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NKS ICP User 2017 Seminar Proceedings

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Final report for NKS-B Nordic ICP (AFT/B(17)4)

NKS ICP User 2017 Seminar

Proceedings

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Abstract

The ICP User seminar was held at Risø, Denmark, during 25-27 September 2017. The seminar consisted of 2-day lectures given by six invited professionals and thirteen seminar participants, as well as 1-day lab training by invited expert. The lectures covered different topics including theoretical principles of ICP, methodology development and application of ICP techniques for different isotopes/isotopic ratios, recent development of new ICP technology and its application, etc. The results obtained from the inter-comparison exercise in 2016 were presented and discussed. The lab training covered demonstration of the operation of ICP-MS, software and technical instructions. The seminar was attended by 51 individuals from 26 organisations.

Key words

ICP-MS, ICP-OES, User, Seminar, Lab training

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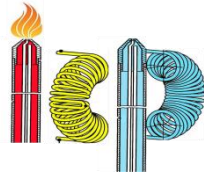
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USER 2017



NKS-B ICP User Seminar
Development and Exploration
of Inductively Coupled Plasma
Spectrometry

25-27 SEPTEMBER 2017

DTU Risø Campus
Frederiksborgvej 399, Roskilde, Denmark

Seminar Sponsors:

Nordic Nuclear Safety Research (NKS), Program B

Thermo Fisher Scientific | www.thermofisher.com

Agilent Technologies | www.agilent.com



Seminar Organizers:

Center for Nuclear Technologies, Technical University of Denmark (DTU Nutech), Denmark

Swedish Defence Research Agency, Sweden

University of Helsinki, Finland

ALS Scandinavia AB, Sweden

Norwegian University of Life Sciences, Norway

Swedish Radiation Safety Authority, Sweden

Organizing Committee:

Jixin Qiao, Technical University of Denmark (Chairman)

Petra Lagerkvist, Swedish Defence Research Agency, Sweden

Susanna Salminen-Paatero, University of Helsinki, Finland

Karl Andreas Jensen, Norwegian University of Life Sciences

Patric Lindahl, Swedish Radiation Safety Authority, Sweden

Szabolcs Osvath, Technical University of Denmark

Iliia Rodushkin, ALS Scandinavia AB, Sweden

Per Roos, Technical University of Denmark

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Introduction

Among different measurement techniques, inductively coupled plasma optical emission spectrometry (ICP-OES) and mass spectrometry (ICP-MS) are the most commonly exploited techniques for determination of trace or ultra-trace stable and radioactive elements, with advantages of short analytical time and multi-element detection. Over the past years, especially ICP-MS has become the technique of choice in nuclear industry, nuclear forensics, radioactive waste disposal and management, environmental monitoring, emergency preparedness and radioecology for providing accurate and precise measurements.

In 2016, an inter-comparison exercise was performed within the NKS-B Nordic ICP project to evaluate the performances and capacities of different types of ICP instrumentations used among the Nordic laboratories for nuclear and radiochemical analyses. Through the inter-comparison exercise, it is clearly shown that each institute has different set-up of ICP instruments and distinct experiences in running them due to different application purposes and technical background of the analysts.

In 2017, within the continued NKS-B Nordic ICP project, a professional seminar (ICP User) was organized during 25-27 September, 2017 at DTU Risø campus, Denmark with the aim to improve collaboration, communication and knowledge sharing among the Nordic countries.

51 individuals from 26 organizations were present at the ICP User 2017 seminar (Appendix 1). The participants were a good mixture of scientists from universities as well as users from industry and authorities.

The seminar programme is listed in Appendix 2 and it consisted of two major parts:

1) 2-day lectures given by 6 invited professionals and 13 seminar participants. The lectures covered different topics including theoretical principles of ICP techniques (plasma generation, measurement techniques, sample introduction, spectral interferences, line selection, and diagnostic studies and fundamental characteristics), methodology development and application of ICP techniques for different isotopes/isotopic ratios, recent development of new ICP technology (e.g., ICP-QQQ) and its application, etc. The results obtained from the inter-comparison exercise 2016 were presented and discussed together with many other results of on-going researches relevant to the application of ICP techniques. Future development of novel ICP technologies and related radiochemical methods was also discussed.

2) 1-day lab training by invited expert. It covered demonstration of the operation of ICP-MS, software and technical instructions.

An abstract book for the seminar was prepared and handed out to each participant at the seminar (Appendix 3). All the presentations at the seminar have been made available on the NKS web site (<http://www.nks.org/en/seminars/presentations/nks-b-icp-user.htm>).

Questionnaires were prepared and filled by participants to evaluate the result of the seminar organization. The overall feedbacks from participants were very positive, see Appendix 4. The participants found the ICP User seminar-to be beneficial to aid education and knowledge sharing among different user and necessary for boosting efficient application of ICP. All participants are looking forward to future activities relevant to the exploration of ICP techniques.

Appendix 1 - List of Participants

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Appendix 2 - Seminar Programme

Welcome Message

Dear colleagues,

I am delighted to welcome you to the NKS-B ICP User Seminar (ICP User 2017). It takes place during September 25-27, 2017 at the cosy Roskilde, Denmark.

This is the first Nordic Nuclear Safety Research (NKS) granted seminar with focus on development and application of inductively coupled plasma (ICP) techniques. The seminar is organised by the Technical University of Denmark, in collaboration with Swedish Defence Research Agency; University of Helsinki; ALS Scandinavia AB; Swedish Radiation Safety Authority and Norwegian University of Life Sciences.

Over the past years, inductively coupled plasma techniques has become the technique of choice in nuclear industry, nuclear forensics, radioactive waste disposal and management, environmental monitoring, emergency preparedness and radioecology for providing accurate and precise measurements. Proficient application of different ICP techniques requires long-term dedicated effort and experiential learning. Facing the worldwide challenges related to nuclear safety, disposal of radioactive wastes, decontamination and decommissioning of closed nuclear facilities, we have seen increased demands for applying ICP techniques in various sectors for effective radiochemical analysis as an alternative of traditional radiometric measurement.

I hope that ICP User Seminar is successful to share knowledge, exchange experience and further promote international collaborations, while you also enjoy the lovely autumn in Denmark!

With best regards,



Dr. Jixin Qiao
Technical University of Denmark
Chairman, ICP-User 2017 Organizing Committee

Program Overview

| Monday, 25th September | Tuesday, 26th September | Wednesday, 27th Sept. |
|--|---|---|
| H. H. Koch, building 112 | H. H. Koch, building 112 | Buildings 202 and 204 |
| | 8:00 Registration | |
| | Radioanalytical method development and comparison (Chairman: Kasper Andersson) | Lab Group 1+2 |
| | 8:30 Jian Zheng (Invited talk 3) | 8:00 Introduction lecture Karl Andreas Jensen |
| | 9:05 Oral-4 Mats Eriksson | 9:30 Coffee break |
| | 9:30 Oral-5 Jixin Qiao | Lab Group 1 |
| | 9:55 Oral-6 Pablo Lebed | |
| 10:00 Registration and coffee | 10:20 Coffee break | |
| Opening session (Chairman: Jixin Qiao) | Application in environmental radioactivity and nuclear forensics (Chairman: Mats Eriksson) | 9:45 Lab training Karl Andreas Jensen Per Roos |
| 11:00 Welcome (Jens-Peter Lynov, Kasper Andersson, Jixin Qiao) | 10:40 Walter Geibert (Invited talk 4) | |
| 11:45 Frank Vanhaecke (Opening lecture) | 11:15 Oral-7 Xiaolin Hou | |
| | 11:40 Oral-8 Youyi Ni | |
| | 12:05 Oral-9 Anna Vesterlund | |
| 12:30 Lunch | 12:30 Lunch | 12:45 Lunch |
| Quadrupole and Sector Field ICP-MS (Chairman: Petra Lagerkvist) | Determination of stable elements (Chairman: Per Roos) | Lab Group 2 |
| 13:30 Douglas Baxter (Invited talk 1) | 13:30 Oral-10 Susanna Salminen-Paatero | |
| 14:05 Oral-1 Ilia Rodushkin | 13:55 Oral-11 Zilvinas Zacharuskas | 13:15 Lab training Karl Andreas Jensen Per Roos |
| 14:30 Oral-2 Per Roos | 14:20 Oral-12 Szabolcs Osvath | |
| 14:55 Seminar photo and Coffee break | 14:45 Coffee break | |
| Triple Quadrupole ICP-MS (Chairman: Ilia Rodushkin) | New instrumental development (Chairman: Susanna Salminen-Paatero) | |
| 15:30 Phil Warwick (Invited talk 2) | 15:10 Mikael Axelsson (Invited sponsor lecture) | 16:15 Wrap-up |
| 16:05 Oral-3 Karl Andreas Jensen | 15:45 Søren Dalby (Sponsor lecture) | |
| 16:30-18:00 Lab tour for participants at Nutech | 16:00 Closing ceremony | |
| 16:30-18:00 Nordic ICP internal committee meeting | | |
| 19:00-21:00 Nordic ICP dinner (Flammen Roskilde) | | |

Seminar Program

MONDAY, 25 SEPTEMBER

10:00 Registration and Coffee

Opening session – Chair: Jixin Qiao

11:00 Welcome speech

(Jens-Peter Lynov, Center for Nuclear Technologies, Technical University of Denmark, Denmark)

11:15 General introduction of NKS and a view on the use of measurement techniques in emergency management

(Kasper Grann Andersson, Technical University of Denmark, Denmark / Nordic Nuclear Safety and Research (NKS))

11:30 General introduction of Radioecology section and the ICP User 2017 program

(Jixin Qiao, Radioecology, Center for Nuclear Technologies, Technical University of Denmark, Denmark)

11:45 Exploiting the isotopic information provided by ICP-mass spectrometry

(Frank Vanhaecke, Department of Analytical Chemistry, Ghent University, Belgium)

12:30 Lunch

Quadrupole and Sector Field ICP-MS – Chair: Petra Lagerkvist

13:30 Developing ICP-MS-based methods for regulatory purposes: Examples from a contract laboratory

(Douglas Baxter, ALS Scandinavia AB, Luleå, Sweden)

14:05 Determination of trace and ultra-trace elements by ICP-SFMS

(Iliia Rodushkin, Division of Geosciences, Luleå University of Technology / ALS Laboratory Group, ALS Scandinavia AB, Luleå, Sweden)

14:30 Analysis of U and Pu isotopes in Thule debris

(Per Roos, Center for Nuclear Technologies (Nutech), Technical University of Denmark (DTU), Denmark)

14:55 Taking the official seminar photo
Coffee break

Triple Quadrupole ICP-MS – Chair: Ilia Rodushkin

- 15:30 Advantages of ICP-QQQ for nuclear waste characterization
(*Phil E Warwick, GAU-Radioanalytical, University of Southampton, UK*)
- 16:05 Spectral interferences on the radionuclides: Method development ICP-MS-MS
(*Karl Andreas Jensen, Centre for Environmental Radioactivity (CERAD CoE), Norwegian University of Life Sciences (NMBU), Norway*)

16:30-18:00

For participants

Lab tour at Nutech
Guides (5-10 people in each group):
Group A. Xiaolin Hou
Group B. Sven Nielsen
Group C. Szabolcs Osvath
Group D. Kasper Andersson

For the committee

Internal meeting at H. H. Koch room
(members: Jixin Qiao
Petra Lagerkvist
Susanna Salminen-Paatero
Karl Andreas Jensen
Per Roos
Mats Eriksson
Ilia Rodushkin
Anna Vesterlund
Emma Engstrom
Patric Lindahl)

19:00-21:00 Nordic ICP dinner – **for all**

[Flammen Roskilde, RO's Torv, Københavnsvej 29, 4000 Roskilde –
2 stops by bus 600S]

TUESDAY, 26 SEPTEMBER

8:00 Registration

Radioanalytical method development and comparison

Chair: Kasper Andersson

8:30 Development and application of radiochemical methods based on the use of ICP-MS instruments

(Jian Zheng, National Institute of Radiological Sciences, National Institute for Quantum and Radiological Science and Technology, Japan)

9:05 Some aspects of analysing mixed U/Pu particles by different mass-spectrometric techniques

(Mats Erikson, Swedish Radiation Safety Authority, SSM, Sweden)

9:30 An inter-comparison exercise on the application of ICP-MS techniques for measurement of long-lived radionuclides

(Jixin Qiao, Center for Nuclear Technologies (Nutech), Technical University of Denmark (DTU), Denmark)

9:55 Radiochemistry laboratory possibilities offered by modern ICP-MS: protactinium, lanthanides and strontium

(Pablo J Lebed, Centre for Environmental Radioactivity (CERAD CoE), Norwegian University of Life Sciences (NMBU), Norway)

10:20 Coffee break

Application in environmental radioactivity and nuclear forensics

Chair: Mats Eriksson

10:40 The specific challenges of short-lived U and Th-series isotope analyses by ICP-MS

(Walter Geibert, Alfred Wegener Institute for Marine and Polar Research, Bremerhaven, Germany)

11:15 Determination of Radionuclides by mass spectrometry

(Xiaolin Hou, DTU Nutech, Risø, Denmark)

11:40 Determination of potentially bioavailable Pu in Japanese rice paddy soils with sector field inductively coupled plasma mass spectrometry

(Youyi Ni, Peking University, China / National Institutes for Quantum and Radiological Science and Technology, Japan)

12:05 Implementing a method for age determination of uranium using the $^{230}\text{Th}/^{234}\text{U}$ chronometer

(Anna Vesterlund, CBRN Defence and Security, Swedish Defence Research Agency (FOI), Sweden)

12:30 Lunch

Determination of stable elements – Chair: Per Roos

- 13:30 Heavy metals (Zn, As, V, Cu, Co, Ni, U, Pb, and Cr) in archived human samples
(Susanna Salminen-Paatero, Department of Chemistry, Radiochemistry, University of Helsinki, Finland)
- 13:55 Using ICP-QQQ to measure precise stable Pb isotope ratios in environmental materials to assess pollution source terms
(Zilvinas Zacharauskas, University of Southampton, National Oceanography Centre, Southampton, UK)
- 14:20 ICP-OES measurement of some transition metals in HF acid media
(Szabolcs Osváth, Center for Nuclear Technologies, Technical University of Denmark, Denmark / National Public Health Institute, Department of Radiobiology and Radiohygiene, Hungary)
- 14:45 Coffee break

New instrumental development – Chair: Susanna Salminen-Paatero

- 15:10 Latest developments in elemental analysis
(Mikael Axelsson, Trace Elemental, ICP-MS and Discrete Analyzers, ThermoFisher Scientific Sweden)
- 15:45 Recent developments of multicollector inductively coupled plasma mass spectrometry (MC-ICPMS) Faraday cup amplifiers.
(Søren Dalby, Isotope Ratio Mass Spectrometry and High Resolution ICP-MS Sales, Nordic and Iceland, Thermo Fisher Scientific)
- 16:00 Closing ceremony

WEDNESDAY, 27 SEPTEMBER

8:00 Introduction lecture for ICP-MS lab training – **for all**
(Karl Andreas Jensen, Centre for Environmental Radioactivity (CERAD CoE),
Norwegian University of Life Sciences (NMBU), Norway)
[In Building 202, meeting room]

9:30 Coffee break

9:45-12:45 Lab training (Karl Andreas Jensen, Per Roos) – **for Group 1**
Free time for Group 2

Lunch [In Building 202, meeting room]

13:15-16:15 Lab training (Karl Andreas Jensen, Per Roos) – **for Group 2**
Wrap up for Group 1

| Group 1 | Group 2 |
|----------------------------|--------------------|
| Annemieke Vos van Avezathe | Mikael Axelsson |
| Hanneke Brust | Lejla Basic |
| Viktoria Damberger | Søren Dalby |
| Alex Hölzer | Violeta Hansen |
| Fabian Köhler | Pentti Hietala |
| Petra Lagerkvist | Maria Karlsson |
| Patric Lindahl | Youyi Ni |
| Satu Meriläinen | Daniela Pittauer |
| Mila Kristiina Pelkonen | Siebren van Tuinen |
| Ilkka Välimaa | Linda Wadman |
| Anna Vesterlund | Yihong Xu |
| Elmo Wiikinkoski | Jian Zheng |
| Zilvinas Zacharauskas | |

Laboratory training

In the practical part of the ICP-MS course, the focus will be on how an ICP-MS works and spectral interferences. Spectral interferences are divided into two groups: Polyatomic and doubly charged. Polyatomic interferences are due to the recombination of sample and matrix ions with Ar and other matrix components such as O, N, H, C, Cl, S, F, etc. For example, $^{40}\text{Ar}^{35}\text{Cl}^+$ will have the same mass/charge (m/z) as $^{75}\text{As}^+$. Doubly charged ion interference is due to doubly charged element isotopes with twice the mass of the analyte isotope. For example, interference from $^{150}\text{Sm}^{++}$ ($m/z=75$) will also interfere on $^{75}\text{As}^+$.

In triple quadrupole ICP-MS (ICP-QQQ) two quadrupoles (Q1 and Q2) are separated by a reaction cell. The analyte and the “on mass” interference exits the Q1 and enters the reaction cell. The use of reactive gases can, in most cases, remove the interference by reaction, reduction or atom transfer. If the analyte has an exothermic reaction with for example oxygen, the mass of the product (M^{16}O^+) can be measured as $\text{Q2} = \text{Q1} + 16$. This is called a mass-shift reaction. If the analyte is unreactive and the interference reactive, the $\text{Q1} = \text{Q2}$. The interference will react to a new m/z , which is different from the analyte m/z . Typical reaction gases are NH_3 , O_2 and H_2 .

In the exercise, the elements of interest will be P, As, Cd, and U. All the elements suffer from spectral interferences. A method for quantifying these elements and isotope ratio measurement for uranium will be developed. The exercise ends with analysis of real samples, interference solutions and a discussion of the results.



Appendix 3 – Abstracts

Exploiting the isotopic information provided by ICP-mass spectrometry

FRANK VANHAECKE

Department of Analytical Chemistry, Ghent University, Belgium

As a mass spectrometric device, an ICP-MS instrument also provides information on the isotopic composition of the elements. This information is, e.g., relied on for assuring absence of spectral overlap – by comparing the relative intensities of the signals for the isotopes of an element (the signal pattern) to the corresponding natural relative abundances – and in mathematical approaches to correct for spectral overlap. This capability also permits the use of isotopic tracers for accurate and precise elemental assay (isotope dilution MS) and for studying the fate of an element in various processes (tracer experiments).

While the applications described above rely on the approximation that all elements have an “invariant isotopic composition in nature”, another group of applications focuses on natural variations in the isotopic composition of the elements. Elements such as Sr and Pb are characterized by the presence of one or more radiogenic nuclides, formed as a result of the decay of naturally occurring long-lived radionuclides, and therefore show a quite pronounced natural variation in their isotopic composition. In addition, *all* elements with ≥ 2 isotopes show natural variation in their isotopic composition due to kinetic and/or thermodynamic isotope fractionation effects accompanying physical processes and (bio)chemical reactions.

The demands in isotope ratio precision for these various types of applications will be addressed. While in less demanding applications, the isotope ratio precision offered by single-collector ICP-MS suffices, the superior precision of multi-collector ICP-MS (MC-ICP-MS) is required for revealing and quantifying more modest isotope ratio variations. As the extent of instrumental mass discrimination, causing a bias between an isotope ratio measurement result and the corresponding true value, is measurably affected by differences in matrix composition in MC-ICP-MS, sample preparation typically includes chromatographic isolation of the target element. In all types of instrumentation, one also needs to be aware of the potential occurrence of spectral interference.

This lecture will not only address the basic principles of isotopic analysis with ICP-MS, but will also cover more ‘adventurous’ applications, e.g., involving the use of alternative sample introduction strategies (such as cold vapor generation and laser ablation), of chemical resolution to overcome isobaric overlap in tandem ICP-mass spectrometry (ICP-MS/MS) or of the hyphenation of an HPLC or GC-unit to the MC-ICP-MS instrument for speciated isotopic analysis.

Isotope ratio applications in archaeology, environmental studies and medical diagnosis will be briefly discussed to illustrate the wide application range of isotopic analysis via ICP-MS.

Developing ICP-MS-based methods for regulatory purposes: Examples from a contract laboratory

DOUGLAS BAXTER, EMMA ENGSTRÖM, DIEKE SÖRLIN, ILIA RODUSHKIN

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When new analytical methods are developed, they can be used to generate data on the turnover, distribution and environmental consequences of elements and compounds. This data can then be used to fuel debate, draft legislation and enforce monitoring programs to ensure public safety or preserve the environment. One example is the measurement of U isotopes in natural water samples by ICP-MS.

Analytical method development ultimately became the driving force behind implementation of EU directive 2013/51/Euratom “laying down requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption” (<http://extwprlegs1.fao.org/docs/pdf/eur128382.pdf>). Indeed, if not for the growing availability and capabilities of ICP-MS it is unlikely that this drinking water directive would have been revised to include monitoring of U-238 and U-234.

In the wake of this revision, the international standards organization recently updated ISO 17294 to extend the scope to the “determination of selected elements including uranium isotopes”. This has, in turn, proven to be a powerful incentive for commercial laboratories to implement the latest version of ISO 17294 among their analytical offerings, since potential customers in all EU member states are now required to demonstrate compliance with the EU directive. So the development of new analytical methods can have a profound impact on the world.

So what are the lessons to be learned from this example? Or in other words, how are analytical methods for regulatory purposes developed?

Determination of trace and ultra-trace elements by ICP-SFMS

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State of the art inductively coupled plasma mass spectrometry (ICP-MS) instrumentation possesses the prerequisite sensitivity to carry out multi-elemental trace analyses at sub-ng l⁻¹ to sub-pg l⁻¹ levels in solution making the technique ideally suitable for determination of trace and ultra-trace elements in wide range of matrices. However, constraints mainly imposed by spectral interferences as well as various sources of contamination ultimately limit the detection capabilities and accuracy of results obtained. Here we review ICP-SFMS (also known as HR-ICP-MS) figures of merit for multi-elemental trace analyses in environmental and clinical matrices.

Analysis of U and Pu isotopes in Thule debris

PER ROOS

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The Thule accident in 1968, northwest Greenland, caused dispersion of actinides and tritium in the environment around the impact point. Several investigations on the magnitude and distribution of the contaminated area have been conducted in the past 40 years, mainly focusing on Pu-isotopes using radiometric determinations via alpha spectrometry. Since the main part of the contamination is made up of micrometer sized particles it is essential to try and determine their composition, morphology and general behavior. Analysis of Pu and U content and spatial distribution in individual Thule-particles has been done previously using SEM/EDX, X-ray tomography and synchrotron based micro-beam techniques and isotopes of Pu and U have been analysed using ICP-MS and SIMS. Due to the low amount of material available in individual particles contamination of particularly uranium from ambient materials is a risk. The analysis of U- and Pu-isotope composition in individual particles using SIMS is an excellent way of minimizing influence from environmental material but isotope ratios obtained have yet been uncertain mainly due to unclear need of correction procedures for oxides and hydrides. Likewise, handling of particles in past ICP-MS analysis of individual particles has likely biased the U-isotope ratios.

The presentation describes some of the analysis done on U- and Pu-isotopes in the Thule material during the last two decades.

Advantages of ICP-QQQ for nuclear waste characterisation

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Nuclear waste characterisation poses significant challenges to the Radioanalytical laboratory. Characterisation requires the measurement of a wide range of radionuclides in diverse matrices and must often be completed to demanding timescales. Mass spectrometry provides an invaluable tool for the quantification of radionuclides, finding particular application to the measurement of long lived radionuclides where mass spectrometry has significant advantages in terms of sensitivity compared with decay counting. Effective and precise measurement of isotope ratios is also of importance in assessing source terms. Recent developments in tandem mass-spectrometry have offered the potential to expand the application of mass spectrometry through improvements in on-line interference suppression and abundance sensitivity. The application of tandem mass spectrometry for radionuclide measurement in the context of nuclear waste characterisation is discussed and demonstrated through the presentation of case studies.

Spectral interferences on the radionuclides: Method development ICP-MS-MS

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and Natural Resource Management (MINA), Norwegian University of Life Sciences (NMBU),
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Detecting radionuclides on an ICP-MS can be challenging due to low concentrations, spectral interferences and poor abundance sensitivity. This presentation will show how to proceed in method development working on an ICP-MS triple quadrupole. The following questions will be addressed:

- How can the interferences be avoided?
- Is it possible to detect a radionuclide with an isobar overlap from another element?
- Does the interference or the analyte have an exothermic reaction with the added gas?
- Which gas is suitable for the element of interest?
- Is it possible to react an element even if the reaction, under standard conditions, is endothermic?

These questions will be answered with examples on $^{239,240}\text{Pu}$, ^{226}Ra with ^{133}Ba as internal standard and ^{236}U .

Development and application of radiochemical methods based on the use of ICP-MS instruments

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In recent years, there has been an increasing demand of ultra-trace determination of radionuclides in studies of nuclear emergency preparedness, safeguards and nuclear forensics, and in environmental studies, such as tracing the sources of radioactive contamination, revealing the transport process of aerosols in the atmosphere, and in study of the ongoing environmental changes, for instance, soil erosion and desertification. Mass spectrometry, especially ICP-MS, as an atom-counting approach that counts the atoms themselves, irrespective of their decay mode, specific activity, or half-life, is gradually replacing/has replaced conventional radiometric methods, such alpha spectrometry, LSC etc. as a mainstream technique for hard-to-detect radionuclides analysis. Due to the strength of argon plasma source of ICP-MS, extremely high, almost 100% ionization efficiency can be achieved for actinides, thus, highly sensitive determination of actinides can be realized. For example, recent development of ICP-MS techniques, especially the sector-field ICP-MS, has pushed the detection limit for analysis of Pu isotopes down to sub-fg level.

To improve the sensitivity of ICP-MS for ultra-trace radionuclide analysis, two approaches can be considered. First, is improvement of the sample introduction efficiency of ICP-MS; for example, the hyphenation of a membrane desolvation system, such as the APEX and Aridus systems, to ICP-MS has proved to be effective for enhancing radionuclide analysis sensitivity by a factor of 5 to 10. APEX and Aridus systems both use a membrane desolvation unit to enhance the sensitivity and lower formation of H₂O-origin background species, such as oxides and hydrides (MO⁺ and MH⁺). The improved sensitivity of the APEX and Aridus sample introduction systems are due to the increase of both sample transport efficiency and the aerosol quality. However, the introduction of dry aerosol sample into the ICP torch by using the membrane desolvation system could result in an enhanced matrix effect. Second is the use of larger sample sizes from 1-5 g to several tens of grams to increase the concentration factor of the target radionuclides in low-level environmental samples. With the increase of sample size, however, reducing the enhanced sample matrix effect is a great challenge for the ICP-MS-based method development, which requires a powerful chemical separation capability.

This lecture will discuss the determination of hard-to-detect radionuclides, Pu isotopes (²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu), ²⁴¹Am and radioactive Cesium isotopes (¹³⁵Cs, ¹³⁷Cs) using SF-ICP-MS and ICP-MS/MS in different environmental samples, such as soil/sediment, seawater and plants. Chemical separation/purification methods for removal sample matrix (for example, separation of alkali and alkaline earth elements and REEs for ultra-trace ¹³⁵Cs and ¹³⁷Cs, and ²⁴¹Am analysis), separation of interfering elements to reduce/eliminate polyatomic and isobaric interferences (for instance, separation of U, Sb, Mo, Sn, Bi, Pb, Tl, Hg for Pu, radioCs and ²⁴¹Am analysis), will be highlighted.

Some aspects of analysing mixed U/Pu particles by different mass-spectrometric techniques

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Analysis of actinide particles have applications in both radiological and safeguard studies with a focus on the determination of isotopic and nuclide ratios. These ratios are essential in the characterisation of the sources from where the particles originates. There is a challenge in analysing micro-meter sized particles typically containing some nano-gram of material. We will give some examples on different mass spectrometric analysis of such particles. Advantages and challenges with the three different techniques ICP-MS, SIMS and AMS in such analysis will be highlighted.

An inter-comparison exercise on the application of ICP-MS techniques for measurement of long-lived radionuclides

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Inductively coupled plasma (ICP) spectrometry techniques are widely used in the fields related to environmental monitoring, nuclear waste disposal and management, radioecology and tracer studies, as well as nuclear forensics and nuclear emergency preparedness. Especially ICP mass spectrometry (ICP-MS) is playing an important role for determination of low-level long-lived radionuclides and their isotopic ratios.

Within the Nordic-ICP project, an inter-comparison exercise was performed during 2016, which was focused on the measurement of uranium and plutonium isotopes in certified reference material by ICP-MS in combination with radiochemical separation. The participating institutes in this inter-comparison exercise included FOI CBRN Defence and Security, Sweden; Center for Nuclear Technologies (Nutech), Technical University of Denmark (DTU), Denmark; Laboratory of Radiochemistry, University of Helsinki (UH), Finland; Centre for Environmental Radioactivity, Norwegian University of Life Sciences (NMBU) Norway; ALS Life Sciences Division, ALS Scandinavia AB, Luleå, Sweden, and Department of Chemistry, Norwegian University of Science and Technology (NTNU), Norway. This presentation summarizes the results and conclusions obtained base on this inter-comparison exercise.

Radiochemistry laboratory possibilities offered by modern ICP-MS: protactinium, lanthanides and strontium.

PABLO J LEBED

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Current research efforts in development at CERAD/NMBU, Norway that combine radiochemistry and modern ICP-MS will be discussed.

Protactinium-231. Efforts on the development of microwave-digestion extraction methods of uranium ores, description of the elution and retention behaviour of protactinium on modern commercially available extraction chromatography resins and capabilities of tandem ICP-MS will be presented.

Lanthanides fingerprints. The presence of fission products, which include numerous isotopes of lanthanides, can change the isotopic ratios of these elements in the environment. Isotopic ratios were measured in San Joaquin soil (NIST-2709a), an area with little contamination of nuclear origin. Samples collected from three sites with known nuclear activities (Fangataufa Lagoon in French Polynesia, Chernobyl and the Ottawa River near Chalk River Laboratory) were analysed and all exhibited altered isotopic ratios for $^{143/145}\text{Nd}$, $^{147/149}\text{Sm}$, and $^{151/153}\text{Eu}$.

Strontium-90. The combined performance of a sample clean-up method of contaminated fish bones from Glubokoye, Ukraine using extraction chromatography resins combined with the new Agilent ICP-MS 8900 will be discussed and compared with Sr-90 radiometric and ICP-MS 8800 measurements.

The specific challenges of short-lived U and Th-series isotope analyses by ICP-MS

WALTER GEIBERT

Alfred Wegener Institute for Marine and Polar Research, Bremerhaven, Germany

The analysis of many radioactive decay products of the natural uranium and thorium decay chains has undergone a revolution in the past two decades. With increasing ion transmission of ICP-MS systems, ever smaller quantities of the analytes are required, shifting standard measurement techniques from traditional decay- based counting techniques towards ICP-MS.

Smaller absolute quantity requirements translate to isotopes of shorter half-life entering the analytical window. However, a range of specifics of ICP-MS systems as well as elemental properties need to be considered for accurate and precise analyses of isotopes with short half-lives. Examples for important practical aspects to consider are the availability of spikes for isotope dilution techniques, measuring extreme isotope ratios that are challenging abundance sensitivities, interferences from more abundant elements and isotopes, or demanding sample processing steps.

This talk will give an overview of the limiting parameters for uranium-234, protactinium-231, thorium-230, actinium-227 and radium-226 and -228 isotopes by ICP-MS, discussing the individual challenges of each isotope from a practical perspective, possible analytical solutions and future perspectives. A particular focus of the presentation will be placed on examples from the marine environment.

Determination of potentially bioavailable Pu in Japanese rice paddy soils with sector field inductively coupled plasma mass spectrometry

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Plutonium (Pu) in the environment has caused public concern due to its radiotoxicity and long half-lives of its main isotopes ²³⁹Pu (T_{1/2}=24110 years) and ²⁴⁰Pu (T_{1/2}=6563 years). The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident has once more highlighted the necessity to study the potential bioavailability of Pu in cultivated soils since it governs the transfer of Pu from soil to crops therefore relates much to food safety.

The bioavailability of a radionuclide is largely influenced by its physicochemical form (or speciation) presented in soil. As it's often impractical to directly determine radionuclides' speciation(s) in an environmental level, a general solution is to classify them by extraction with certain types of reagents, resulting in operational fractions.

In this work, we investigated the level of water soluble fraction and exchangeable fraction of Pu in Japanese rice paddy soils. These two fractions are weakly associated to soil particles thus might be potentially bioavailable for rice plant uptake. H₂O and 1M NH₄OAc solution were employed successively to extract Pu from rice paddy soils. Different pretreatments of the extracted solution were compared. The whole chemical procedure consists of H₂O/NH₄OAc extraction, HNO₃-H₂O₂ wet digestion, Fe(OH)₃ co-precipitation and a two-stage ion exchange chromatography. Pu in the final sample was measured with SF-ICP-MS (Element XR) equipped with APEX introduction system.

The water soluble fraction of Pu in Japanese rice paddy soils ranged from 0.01% to 0.22%, while the exchangeable fraction was between 0.01% and 0.08%. The sum of these two fractions (0.03% to 0.27%) was assumed to be potentially bioavailable for rice plant. Complete sequential extraction was conducted to identify the association of Pu in various physicochemical forms. Organic matter was the most important sink of Pu in rice paddy soils investigated in this study with more than 50% of Pu associated with them and followed by the residual fraction. The percentage of Pu associated with these two least labile fractions was in total over 93%, partly explained the low potential bioavailability of Pu in Japanese rice paddy soils.

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Implementing a method for age determination of uranium using the $^{230}\text{Th}/^{234}\text{U}$ chronometer

ANNA VESTERLUND, PETRA LAGERKVIST

Swedish Defence Research Agency – CBRN Defence and Security

One aim of nuclear forensics is to establish the origin of nuclear and other radioactive material out of regulatory control. For uranium, the age, i.e. the time passed since last chemical separation, can be a useful discriminator between different materials and may give a hint about from where the material originates.

^{230}Th is a daughter nuclide in the uranium series and therefore the amount of this nuclide is dependent on the age of a uranium material and the age can be determined by measuring the $^{230}\text{Th}/^{234}\text{U}$ ratio. If the material is relatively young, i.e. the time since last separation is short, the ^{230}Th content is low and therefore sensitive measurement techniques such as ICP-SFMS is suitable for this application.

The aim of this presentation is to show the implementation of a method for age determination of uranium at the Swedish Defence Research Agency using isotope dilution and provide some results and reflections on the method.

Heavy metals (Zn, As, V, Cu, Co, Ni, U, Pb, and Cr) in archived human samples

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A sample set of human livers, lungs, bones (ribs), flesh and lymph nodes was analyzed and concentration of heavy metals (Zn, As, V, Cu, Co, Ni, U, Pb, and Cr) was determined in different organs. This work was continuing previous investigations of enrichment of radionuclides and heavy elements along the food chain “lichen – reindeer – man” in Finland (Jaakkola et al. 1981, Mussalo-Rauhamaa 1981).

The samples were obtained from autopsies of 67 persons, who died in 1976-1979. The samples were dried at 105 °C for 2 days. Ribs, lungs, and livers were ashed in an oven at 450 °C overnight. The dried and/or ashed samples were digested in conc. HNO₃ on a hot plate for 6 hours. H₂O₂ was added dropwise to the samples for ensuring the complete oxidation of the samples before the last hour of wet-ashing. The solutions were filtered and diluted with mQ-water for making measurement samples of 5% - 33% HNO₃ (v/v). The concentrations of heavy metals were determined with Agilent 4100 Microwave Plasma-Atomic Emission Spectrometer.

In general, heavy metal concentrations had the same magnitude as other reported values in human samples (Kollmeier 1990, Verma 2013, Kathren & Tolmachev 2015), although 10-fold variation may appear among concentration of a certain heavy metal (Table 1). Cobalt was not detected in any of the samples.

Table 1. Concentration of heavy metals in human samples (ng/g wet weight). The number of representatives of each sample type is in parentheses.

| | Zn | As | V | Cu | Ni | U | Pb | Cr |
|-----------------|--------|-----------|---------|---------|---------|-----------|-------------|-----------|
| liver (2) | 79±16 | | 2.4±0.5 | 6.4±1.3 | 1.3±0.3 | 2.9±0.6 | | |
| | - | - | 5.2±1.0 | - | - | - | - | - |
| | 141±28 | | | 7.9±1.6 | 1.6±0.3 | 4.5±0.9 | | |
| lung (16) | < DL | < DL | 1.3±0.3 | < DL | < DL | 0.7±0.1 | < DL | < DL |
| | - | - | - | - | - | - | - 0.99±0.20 | - |
| | 52±10 | 0.11±0.02 | 13±3 | 2.0±0.4 | 3.7±0.8 | 7.3±1.5 | | 14±3 |
| lymph nodes (7) | < DL | < DL | 3.0±0.6 | < DL | < DL | < DL | | < DL |
| | - | - | - | - | - | - | - | - |
| | 29±6 | 0.07±0.01 | 45±9 | 6.8±1.4 | 1.6±0.3 | 3.4±0.7 | | 2.6±0.5 |
| ribs (3) | < DL | | 3.1±0.6 | | | 0.18±0.04 | | |
| | - | - | - | - | - | - | - | - |
| | 15±3 | | 4.1±0.8 | | | 0.46±0.09 | | |
| flesh (2) | 88±18 | | 1.5±0.3 | 1.6±0.3 | < DL | 0.97±0.19 | | < DL |
| | - | - | - | - | - | - | - | - |
| | 165±33 | | 1.7±0.4 | 2.2±0.5 | 0.5±0.1 | 2.2±0.4 | | 0.13±0.03 |
| collarbone (1) | 99±20 | 0.04±0.01 | 1.9±0.4 | 1.9±0.4 | 1.4±0.3 | 2.7±0.5 | - | 0.24±0.05 |

A larger sample set is needed after these preliminary test samples, to get better statistics for concentration values and variation.

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Using ICP-QQQ to measure precise stable Pb isotope ratios in environmental materials to assess pollution source terms

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Traditionally, high precision Pb isotopic analysis has been carried out using thermal ionisation MS (TIMS) and multi-collector MS (MC-ICPMS) instruments with stable isotope spiking being used to correct for isotopic mass fractionation. In this study the Agilent 8800 tandem mass spectrometer (ICP-QQQ) was investigated to evaluate its capability in determining precise Pb isotope data for environmental samples. Although the instrument is equipped with two quadrupole mass filters positioned between a collision/ reaction cell and this design is well suited for interference removal it was decided instead to purify samples to optimise measurements. Any potential interferences, such as Hg-204 on Pb-204, were therefore eliminated through chemical separation.

It is well established that the relative abundance of the four stable Pb isotopes can be used to infer lead pollution source terms from industrial and other anthropogenic processes (e.g. lead ore extraction, lead pipes used for water supplies, lead paint, lead added to gasoline). The addition of tetraethyl lead to gasoline as an anti-knock agent was widespread from the 1920s to the 1980s and had a significant environmental impact. Regulatory pressures eventually led to reductions and final elimination of leaded gasoline in the USA and Europe driven by cogent evidence of proven neuro-toxicological effects.

A set of sediment core samples collected from UK saltmarshes and lakes were chosen to study temporal variations in Pb isotopes. These were investigated using a Thermo Neptune MC-ICPMS to evaluate the potential of the ICP-QQQ as a suitable tool for isotope ratio analysis. All sample handling and chemical processing was carried out in HEPA filtered workstations to limit any environmental contamination. Sub-boiled reagents were used for all digestions and chemical separations. Samples were first leached with aqua regia for 8 hours then centrifuged. Small aliquots of leachate were evaporated and converted to bromides using sub-boiled HBr before separating the Pb using an HBr-anion exchange procedure.

Temporal variations in Pb isotopic composition were determined for sequences of dated sediment cores. The profiles from Southampton saltmarshes and Windermere (English Lake District) reveal distinct variations relating to major historical events and known changes in source terms.

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ICP-OES measurement of some transition metals in HF acid media

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A method for determination of ^{93}Mo and ^{94}Nb from nuclear power plant wastes (stainless steel samples) is under development in our laboratory. (As a long-term goal, method could be extended for determination of ^{93}Zr as well.)

These radionuclides have not any easy-to-determine gamma-emitting isotopes; except of ^{95}Zr - ^{95}Nb , which were very convenient to use, but in the absence of a working nuclear reactor, they would be very expensive to purchase. This is why the stable isotopes of natural composition of these analytes are used for the preliminary experiments of method development; and their concentrations are measured using ICP-OES.

The main difficulty is that HF acid is practically always needed when dissolving and separating these elements, so it is present in all of the samples to be measured. However, HF damages the glassware of the ICP-OES equipment. To avoid this damage, dilution, evaporation and complexation can be used.

As some papers (e. g. [1]) do not take care of presence of F^- when its concentration is below 0.005 M, dilution might be used in certain cases (typically when HF concentration is not much higher than the given level). But it also decreases the concentration of the analytes, so it cannot be widely used.

HF can be eliminated by evaporation easily, but the residue is typically impossible to be taken up in any other solution, as Nb and Zr are “fluorophile” elements [2], which are unstable in a solution without F^- . However, evaporation can be used to decrease the amount of F^- significantly.

For complexation of excess F^- , boric acid can be used:



In this case it has to be taken into account, that boric acid causes false signals (for example at 201.512 nm line of Mo, 343.823 nm line of Zr, 344.199 nm line of Mn, 230.299 nm line of Ni, 234.830, 273.358 and 373.713 nm lines of Fe).

Some experiments performed with simulated samples and stainless steel samples as well as their measurements by ICP-OES will be presented and discussed.

References:

1. Prog Nucl En 93 (2016) 362-370
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Latest developments in elemental analysis

MIKAEL AXELSSON

Trace Elemental, ICP-MS and Discrete Analyzers, Thermo Fisher Scientific, Sweden

This presentation will focus on our latest innovation in trace elemental analysis, the Thermo Scientific iCAP™ TQ ICP-MS, a triple quadrupole instrument. The presentation will describe the iCAP TQ hardware and briefly the software and will discuss various applications of the instrument, including trace analysis in metallurgical samples, enhanced accuracy analysis of As and Se in environmental samples and low level detection of Ti in biological samples for biomedical research applications. Some benefits of going high resolution ICPMS will also be covered.

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S C I E N T I F I C

Recent developments of multicollector inductively coupled plasma mass spectrometry (MC-ICPMS) Faraday cup amplifiers

SØREN DALBY

*Isotope Ratio Mass Spectrometry and High Resolution ICP-MS Sales, Nordic and Iceland,
Thermo Fisher Scientific, Stamholmen 193, 2650 Hvidovre, Denmark*

One of the major challenges in Geosciences is the analysis of small ion beams. Especially in studies that focus on isotope compositions of scarce materials or materials that have ultra-low concentration of the isotopes of interest, but also in studies that aim to resolve isotopic variability on a small spatial scale. Ultimately, the analytical precision of such studies is limited by the detection system of your mass spectrometer. Thermo Fisher Scientific™ has developed a new design of amplifiers equipped with 1013 Ohm Amplifiers, a revolutionary new resistor design guarantees fast response times and extremely low noise/signal characteristics. Here we present newly developed amplifiers equipped with 1013 Ω feedback resistors for the Thermo Scientific™ Neptune Plus™ MC-ICP-MS product line. A revolutionary new current amplifier design compensates for settling time effects observed with high-gain current amplifiers using high ohmic resistors in the feedback loop. The new design concept guarantees fast response times and very low noise characteristics.

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S C I E N T I F I C

Important places and where to find them

Lectures & committee meeting: Building 112, H. H. Koch room



Introduction lecture to lab training: Building 202, meeting room

Laboratory training: Building 204, laboratory room S15-17



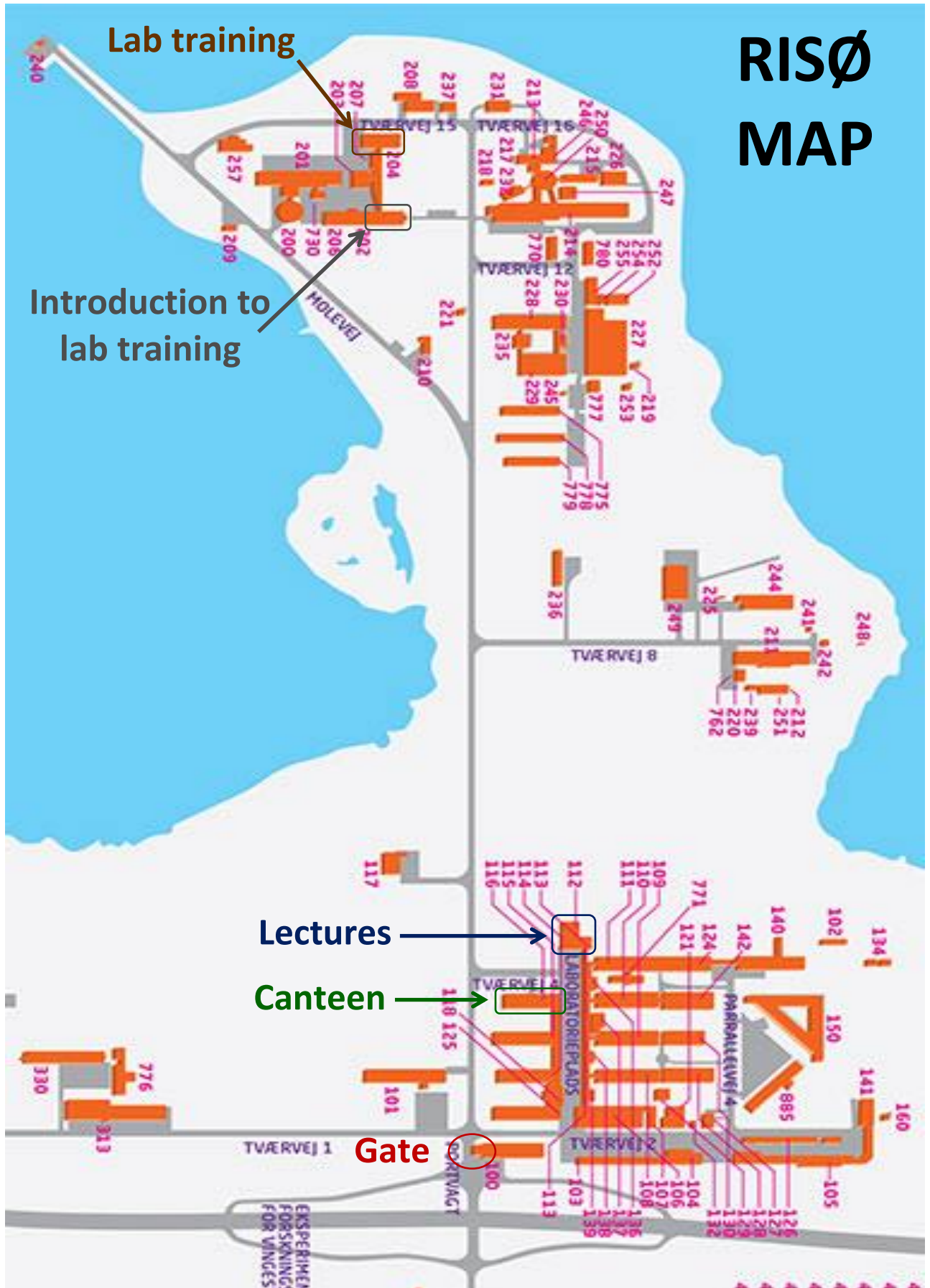
Lunches on Monday and Tuesday: Building 116, Canteen



Flammen Roskilde (Nordic ICP dinner): RO's Torv, Københavnsvej 29, 4000 Roskilde
Take a southbound bus 600S (towards Hundige St.) and take off at Ro's Torv (2 stops, ca. 12 min.). If you are lost, call Jixin Qiao (+45-2179-8724) or Szabolcs Osvath (+36-20-824-9647).



RISØ MAP



Lab training

Introduction to
lab training

Lectures

Canteen

Gate





Introduction of Radioecology Section of DTU Nutech

Characterization of decommissioning materials — All in one place

DTU Nutech is capable of conducting all steps of sample processing, data analysis and customer support. Our state-of-the-art methods have been applied successfully for the determination of difficult-to-measure radionuclides including ^3H , ^{14}C , ^{36}Cl , ^{45}Ca , ^{55}Fe , ^{63}Ni , ^{65}Ni , ^{90}Sr , ^{99}Mo , ^{90}Zr , ^{90}Nb , ^{99}Tc , ^{129}I , ^{210}Po , ^{210}Pb , ^{226}Ra , ^{237}Np , ^{234}U , ^{235}U , ^{236}U , ^{238}U , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{241}Am and ^{244}Cm as well as gamma emitters in various samples from nuclear decommissioning.

Outstanding sample capacity

The laboratories include facilities suited for chemical and radiochemical analytical purposes. The facilities include the following equipment:

- 32 x low-level alpha spectrometers
- 8 x low-level gamma spectrometers
- 35 x low-level gas flow beta counters
- 1 x TriCarb and 1 x Quantulus, low-level LSCs
- 1 x ICP-OES and 1 x ICP-MS



Analysis under Accreditation — Clear and Transparent

Since 2012 the radioanalytical laboratory at DTU Nutech carries out radiochemistry and chemical testing for selected elements and radionuclides under accreditation standard DS/EN ISO/IEC 17025:2005.

Training on radiochemistry and radioecology

DTU Nutech offers intensive courses to train students and laboratory staff with technical knowledge and skills to develop sampling, sample preparation and analytical protocols for analysis of radioisotopes of interest. Moreover, the courses are individually tailored to meet the participants' needs.



DTU Nutech:

DTU Nutech is the national competence center for nuclear technologies conducting research in the following scientific areas:

- Radioecology (environmental radioactivity and associated assessments)
- Radiochemistry and radiopharmacy
- Emergency preparedness
- Luminescence physics
- Ionizing radiation dosimetry
- Neutronics (neutron transport through materials)
- Isotope production and accelerator technology

DTU Nutech has a long tradition of transferring research results into numerous services to authorities, hospitals, industry and media. The Center has a solid, international position in environmental research based on nuclear technologies. It has vast experience in characterization and radiochemical analyses of various waste materials and environmental samples from nuclear decommissioning activities.

Analyses we offer in the Radioecology Section:

- 1) Expertise since 1956 on characterization and radiochemical analyses of materials from nuclear facilities
- 2) Complete analytical services including sample preparation, analyses, reporting and advisory support
- 3) Customized analyses to meet individual requirements
- 4) Analytical work performed under accreditation (ISO/IEC 17025:2005)
- 5) Comprehensive training programmes in radiochemistry and radioecology

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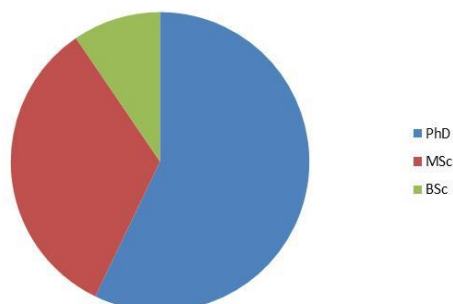


USER 2017

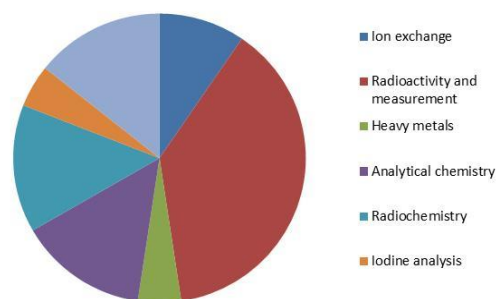
Appendix 4 - Questionnaire Summary

After the NORDIC ICP workshop, the participants were asked a number of questions that were thought to be helpful to know the answers to, when planning the way forward for further integration and use of ICP technologies in the Nordic countries. A summary of the questions and answers is given below.

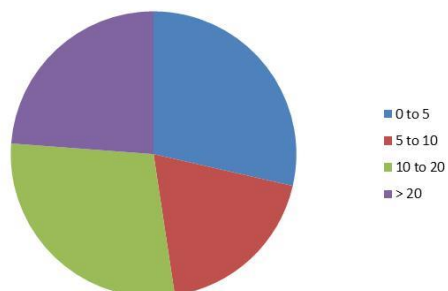
Question: What is the level of your highest education?



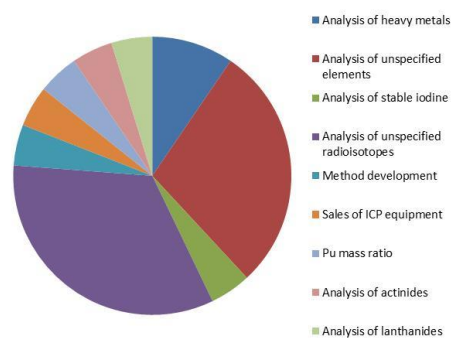
Question: Which work area do you specialise in?



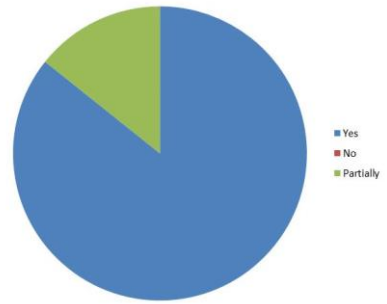
Question: How many years work experience do you have with this?



Question: How do / would you use ICP techniques in your work?



Question: Did the workshop meet your expectations?



Question: Did the lectures give you new knowledge?

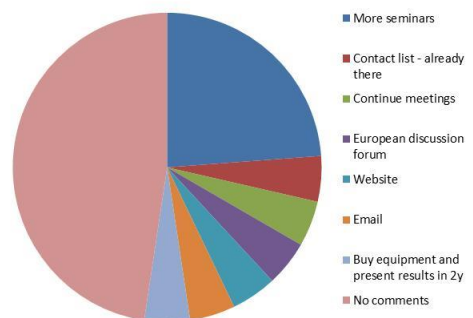


Question: Where do you see the future of ICP work?

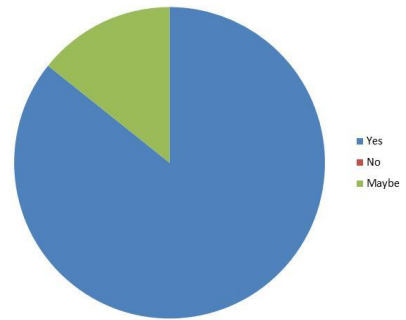
AREAS MENTIONED:

- New applications
- More precise measurements
- Development of even faster routine methods
- Creating larger databases
- Speciation
- Radio decommissioning
- It will expand and grow and become more available
- QQQQ
- Sector field instrument with collision cell
- Increasing amounts of analyses of radioisotopes

Question: What would be the best way to follow up?



Question: Did you make new useful contacts?



Question: Did you find the seminar well arranged?

Answer: yes (100%)

Question: Was the amount of lab work OK?

Answer: yes (55%), no reply (45%), no (0%)

Question: Would you consider writing an NKS funding application?

Answer: yes from 60% of the eligible participants

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|----------------------------------|---|
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| No. of references | 0 |
| Abstract max. 2000 characters | The ICP User seminar was held at Risø, Denmark, during 25-27 September 2017. The seminar consisted of 2-day lectures given by six invited professionals and thirteen seminar participants, as well as 1-day lab training by invited expert. The lectures covered different topics including theoretical principles of ICP, methodology |

development and application of ICP techniques for different isotopes/isotopic ratios, recent development of new ICP technology and its application, etc. The results obtained from the inter-comparison exercise 2016 were presented and discussed. The lab training covered demonstration of the operation of ICP-MS, software and technical instructions. The seminar was attended by 51 individuals from 26 organisations.

Key words

ICP-MS, ICP-OES, User, Seminar, Lab training