NKS-489
ISBN 978-87-7893-585-4

NKS-B RadWorkshop 2024 proceedings

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November 2024

Abstract

The NKS-B RadWorkshop 2024, held during 9-13 September 2024, at the DTU Risø Campus in Roskilde, Denmark, brought together 104 participants from nearly 60 organizations in 20 countries to focus on advancements in radioanalytical chemistry and its application in various fields. The workshop featured a blend of 3-day keynote lectures, presentations, posters and 2-day hands-on laboratory training, covering topics such as radiochemical separation techniques, measurement methods, and applications in environmental monitoring, nuclear decommissioning, emergency preparedness, radioecology and tracer studies. The event successfully enhanced the education and expertise of attendees, fostering international collaboration and innovation in the field.

Key words

Radioanalytical chemistry, workshop, environmental monitoring, nuclear decommissioning, emergency preparedness, radioecology, tracer studies

NKS-489
ISBN 978-87-7893-585-4
Electronic report, November 2024
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Final report for NKS-B Radworkshop 2024 (AFT/B(24)2)

NKS-B RadWorkshop 2024 proceedings

Workshop on radioanalytical chemistry for environmental monitoring, nuclear decommissioning, emergency preparedness, radioecology and tracer studies

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Acknowledgements

The project partners of NKS-B RadWorkshop 2024 wish to express their profound gratitude to NKS for the funding support. We also extend our thanks to all the participants, collaborators, and our generous sponsors—GammaData Instrument AB, metorX B. V., PerkinElmer, Thermo Fisher Scientific, and Triskem International.

NKS conveys its gratitude to all organizations and individuals who, through direct financial support or in-kind contributions, have made the work presented in this report possible.

Disclaimer

RadWorkshop 2024

Workshop on Radioanalytical Chemistry for Environmental Monitoring, Nuclear Decommissioning, Emergency Preparedness, Radioecology and Tracer Studies

9-13 September 2024

DTU Risø Campus
Frederiksborgvej 399, Roskilde, Denmark

Workshop Organizers:

Technical University of Denmark (DTU)
Norwegian Life Science University (NMBU)
University of Helsinki (UH)
Swedish Food Agency (SLV)
Norwegian Radiation and Nuclear Safety Authority (DSA)
Radiation and Nuclear Safety Authority, Finland (STUK)

Organizing Committee:

Jixin Qiao, DTU (Chairman)
Karl Andreas Jensen, NMBU
Lindis Skipperud, NMBU
Ole Christian Lind, NMBU
Susanna Salminen-Paatero, UH
Marion Grange, SLV
Maria Kaipiainen, STUK
Aslak Roalkvam Skåra, DSA
Torbjørn Gäfvert, DSA
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Matic Dokl, DTU

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Table of Contents

Abstract

The NKS-B RadWorkshop 2024, held during 9-13 September 2024, at the DTU Risø Campus in Roskilde, Denmark, brought together 104 participants from nearly 60 organizations in 20 countries to focus on advancements in radioanalytical chemistry and its application in various fields. The workshop featured a blend of 3-day keynote lectures, presentations, posters and 2-day hands-on laboratory training, covering topics such as radiochemical separation techniques, measurement methods, and applications in environmental monitoring, nuclear decommissioning, emergency preparedness, radioecology and tracer studies. The event successfully enhanced the education and expertise of attendees, fostering international collaboration and innovation in the field.

Key words

Radioanalytical chemistry, workshop, environmental monitoring, nuclear decommissioning, emergency preparedness, radioecology, tracer studies

Introduction

A number of nuclear (e.g., nuclear weapon tests, nuclear power plants, spent fuel reprocessing, nuclear accidents/incidents) and non-nuclear industrial (e.g., mining of uranium and other elements, geothermal installations, oil and gas production [1,2]) activities and legacies have increased the dispersion of natural and artificial radionuclides in the environment. Current situation in Europe is also sensitive due to the recent events in Ukraine, where radioactive substances were measurable in many Nordic and European countries. The dispersed radionuclides are continuously transferred into the food chain through biological processes. Continuous and accurate monitoring of radioactive contaminants in the environment and food products is important to meet the increasing regulatory demands in radiation protection and food safety.

Many nuclear reactors have already reached their lifetime of 30-40 years. Europe is predicted to become the world's largest market for Decontamination & Decommissioning by 2025. Over half of the European Union's 223 nuclear power plants are due to close by 2025, requiring expenditure on subsequent decommissioning of €60BN+ [3]. In recent years, more and more nuclear reactors in Nordic countries are under decommissioning or going to be decommissioned. According to Nuclear Energy Agency [4], radiological characterization efforts are needed during all stages of a nuclear facility's life cycle, in order to plan and perform a safe and efficient decommissioning. On the other hand, many countries in Europe (e.g., UK, Finland) and Asia (e.g., China, India) have set ambitious targets to grow nuclear capacity [5]. Upon national and international regulations, comprehensive environmental monitoring has to be established within and around each nuclear facility to ensure a safe operation.

The COVID-19 pandemic had a serious impact on our health, livelihoods and economies. It has created a moment to contemplate our capacity for handling emergencies similar to COVID-19 in the future, including nuclear emergencies. Fast and correct countermeasures from decision makers to mitigate radiation risk and economic loss rely on establishment of powerful analytical tools to quantify radioactive contamination to people, environment and food.

In contrast to most gamma emitters (so-called easy-to-measure (ETM) radionuclides), alpha and beta emitters (sometimes low concentration gamma emitters) cannot be quantified directly due to interferences from sample matrix and other radionuclides. They are so-called difficult-to-measure (DTM) radionuclides, which often require complex chemical purification

prior to the measurement [6]. To facilitate sound environment/food monitoring, optimize waste characterization and support radioecology and tracer studies, it requires rapid and cost-effective determination of both ETM and DTM radionuclides. In all cases, the high capacity and multi-element measurement ability is needed to ensure high sample throughput with reliable analytical results.

With the increased number of nuclear related activities, Nordic countries are facing increased challenges and needs to boost radioanalytical capacities, especially for DTM analysis. To cope with these demands, many students and young scientists from other disciplines, as well as staff working at the nuclear facilities have joined the community involving radiochemical analysis. Many organizations including nuclear industries, radiation protection authorities, research institutes and universities are taking part in method development for radiochemical analysis of a wide range of DTM radionuclides.

Even though there are already some methods and experiences available from previous efforts in Nordic countries, new challenges in different application areas are continuously appearing. Therefore, no uniform radiochemical methods can apply to all different situations. Specific method development is necessary with focus on individual analytical needs and obstacles. In the recent years, demands in determining DTM radionuclides, e.g., several new-appearing long-lived radionuclides (e.g., ^{93}Zr, ^{93}Mo, ^{79}Se, ^{126}Sn, ^{135}Cs in nuclear decommissioning, ^{236}U and ^{233}U in environmental tracer studies) are increasing steadily [7]. Development of effective radioanalytical methods is necessary in combination of modern measurement techniques, e.g., triple quadrupole inductively coupled plasma mass spectrometry (ICP-MS/MS) [8] and compact (0.3 MV) accelerator mass spectrometry (AMS) [9], triple-double coincidence ratio liquid scintillation counting (TDCR-LSC) [10], as well as automated radiochemical methodology by coupling flow-based system with on-line detection systems [11].

To enhance radioanalytical competences, there is a need for communication, knowledge/experience sharing and cooperation among different parties in the Nordic nuclear communities. This requires collective inputs from stakeholders, regulators and researchers to better foresee the challenges in radiochemical analysis and develop fit-for-purpose methods which can be used in real situations. The students and young scientists need to actively gather knowledge and experience from the senior experts in the field and seek for education and training opportunities in order to quickly cope with the demands in the work.

With this aim, we organized a one-week workshop (RadWorkshop 2024) at DTU Risø Campus, Denmark, during 9-13 September 2024. The workshop attracted a diverse group of 104 participants from nearly 60 organizations in 20 countries worldwide (Appendix 1). The audiences were a good mixture of all parties involved in the field including regulators, operators, service partners, researchers, young scientists and students. This international gathering provided a unique platform for knowledge exchange and professional development.

The workshop's program (Appendix 2) was designed to address key challenges and innovations in radiochemistry. The RadWorkshop 2024 consisted of two major parts:

1) 3-day lectures given by 10 invited professionals and 24 seminar participants. 7 posters were also presented at the workshop. The presentations covered a broad spectrum including theoretical principles of radiochemistry and measurement techniques (alpha, beta and gamma), recent advancement in separation techniques, materials and detection instruments. Specific highlights included ultra-trace analyses with AMS, optimizing ICP-MS/MS for radionuclide measurement, applications of radioanalytical chemistry in nuclear decommissioning, environmental tracer applications and emergency preparedness.

2) 2-day lab training by experienced professionals, with nearly 45 participants engaging in this hands-on session. The lab training focused on the radiochemical analysis of Sr-90 and advanced measurement techniques using ICP-MS/MS, as well as liquid scintillation counting (LSC) & beta counting. This session provided participants with practical skills and in-depth knowledge on advanced analytical techniques, specifically in the use of triple quadrupole ICP-MS.

An abstract book for the seminar was prepared and handed out to each participant at the seminar (Appendix 3). The photos (Appendix 4) and presentations at the workshop have been made available on the NKS web site (https://www.nks.org/en/seminars/presentations/nks-b-radworkshop-2024.htm).

The overall feedbacks from participants were very positive. The participants found RadWorkshop 2024 to be an excellent forum to exchange experience, discuss challenges, seek solutions and potentially form new collaborative projects. The lab practical training was beneficial to the participants, particularly for young students and scientists, providing valuable hands-on experience in ICP-MS and radioanalytical methodology development. All participants are looking forward to future activities relevant to the Radioanalytical Chemistry. Some aspects and findings of the event will be presented in a future conference.

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Appendix 1 - List of Participants

First name	Last name	Affiliation
Aaron	Lehnert	Leibniz University Hanover, Institute of Radioecology and Radiation Protection
Alessandro	Nozza	ARPA Lombardia - Milan - Italy
Angela	Hj Tan	NEA
Anna	Banel	Institute for Energy Technology, Kjeller, Norway
Annemieke	Vos van Avezathe	wageningen food safety research
Antje	Ebert	PerkinElmer
Anu	Bhaisare	National Physical Laboratory
Aslak	Roalkvam Skåra	DSA
Camillo	Bruhn	Institutt for energiteknikk
Casper	Boe Jensen	Danish Decommissioning/Dansk Dekommissionering
Charlotte	Scheutz	DTU
Chiaoting	Lo	Taiwan Power Company
Christian	Vig Dalsgaard	DTU
Christina	Pannach	DTU
Dag	Stinus	DTU
Dave	Bubberman	Wageningen Food Safety Research
Dina	Rindorf	DTU
Duoqiang	Pan	Lanzhou University
Eike	Rades	DTU
Eirik	Gundersen	Gammadata Instrument AB
Elżbieta Anna	Stefaniak	European Commission Joint Research Centre JRC-Geel
Esperanza	Lara	CIEMAT
Filip	Jedrzejek	AGH University of Krakow
Francisco Javier	Guillén Gerada	University of Extremadura
Franck	Dal Molin	Centre for Environment, Fisheries and Aquaculture Science, UK
Galia	Selikowitz	DSA
Georg	Steinhauser	TU Wien
Guillaume	Lutter	DTU
Habacuc	Pérez-Tribouillier	Laboratory for Ion Beam Physics, ETH Zürich
Hanne	Wojtaszewski	DTU
Helena	Fajfar	JSI
Hyuncheol	Kim	Korea Atomic Energy Research Institute
Ines	Llopart-Babot	Triskem International
Ingunn	Isdahl	Institutt for energiteknikk
István	Papp	Isotoptech Zrt.
Jesper Skov	Andersen	DTU
Jixin	Qiao	DTU
Johanna	Lechle	NUCLEAR ENGINEERING SEIBERSDORF GMBH
Jonathan	Sundström	University of Gothenburg
José Antonio	Corcho Alvarado	Spiez Laboratory
Kai Mikael	Jensen	DTU Health Tech
Karl Andreas	Jensen	NMBU
Kasper	Andersson	DTU
Keliang	Shi	Lanzhou University
Li	Bao	China Institute for Radiation Protection

First name	Last name	Affiliation
Liga	Bech	DTU Sustain
Linda	Corneliusson	Vattenfall AB - Ringhals
Lindis	Skipperud	NMBU
Luyuan	Zhang	Institute of Earth Environment Chinese Academy of Sciences
Magnus	Mortensen	Swedish Food Agency
Mahrosh	Javed	Nacionalinis Fizinių ir technologijos mokslų centras (NFTMC)
Maoyi	Luo	China Institute for Radiation Protection
Marcus	Christl	ETH Zurich
Maria	Davidsson	Ringhals
Marion	Grange	SLV
Matic	Dokl	DTU
Mats	Eriksson	Linköping Uni, Sweden
Maud	Zilbermann	Helmholtz-Zentrum Dresden-Rossendorf
May-Britt	Price	DTU
Mengting	Zhang	Xi'an Institute for Innovative Earth Environment Research
Mette	Ziersen	DTU
Michael	Petrich	PerkinElmer
Mikkel	Øberg	Danish Decommissioning
Minyu	Zuo	PerkinElmer
Ni	Yuan	China Institute for Radiation Protection
Nicolas	Maurissen	IRE
Nienke	Peters	Wageningen Food Safety Research
Ole Christian	Lind	NMBU
Olof	Gottfridsson	Vattenfall AB
Parvine	Naghchbandi	Direktoratet for strålevern og atomsikkerhet
Patric	Lindahl	Cyclife Sweden AB
Pengxiang	Li	Conference and laboratory training participants
Peter	Hoskam	Thermo Fisher
Péter	Kiss	DTU
Petya	Dimitrova	Bulgarian
Qiuju	Guo	Peking University
Rainer	Kadan	Austrian Agency for Health and Foodsafety
Ramahaleotsitohaina	Avoko Ravakiniony	INSTITUT DES RADIOELEMENTS BELGIUM
Sama	Mammadova	University of Helsinki
Sara	Ehrs	Swedish Food Agency
Sarah	Mullins	Radiation Protection Unit, Radiation Sciences, QLD Health
Silja	Thomassen	Bayer AS
Soonhyun	Kim	DGIST, Republic of Korea
Soumya	Gupta	Université Paris-Saclay, CEA, Service de Physico-Chimie (SPC), 91191 Gif sur Yvette, France
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Steffen	Happel	Triskem International
Susanna	Salminen-Paatero	HU
Sven	Nielsen	DTU
Svend	Olsen	DTU
Teija	Hovirinta	Hidex Oy
Thomas	Cole	Ringhals AB
Thor	Andersen	DTU

First name	Last name	Affiliation
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Toon	Meeuwsen	metorX B.V.
Torbjørn	Gäfvert	DSA
Torill	Solheim	Institutt for Energiteknikk, Norge
Tzu-Hao	Wang	DTU
Unni	Augestad Kvitastein	IFE
Valtteri	Suorsa	STUK - Radiation and Nuclear Safety Authority (Finland)
Wangsuo	Wu	Lanzhou University
Wei	Huang	Shanghai Institute of Applied Physics, Chinese Academy of Sciences
Xiaolin	Hou	Lanzhou University
Xue	Zhao	Institute of Earth Environment, Chinese Academy of Sciences
Zhiqiang	Cheng	Shanghai Institute of Applied Physics Chinese Academy of Sciences

Appendix 2 - Seminar Program

Welcome message

Dear colleagues,

I am delighted to welcome you to RadWorkshop 2024 in the historic town of Roskilde, Denmark. This is the fourth RadWorkshop supported by NKS, following three successful RadWorkshops at Risø, Denmark in 2009, 2013, and 2018, each with different focuses within the realm of radioanalytical chemistry. It is also partially supported by MetroPOEM. Different from previous ones, RadWorkshop 2024 will embrace a broader spectrum of topics in radioanalytical chemistry, reflecting the evolving landscape of this field.

The workshop is organized by the Technical University of Denmark (DTU), in close collaboration with Norwegian Life Science University (NMBU), University of Helsinki (UH), Swedish Food Agency (SLV), Norwegian Radiation and Nuclear Safety Authority (DSA), and Radiation and Nuclear Safety Authority, Finland (STUK).

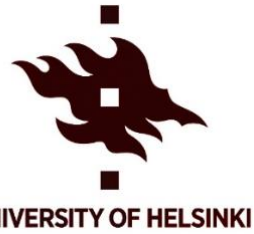
RadWorkshop 2024 aims to foster a network for all participants and stakeholders, emphasizing the pivotal role of radioanalytical chemistry in our research domain. I hope RadWorkshop 2024 will be successful in sharing knowledge, exchanging experiences, and further promoting international collaborations, while you also enjoy the lovely autumn in Denmark!

I would like to extend my sincere thanks to all the participants, collaborators, and our generous sponsors—GammaData Instrument AB, metorX B. V., PerkinElmer, Thermo Fisher Scientific, and Triskem International—whose support has been invaluable in making this event possible.

Jixin Qiao
Chairman of RadWorkshop 2024
Senior Researcher, DTU Sustain

Workshop collaborators and sponsors

Collaborators



Sponsors

Programme overview

Monday, 9th September+B2:OB2:O29		Tuesday, 10th September		Wednesday, 11th September		Thursday, 12th September				Friday, 13th September			
8:30	Registration	8:30	Registration	8:30	Registration								
Opening session (Chairman: Jixin Qiao)		**Application (Chair: Susanna Salminen-Paatero)**		**Methodology (Chair: Torbjørn Gäfvert)**		**Lab training**				**Lab training**			
9:00	Claus Hélix-Nielsen, Head of Department, DTU Sustain	9:00	Mikkel Øberg, DD (Keynote)	9:00	Hyuncheol Kim, KARERI (Keynote)	09:00	**Group 1**	**Group 2**	**Group 3**	09:00	**Group 1**	**Group 2**	**Group 3**
9:15	Charlotte Scheutz, DTU	9:35	Jixin Qiao, DTU	9:35	Francisco J. G. Gerada, UNEX	**Lectures about Sr radiochemical analysis and ICP-MS** Karl A. Jensen, NMBU Eike Rades, DTU Jixin Qiao, DTU							
9:30	Kasper Andersson, NKS	9:55	Valtteri Suorsa, STUK	9:55	Johanna Lechle, NES								
9:45	Jixin Qiao, RadWorkshop 2024	10:15	Angela H. J. Tan, DTU	10:15	Li Bao, CIRP								
10:00	Xiaolin Hou, LZU (Keynote)												
10:35	Coffee break	10:35	**Coffee break (Group photo)**	10:35	Coffee break	10:30	Coffee break						
Methodology (Chair: Charlotte Scheutz)		**Application (Chair: Jixin Qiao)**		**Methodology (Chair: Eike Rades)**		**Sr chemical separation** Group 1: Susanna Salminen-Paatero, UH Group 2: Marion Grange, SLV Group 3: Eike Rades, DTU					**ICP-MS** Karl A. Jensen	**Sr chemical separation**	**LSC & Beta counting** Matic Dokl
11:00	Marcus Christl, ETH (Keynote)	11:00	José A. C. Alvarado, Spiez Lab (Keynote)	11:00	Mikael Jensen, DTU (Keynote)								
11:35	Karl A. Jensen, NMBU	11:35	Ole Christian Lind, NMBU (Keynote)	11:35	Casper Boe Jensen, DD								
11:55	Marion Grange, SLV	12:10	Susanna Salminen-Paatero, UH	11:50	**Site tour at Risø-1**								
12:15	Anu Bhaisare, NPL	12:30	Xue Zhao, IEECAS	**Warehouses/storage facilities-hot cells-DR3 and DR2**									
13:00	Lunch	13:00	Lunch	13:00	Lunch	12:00	Lunch			12:00	Lunch		
Methodology (Chair: Marion Grange)		**Application (Chair: Ole Christian Lind)**		**Application (Chair: Kasper Andersson)**		**Lab training**				**Lab training**			
14:00	Qiuju Guo, PKU (Keynote)	14:00	Georg Steinhauser, TU Wien (Keynote)	14:00	Franck Dal-Molin, CEFAS (Keynote)	13:00	**Group 1**	**Group 2**	**Group 3**	13:00	**Group 1**	**Group 2**	**Group 3**
14:35	Sven Nielsen, DTU	14:35	Mengting Zhang, IEECAS	14:35	Tzu-Hao Wang, DTU								
14:55	Soumya Gupta, CEA	14:55	Habacuc Pérez-Tribouillier, ETH Zurich	14:55	Sarah Mullins, QLD Health								
15:15	Aaron Lehnert, IRS			15:15	Soonhyun Kim, DGIST		**Sr chemical separation**	**LSC & Beta counting** Matic Dokl	**ICP-MS** Karl A. Jensen		**LSC & Beta counting** Matic Dokl	**ICP-MS** Karl A. Jensen	**Sr chemical separation**
15:35	Coffee break	15:15	Coffee break	**15:35**	**Closing ceremony**								
Sponsor Workshop		**Methodology (Chair: Valtteri Suorsa)**		**Site tour at Risø-2** **Risø Laboratory, DTU sustain**									
		15:40	Keliang Shi, LZU										
		16:00	Guillaume Lutter, DTU										
16:00-18:00	**Triskem International**	16:20	Pengxiang Li, CIRP			15:30	Joint Q&A			15:30	Joint Q&A		
		16:40	Michael Petrich, PerkinElmer										
		17:15	Departure for social event										
		18:00-22:00	**RadWorkshop social event at Herthadalen**	**17:30**	**Close of day 3**	16:00	Close of day 4			16:00	**Close of the workshop. See you next time!**		

Detailed programme

Monday, 9th September
(Building 112, DTU Risø Campus)

8:30 Registration

Opening session (Chairman: Jixin Qiao)

9:00 Claus Hélix-Nielsen, Head of Department, DTU Sustain
9:15 Charlotte Scheutz, DTU Sustain
9:30 Kasper Andersson, NKS
9:45 Jixin Qiao, RadWorkshop 2024
10:00 Radioanalysis of long-lived radionuclides, the present status and the perspectives
(Xiaolin Hou, LZU)
10:35 Coffee break

Methodology (Chair: Charlotte Scheutz)

11:00 Ultra-trace analyses with compact accelerator mass spectrometry (AMS) - technology and applications
(Marcus Christl, ETH)
11:35 Tuning an ICP-MSMS to obtain the optimum signal to noise ratio for radionuclide measurement
(*Karl A. Jensen, NMBU)*
11:55 Analysis of Sr-90 in milk and drinking water with ICP-MS/MS
(*Marion Grange, SLV)*
12:15 Assessing the potential of ICP-MS/MS for analysing ^{210}Pb in sediment
(*Anu Bhaisare, NPL)*
13:00 Lunch

Methodology (Chair: Marion Grange)

14:00 Radon in the environment
(*Qiuju Guo, PKU)*
14:35 Analysis of Ra-226 and short-lived daughters by gamma spectrometry
(*Sven Nielsen, DTU)*
14:55 Uranium isotope measurements by mass spectrometry to characterize reference materials
(*Soumya Gupta, CEA)*
15:15 Production and characterization of synthetic homogenous multi-element actinides samples via Sol-Gel as standards for mass spectrometry
(*Aaron Lehnert, IRS)*
15:35 Coffee break

Sponsor Workshop

16:00 Triskem International

18:00 Close of day 1

Tuesday, 10th September
(Building 112, DTU Risø Campus)

8:30 Registration

Application (Chair: Susanna Salminen-Paatero)

9:00 The Use of Sampling in Decommissioning Work
(*Mikkel Øberg, DD)*

9:35 Tracer application using anthropogenic uranium isotopes (U-233 and U-236) in the marine system
(*Jixin Qiao, DTU)*

9:55 Radioanalytical and emergency preparedness activities at STUK
(Valtteri Suorsa, STUK)

10:15 Levels of natural and anthropogenic radionuclides in selected environmental radioactivity monitoring sites in Singapore
(Angela H. J. Tan, DTU)

10:35 Coffee break (Group photo)

Application (Chair: Jixin Qiao)

11:00 Determination of Pu and U radionuclides in environmental samples
(*José A. C. Alvarado, Spiez Lab)*

11:35 Reducing overall uncertainties radioecological models
(*Ole Christian Lind, NMBU)*

12:10 U, Np and Pu in lichen: temporal and regional variation in Finland
(*Susanna Salminen-Paatero, UH)*

12:30 Plutonium marker for the Great Acceleration by intensified human activities
(*Xue Zhao, IEECAS)*

13:00 Lunch

Application (Chair: Ole Christian Lind)

14:00 Understanding the "wild boar paradox"
(Georg Steinhauser, TU Wien)

14:35 Plutonium isotopes in biological samples along the coast of China: level, distribution and partitioning behavior
(Mengting Zhang, IEECAS)

14:55 Combining AMS and MC-ICP-MS to study the entrance of Atlantic Waters into the Arctic Ocean
(Habacuc Pérez-Tribouillier, ETH Zurich)

15:15 Coffee break

Methodology (Chair: Valtteri Suorsa)

15:40 Research on analytical techniques for trace radionuclides in complex matrices
(Keliang Shi, LZU)

16:00 Gamma-ray spectrometry with Liquid Scintillation Counting: Techniques and Benefits
(Guillaume Lutter, DTU)

16:20 Determination of $^{239+240}$Pu in Environmental Aerosol
(Pengxiang Li, CIRP)

16:40 Direct Determination of Radium-226 and Radium-228 in Groundwater with the NexION 5000 Multi-quadrupole ICP-MS
(Michael Petrich, PerkinElmer)

17:15 Departure for social event

18:00 RadWorkshop social event at Herthadalen

22:00 Close of day 2

Wednesday, 11th September
(Building 112, DTU Risø Campus)

8:30 Registration

Methodology (Chair: Torbjørn Gäfvert)

9:00 Automated Radionuclide Separation for Radioactivity Analysis
(Hyuncheol Kim, KARERI)

9:35 Validation of a rapid method for ^{90}Sr determination in biota samples
(Francisco J. G. Gerada, UNEX)

9:55 Comparing methods of digestion for the analysis of Strontium, Plutonium and Americium in solid samples
(Johanna Lechle, NES)

10:15 Analysis of Iron-55 in Liquid Effluent of NPP
(Li Bao, CIRP)

10:35 Coffee break

Methodology (Chair: Eike Rades)

11:00 Radionuclidic purity in radiopharmaceuticals. Can 99.9% ever be measured?
(Mikael Jensen, DTU)

11:35 Danish Decommissioning Site introduction
(Casper Boe Jensen, DD)

11:50 Site tour at Risø-1: Warehouses/storage facilities-hot cells-DR3 and DR2

13:00 Lunch

Application (Chair: Kasper Andersson)

14:00 Improving knowledge in marine science at Cefas using radioanalytical tools
(Franck Dal-Molin, CEFAS)

14:35 Reconstructing ^{135}Cs/^{137}Cs signals released from Sellafield reprocessing plant and its potential environmental applications
(Tzu-Hao Wang, DTU)

14:55 Radionuclides in bovine muscle and liver from north-west Australia
(Sarah Mullins, QLD Health)

15:15 Enhancing Radioactive Cs Adsorption: Mechanistic Insights of Prussian Blue Analog with Various Transition Metals
(Soonhyun Kim, DGIST)

15:35 Closing ceremony

16:00 Site tour at Risø-2: Risø Laboratory, DTU Sustain

17:30 Close of day 3

Thursday, 12th September
(Building 730, DTU Risø Campus)

Lab training

09:00 Lectures about Sr radiochemical analysis and ICP-MS
Karl A. Jensen (NMBU), Eike Rades (DTU), Jixin Qiao (DTU)
10:30 Coffee break
11:00 Sr chemical separation
Group 1: Susanna Salminen-Paatero (UH)
Group 2: Marion Grange (SLV)
Group 3: Eike Rades (DTU)
12:00 Lunch

Lab training

13:00 Group 1: Sr chemical separation
Group 2: LSC & Beta counting (Matic Dokl)
Group 3: ICP-MS (Karl A. Jensen)
15:30 Joint Q&A
16:00 Close of day 4

Friday, 13th September
(Building 730, DTU Risø Campus)

Lab training

09:00 Group 1: ICP-MS (Karl A. Jensen)
Group 2: Sr chemical separation
Group 3: LSC & Beta counting (Matic Dokl)

12:00 Lunch

Lab training

13:00 Group 1: LSC & Beta counting (Matic Dokl)
Group 2: ICP-MS (Karl A. Jensen)
Group 3: Sr chemical separation

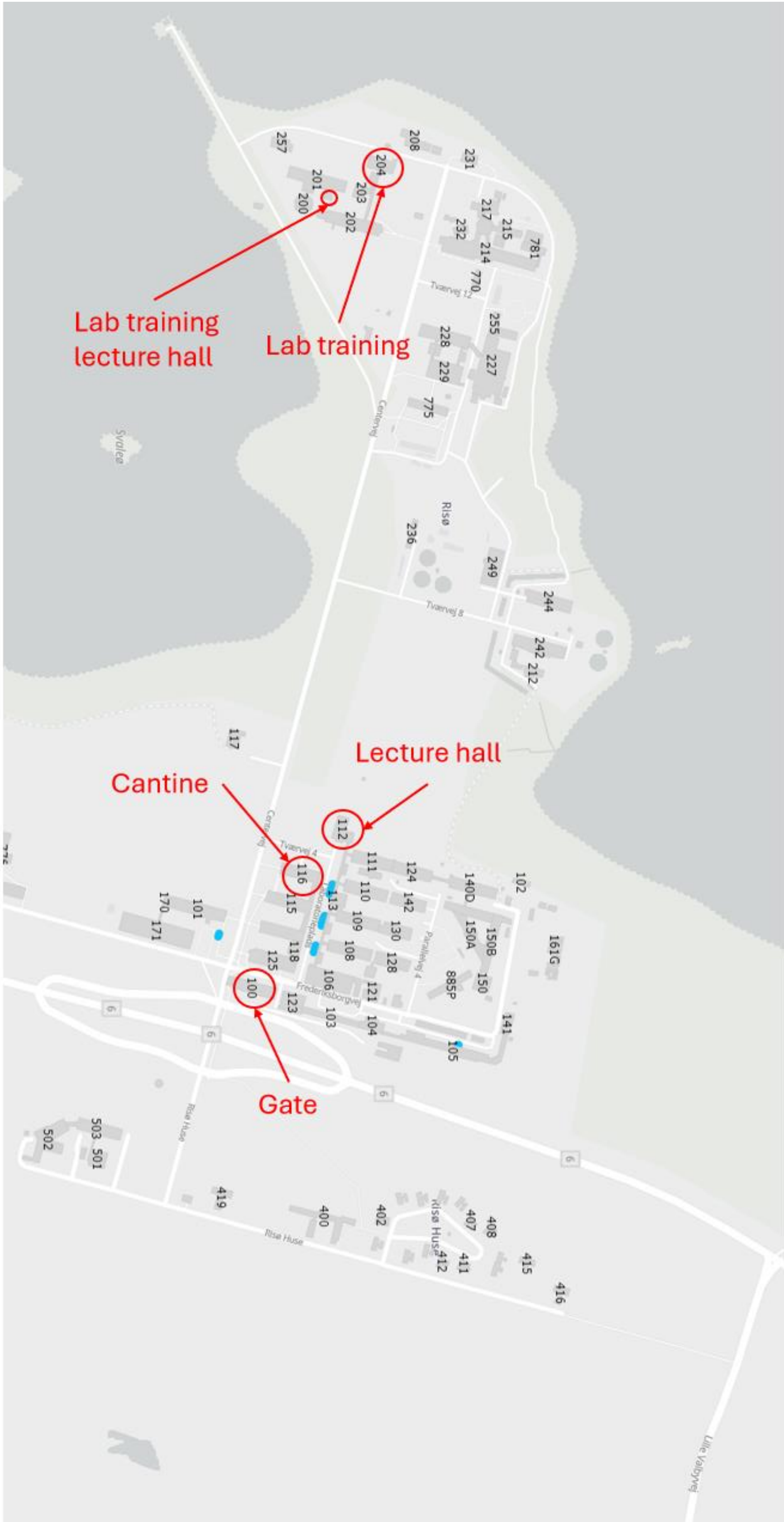
15:30 Joint Q&A

16:00 Close of the workshop

Laboratory exercise groups

Group 1	Group 2	Group 3
Alessandro Nozza	Aaron Lehnert	Camillo Bruhn
Elżbieta Anna Stefaniak	Anu Bhaisare	Filip Jedrzejek
Esperanza Lara Robustillo	Chiaoting Lo	Francisco Javier Guillén Gerada
Habacuc Pérez-Tribouillier	Galia Selikowitz	Ingunn Isdahl
Li Bao	Helena Fajfar	István Papp
Maud Zilbermann	Hyuncheol Kim	Magnus Mortensen
Michael Petrich	Johanna Lechle	Maria Davidsson
Minyu Zuo	Jonathan Sundström	Petya Dimitro
Parvine Naghchbandi	Ni Yuan	Ramahaleotsitohaina Avoko Ravakiniony
Pengxiang Li	Rainer Kad	Teija Hovirinta
Sama Mammadova	Sarah Mullins	Thomas Cole
Sara Ehrs	Silja Thomassen	Torill Solheim
Soonhyun Kim	Wei Huang	Valtteri Suorsa
Zhiqiang Cheng	Xue Zhao	

Map of Risø


Lab training
lecture hall
Lab training
Lecture hall
Cantine
Gate

Appendix 3 – Abstracts

Abstracts of oral presentations

Radioanalysis of long-lived radionuclides, the present status and the perspectives

Xiaolin Hou[1, 2]

[1]*School of Nuclear Science and Technology, Lanzhou University, Lanzhou, China*

[2]*Xi'an AMS Center, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, China*

Accurate determination of radionuclides in various environmental and waste samples is critical for investigation of environmental radioactivity, radioecology, evaluation nuclear environmental safety, nuclear forensics, environmental process tracing using radionuclide, decommissioning of nuclear facilities. Besides relatively easy measurable gamma emitters, pure beta and alpha emitting radionuclides are often hard to measure (HTM). The conventional analytical methods based on radiochemical separation followed by radiometric measurement have been used for decades. But it suffers a few difficulties such as long analytical time up to a few weeks, low sensitivity for long-lived radionuclides (e.g. ^{238}U, ^{235}U, ^{232}Th, ^{126}Sn, ^{99}Tc, ^{237}Np, ^{129}I, unable to measure some low-level and similar decay energy radionuclides, such as ^{135}Cs, ^{239}Pu/^{240}Pu and ^{236}U). Mass spectrometry is a sensitive technique for measurement of long-lived radionuclides. but often suffer from serious interference from high-level stable isotopes and polyatomic ions. The rapid improvement of accelerator mass spectrometry (AMS), especially ICP-MS make them more attractive and competitive for the determination of radionuclides of long-lived.

A critical review of the present status and perspective of AMS and ICP-MS on the determination of long-lived radionuclides in environmental and waste samples against radiometric techniques is presented, with some examples on the determination of important radionuclides such as ^{90}Sr, ^{99}Tc, ^{126}Sn, ^{129}I, ^{135}Cs, ^{234}U, ^{235}U, ^{236}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am. The main challenge and strategy on the determination of these radionuclides in different environmental and waste samples are discussed, focusing on the pros and cons of each measurement techniques. The requirement and strategies on radiochemical separation for the determination of these radionuclides with different techniques are also discussed. A special effort on elimination of isobaric and polyatomic ion interference in ICP-MS and highly sensitive measurement are presented aiming to obtain a reliable analytical results. Application of these methods on the investigation of environmental radioactivity and evaluation of radiation risk, as well as tracing environmental processes using artificial radionuclides is highlighted.

Acknowledgments:

This work is partly supported by projects of National Nature Science Foundation of China (22341603, U21A20442), The contributions from Yanyun Wang, Weichao Zhang, Shan Xing, Liuchao Zhu, Zhao Huang, Huan Jiang, Qian Wang, Qi Liu are acknowledged.

Ultra-trace analyses with compact accelerator mass spectrometry (AMS) -technology and applications

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Accelerator Mass Spectrometry (AMS) is an ultra-sensitive method for the detection of long-lived radionuclides that uses nuclear and atomic techniques for isobar suppression. Since its invention in 1977 [1-3] it has evolved as the state-of-the-art analytical method not only for radiocarbon dating but also for many other long-lived radionuclides with applications in various fields of sciences [4].

In recent decades, compact AMS systems operating at terminal voltages much below 1 MV became available [5-7] and have proven to outperform larger AMS facilities for most of the traditional AMS nuclides such as ^{14}C, ^{10}Be, ^{26}Al, but also for ^{129}I and the actinides. The reduction in size and complexity of AMS systems was not only key for more stable measurement conditions, which lead to higher accuracy and precision, it also created new applications in various fields of sciences ranging from Geology, Environmental and Climate Sciences to Nuclear Astrophysics and high precision dating [8]. The development of dedicated simulation tools [9], improved detection techniques for low energy ions [10], and the implementation of He-stripping [11; 12] allowed the construction of optimized, next generation multi-isotope AMS systems [13] that often reach better detection limits and/or higher sensitivity than their larger and more complex counterparts.

The Laboratory of Ion Beam Physics (LIP) at ETH Zurich is world renown for the development of state-of-the-art compact low-energy AMS systems. The worldwide first compact AMS system, TANDY, was developed at LIP in the late 1990ies [5] and culminated more recently in the development of the MICADAS and LEA AMS systems that have revolutionized radiocarbon analyses and are still setting the standards for state-of-the-art, fast, precise, highly efficient and therefore cost efficient radiocarbon analyses [14-16]. The development and commissioning of a new compact multi-isotope 300 kV AMS system MILEA at LIP together with our ETH spin off company Ionplus AG in 2017/18, represents another milestone in the long and successful history of AMS developments at LIP [13; 17]. The AMS system MILEA is optimized for the highly sensitive, and low background analysis of heavy ions such as the actinides [9; 17] and therefore often outperforms much larger AMS systems [18].

This keynote lecture first gives an overview over the technical developments at LIP, ETH Zurich that led to the development of compact AMS systems such as the compact multi isotope system MILEA, and finally shows some selected applications of this ultra-sensitive technique.

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Tuning an ICP-MSMS to obtain the optimum signal to noise ratio for radionuclide measurement.

KARL ANDREAS JENSEN

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This presentation will describe strategies for method development in radionuclide measurement with an ICP-MSMS (triple quadrupole) instrument.

The presentation is aimed at users of ICP-MSMS who would like to gain more knowledge about gas-phase reactions in the reaction cell of such instruments, and how to select and optimise such reactions. These questions will be addressed in the presentation:

- Which gases are suitable for different radionuclides and their mass interferences? What are the reaction products?
- Does the radionuclide of interest or it's interference(s) have an exothermic or endothermic reaction with the added gas?
- How does the collision energy affect the reactions in the reaction cell?
- How can mixing of gases help with better signal to noise ratios for radionuclide measurement?

Examples including polyatomic and isobar mass interferences for some radionuclides will be used to answer these points. Participants are encouraged to bring examples of their measurement needs for discussion during the course.

Analysis of Sr-90 in milk and drinking water with ICP-MS/MS

Marion Grange[1], Magnus Mortensen[1], Marcus Östman[2], Petra Lagerkvist[2], Annika Tovedal[2], Mats Eriksson[3], Mårten Dario[3], Marie Carlsson[3]

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[2] *Swedish Defence Research Agency, Stockholm*

[3] *Linköping University, Linköping*

We present a fast method to analyse Sr-90 in both milk and drinking water (DW), using ICP-MS/MS after a nuclear emergency.

In case of a radioactive fallout following a nuclear power plant accident or a nuclear weapons explosion both Sr-89 and Sr-90 might contribute to the radiation dose to the population through food and water. Strontium-89, with a half-life of 50 days is most abundant the first months after a nuclear event. However, it decays quickly and Sr-90, with a half-life of 28.8 years, is one of the most significant radionuclides responsible for potential internal radiation dose in the long term. Sr-90 is a pure-beta emitter and is traditionally analysed using liquid scintillation counting (LSC). This method is time consuming and not the best suited in case of an emergency, in which rapid analyses are needed to meet the demand of data to decision-making. Therefore, in order to maintain the control of food and water supply chains over a long period of time it is crucial to increase measurement capabilities for Sr-90.

The Euratom directive 2016/52 stipulates that levels of strontium isotopes in water and liquid foodstuff should not exceed 125 Bq/kg. The biggest challenge is to analyse low concentrations of Sr-90 within a reasonable time frame, *i.e.* within a few hours after samples arrive at the lab. We also want the method to be easily transferable between laboratories to increase the analytical capabilities in case of a nuclear emergency, which could generate the need of analysing hundreds of samples within a short period of time.

Our milk method includes the digestion of milk with nitric acid in a microwave oven, separation and purification of Sr with Sr-specific resin, and concentration through evaporation. Our water method consists of acidification of the samples with nitric acid followed by separation and purification of Sr with Sr-specific resin. We are also developing a quick screening method of water samples, without the purification step, that would allow screening of the drinking water samples with the highest level of Sr-90. The methods are under validation, and the estimated limits of quantification are 30 Bq/kg for the milk and drinking water method (with resin separation) and 60 Bq/kg for the screening method. The turnaround time (time between the arrival at the lab and the final result) is approximately 7 hours for the milk and drinking water method and 1 hour for the screening method.

The analysis with ICP-MS/MS is performed in presence of collision and reaction gases (He and O_2). Two analyses of each sample are required for the milk- and drinking water method in order to calculate the recovery of the method for each sample. This is done by analysing stable Sr naturally present in the samples before and after separation on Sr-specific resin.

***Table 1**: Overview of the developed methods for the analysis of Sr-90 with ICP-MS/MS*

Parameters / **Method**	**Digestion**	**Separation with resin**	**Evaporation**	**ICP-MS/MS mode**		**Approx. time between arrival and results**
				Stable Sr	*Sr-90*	
Milk method	x	x	x	He-mode	$He+O_2$ mode	7 hrs
DW method	-	x	-			5 hrs
Screening method (DW)	-	-	-	-	$He+O_2$ mode	1 hr

This project is funded by the Swedish Civil Contingencies Agency and led by the Swedish Food Agency, with work in collaboration with the Swedish Defence Research Agency and Linköping University, and past contribution from the Swedish Radiation Safety Authority.

Assessing the potential of ICP-MS/MS for analysing ^{210}Pb in sediment

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Lead-210 (^{210}Pb) is extensively employed in environmental science, geology, and archaeology for chronological research. The isotopic dating method provided by ^{210}Pb is valuable in offering insights into past environmental changes and anthropogenic activities. Measurement of ^{210}Pb by inductively coupled plasma-mass spectrometry[2] (ICP-MS) is challenging given the relatively short half-life for mass spectrometry[1], as well as interferences from polyatomic ^{209}Bi^{1}H and ^{208}Pb^{1}H$_2$, and tailing from ^{209}Bi and ^{208}Pb.

This study focuses on the ability of tandem ICP-MS/MS to measure ^{210}Pb in two sediment samples, with expected activity levels from 0.01 – 0.3 Bq g^{-1} (0.004-0.11 pg/g. The potential for ICP-MS/MS measurement of ^{210}Pb and the impact of interferences was initially assessed using stable Pb and Bi standards using different sample introduction methods (standard and Apex Q systems).

In all cases, the sensitivity was significantly improved using the Apex Q compared to the standard sample introduction. Using the Apex Q in MS/MS mode, for ^{208}Pb concentrations up to 10 µg g^{-1}, there was no increase in signal at mass 210, suggesting limited impact from ^{208}Pb or ^{208}Pb^{1}H$_2$. Using the same setup, an increase in the count rate at m/z = 210 was observed at ^{209}Bi concentrations as low as 10 ng g^{-1}.

To achieve environmental level ^{210}Pb measurement required in this study, offline chemical separation of ^{209}Bi was needed, along with pre-concentration of ^{210}Pb. This was initially tested on a ^{210}Pb standard, which had previously been spiked with ^{209}Bi carrier. An extraction chromatography technique using SR-resin (Triskem International) originally developed for separation of ^{212}Pb from ^{212}Bi for nuclear medicine applications was applied. This reduced the ^{209}Bi concentration to a level that did not contribute to the background at m/z = 210. Post-separation, ^{210}Pb calibration standards from 0.3 – 5.0 Bq g^{-1} (0.2 – 1.8 pg g^{-1}) were measured to assess the optimal instrument setup. A sensitivity of approximately 20 CPS for 1 Bq g^{-1} (0.4 pg g^{-1}) ^{210}Pb was achieved with the improvement offered by the Apex Q sample introduction. Recommendations are made as to how ^{210}Pb measurement by ICP-MS/MS can be further improved.

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Radon in the environment

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Radon (^{222}Rn) is a radioactive gas formed from radium (^{226}Ra), which is a decay product of Uranium (^{238}U). Uranium and radium occur naturally and present in all terrestrial materials, like soils and rocks, so radon is ubiquitous in the environment. Radon gas, which has a half-life of 3.8 days, might escape from radium-bared grains into the pore space(emanation). Radon atoms entering the pore space are then transported, by diffusion and advection through this space until they in turn decay or are released into the atmosphere(exhalation).

Radon and its short-lived decay products in the atmosphere are evaluated to be the most important contributors to the ionizing radiation dose received by the general population from natural sources[1]. The health risks associated with high radon exposures in Uranium mine and underground mines have been known for a long time, while much attention has been paid to environmental radon exposure since people began to realized that indoor radon exposures could be quite high. The risk of lung cancer associated with radon arises from the inhalation of the airborne radon progeny and the resulting dose to the lung, mainly from alpha radiation. Up to now, radon exposure in indoor environment has been epidemiologically proved to be the second cause of lung cancer in the general population after smoking[2]. Current estimates of the proportion of lung cancers attributable to radon range from 3 to 14%, depending on the average radon concentration in the country concerned and the calculation methods[2].

From the perspective of public health to reduce radon risk, a reference level for radon represents the maximum accepted radon concentration in a residential dwelling is recommended by both related international organizations and national authorities today as a national radon police, and measurement of radon concentration in dwellings or so-called radon survey should be the first step. The most popular measurement monitors or devices will be introduced in my presentation, quality assurance and measurement scenarios are also included as well.

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1.

Analysis of Ra-226 and short-lived daughters by gamma spectrometry

Sven Nielsen[1]

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Concentrations of ^{226}Ra (T½ = 1600 y) in environmental samples may be determined by gamma spectrometry using the characteristic gamma-ray energy of 186.2 keV. This may be difficult, however, due to interference from a gamma ray of similar energy at 185.7 keV from ^{235}U (T½ = 708 My), which is often present in environmental samples, particularly in sediment and soil. The two radionuclides ^{214}Pb (T½ = 27 min) and ^{214}Bi (T½ = 20 min) are short-lived decay products of the longer-lived radon, ^{222}Rn (T½ = 3.8 d), which is produced by the decay of ^{226}Ra. The isotopes ^{214}Pb and ^{214}Bi each have several characteristic gamma rays that do not suffer from interference with gamma rays from other radionuclides typically found in environmental samples. These two radionuclides are therefore well suited for analysis by gamma spectrometry and their concentrations will correspond to that of ^{226}Ra in case of secular equilibrium. This condition is obtained if the sample is placed in a container from which there is no escape of gaseous ^{222}Rn and time for build-up of the two daughter radionuclides since closing of the container has been sufficient (3 weeks or more).

This presentation describes results of tests of vacuum packaging for radon tightness with different types of plastic bags. The tests were carried out during November 2018 to January 2019.

Uranium isotope measurements by mass spectrometry to characterize reference materials

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The critical challenge in the field of trace or ultra-trace analysis of radionuclides in environmental matrices lies in the lack of reference materials. This is a major need in many fields, including environmental monitoring, the nuclear industry and nuclear forensics. One of the aims of the European MetroPOEM[1] project is to produce two reference materials: a liquid seawater-based material and a solid synthetic silicate material manufactured at the Laboratoire National Henri Becquerel (LNHB). These two materials will be doped with radionuclide solutions that have previously been characterised using both radiometric techniques at the LNHB and mass spectrometry at the Laboratoire de développement Analytique Nucléaire Isotopique et Elémentaire (LANIE). The doping solutions will be characterised in terms of isotope ratios and concentration by Thermal Ionisation Mass Spectrometry (TIMS) and by Multi-collector Inductively Coupled Plasma Mass Spectrometry (MC ICP-MS). Furthermore, Quadrupole Inductively Coupled Plasma Mass Spectrometry (Q ICPMS) measurements have been performed in order to check the presence of impurities.

We present here the preliminary results obtained on the initial uranium isotopic composition of seawater taken from the North Sea and subsequently acidified. This sample was first purified on a UTEVA-type extracting resin.[2] The protocol for washing the resin to obtain the lowest possible blanks will be described in detail. The first isotopic results for U, Pu and Am obtained by TIMS and ICPMS MC on the initial doping solutions of the two reference materials and the final uncertainty budget will be discussed and compared.

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Production and characterization of synthetic homogenous multi-element actinides samples via Sol-Gel as standards for mass spectrometry

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MetroPOEM[12] is committed to developing SI-traceable mixed element reference materials for the calibration of mass spectrometric devices. In nuclear forensics, elemental selectivity and precise spatially resolved mass spectrometry is essential for ultra-trace analysis of environmental samples. Resonant laser secondary neutral mass spectrometry (rL-SNMS) combines both element selective isotope ratio measurements and spatial resolution on the micrometer scale. Multi-element reference materials are needed to quantitatively examine elements important for environmental analytics.

In this work we present a production method of mixed actinide samples such as U, Pu and Am via Sol-Gel. These samples consist exclusively of the respective metal and fulfil the conditions for inter and intra homogeneity confirmed by EDX and SIMS. The spatially resolved element distribution is determined using rL-SNMS. ICP-MS is also used to determine the element composition and verify the traceability. The particles are used to estimate the efficiency of the ToF-SIMS used in this work.

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The Use of Sampling in Decommissioning Work

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The nuclear facilities at the Risø site are undergoing decommissioning. Before, during and after decommissioning sampling is used. Sampling itself comprises the following: the determination of the number of samples, the places where the samples are taken, the size of the samples, the method of obtaining and storing the samples and the method of analysis. Sampling is used for four specific purposes:

- Planning of dismantling
- Monitoring during dismantling
- Characterization of radioactive waste
- Clearance of materials, buildings and land areas

Sampling is normally done for one of the specific purposes because the different purposes have different objectives; however some overlap does exist. In the planning phase results from sampling primarily help guide the health physics measures necessary during the dismantling, but results can also be used to help determine the best way of executing the actual decommissioning. During dismantling sampling is used to further substantiate assumptions made during the planning phase or to explore abnormalities. Characterization of radioactive waste is made to provide data for safe storage (short term as well as long term) and inventory calculations. In clearance measurements sampling is used when it is impractical or impossible to make a total measurement on the item to be cleared. It is also used when nuclide concentration ratios are to be determined. Examples of the use of sampling including the analyses used will be given.

Tracer application using anthropogenic uranium isotopes (U-233 and U-236) in the marine system

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The global oceans are a repository of radionuclides, both naturally occurring and anthropogenic, the latter originating predominantly from fallout from past atmospheric nuclear weapons tests, discharges from nuclear reprocessing plants and releases following accidents at nuclear facilities. In recent years, the application of anthropogenic U-233 and U-236 as a new tracer pair has been increasingly adopted, especially in the marine environment. This is due to their conservative behavior, long residence time in the ocean (10^5y) and unique fingerprint of their isotopic ratios. Here, a series of studies on anthropogenic U-233 and U-236 in the Baltic Sea[13–16], the Pacific Ocean[17], and the Arctic Ocean[18,19] are presented to demonstrate the promises of this tracer pair in identifying radioactive source terms, reconstructing historical nuclear activities, benchmarking age-depth model in sediment chronology, tracking ocean circulation pathways and transit times, quantifying water mass composition and estimating pollutant/nutrient dynamics.

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Radioanalytical and emergency preparedness activities at STUK

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STUK is the corresponding authority in Finland for radiation and nuclear safety. Our mission is to promote and supervise the safe use of radiation and nuclear energy. Our duty is to provide expert knowledge and special services regarding the radioactivity and radiation safety to ensure the overall security of society.

Measurements and environmental monitoring unit corresponds of the radiation measurements (field and laboratory) performed at STUK. The laboratory is an accredited testing laboratory (T167, ISO/IEC 17025:2017). The fields of testing (both accredited and non-accredited) include gamma-spectrometric analyses of different objects (e.g., environmental and industrial samples, direct measurements of people), radiochemical analyses (Pu, Am, Cu, Sr, ^{3}H, U, Pb, Po, Rn, gross alpha/beta), ICP-MS analyses (U, Th, Sr), airborne Rn measurements and sampling for the environmental surveillance of radioactivity.

STUK provides testing services for both internal and external customers. The major surveillance programs consist of the environmental radiation monitoring in the vicinities of the Finnish nuclear power plants, and the monitoring of environmental radiation in Finland. In addition, laboratory radioactivity analyses are provided for e.g., CTBT (comprehensive Nulear-Ban Treaty), citizens and industry (e.g., building materials, mining products).

Besides the routine analyses, the laboratory serves as the national emergency preparedness laboratory which corresponds of providing radioactivity analyses for the base of decision-making during a possible radiological emergency. As possible treats are multidimensional, the radiation measurements, including the laboratory activities, are closely linked to other responsible authorities. For a fluent operation, the authorities are required to have ability to communicate and give instructions to the population without delay. This also sets requirements for the authorities corresponding of the radiation and nuclear safety.

This presentation focuses on the radioanalytical laboratory and emergency preparedness activities at STUK. It discusses major surveillance programs conducted in Finland and provides an overview of the different analytical services and capabilities of the laboratory. In addition, the importance of emergency preparedness and the Finnish national measurement strategy is highlighted.

Levels of natural and anthropogenic radionuclides in selected environmental radioactivity monitoring sites in Singapore

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This study provides insights into the radioactivity levels in soil, sediment and surface seawater from selected monitoring sites in Singapore. The environmental samples were measured for naturally occurring and anthropogenic radionuclides. In soils, the decay series of ^{232}Th and ^{238}U are the highest contributors to the total radioactivity, while in sediments, ^{40}K is the main contributor to the total radioactivity. The levels of ^{90}Sr, ^{137}Cs and $^{239+240}$Pu are also reported. The data demonstrates that the main source of anthropogenic radionuclides in soils is the global fallout from nuclear weapons testings; in sediments, contributions from the Pacific Proving Grounds have been identified. This study also highlights significant differences in radioactivity levels in soils between the northern and central regions of Singapore, which are likely attributed to the histories of the monitoring sites.

Determination of Pu and U radionuclides in environmental samples

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The validation of radiochemical methods for the determination of Pu and U radionuclides in different environmental matrices is presented. Soil and sediment samples are digested by borate fusion. For water samples, these radionuclides are pre-concentrated by co-precipitation with an iron hydroxide precipitate. Acid leaching is used for the digestion of vegetation and biological samples. Pu and U radionuclides are then separated and purified in extraction chromatography resins TEVA and UTEVA, respectively. The Pu and U radionuclides are finally measured by ICP-MS.

A summary of the validation results is presented. The application of the validated methods in different projects is also shown. The projects include: i) studies in Swiss glaciers; ii) studies of sediments in freshwater systems in Switzerland; iii) Deep sediments in the Southern Gulf of Mexico; iv) environmental samples from the Marshall Islands; v) sediments samples from South America; etc.

Reducing overall uncertainties radioecological models

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In case of a significant nuclear event, decision makers in any affected country would need to act promptly to protect people, the environment and societal interests from harmful impacts of radioactive fallout. Assessing dose estimates and what mitigating actions to implement are usually facilitated by employing an emergency decision support system (DSS) that are based on input from a combination of model prognoses, measurements, and expert judgements. Radioecological models used to make predictions of the radionuclide distribution in environmental compartments, radionuclide fluxes and radiation exposures must be sufficiently robust and fit for purpose with uncertainties reduced as much as reasonable. A series of factors will contribute to the overall uncertainties of radioecological models. The present work focuses on research performed to quantify and reduce uncertainties in various models with emphasis on the use of advanced analytical methods.

U, Np and Pu in lichen: temporal and regional variation in Finland

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Two sample sets of whole lichen and top lichen were analysed. The first sample set was the time series of lichen sampled in upper Lapland in 1960-2020. There, the global fallout from atmospheric nuclear weapons testing has been the only nuclear contamination source in practice. The second sample set included lichens collected around Finland soon after the Chernobyl accident in 1986, containing two possible nuclear contamination sources, i.e., Chernobyl deposition and global fallout. Long-term behavior and isotopic composition of global nuclear fallout in upper Lapland was sought, as well as isotopic composition of the Chernobyl-derived deposition in Finnish lichen. The lichen samples were digested and after radiochemical purification, isotopic concentrations of U, Np and Pu were determined by MC-ICP-MS and SF-ICP-MS.

For example, the Pu content in lichen in Lapland has decreased four orders of magnitude from the 1960´s to the present day. The regionally highest Pu concentrations were found in lichen samples collected in southwestern Finland in 1986. In the seminar presentation, concentrations and mass ratios of U, Np and Pu isotopes will be presented and the results compared to the corresponding values for nuclear contamination sources in the literature.

Plutonium marker for the Great Acceleration by intensified human activities

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Human activities have been drastically altering the earth since 1950s, nuclear activities are one of the strongest ways.[1] ^{239,240}Pu released by atmospheric nuclear weapons tests provides an ideal geo-marker for labeling this change due to its global fallout feature, unified temporal variation, temporal mutation, and long half-lives.[2] However, behaviors of plutonium from atmospheric deposition to the reservation in the sediment are still controversial.[3] Three sediment cores collected from two maar lakes with high-quality chronology were analyzed for ^{239,240}Pu. The first high-yield thermonuclear test (31st Oct. 1952) and the highest global tests in 1961–1962 were identified respectively as the first rapidly increased ^{239,240}Pu in 1953 and peak in 1963. In anoxic Lake Sihailongwan, scavenging Pu from the water to sediment was accelerated by the formation of insoluble PuO_2. The desorption of Pu from beneath sediment to the lake water and re-settlement was stronger in shallow and toxic Lake Huguangyan.

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Understanding the "wild boar paradox"

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Wild boars in the Alpine region are frequently burdened with remarkably high levels of radioactive ^{137}Cs ($T_{1/2}$ = 30 y). Exceedances of the regulatory limits by factors of up to 30 are not uncommon. Even more astonishing is the fact that the half-life of 30 years is not reflected in the "wild boar system." Contaminations decrease more slowly than the half-life would suggest. Wild boars are thus often more radioactive than physics permits. Thanks to new analytics of the isotopic fingerprint $^{135}Cs/^{137}Cs$,[1] we understand how this apparent "violation" of the law of radioactive decay comes about: a

source of radioactive cesium in the environment had been virtually "forgotten." Wild boars in Central Europe not only carry the fallout of Chornobyl in their muscle tissue, they are also heavily contaminated by “old” fallout from atmospheric nuclear weapons tests.

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Plutonium isotopes in biological samples along the coast of China: level, distribution and partitioning behavior

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Environmental pollutants such as radionuclides released into the marine system will continue to migrate and transform, be taken up and accumulated by marine organisms, and be transmitted to higher phic levels through food chain amplification (Qiao et al., 2024). Among the released radioactive isotopes, plutonium has attracted particular attention, which can significantly impact the behaviour of various aquatic organisms and poses a substantial risk to human health and well-being via the food chain (Deblonde et al., 2020). However, due to the lack of accurate analysis methods for ^{239}Pu and ^{240}Pu in biological samples, the environmental ecological impact assessment and environmental behaviour of plutonium isotopes in biological samples has not been systematically investigated. The main challenge belongs to the extremely low content of plutonium isotopes in organisms, the complexity of matrix and interfering elements, and the inability to accurately ^{239}Pu and ^{240}Pu concentration and reliable ^{240}Pu/^{239}Pu atomic ratio in environmental samples.

In this study, A method has been successfully established with high-efficiency pretreatment, separation, purification, and high-sensitivity measurement for accurate determination of ^{239}Pu and ^{240}Pu in large weight of various biological samples. First, sodium carbonate was used as saponification reagent and magnesium acetate as assisted ashing reagent. Then $Fe(OH)_2$-$Fe(OH)_3$ was used for co-precipitation to separate and enrich the acid-leached plutonium from the ashing sample. For the purification step, aluminium nitrate was used as masking reagent to eliminate the interference of a large amount of phosphorus in biological samples on plutonium in the anion exchange separation process. The average recovery of plutonium in the whole process is more than 73%, and the decontamination factor of ^{238}U is 3.3×10^{4}-5.4×10^{5}. The high-efficiency sampling system (APEX-Ω) was equipped with inductively coupled plasma mass spectrometry (ICP-MS/MS) for the determination of plutonium isotopes using NH_3-He as the collision/reaction gas. ^{239}Pu and ^{240}Pu concentrations in the samples were calculated by spiking ^{242}Pu as the yield tracer and isotope diluent. This method not only effectively inhibits the ^{238}U peak tail and $^{238}U^{1}H^{+}$ interference, but also greatly improves the analytical sensitivity of plutonium to 8000 cps/(pg /g). The detection limits of ^{239}Pu and ^{240}Pu in fish and prawn samples (3.5kg wet weight) were 2.56×10^{-4} mBq/kg (wet weight) and 7.33×10^{-4} mBq/kg (wet weight), respectively. For 300 g wet weight (30 g dry weight) seaweed samples, the detection limits of ^{239}Pu

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and ^{240}Pu were 2.98×10^{-3} mBq/kg (wet weight) and 8.55×10^{-3} mBq/kg (wet weight), respectively, which were more than 20 times lower than those in previous reports. The accuracy and stability of the method were verified by analysing the samples added reference materials.

The spatial distribution of plutonium isotopes (^{239}Pu, ^{240}Pu) in macroalgae, fish and shrimp collected from the coast of China Sea in December 2022-May 2023 was investigated. The ^{239,240}Pu concentrations in biological samples vary from $(2.28\pm0.16)\times10^{-3}$ to (11.99±0.33) mBq/kg. wet (with an average of 1.52±2.30 mBq/kg. wet), basically showing a increasing tendency from fish, shrimp to macroalgae, mainly depending on different biological species and growth environment. High ^{240}Pu/^{239}Pu atomic ratios that ranged from 0.158 - 0.275 (with an average of 0.215±0.036) in biological samples of the China sea, indicative of non-global fallout Pu sources were observed. The distribution of higher ^{240}Pu/^{239}Pu atomic ratios in the coastal of the East China Sea and South China Sea than the Yellow Sea and the Bohai Sea was in consistent with the introduction pathway of the Kuroshio Current with higher ^{240}Pu/^{239}Pu atomic ratios from the Pacific Proving Grounds (PPG). The contribution of the PPG derived plutonium in biological samples off the China Coast was estimated to be 0% - 61% (with an average of 26% ± 21%) using a simple two-end member mixing model. The concentration factors (CFs) for ^{239,240}Pu in this work indicated that the mean CFs for ^{239,240}Pu were significantly different across the three biological types, following the order of fish＜shrimp＜ macroalgae (green＜brown algae). *Ulva lactuca* (green algae) has extremely high accumulation ability for plutonium, which is a good environmental indicator species for plutonium isotopes. The dose assessment demonstrates that the annual per capita intake dose of plutonium from seafood consumed by public in China was 3.33×10^{-6}-4.32×10^{-6}, which is six magnitude lower than the Basic Standards, and was in a secure level at present under the impact of global fallout of atmospheric nuclear weapon tests and long transportation of PPG. The PPG derived dissolved and colloidal plutonium dominates the enrichment of plutonium in the same macroalgae species (*Ulva linza*).

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1.

Combining AMS and MC-ICP-MS to study the entrance of Atlantic Waters into the Arctic Ocean

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Recent studies indicate that the Arctic is warming up to four times faster than the global ocean. This accelerated warming is partly due to an increased influx of warm Atlantic waters into the Arctic basin. The Santa Anna Trough (SAT) is a strategically important location within the Arctic Ocean, where the two main Atlantic water branches—the Barents Sea Atlantic Waters (BSAW) and the Fram Strait Atlantic Waters (FSAW)—converge and mix. Additionally, freshwater from Siberian rivers, and ice melt contributes to the surface layers of this region. The interactions between these three components are crucial for the downstream flow of the Transpolar Drift system. Despite the SAT's significant role, studies in this region remain scarce.

In this study, we aim to address the following research questions: (i) What are the sources, pathways, tracer ages, and mixing of surface waters? and (ii) What are the mixing dynamics and trajectories of the two Atlantic branches? To address these questions, seawater samples collected during the Arctic Century Expedition in 2021, onboard the icebreaker Akademik Tryoshnikov, were processed and measured for ^{129}I (AMS), ^{236}U (AMS), and neodymium isotopic composition (MC-ICP-MS).

Based on our results, we identify minimal mixing between BSAW and FSAW in the SAT. Additionally, we trace the entrance of FSAW almost all the way to Novaya Zemlya island. This is confirmed by the distinct trajectories of both water masses, with BSAW following a northward path along the seafloor, while FSAW follows a cyclonic trajectory above BSAW. Surface waters also follow a northward trajectory, likely influenced by seasonal wind patterns in the area. Finally, the Nd isotopic composition confirms its potential as a tool to provide additional information on freshwater inputs to the surface layers of the Arctic Ocean. Although this region is likely affected by inter-annual variability, our findings enhance the understanding of the mixing dynamics and circulation patterns in the SAT. These insights improve transit time distribution calculations and underscore the importance of further research in this region.

Research on analytical techniques for trace radionuclides in complex matrices

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The continuous development and utilization of nuclear energy will produce a large number of radioactive substances, some of which will be released into the environment due to man-made or natural causes, resulting in a certain degree of environmental radioactive pollution. Aiming at the problems of environmental radionuclide analysis involved in spent fuel reprocessing, decommissioning of nuclear facilities, and environmental radiological safety assessment, as well as the separation and extraction of trace radionuclides in complex systems, out research work mainly focused on the analytical techniques for trace radionuclides in complex matrices. For uranium and transuranic elements (Np, Pu and Am), long-lived fragment nuclides (^{79}Se, ^{99}Tc, ^{90}Sr and ^{129}I), a series of analytical methods for target elements or nuclides in environmental samples such as soil, sediments, and water samples have been established. High-efficiency separation resin materials have been developed for actinides and other difficult-to-separate radioactive elements, and some materials are expected to achieve domestic substitution. Aiming at the complex manual operation of the analysis process, an automatic separation system was independently developed to achieve efficient and rapid separation of target elements. The related research work has potential application value in the accurate，rapid and automatic determination of trace radionuclides in complex matrices.

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Gamma-ray spectrometry with Liquid Scintillation Counting: Techniques and Benefits

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Many radionuclides present in environmental or decommissioning samples are not purely alpha or beta emitters; they are often accompanied by the emission of one or several gamma rays. Gamma-ray spectrometry is a widely used technique for identifying and quantifying gamma-ray radionuclide emitters. This method has the advantage of being non-destructive and allows for the measurement of almost any type of sample, from a bulk raw sample to a prepared sample in a vial for liquid scintillation counting (LSC). Additionally, multiple radionuclides can be measured simultaneously.

LSC measurements have very low energy resolution, and due to the continuous spectrum energy emission of beta particles, discriminating between different radionuclides in the sample can be challenging or even impossible. Therefore, radiochemical separation is often necessary before measuring the sample.

To minimize interferences in the sample measurement, DTU performs gamma-ray screening of the ready-to-measure liquid scintillation samples prior to LSC measurement. If gamma-ray emitters are detected, further chemical separation is conducted, followed by additional gamma-ray screening. This process is repeated until the interfering radionuclides are reduced below the detection limit.

This presentation will focus on the gamma-ray spectrometry of ready-to-measure liquid scintillation samples. It will describe the qualitative and quantitative measurement methods used at DTU and provide advice for users unfamiliar with gamma-ray techniques. Additionally, it will highlight the possible benefits of incorporating gamma-ray screening alongside radioanalytical chemistry to enhance the accuracy and reliability of measurements.

Determination of $^{239+240}$Pu in Environmental Aerosol

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The content of $^{239+240}$Pu in environmental aerosols in China was analyzed by using anion exchange resin separation and α spectrometer. The total recovery rate was 60.8%~94.6%, The minimum detectable limit of $^{239+240}$Pu in aerosols was 0.008μBq/m^3 (volume V=30 000m^3, recovery Y=0.8, measurement time t=72 h). The sampling amount of aerosol in common environment needs more than 30 000 m^3. In the emergency situation and the post-treatment plant aerosol sampling amount is 10000 m^3, The ^{210}Po content in aerosol is 4~6 orders of magnitude higher than $^{239+240}$Pu, In the analysis, we should pay attention to the influence of ^{210}Po on $^{239+240}$Pu measurement. The experimental results show that the content of $^{239+240}$Pu in common environmental aerosols in China is 10^{-3}μBq/m^3 level .

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Direct Determination of Radium-226 and Radium-228 in Groundwater with the NexION 5000 Multi-quadrupole ICP-MS

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The WHO drinking water guidance level for 226Ra is 1 Bq/L and for 228Ra 0.1 Bq/L. The U.S. Environmental Protection Agency (USEPA) has set a combined MCL (maximum contaminant level) for 226+228Ra by 0.185 Bq/L. In the EU, the Council Directive 2013/51/EURATOM specifies activity concentrations of 0.5 Bq/L 226Ra (14 pg/L) and 0.2 Bq/L 228Ra (0.02 pg/L). The required detection limits (LOD) of 226Ra in drinking water is 0.04 Bq/L (1 pg/L).

Measurement of 226Ra in environmental samples can be done by alpha spectrometry, gamma spectrometry, liquid scintillation spectrometry (LSS) and mass spectrometry (ICP-MS, TIMS, AMS). Radiometric determination usually involves lengthy, work-intensive sample preparation. Counting time in these techniques is typically hours to days for one sample, which together with the potential need for prior concentration, radiochemical separation, and/or waiting for equilibration with daughter nuclides for up to 30 days notoriously impedes rapid processing of samples.

The ISO draft standard ISO/DIS 4685 describes an ICP-MS method for the determination of 226Ra applicable in the event of an emergency. It recommends a first preconcentration of 226Ra before undergoing purification from potential interferent by passing solution through a cation exchange resin followed by a crown-ether based extraction.

This work describes the direct measurement of 226Ra in groundwater samples in less than 3 min without any preconcentration or matrix separation. Detection can be performed on mass using NH3 at 226 amu or alternatively via a mass shift reaction with N2O at 242 amu. Both cell gas reactions are explained using product ion and precursor ion scans. The calibration range extends from 1 - 500 ppq, MDL 0.6 ppq with both reaction gases. Groundwater samples with elevated Ra concentrations from the area of former East German uranium mining as well as groundwater from Norwegian deep wells were measured. The results of the ICP-MS/MS measurement are in excellent agreement with those of alpha spectrometry for all samples.

Despite comparable sensitivity and detection limits, this agreement can neither be achieved with NH3 nor with N2O in single quad mode for all real samples, even under optimized cell conditions. The relevant interferents are discussed for both cell gases.

228Ra has a factor 276 shorter half-life than 226Ra and therefore requires monitoring in the ppqt (fg/L) range. This is not possible without prior enrichment. Nevertheless, the method also provides initial information on 228Ra within the rapid emergency response.

Automated Radionuclide Separation for Radioactivity Analysis

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Radioactive analysis can be practiced with non-destructive and destructive methods, determined by the detection techniques which define the characteristics required for sample preparation. Destructive analysis, in particular, necessitates the radiochemical separation of radionuclides of interest from the sample matrix and interferences. This separation is crucial for alpha and beta emitters due to their radiological characteristics, why they are called with difficult-to-measure radionuclides.

Automation in radiochemical analysis offers numerous advantages, including increased reliability through reduced human error, improved safety, shorter analysis times, and high throughput. By integrating mass spectrometry[1] or radiometric sensors[2] with automated radiochemical separation techniques, the process from separation to measurement can be completed in a single run, facilitating on-line monitoring.

Currently, various extraction chromatographic materials are employed for radiochemical analysis in environmental samples and radioactive wastes. The general sequence in extraction chromatography includes conditioning, sample loading, rinsing, and elution, as shown in Fig. 1. The volume and type of reagent used depend on the radionuclides of interest and the extraction chromatographic resin. Traditionally, gravimetric or vacuum box approaches are used, which are time-consuming and labor-intensive. Although the vacuum box method provides a higher flow rate than the gravimetric method, it still requires manual labor for selecting solutions for each process.

Chung et al.[3] introduced automated radiochemical separation techniques prior to measurement, utilizing a multi-port flow selection valve (FSV) and peristaltic pump to separate 99Tc from samples. Kim et al. (2020)[4] developed a custom-made automated separation instrument comprising a peristaltic pump, multi-port flow selection valve, column part, sample tube, collection tube, and a control board, as illustrated in Fig. 2. All components are highly resistant to strong acids. The multi-port flow selection valve switches between the sample and reagent solutions, while the peristaltic pump ensures their flow. Unlike manual methods, the automated process—including conditioning, loading, rinsing, and elution—is executed based on a customized protocol controlled by a PC.

The automated separation instrument is currently applicable for large volumes of seawater samples. Kim et al. (2022)[5] demonstrated that the analysis of ^{90}Sr in a 60 L seawater sample could be completed within 4 hours, excluding measurement time. The study used DGA resin to purify ^{90}Y in equilibrium with ^{90}Sr at a loading rate of 80 mL min^{-1}. Additionally, Kim et al. (2023)[6] presented a new application of an automated sample loading instrument, which included a peristaltic pump and column parts, to concentrate radiocesium in a 60 L seawater sample to 0.08 L using AMP-PAN resin, achieving this within 4 hours.

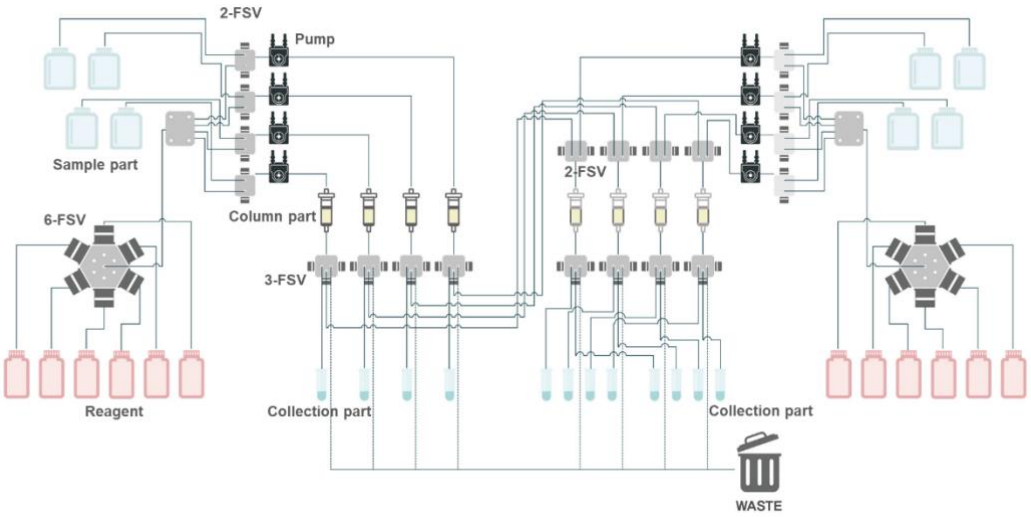

Figure 1. Diagram of automated radionuclide separation instrument

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Validation of a rapid method for ^{90}Sr determination in biota samples

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The establishment of rapid method for radiochemical separations is related to the need to assess the radionuclide concentration of a given sample in scenarios when a fast response is essential for decision making. In this work, we present the validation of a rapid method for ^{90}Sr determination in water and organic samples, including whole milk, which can be considered a complex matrix due to the occurrence of fat. This rapid method consists on a pretreatment step, in which strontium is co-precipitated along with calcium phosphate. The precipitate is dissolved and purified in order to remove interferents. Then, strontium separation is carried out using a Sr-resin column, precipitated as strontium carbonate and measured by Liquid Scintillation Counter. The validation of the method took into account the elapsed time for sample preparation and measurement, the reutilization of the Sr-resin column, and the assessment of quality parameters, such as recovery, accuracy and precision.

Acknowledgements:

This work was funded by Junta de Extremadura through the project “Realización de actividades relativas a la protección radiológica ambiental, a la vigilancia de las emisiones industriales y al mantenimiento de la red de medición de calidad del cielo nocturno y contaminación lumínica en Extremadura. Anualidades 2024-2027”. Expediente: 241803FD001

Comparing methods of digestion for the analysis of Strontium, Plutonium and Americium in solid samples

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At Nuclear Engineering Seibersdorf GmbH, the treatment of Austria's radioactive waste is managed. As there are no nuclear power plants in our country, only low and intermediate level active waste is treated in our facilities. Samples of the waste are analyzed in our laboratories for their elemental and radioactive properties. They appear in liquid, solid, and gaseous state. If the analysis requires, samples are processed and converted into the desired form.

In a first step, gamma measurement is carried out. For alpha and beta emitting HTM nuclides, predominantly Plutonium and Strontium, LSC and alpha counting is utilized. Also, Cerenkov-counting is applied for 90Sr.

For the analysis, solid and gaseous samples are brought into a liquid state. If the dissolved sample presents impurities, whether from chemical or physical contaminants or other nuclides, chromatographic separation is accomplished. The separation of Plutonium requires anion exchange chromatography with 242Pu added as a tracer. Dissolved in 8M HNO3, the sample is run through the column. After rinsing with additional 8M HNO3, Thorium is eluted with 10M HCl, and finally Plutonium with HCl/HI. This fraction, containing Plutonium only, is evaporated and taken in 0.05M HNO3. In this quality, LSC can be performed. Subsequently, for alpha counting, Plutonium is precipitated with NdF3.

Cerenkov-counting for 90Sr can be applied immediately after digestion of the sample. Correction for beta emitters, which can be identified by gamma spectrometry (e.g. 137Cs, 60Co), is often necessary.

The solid samples in our facilities discussed here are concrete, soil or sediments from our wastewater system. To dissolute a solid sample prior to analysis, two methods for digestion are implemented in our laboratories. For several years, microwave digestion with adding HNO3, applying pressure and elevated temperatures (220°C) to the dried sample was the method of choice. In 2021, Lithium borate fusion digestion ("*LiBO*") was introduced to our operational work. For this method, Lithium tetraborate as melting agent and Lithium bromide as wetting agent are added to the grinded and ignited sample. At 1150 °C the mixture is melted and finally poured into a solution of 8M HNO3 which is stirred until complete dissolution. To free the now obtained solution from silica, polyglycol is added which precipitates the accompanying compound. With centrifuging, the dense sludge can be removed. In this composition, silica is undesired as it clogs the column in the subsequent step of chromatographic separation.

For microwave digestion, the sample is leached with silica remaining unprocessed. When checking the residue with gamma spectrometry and elemental analysis, it was found that Iron, heavy metals, and gamma emitters are not completely dissolved.

In contrast, the complete dissolution in *LiBO* could be confirmed as we apply this method likewise to produce glass beads for XRF, resulting in a completely clear and homogeneous glass.

It was observed that comparing analysis for the two methods of digestion results in the finding of higher activity concentrations for *LiBO* than for microwave process for the same sample. This finding applies more for Plutonium than for Americium and Strontium. A possible explanation for higher findings might be the complete dissolution of the sample in *LiBO*.

Another effect is the inhomogeneous distribution of nuclides in the sample. Plutonium may be present in fine particles so that even grinding the sample to finest grain size prior to digestion does not guarantee a homogenous blend.

Comparing the ratios *LiBO*/microwave, not all isotopes of Plutonium present the same results. 238Pu, originating from subsequent neutron capture on 235U, provides different findings than 239+240Pu and 241Pu, originating from 238U. These different paths of origin may induce dissimilar solubilities of isotopes. In geological samples, this observation on 234U is known as radiogenic isotope effect.

In Table 1, a summary of the results is presented.

Table 1 Ratios LiBO/Microwave and alteration for findings

Nuclide	**Average Ratio**	**Median Ratio**	**Average Alteration**
238Pu	1,96	1,32	+ 96%
239+240Pu	1,95	1,32	+ 95%
241Pu	2,46	1,43	+ 146%
241Am	1,80	1,45	+ 80%
90Sr	1,22	1,14	+ 22%

Analysis of Iron-55 in Liquid Effluent of NPP

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As a metal material, iron is widely used in various components of reactors, and the corresponding activation products exist in the wastewater generated by reactors and are discharged to the environment in the form of liquid effluent. Radioactive iron discharged into the environment is easily absorbed into the food chain, and 70% of iron in organisms is combined with hemoglobin. Iron -55 is a large radioactive nuclide in the liquid effluent of nuclear power plants, and it has been reported that its emission in the liquid effluent can reach more than 12%, so many nuclear power plants have carried out monitoring of iron -55 in the liquid effluent.

Iron -55 emits low-energy X-rays and Auger electrons through EC decay, and is determined by liquid scintillation counting method after radiochemical separation. Aiming at the iron -55 in the liquid effluent of PWR nuclear power plant, based on the separation of anion exchange resin, the separation effects of hydroxide precipitation method and evaporation method were compared and analyzed, and the influence of TRU resin purification was also analyzed.

Iron -55 in the liquid effluent of PWR nuclear power plant was separated by three technical routes. a) After the liquid effluent was concentrated by hydroxide precipitation, it was separated by strong alkaline anion exchange resin, and the desorption solution was further purified by TRU resin to collect the desorption solution; b) After the liquid effluent is concentrated by hydroxide precipitation, it is separated by strong alkaline anion exchange resin, and the desorption solution is collected; c) After the liquid effluent is concentrated by evaporation, it is separated by strong basic anion exchange resin, and the desorption liquid is collected. After the desorption solutions obtained by different methods were sampled, the recovery rate of iron was determined by ICE 3000 flame atomic absorption spectrometer, and the radioactivity of iron -55 was determined by Quantulus 1220 ultra-low level liquid scintillation spectrometer.

The experimental results show that the detection efficiency of liquid scintillation spectrometer for iron -55 is 36%, and MDA is 0.32 Bq/L when the sample volume is 100mL. There is no obvious difference in the recovery rate of iron in liquid effluent among the three separation methods, all very high, ranging from 97% to 100%. The results of iron -55 in the liquid effluent obtained by the three separation methods were different. The activity concentration of iron -55 separated by hydroxide precipitation and anion exchange resin is 0.84 Bq/L, further purified by TRU resin is 0.71 Bq/L, and the activity concentration of iron -55 separated by anion exchange after evaporation concentration is 0.91 Bq/L.

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Radionuclidic purity in radiopharmaceuticals. Can 99.9% ever be measured?

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The widespread medical use of short lived radiopharmaceuticals for diagnosis (PET-scans) has put even short-lived compounds under pharmaceutical regulation. A very important example is the F-18 labelled glucose analog "FDG" with a physical half-life of just 110 minutes. According to current regulations, this product should have a verified radionuclidic purity (RNP) better than 99.9% … even at end of the shelf life of the product, typically 10 hours after production.

At face value, this requires that the producer has to exclude any foreign activity other than F-18 at a level of 3E-5 Bq/Bq at the time of manufacture. Some gamma spectroscopic methods are available, but not always with the necessary sensitivity, and the principal shortcomings here will be exposed. Beta spectroscopy and LSC has too low spectroscopic resolution for the job, leaving only half-life analysis as a universal tool.

Such method can however only capture impurities that are counted with the same or better sensitivity than the principal component (F-18).

An experimental implementation that uses a small, self-contained NaI scintillation detector has been developed and tested.

It can do the analysis and meet the requirement with measurements over less than 30 minutes, but certainly not for all impurities.

The relevance of this radio-analytical blind spot will be discussed.

Improving knowledge in marine science at Cefas using radioanalytical tools

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As an international leader in marine science, the UK Centre for Environment, Fisheries and Aquaculture Science (Cefas) have always been at the forefront of research activities aiming to keep our seas, oceans, and rivers healthy and productive and ensure our seafood remains safe and available, by providing robust data, evidence, and advice to diverse stakeholders.

Since the late 1940's, many radioanalytical methods have been developed at the Cefas Lowestoft laboratory to support these motivations. Initially, they were designed to enable scientists to study the direct effect of nuclear- and NORM-derived radioactivity on animal and human health and advise on the safe disposal of nuclear wastes The continuous development of new radiochemical preparative techniques, more sophisticated radiometric instrumentations, and more recently, benchtop state-of-the-art mass spectrometric techniques have allowed the exploration of new research horizons within the wider field of nuclear science and aligning our expertise with projects aiming to tackle global threats such as climate change or the presence of contaminants of growing concern, partially responsible for biodiversity loss.

In this presentation, major Cefas radioanalytical developments undertaken in the last seven decades and associated scientific contributions in marine science will be described. Future inspirations and directions, particularly where novel ICP-MS technologies can play an important part will also be shared with the audience.

Reconstructing $^{135}Cs/^{137}Cs$ signals released from Sellafield reprocessing plant and its potential environmental applications

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Isotope ratio of fission products $^{135}Cs/^{137}Cs$ is an emerging fingerprint for identifying the source of radioactive contamination and tracing the circulation of water masses in the ocean. The $^{135}Cs/^{137}Cs$ ratios in natural samples such as soil, sediments and seawater record a mixed signal from both the regional/local sources (e.g., nuclear power plant accidents, reprocessing plants) and the global fallout of nuclear weapon tests peaking in early 1960s, which are highly dependent on reactor and weapon type, fuel composition and operating condition (e.g., neutron flux, energy and irradiation time).

To estimate the contribution from different end-members, well-representative initial values and their evolving history are of necessity. However, available values for local nuclear installations are currently sparse and the signals are commonly assumed to be constant with time which is unlikely to be the case. Here we report the first time-series $^{135}Cs/^{137}Cs$ ratios derived from sediment cores collected in the vicinity of Sellafield reprocessing plant, providing the possibility to reconstruct the historical discharge of ^{135}Cs and end-member values of $^{135}Cs/^{137}Cs$ from Sellafield. With $^{135}Cs/^{137}Cs$ ratios obtained in the sediments from the North Sea, Greenland Sea and Lake Geneva, we identify the source term of radiocesium and estimate the contribution from different sources.

The ultra-trace level of ^{135}Cs and ^{137}Cs in these sediments requires well-controlled digestion, improved chemical separation procedure and measurement on triple quadrupole inductively coupled plasma mass spectrometry (ICP-QQQ). The $^{135}Cs/^{137}Cs$ ratio in the sediments from Lake Geneva is shown to be higher than the signals from Sellafield, which suggests a contribution from global fallout of early nuclear weapon test. While the $^{135}Cs/^{137}Cs$ signals in the North Sea sediments are within the range of Sellafield signals which may be the main source of radioactive contamination in the region. The Greenland Sea sediments show more variable $^{135}Cs/^{137}Cs$ ratios, which implies a changing dominance of the radiocesium source. This study provides the first time-series $^{135}Cs/^{137}Cs$ signals near Sellafield reprocessing plant, demonstrates the potential of $^{135}Cs/^{137}Cs$ ratio as a powerful forensic tool, and prompts the understanding of historical discharges from local point-sources.

Radionuclides in bovine muscle and liver from north-west Australia

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This presentation explores the determination of radionuclide levels in the typical Australian diet, with a specific focus on meat products. Cattle samples from Western Australia were analysed for radionuclides using an array of techniques. This study aims to add data to national baseline radionuclide levels in the Australian diet, an important area of public health protection.

Key points of the presentation include:

1. Development of sample preparation methods to concentrate sample volume and achieve lower detection levels.

2. Analysis of radium-226, caesium-137, lead-210, and uranium-238 to generate relevant radiological data for exposure assessment in the Australian diet.

The research achieved success in improving the available data on radionuclides in food, although it also encountered significant challenges. The process of sample pre-concentration for low-level radioactivity analysis required large sample volumes, which presented difficulties. Furnace ashing temperatures had limitations due to sample volatility, and balancing sample volume, with reactivity, proved challenging. Additionally, the inability to visually inspect samples during preparation led to error identification issues.

Specific problems included solubility and precipitation issues during separation chemistry and matrix issues affecting ICP-MS analysis. The conversion of lead to lead sulfate for liquid scintillation counting was only partially successful due to uneven heating, vessel overflow, and reconstitution difficulties.

Despite these challenges, plasma ashing emerged as a promising method for sample concentration, avoiding volatility and the need to sulfate volatile elements. Future research could focus on diluting plasma ashed samples into a larger volume of water and analyzing them using alpha spectrometry or liquid scintillation counting.

The significance of this research lies in its contribution to the limited data on radiation doses from the average Australian diet, with potential for interstate and international comparison. The study's ultra-low detection limits enabled by sample preconcentration steps open new avenues for examining matrices previously subject to higher detection limits. Additionally, the focus on specific Australian cattle tissues, such as muscle and liver, provides valuable insights compared to broader categories such as 'beef mince,' which could benefit organ-based uptake studies in animal models and dose assessments for Australian livestock.

Enhancing Radioactive Cs Adsorption: Mechanistic Insights of Prussian Blue Analog with Various Transition Metals

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Various materials such as zeolites, polyoxometalates, and sulfides have been used to remove ^{137}Cs. Among these, Prussian blue analogues (PBAs) have been highlighted for their ability to selectively and efficiently capture ^{137}Cs. Previous research confirmed that the combined effects of TiO_2 photocatalysis under ultraviolet (UV) light[1]. When exposed to UV light, TiO_2 absorbs light and generates electrons that convert Fe(III) to Fe(II) within PBAs, making it easier to capture additional Cs^+ ions from the solution. PBAs can also accommodate various transition metal ions instead of Fe(III), changing their ability to absorb light depending on the type of transition metal ion. NiFe PBAs, for example, doubled their cesium ion absorption under light exposure[2]. Furthermore, NiFe embedded in alginate hydrogel beads (NiFe-AH) showed a 1.5-fold increase in adsorption efficiency under light exposure. NiFe-AH also effectively removed ^{137}Cs from real seawater samples[3].

In this study, PBAs modified with different transition metals (Co, Cu, Fe, Mn, Ni, Zn) were synthesized and evaluated for their physical and chemical properties influenced by water molecules, such as surface area, ion exchange capacity, lattice parameters, and defect sites. Cs^+ adsorption varied significantly depending on the transition metal ions present in the PBAs. CoFe and FeFe PBAs predominantly exchanged Cs^+ with K^+ due to higher K^+ content, while CuFe and MnFe PBAs, characterized by elevated defect levels, primarily underwent H^+ and Cs^+ ion exchange. NiFe and ZnFe PBAs exhibited enhanced Cs^+ adsorption under light irradiation, attributed to their light-absorbing properties facilitating reduction reactions that enhance Cs^+ adsorption. Additionally, the application of PBAs in filter form for evaluating their efficiency in analyzing ^{137}Cs in seawater underscores their potential utility in rapid pre-treatment applications. In conclusion, PBAs modified with varied transition metals offer diverse Cs^+ adsorption mechanisms, showing promise for effective ^{137}Cs removal across different environmental contexts.

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Abstracts of posters

Validation of rapid method for ^{134}Cs single nuclide experiments

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The work presented here was developed for a larger project related to the evaluation of the stability of the structure of finely crystalline mica (illite) substituted with radioactive caesium cations. Cs-substituted illite like phase is prepared based on high-charge dioctahedral Al-bearing clay minerals.

The experiments used isotope ^{134}Cs, a commercially available analogue of isotope ^{137}Cs. The research aimed to identify new sorbents for environmental decontamination, e.g. water treatment of this important anthropogenic radionuclide. In experiments related to the sorption conditions of caesium, we required a rapid radioactivity measurement analysis for a single nuclide water solution of the isotope. The need was related to the characteristics of the experiments, the necessity for quick result verification and the limitation of radioactive and organic waste created after the analyzes using LSC.

^{134}Cs is a isotope emitting beta and gamma radiation with a short half-life of 2.06 years. ^{134}Cs decays primarily through beta decay, resulting in the formation of a stable barium isotope (^{134}Ba)[1]. This decay includes transitions to 3 different energy levels, connected with electron emissions of end-point energies 88.8 keV, 415.4 keV and 658.1 keV[2]. Successively different gamma-ray photons are emitted, with the highest probability of 97.63% and 85.47% corresponding to energies of 604.7 keV and 795 keV[1]. In gamma-ray spectrometry, the summation peaks that occur in the spectrum are also important, due to complexity of decay that emits cascades of single photons.

According to a standard laboratory procedure, gamma-ray spectrometry with an HPGe semiconductor detector or liquid scintillation counter (LSC) is used to measure the ^{134}Cs radioactivity for solid and liquid (or digested) samples, respectively. In this work, samples of aqueous solutions of the ^{134}Cs radioisotope were the object of research. The goal was to develop quick measurements of the ^{134}Cs content in aqueos solution using two measurement techniques. The radioactivity was determined using LSC. In addition, a method of analysis using a gamma-ray detector with a well-type NaI scintillation crystal was implemented. A diagram of the experiment setup is presented in Figure 1.

The LSC technique is exceptionally good for measuring caesium and beta decay isotopes in general, mainly due to its ability to measure in 4π geometry. This means a high counting efficiency, with a relatively low background compared to gamma spectrometry. However, the sample preparation procedure involves the use of scintillation cocktails characterised by the content of environmentally toxic compounds, e.g. nonylphenol ethoxylates. The average cocktail consumption for a single sample is 15 cm^3. Additionally, the procedure is destructive to the aqueous solution and the volume of radioactive waste increases. In the present work, the HIDEX LSC 300 SL measuring system was used, equipped with a low-level lead shielding. The absolute efficiency was determined on the quench curve. The Triple-to-Double Coincidence Ratio (TDCR) parameter was used as the quenching factor, since the measurement system was equipped with three photomultiplier tubes (PMT) operating in coincidence.

Additionally, a scintillation spectrometer equipped with a well-type NaI(Tl) detector and SSU-70-2 scintillation probe with Polon-alpha's RUM-2 analyser, and 10 cm thick lead shields was used. For

calibration purposes, a performance curve was prepared to avoid the influence of summation peaks, the intensity of which increases for close sample-detector geometry[3].

The measurements with the use of the NaI(Tl) detector are quite fast, did not require any special sample preparation or additional reagents and the method is nondestructive. However, because of the low interaction probability of gamma photons with matter, the absolute efficiency is much lower compared to that of LSC. The background spectrum is also at a higher level. Nevertheless such equipment is very useful and can be treated as a check point in preliminary experiments.

The result of this work is a range of caesium concentrations in the aqueous solution, in which the results are statistically correct.

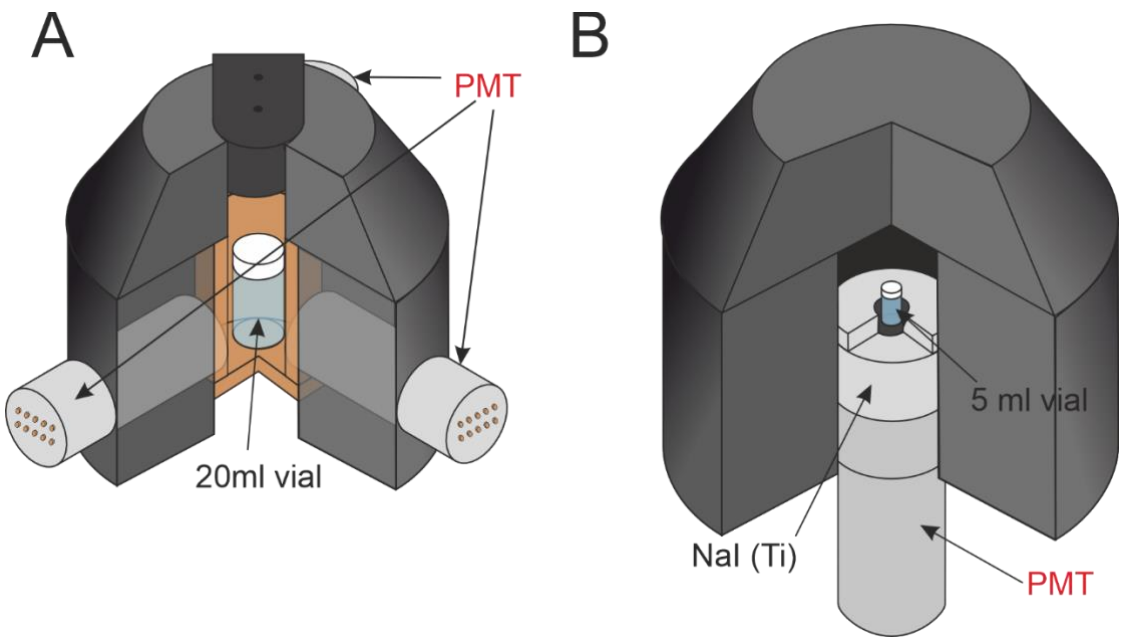

Fig 1. Diagram of measurement systems: A. Liquid scintilation counter with triple coincidence detector; B. well-type NaI scintillation detector

This work was supported by NCN grant 18.18.210.05850.

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Ultra-Sensitive Determination of Atmospheric Iodine-129 and Its Species Using Accelerator Mass Spectrometer

Luyuan Zhang [1*], Xiaolin Hou [1], Tong Zhang [2], Miao Fang [1], Huan Jiang [2], Ning Chen [1], Peng Cheng [1], Yunchong Fu [1], Qi Liu [1] and Hyuncheol Kim [3]

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Airborne radioactive iodine is a key concern for the transport and dispersion of radioactive pollutants and radiation exposure evaluation during nuclear accidents and nuclear emergency preparedness, as well as for understanding the atmospheric cycling of iodine. However, atmopsheric ^{129}I is difficult to measure due to its low concentration in remote areas, away from nuclear pollution sources. Here we have establised a series of methods for sensitive determination of atmopsheric ^{129}I and its species. A novel method for the determination of ^{129}I in aerosols collected on a glass fiber filter was developed using high-temperature pyrolysis and AgI-AgCl coprecipitation for separation coupled with highly sensitive accelerator mass spectrometry (AMS) measurements. It is worth noting that even though the pyrolysis behaviors of various iodine species were investigated and found to be different, all of the iodine can be quantitatively recovered. And iodide, iodate, NaOH soluble iodine, and insoluble iodine were separated from aerosols using sequential extraction, chromatography separation, and alkaline ashing and measured using AMS for ^{129}I. Recently, an ultra-sensitive analytical method was established for determination of ^{129}I in particulate, gaseous inorganic, and gaseous organic species, which was conducted with a self-designed cascade sampling apparatus, followed by their separation with a pyrolysis system and accelerator mass spectrometry. The detection limits were 0.05-0.22 × 10^5 atoms m^{-3} for ^{129}I. These established methods have been successfully applied to analyze the levels and species of ^{129}I in ambinet air from China and Demark, and to investigate transport of atmospheric iodine.

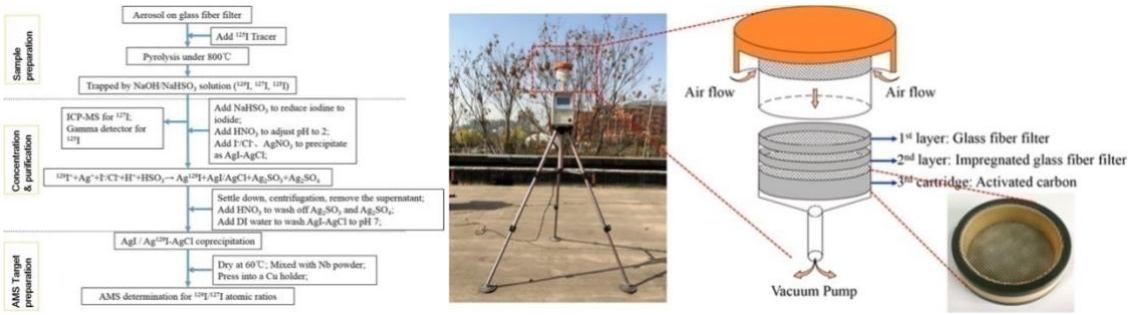

Fig. 1. A flowchart for the determination of aerosol ^{129}I using AMS, and schematic structure of the sampling collector for particulate, gaseous inorganic, and gaseous organic iodine.

Acknowledgement

This work was supported by the National Natural Science Foundation of China, the West Light Foundation, the Youth Innovation Promotion Association of the Chinese Academy of Sciences.

Simultaneous determination of actinides and ^{90}Sr in large-size soil and sediment samples

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There is a growing demand for ultra-trace determination of actinides in environmental samples for environmental monitoring, environmental tracing and nuclear forensic reasons. In this work, a total sample dissolution method based on lithium metaborate fusion, followed by sequential column chromatographic separation, was developed for simultaneous determination of Pu, Np, Am, Cm and 90Sr isotopes in large-size environmental samples by alpha spectrometry and mass spectrometric techniques.

The sample was melted using lithium metaborate fusion to ensure adequate isotopic exchange between potential refractory actinide species in the sample and the yield tracers. successive co-precipitation steps were firstly conducted to remove matrix elements, then sequential column separation method was applied for simultaneous separation and purification of actinides and ^{90}Sr/^{90}Y. The activity of ^{90}Sr could be obtained immediately by measuring its daughter radionuclide (^{90}Y) with triple-to-double coincidence ratio (TDCR) Cherenkov counting. The overall recoveries of Pu, Am, Sr and Y were higher than 70% for the entire procedure, the recovery ratio of Sr/Y were between 0.95 and 1.04 before chromatographic separation.

The developed method was verified using 20 g and 50 g of environmental soil samples spiked with certified reference materials IAEA-384 or IAEA-385 and standard solution of ^{90}Sr/^{90}Y, good agreement between the expected values and measured results has been achieved. The MDAs were found to be 0.31±0.10 mBq for $^{239+240}$Pu, 0.26±0.08 mBq for ^{238}Pu and 0.30±0.07 mBq for ^{241}Am for 20 g of soil by alpha spectrometry; and 0.08±0.07mBq for ^{239}Pu for 5g of soil by ICP-MS.

Due to perform the sample preparation (e.g. ashing, digestion, co-precipitation, etc.) only once, this method has particular advantages when several actinides and ^{90}Sr/^{90}Y must be simultaneously determined in a limited amount of environmental sample (e.g., soil or sediment core sample). The time required for analysis of a batch of 12 samples from the coprecipitation to the preparation of the final samples for the measurements is typically 2.5 days. Batch processing allows analyzing 24 samples in one week.

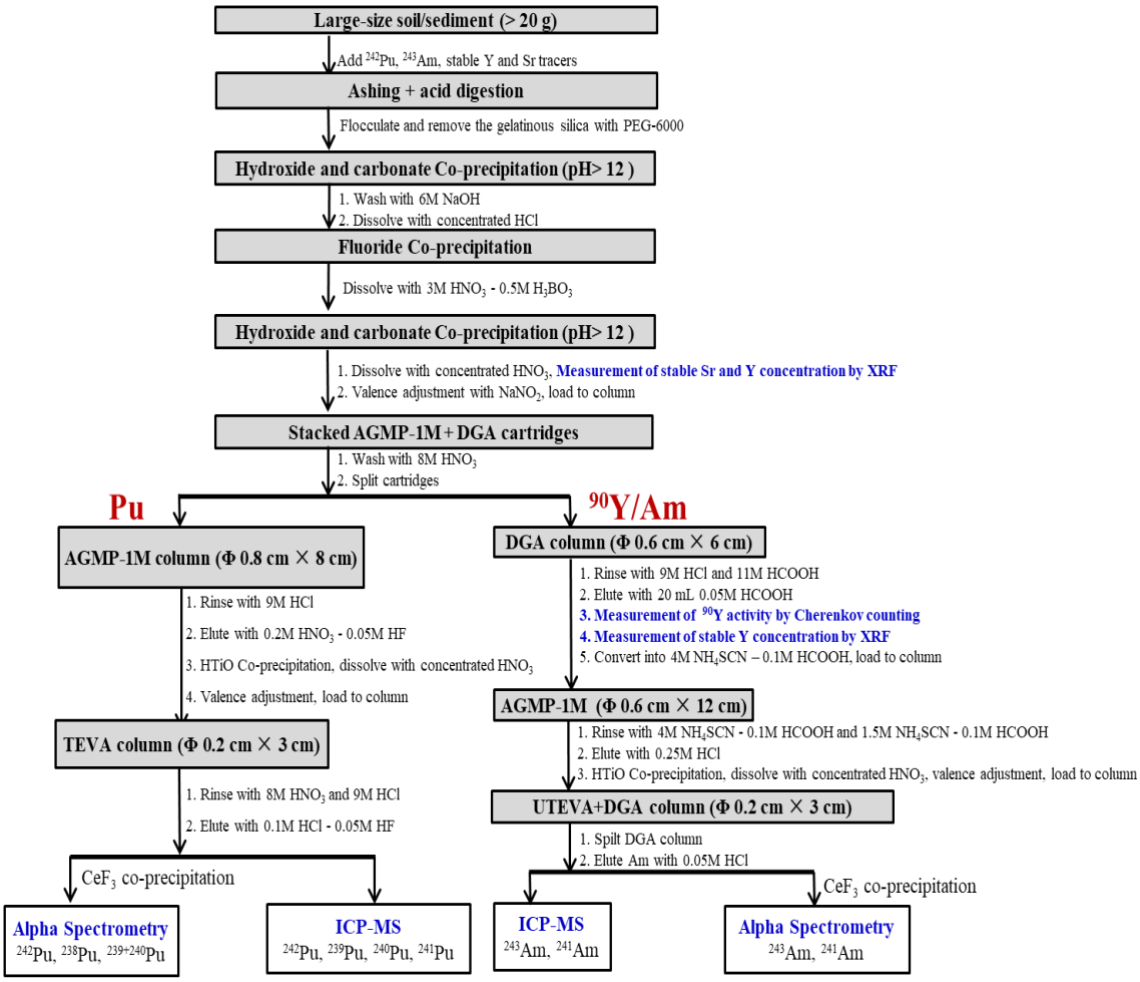

Fig. 1. Flow diagram of the sample preparation method for Pu, $^{90}Sr/^{90}Y$, Am in large-size soil and sediment sample

Rapid determination of ^{99}Tc in water samples using $Ti(OH)_3$-TcO_2 co-precipitation and TK200 resin by LSC

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Technetium-99 (^{99}Tc) is an artificial radionuclide with long half-life (2.11×10^5 a) and high accumulated fission yield from ^{235}U and ^{239}Pu. ^{99}Tc discharged into the environment originates mainly from nuclear fuel reprocessing plants (NFRPs). In the oxidation conditions, Tc is mainly present as the pertechnetate ion (TcO_4^-), which is highly soluble and mobile in the environment. Therefore, long-lived ^{99}Tc is considered to be one of the most important radionuclides in environmental safety assessment, nuclear waste management as well as radiation protection

A novel method for the determination of ^{99}Tc in water samples was developed using stable Re as a chemical yield tracer. The key to the utilization of stable Re tracer is to ensure consistent behaviors of Re and Tc during the entire chemical separation process, especially the co-precipitation and the chromatographic purification steps.The influences of several experimental parameters, including $TiCl_3$ concentration, HCl concentration and reaction time, on the reduction of TcO_4^- and ReO_4^- as well as $Ti(OH)_3$-TcO_2- ReO_2 co-precipitation were investigated. Tc(VII) and Re(VII) retained on TK200 resin were effectively eluted by 5 mL of 1 mol/L NH_4SCN, which can be directly mixed with the scintillation cocktail for liquid scintillation counting.. The results show that the chemical behaviors of Tc and Re are very consistent in the whole procedure. The decontamination factors of potential interferences from β-emitting nuclides mainly released from nuclear fuel reprocessing plants were also evaluated, and the minimum detectable activity concentration was calculated to be 0.08 Bq/L for ^{99}Tc in water samples with a counting time of 2 h.

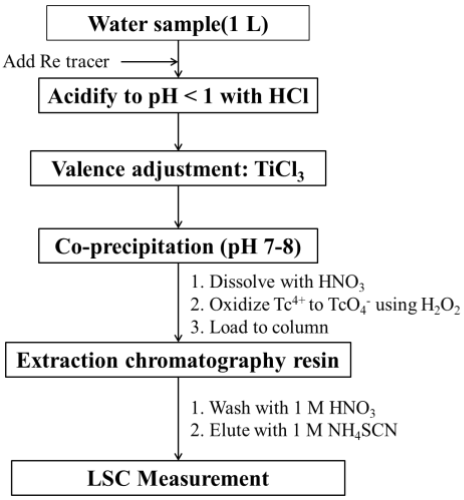

Fig. 1. Schematic diagram of the chemical procedure for separating ^{99}Tc from the water sample.

Using radiochemical fast methods for the characterisation of environmental samples during an emergency exercise

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The Austrian Agency for Health and Food Safety (AGES) conducts the environmental radioactivity monitoring in Austria. As for environmental monitoring the purpose is to determine even the smallest amount of radioactivity. Hence, the used methods and measurements are optimized to receive values with high accuracy close to the lower limit of detection appropriate for monitoring. In order to achieve that, high sample amounts and long measuring times are needed. In contrast to the environmental monitoring, there are situations where fast results are preferred over lower detection limits, e.g., in the case of a radiological emergency. In such situations the achievable lower limit of detection can be higher than for monitoring. For such circumstances, the AGES developed radiochemical fast methods that are tested on a regularly basis. For that purpose, our laboratory conducts a radiochemical emergency exercise once a year.

During such an exercise the AGES' laboratory determines the activity concentrations of Sr-89, Sr-90, Pu-238, Pu-239, Pu-240, Pu-241, Am-241 and Cm-244 in different environmental samples. For this year's emergency exercise soil, vegetation, potatoes, and drinking water have been analysed. The used fast methods were based on extraction chromatographic resins by TrisKem®. After the chemical separation, the activity concentration of Sr-89 was determined by Cherenkov counting. Subsequently, a cocktail was added to the Sr fraction in order to determine the Sr-90 activity by LSC. The alpha emitting radionuclides were analysed via alpha spectroscopy. Additionally, Pu-239 was also measured via ICP-MS. In the final step, Pu-241 was determined via LSC. In order to monitor the chemical recovery of the procedure, the radioactive tracers Pu-242 and Am-243 were used. The chemical yield for Sr was monitored by measuring the stable isotopes Sr-86 and Sr-88 via ICP-MS. For the purpose of verifying the used fast method, a reference material and spiked samples with known activities were analysed. Furthermore, the obtained results were confirmed by analysing the same samples with the methods used for the routine monitoring.

The details of the used fast method and the results of the conducted emergency exercise will be presented at the RAD Workshop. This project was funded by the Federal Ministry of Climate Action, Environment, Energy, Mobility, Innovation and Technology.

Reactor chemistry and pyroprocessing issues of TMSR

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Molten salt reactor is a homogeneous liquid reactor, and the fuel and fission products are dissolved in salt. But in fact, there are gases (gas fission products, volatile fission products, tritium, Ar gas), solids (noble metals and other fission products that may be deposited), graphite and alloy materials that are in contact with the fuel salt. Due to the coupling effect of their chemical state and the characteristics of nuclide decay in the reactor, therefore, the behavior of the fuel salt and nuclides in it is very complicated. The interaction and dynamic change between them and the components of fuel salt are very important for the operation and safety of the reactor. Some important issues must be given attention: (1) U^{4+}/U^{3+} ratio: the rate of corrosion of Hastelloy N, and the distribution of certain fission products (e.g. the noble metals Nb, Mo, Ru, Sb, Te, Tc). Knowing and controlling the redox potential of fuel salt is very important for maintaining long-term stable operation of MSR; (2) Oxygen content in molten salt: reduce the solubility of nuclear fuel, for example, uranium oxide would slowly precipitate, and cause local overheating of the fuel circuit; or affect the chemical behaviors of fission products and some corrosion products. The requirement of oxygen content in molten salt reactor is less than 200 ppm.; (3) Radioactive nuclide quantification: neutron poison for the reactor operation (fission products in reactor), critical parameters and reactor reactivity for the fuel cycle and reactor safety (actinide nuclides in reactor), and volatile fission products distribution and reactor/gas filter emission for the environmental safety (volatile fission products).

Another objective of the TMSR is the utilization of thorium and the realization of the closed Th-U fuel cycle. During ^{232}Th breeds to ^{233}U, there is an intermediate nuclide ^{233}Pa. The half-life of ^{233}Pa is about 27 days, which is an order of magnitude longer than the counterpart of U-Pu fuel cycle, ^{239}Np (half-life of 2.35 days). It means that the accumulation of ^{233}Pa in the Th-U cycle reactor is more easily as comparing to U-Pu cycle. Since ^{233}Pa has significant thermal neutron absorption cross-section, large amount of ^{233}Pa accumulating in the reactor core will drastically affect the reactivity via (n, γ) reaction to form ^{234}Pa, and then decay to ^{234}U. As a result, it consequently decreases the breed ratio by consuming the precursor of ^{233}U; and also decreases the reactivity of the reactor by consuming the neutrons. Therefore, in order to maintain the reactivity of the reactor at high level, ^{233}Pa should be isolated interval or continuously from the fuel salt to ensure the effectively breeding of ^{232}Th to ^{233}U. This is a key factor of designing and running TMSR. As a result, from the economic viewpoint, the treatment of out-reactor spent fuel includes recovery fission materials, isolation ^{233}Pa, removing FPs as well as reuse the coolant, namely, the majority of the $^{7}LiF\text{-}BeF_2$.

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1.

The distribution and behavior of actinides and fission products in molten fluoride salt

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Molten salt reactor (MSR) is a unique liquid fuel reactor among the Fourth-Generation reactors. The distribution and behavior of actinides, fission products, and corrosion products in the presence of high-flux neutrons, gamma rays, and high-temperature molten salt have an important impact to the neutron economics, the efficiency of fuel, the breeding rate, and the control of the reactor. Thereby, it is necessary to fully understand the physico-chemical behavior of actinides and fission products in MSR. For this purpose, a radiochemical method of actinides and fission products in molten salt was established using gamma spectrometry, and their behavior and distribution in molten salt were investigated.

As shown in figure 1 and figure 2, the results indicated that ^{237}U, ^{239}Np, ^{143}Ce, ^{95}Zr, and ^{140}Ba were almost all dissolved in molten salt. The vast majority of ^{99}Mo, ^{132}Te and ^{103}Ru settled to the bottom of the molten salt, and the rest were deposited on the graphite and GH3535 alloy. In addition, a small amount of ^{131}I, ^{132}I, ^{115m}In, and ^{115}Cd were volatilized and deposited on nickel foil. Interestingly, it was also found that the behavior of certain fission products, such as ^{95}Nb, ^{99}Mo, and ^{131}I, is particularly sensitive to the oxidation-reduction state of molten salt, which implied that they could be used as redox indicator for MSR. In practice, MSR is a kind of chemical reactor, or so-called chemists' reactor. More experimental studies are needed to understand the various complex chemical reactions and their effects in MSR.

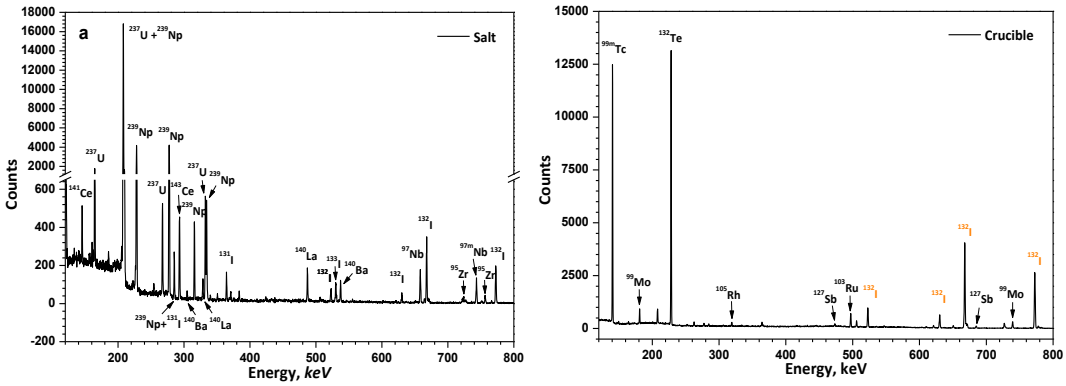

Figure 1 γ-ray spectra of molten salt and graphite crucible

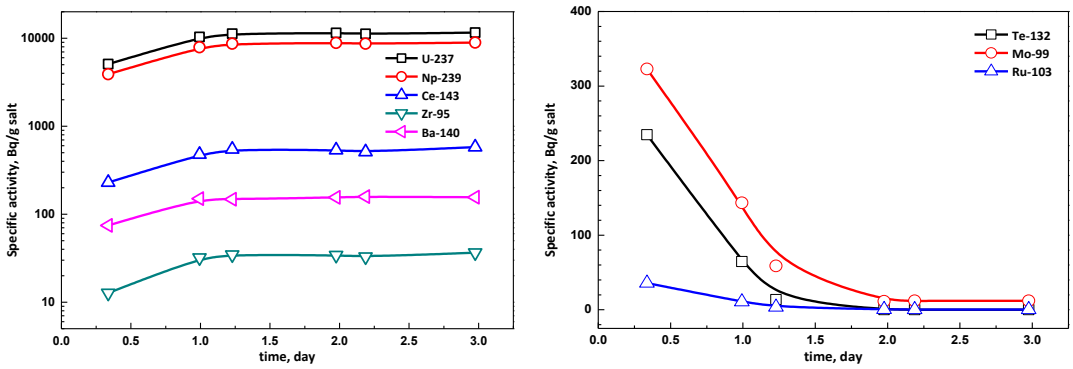

Figure 2 Specific activities of actinides (^{237}U, ^{239}Np) and fission products (^{143}Ce, ^{95}Zr, ^{140}Ba, ^{99}Mo, ^{103}Ru and ^{132}Te) in molten salt, (after correction for its decay)

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Appendix 4 – Workshop photos (selected)

Bibliographic Data Sheet **NKS-489**

Title	NKS-B RadWorkshop 2024 proceedings
Author(s)	Jixin Qiao [1], Matic Dokl [1], Eike Rades [1] Karl Andreas Jensen [2], Ole Christian Lind [2], Lindis Skipperud [2] Susanna Salminen-Paatero [3] Marion Grange [4] Torbjørn Gäfvert [5], Aslak Roalkvam Skåra [5] Maria Kaipiainen [6], Valtteri Suorsa [6]
Affiliation(s)	[1] Technical University of Denmark (DTU) [2] Norwegian Life Science University (NMBU) [3] University of Helsinki (UH) [4] Swedish Food Agency (SLV) [5] Norwegian Radiation and Nuclear Safety Authority (DSA) [6] Radiation and Nuclear Safety Authority, Finland (STUK)
ISBN	978-87-7893-585-4
Date	November 2024
Project	NKS-B Radworkshop 2024 (AFT/B(24)2)
No. of pages	92
No. of tables	1
No. of illustrations	2
No. of references	19
Abstract max. 2000 characters	The NKS-B RadWorkshop 2024, held during 9-13 September 2024, at the DTU Risø Campus in Roskilde, Denmark, brought together 104 participants from nearly 60 organizations in 20 countries to focus on advancements in radioanalytical chemistry and its application in various fields. The workshop featured a blend of 3-day keynote lectures, presentations, posters and 2-day hands-on laboratory training, covering topics such as radiochemical separation techniques, measurement methods, and applications in environmental monitoring, nuclear decommissioning, emergency preparedness, radioecology and tracer studies. The event successfully enhanced the education and expertise of attendees, fostering international collaboration and innovation in the field.
Key words	Radioanalytical chemistry, workshop, environmental monitoring, nuclear decommissioning, emergency preparedness, radioecology, tracer studies