NEC	0.5	^{14}C
3	IAA-Shirakawa	Fukushima	2000	NEC	3.0	^{14}C
4	IAA-Koriyama	Fukushima	2005	NEC	0.5	^{14}C
5	Paleo Labo	Gumma	2004	NEC	0.5	^{14}C
6	NIES-TERRA	Ibaraki	1996	NEC	5.0	^{14}C , ^{129}I (?), ^{10}Be (?)
7	NIES-Compact	Ibaraki	2014	NEC	0.5	^{14}C
8	Univ. Tsukuba	Ibaraki	2016	NEC	6.0	^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{90}Sr , ^{129}I
9	AORI, Univ. Tokyo	Chiba	2013	NEC	0.3	^{14}C
10	Museum, Univ. Tokyo	Tokyo	2015	NEC	0.5	^{14}C
11	MALT, Univ. Tokyo	Tokyo	1993	NEC	5.0	^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{129}I , ^{236}U
12	JAEA-Tono	Gifu	1996	NEC	5.0	^{10}Be , ^{14}C , ^{26}Al , ^{129}I
13	JAEA-Tono	Gifu	2020	HVE	0.3	multi nuclides, under test operation
14	Nagoya Univ.	Aichi	1996	HVE	3.0	^{14}C

MALT is common use facility and open for scholars of not only in Japan but also from oversea. Main application field of AMS at MALT is earth-environmental sciences.

2. Development of non-radiocarbon AMS at MALT

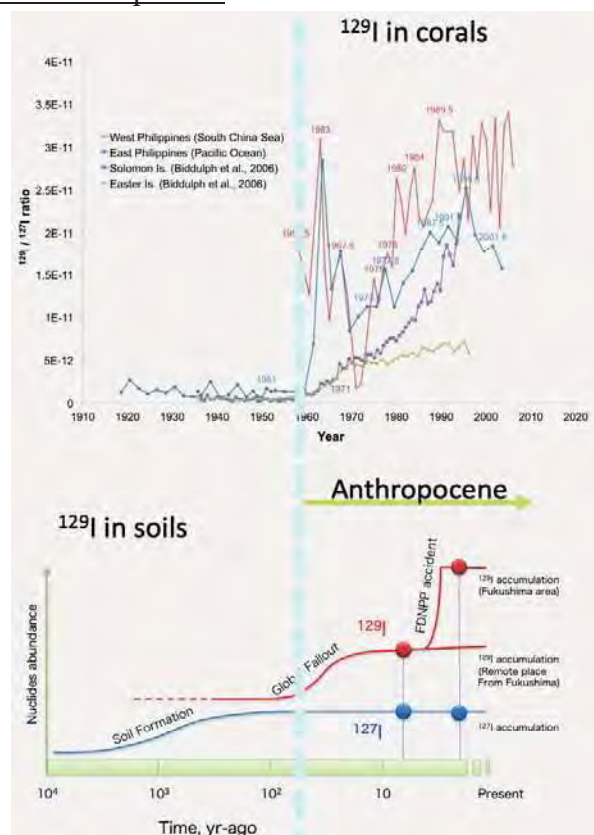
^{10}Be -AMS had been development in early stage of MALT history and one of most-used nuclides. The meteoric ^{10}Be is used for the study of the cosmic-ray intensity variation in past age. In situ ^{10}Be and ^{26}Al are used for the evaluation of surface morphological history. ^{129}I is another most-used nuclide. The natural ^{129}I is measured for the trial of the dating of a kind of

carbon reservoir such as methane hydrates. The anthropogenic ^{129}I is widely used for various studies. The time course variation of ^{129}I recorded in natural archives such as ice core and coral is important to evaluate the dynamics of ^{129}I and also the elemental iodine. It is interesting that the ^{36}Cl in ice core shows entirely different time-course profile due to the difference of production mode and transfer processes between two nuclides. ^{129}I had also measured well for the purpose of environmental assessment evaluation related with the Fukushima Daiichi Nuclear Power Plant accident. Recently a detection method of ^{41}Ca was examined. There is still a space for the sensitivity improvement, it is used for the evaluation of the clearance level of the waste structural concrete material of nuclear facilities. ^{236}U detection was also developed and now TOF system is ready to be installed for further improvement of the sensitivity. It will be used for the evaluation of the fuel debris in the process of the decommission of the Fukushima Plant.

For the aim to extend the ability of AMS, LPD (Laser Photo Detachment) system is now under development, which is expected to be a new isobar separation method. The pioneer of really applicable LPD for AMS is VERA facility. We are following VERA and preliminary experiment is conducted with a test bench beam line.

3. Geochemistry of Anthropocene

The time-course variation of ^{129}I shows a drastic change at the onset of the nuclear power usage. It is very clear signal of human activity recorded in natural archives. So ^{129}I is one of best indicators of the Anthropocene (Fig.1). From this notice, we are proposing “Geochemistry of Anthropocene” including time-course variation, dynamics of the anthropogenic nuclides. I think ^{129}I is most favorable marker because it is a fission product and its versatility in the environment. ^{36}Cl is also useful indicator in comparison with ^{129}I because it is neutron-activated nuclide and show different information. In the future I expect ^{236}U to be new favorable marker in the view of its versatility.



Development of Non-Radiocarbon Accelerator Mass Spectrometry at the University of Tsukuba

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Keywords: Multi-nuclide AMS, ^{36}Cl , ^{41}Ca , ^{90}Sr , ^{129}I

Abstract: The 6 MV Pelletron tandem accelerator was designed and constructed for multi-nuclide AMS at the University of Tsukuba in 2016¹⁾. It has two MC-SNICSs for the routine measurement. A five-electrode ΔE -E gas ionization detector is installed on the end station of the rare-particle detection system. We have developed ultrasensitive detection techniques for ^{10}Be , ^{14}C (graphite and CO_2), ^{26}Al , ^{36}Cl , ^{41}Ca , ^{90}Sr and ^{129}I in isotopic-ratio ranges of 10^{-10} to 10^{-15} .

In the case of ^{36}Cl AMS, we developed the sulfur removal method to reduce ^{36}S contamination. AgCl samples are pressed into an AgBr backing in a large Cu sample cathode (6 mm diameter) in order to reduce ^{36}S contamination²⁾. ^{36}Cl measurements are performed with a carbon foil ($4.8 \mu\text{g cm}^{-2}$) and an 8+ charge state at 6.0 MV, with $^{36}\text{Cl}^{8+}$ being injected into the detector at 54.0 MeV. $^{36}\text{Cl}^-$ beam transmission is $\sim 10\%$. The background of ^{36}Cl AMS has been improved to $^{36}\text{Cl}/\text{Cl} < 2 \times 10^{-15}$ as the result of improved isobaric separation techniques. ^{41}Ca and ^{90}Sr were also detected by the 6 MV multi-nuclide AMS system for the first time in Japan. Especially, the background of ^{41}Ca AMS was reached to $^{41}\text{Ca}/\text{Ca} \sim 3 \times 10^{-15}$. The ion source can extract CaF_3^- beam currents up to 500 nA from CaF_2 materials. $^{41}\text{Ca}^{5+}$ with a total energy of 32.5 MeV is clearly measured by the ionization detector³⁾.

^{90}Sr is one of the most important fission products due to the potential health risks of its uptake and retention in the human body. Sr in samples was recovered as SrF_2 to provide sufficient negative ions in the cesium-sputtering ion source. In the sample preparation step, 95%–98% of Sr was recovered and 99%–100% of Zr removed by ion-exchange separation. Sr recovery was $\sim 30\%$ in the precipitation process. The recovered SrF_2 was mixed with PbF_2 at a $\text{SrF}_2:\text{PbF}_2$ weight ratio of 4:1. A maximum 500 nA beam current of SrF_3^- ions was obtained from SrF_2 samples. $^{90}\text{Zr}^{8+}$ and $^{90}\text{Sr}^{8+}$ could be separated in two-dimensional spectrum by the gas-ionization detector with the total beam energy of 51.7 MeV. The $^{90}\text{Sr}/\text{Sr}$ atomic ratio background of $\sim 6 \times 10^{-13}$ (equivalent to $\sim 3 \text{ mBq } ^{90}\text{Sr}$) was comparable with that achieved at other AMS facilities.

As applied researches of non-radiocarbon AMS at the University of Tsukuba, ^{36}Cl and ^{129}I are measured most frequently in order to investigate the radioactive contamination in rainwater and soil samples, caused by the Fukushima Daiichi Nuclear Power Plant accident. ^{36}Cl is also applied for the tracer of cosmic ray events in the ice core. In this presentation, we report on the progress of non-radiocarbon AMS techniques with the 6 MV Pelletron tandem accelerator.

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The HVE low-energy multi-element AMS system with charge state selector in its high voltage terminal

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Abstract: High Voltage Engineering Europa B.V. (HVE) has designed a 300 kV AMS system for the measurement of ^{14}C and of non-radiocarbon isotope species like ^3H , ^{10}Be , ^{26}Al , ^{41}Ca , ^{129}I and actinides. The system features an electrostatic analyzer (ESA) as well as a high-resolution analyzing magnet in the low-energy spectrometer, a 300 kV, vacuum insulated tandem accelerator and a high energy (HE) spectrometer comprising two high resolution magnets and an ESA in between. The layout of the system is shown in figure 1. Located after the terminal stripper is an electrostatic deflector acting as charge state selector (CSS) [1]. The CSS fundamentally reduces a specific class of measurement background, especially beneficial for actinides.

This class of background is related to the injection of interfering molecules, like $^{238}\text{UOH}^-$ that is isobaric to $^{239}\text{PuO}^-$ and is formed with a probability of ca. 10^{-2} compared to $^{238}\text{UO}^-$. The molecules are dissociated in the terminal stripper and the fragments are charged positive. In the following HE acceleration $^{238}\text{U}^{4+}$ gains more energy than the $^{239}\text{Pu}^{3+}$ that is used for the analysis. A charge exchange to $^{238}\text{U}^{3+}$ within a few millimeters distance after the terminal results in the 4% energy increase, that gives ^{238}U and ^{239}Pu the same magnetic rigidity. At the critical position, pressure is in the order $5 \cdot 10^{-5}$ mbar, resulting in a probability in the order of 10^{-5} for this process. Without CSS, the $^{238}\text{U}^{3+}$ is suppressed in the HE ESA based on the energy difference but transmitted with a probability in the order of 10^{-6} after ion scattering on remnant gas by ca. 0.5° within the ESA. As consequence, ^{238}U will interfere to the ^{239}Pu measurement at the level of 10^{-13} .

The CSS in the terminal deflects the reference beam by ca. 20° , which results in the angle between the beamlines entering and exiting the accelerator. In the terminal, the difference in electrostatic rigidity between $^{238}\text{U}^{4+}$ and $^{239}\text{Pu}^{3+}$ is 33%, which can be compared to the 4% in the HE ESA. The separation of the charge states before the ions enter the HE acceleration reduces the related measurement background by 3 to 4 orders of magnitude. The CSS is designed for actinide measurements in charge state 3+ but improves the background for other isotope species as well, for example by suppressing background from $^{127}\text{I}^{3+}$ in case of $^{129}\text{I}^{2+}$ measurements or $^{13}\text{C}^{2+}$ background in case of $^{14}\text{C}^+$ measurements [2,3].

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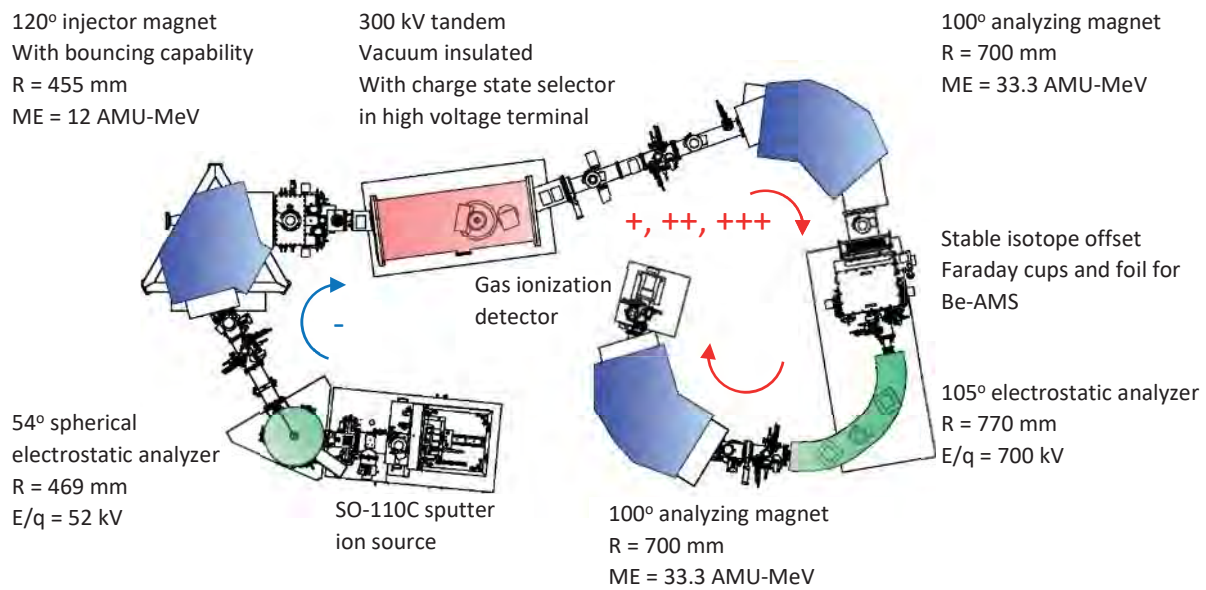


Figure 1: Layout of the HVE 300 kV multi-element AMS system. The arrows indicate direction of negative beams from source to accelerator, and of positive beams from accelerator to final detector.

Accelerator mass spectrometry – an ultra-sensitive radionuclide analysis technology for new frontiers in science

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Abstract: Highly sensitive investigations of rare processes in nuclear physics, in astrophysics, in environmental radioactivity studies, in isotope oceanography and hydrology, in biomedical research and in many other branches of science have had radioanalytical technologies as limiting factors for new discoveries. This has been mainly because of the fact that the available sensitivity was not high enough to get meaningful results, or the required sample size was too big to carry out such investigations. Especially environmental science has been a rapidly developing field, which included studies on behavior of radionuclides in the environment, use of natural and anthropogenic radionuclides as tracers of environmental processes, in marine radioactivity studies, in studying impacts of the Chernobyl and Fukushima accidents on the environment, in radiation protection of environment, in radioecology, etc., to mention at least a few of them. As the levels of radionuclides observed recently in the environment have been very low, highly sensitive radioanalytical systems have been required for carrying out new environmental investigations.

Recent developments in radiometric (RM) and mass spectrometry technologies have been associated in the radiometric sector mainly with underground operations of large volume Ge detectors, while the mass-spectrometry sector, represented mainly by accelerator mass spectrometry (AMS) and inductively coupled plasma mass spectrometry (ICPMS) has become the most sensitive technique for ultra-low-level analyses of long-lived radionuclides. The most sensitive technology till now has been the AMS, reaching detection limits below 1 nBq/g, and approaching new frontiers in radioanalytics — a single atom counting.

We shall review and compare AMS with ICPMS and RM technologies and discuss a few examples of AMS applications for Be-10, Al-26, Th, U and Pu isotope analyses in environmental and material samples. These new developments have had great impacts on investigations of rare nuclear processes and applications of radionuclides in environmental, life and space sciences.

AMS as a tool in marine radioactivity investigations

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Abstract: The Baltic Sea is one of the most studied areas regarding the distribution of radioactive radionuclides. Compared to open oceans, this semi-enclosed and shallow sea has an increased accumulation of radioactive materials coming from diverse sources: i) fallout coming from atmospheric nuclear weapon testing (1945-1980), ii) fallout coming from Chernobyl accident (1986), iii) liquid releases from Sellafield and La Hague nuclear reprocessing facilities transported through the North Sea, iv) authorized radioactive discharges occurring during the routine operation of nuclear installations in the Baltic Sea region, and v) possible leakages from the multiple radioactive wastes dumping sites within this marine region [1]. However, radionuclides long-lived actinides such as ^{233}U , ^{236}U and ^{237}Np , have been scarcely studied and there are still open questions about their sources to the Baltic Sea [2]. AMS has demonstrated competitive limits of detection for the study of such family of radionuclides in environmental samples.

The aim of this work is the study of actinides (i.e. ^{233}U , ^{236}U , ^{237}Np and Pu isotopes, ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{244}Pu) from a series of samples of diverse matrices (i.e. seawater, sediments, seaweed and biota) collected from different regions at the Baltic Sea in order to get better insights into their input sources to this marine environment. AMS analysis were performed at the Centro Nacional de Aceleradores (CNA, Sevilla, Spain) using a 1MV AMS system. The measurement techniques for ^{236}U , ^{237}Np and Pu isotopes (i.e. ^{239}Pu , ^{240}Pu , ^{241}Pu), have been implemented at this facility [3,4], and the possibilities of this system for ^{233}U analysis are being evaluated. Seawater samples were processed by using a chemical procedure, based on the sequential extraction of U, Np and Pu using TEVA® and UTEVA® resins and on the use of ^{242}Pu as tracer for both ^{237}Np and Pu isotopes [5]. This procedure was developed in the frame of the collaboration agreement between the CNA and the IAEA Marine Environmental Laboratories in Monaco. Subsequently, the method has been adapted to expand the target matrixes and to include ^{233}U among the measurable U isotopes at the 1 MV CNA AMS facility. The studied areas are: i) a dumping site placed close to Gothenburg, which has been scarcely documented and not previously studied, being unknown the status and the specific composition

of radioactive materials dumped in 1964, and ii) the Tvären Bay, directly impacted by the liquid releases from Studsvik Nuclear Plant whose actinides isotopic composition have not been documented. We will show that: i) there is no significant evidence of possible leakages from the dumped wastes at the Gothenburg area; and ii) our results from Tvären clearly indicate an important contribution of Studsvik releases, especially for ^{236}U , ^{239}Pu and ^{240}Pu . Dated sediment cores from Tvären Bay have been used to study the historical releases. Furthermore, the study of the isotopic composition of Pu in those sediments is extended to the most minor Pu isotopes, ^{241}Pu and ^{244}Pu .

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The AMS program at the Institute for Structure and Nuclear Astrophysics (ISNAP) at the University of Notre Dame

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Abstract: The Institute for Structure and Nuclear Astrophysics (ISNAP) at the University of Notre Dame in the United States has for many years concentrated its research efforts in experimental Nuclear Astrophysics as well as Nuclear structure and its effects on stellar reaction rates. This research effort centers on the use of three electrostatic accelerators present at Notre Dame (a 10MV FN Tandem, a 5MV single ended high-intensity vertical 5U accelerator and a 3MV Tandem accelerator dedicated to material analysis). Since 2006 an Accelerator Mass Spectrometry (AMS) program based on the 10MV FN Tandem accelerator has been dedicated to Nuclear Astrophysics as well as Nuclear forensics and Environmental science measurements. The available beam energy and experimental beam time combined with the availability of a gas-filled magnet (GFM) and Time-of-Flight (ToF) section and an array of available detectors provides a setup unique in North America dedicated to technical developments and sample measurements.

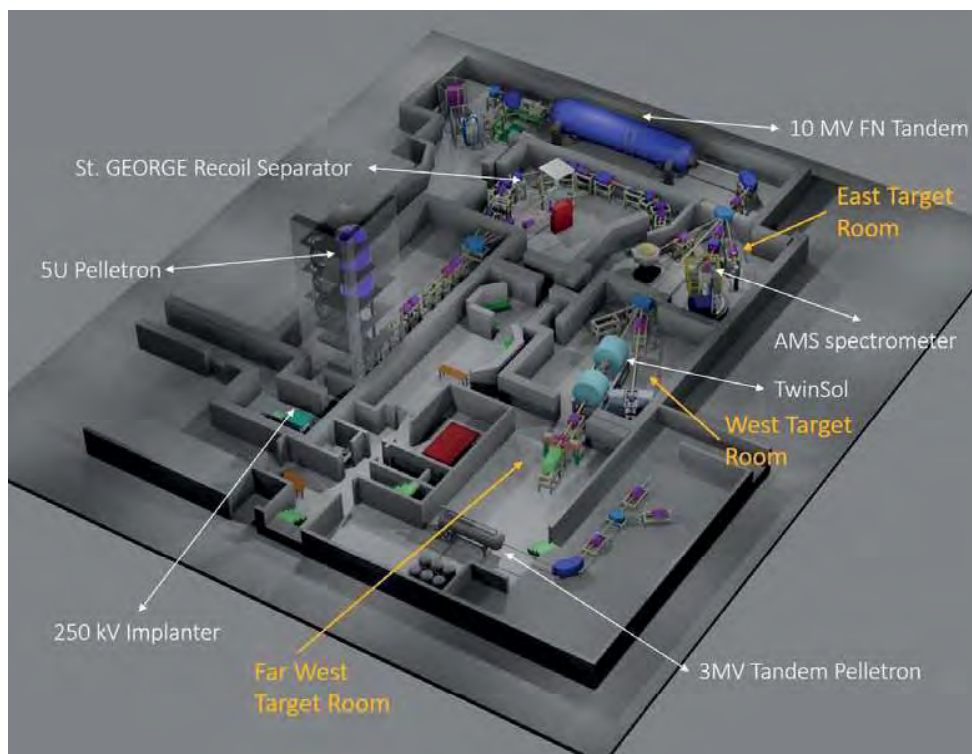


Figure 1: Present layout of the ISNAP facility showing the location of the accelerators and other major research instrumentation.

With accelerated beams ranging from protons all the way up to Actinides the FN provides the AMS program a wide array of available isotopes that allow for an research program dedicated to

developing new techniques as well as wide ranging application fields. In particular, measurements include production cross sections relevant to the synthesis of Short-Lived-Radionuclides (SLRs) in the Early Solar System (e.g., ^{36}Cl , ^{41}Ca , ^{53}Mn).

The on-going push to increase the AMS sensitivities attainable at ISNAP with an array of upgrades ranging from the low-energy injection beamline of the FN to the actual AMS beamline and the developments of new detectors is related to pivoting towards measuring concentrations of certain radionuclides in meteoritic material as well as terrestrial geological archives (^{53}Mn , ^{60}Fe , ^{93}Zr , ^{146}Sm) that provide essential information relevant to planetary formation and differentiation. By also providing a highly sensitive detection method for radioactive probes associated with environmental studies and nuclear forensics (e.g., ^{93}Zr , ^{129}I , actinides), the AMS program of Notre Dame allows a wide range of ongoing research and development efforts. Recently the successful tune of actinide beams at the NSL as well as installation of a TOF section has allowed for the expansion of this program. Initial measurements have focused on the detection of the uranium series $^{233-238}\text{U}$ and will provide an additional technique at ND to study the Ewing Uranium Ore Collection housed at the University. The Ewing collection is probably the largest Uranium ore collection in North America.

The Talk will focus on the recent developments and measurements performed by current PhD students in the group as well as recent technical developments in actinide AMS as well as the in-cathode ^{40}Ca irradiation program, which has allowed for the measurements of nuclear production cross-sections of ^{41}Ca bypassing any need for chemical treatment of the target material prior to the AMS measurement

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Measurements of ^{129}I in coastal Pacific Ocean waters in California and US Pacific Northwest sites.

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Abstract: Radionuclides such as iodine-129 released from nuclear activities and accidents into the ocean become entrained in surface ocean currents and subsequently are transported for great distances. In June 2011, a few months after the Fukushima Daiichi Nuclear Power Plant disaster, we began a surface ocean ^{129}I monitoring program, with samples from Scripps Pier, La Jolla, California, USA, with the expectation that surface currents originating off the east coast of Japan would eventually carry radionuclides to the La Jolla site. The sampling program was ended in early 2020. After 9 years of ocean transport, a distinct signal which can be related to Fukushima was not positively identified, however we have recorded an interesting systematic seasonal ^{129}I time series record that appears to be due surface circulation variations along the California coast, or perhaps other effects. However, $^{129}\text{I}/^{127}\text{I}$ ratios are not correlated significantly with concentrations of ^{127}I . To provide a more comprehensive picture of the ^{129}I budget in coastal surface waters off the west coast of the U.S., we also included ^{129}I data from the Columbia River, and offshore sites along the coast of Washington State. Iodine-129 and other radionuclides are carried by the Columbia River into the Pacific Ocean from the vicinity of the decommissioned Hanford nuclear facility. We find highly elevated $^{129}\text{I}/^{127}\text{I}$ values in the Columbia River, downstream from the Hanford site, but this anthropogenic ^{129}I becomes significantly diluted once it reaches the Pacific Ocean. Nonetheless, the signal persists in surface seawater off the west coast of the U.S. resulting in significantly higher $^{129}\text{I}/^{127}\text{I}$ levels than other surface sites in the Pacific Ocean.

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