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Cores Symposium on Radiation in the Environment

Scientific Achievements and Challenges for the Society

Sisko Salomaa, Merja Lusa, Kaisa Vaaramaa (eds)

Säteilyturvakeskus
Strålsäkerhetscentralen
Radiation and Nuclear Safety Authority

STUK

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Keywords: radioecology, radiochemistry, metrology, transfer of radioactive substances in the environment, modelling, effects on biota

Abstract

The Finnish Consortium for Radiation Safety Research (Cores) organized a symposium "Radiation in the environment – scientific achievements and challenges for the society" in Helsinki on April 16–17, 2018. This report provides a compilation of the abstracts of the presentations, oral as well as posters, given in the symposium. The local organisers were University of Helsinki and STUK – Radiation and Nuclear Safety Authority. The objective of the symposium was to bring together scientists working in the broad field of environmental radioactivity, in particular among the Cores consortium, as well as stakeholders and end-users of the research. The symposium also honored the long and esteemed career of Prof. Jukka Lehto in radiochemistry upon his retirement from the position of professor of radiochemistry at the University of Helsinki. Altogether 109 participants registered in the symposium.

The topics for the symposium covered analytical radiochemistry and various aspects of environmental radiation and radioecological research, ranging from radioactive fallout and deposition of nuclear waste to naturally occurring radioactive materials (NORM) and radon, modelling of transfer of radioactive substances in the environment as well as the effects of environmental radiation exposure on man and the biota. A summary of the presentations in the oral sessions is given in the introduction to the report.

SALOMAA Sisko, LUSA Merja, VAARAMAA Kaisa. Cores-symposium säteilystä ympäristössä – Tieteelliset saavutukset ja haasteet yhteiskunnalle. Tiivistelmät. STUK-A261. Helsinki 2018, 159 s.

Avainsanat: radioekologia, radiokemia, metrologia, radioaktiivisten aineiden kulkeutuminen ympäristössä, mallintaminen, vaikutukset eliökuntaan

Tiivistelmä

Kansallinen Säteilyturvallisuustutkimuksen yhteenliittymä (Cores) järjesti symposiumin ”Radiation in the environment – scientific achievements and challenges for the society” Helsingissä 16.–17. huhtikuuta 2018. Tähän raporttiin on koottuna symposiumin suullisten esitelmien ja postereiden englanninkieliset tiivistelmät. Järjestäjinä toimivat Helsingin yliopisto ja Säteilyturvakeskus. Symposiumin tavoitteena oli saattaa yhteen ympäristön radioaktiivisuuden parissa työskentelevät tutkijat, erityisesti Cores konsortion piirissä, sekä sidosryhmät ja tutkimustiedon loppukäyttäjät. Symposiumilla haluttiin myös kunnioittaa Helsingin yliopiston radiokemian professorin tehtävästä eläkkeelle siirtyvän prof. Jukka Lehdon pitkää ja ansiokasta uraa. Kaikkiaan symposiumiin ilmoittautui 109 osanottajaa.

Symposiumin aihepiiri kattoi analyyttisen radiokemian lisäksi useita ympäristön radioaktiivisuuteen ja radioekologiseen tutkimukseen liittyviä teemoja aina radioaktiivisesta laskeumasta ja ydinjätteen loppusijoituksesta luonnon radioaktiivisiin materiaaleihin (NORM) ja radoniin, sekä radioaktiivisten aineiden kulkeutumisen mallintamiseen ja ympäristön säteilyaltistuksen vaikutuksiin ihmiseen ja eliökuntaan. Raportin johdannossa esitetään järjestäjien yhteenveto symposiumin tieteellisestä annista.

Contents

Introduction

Sisko Salomaa, Merja Lusa, Kaisa Vaaramaa

The Cores Symposium on Radiation in the Environment – Scientific Achievements and Challenges for the Society was designed to address one of the main research areas of the national programme for radiation: the environment. The symposium had five sessions that are briefly summarized below. More details can be found in the abstracts.

The first session on Analytics and metrology was chaired by Dr. Merja Lusa. In his Honorary Lecture, prof. Jukka Lehto reviewed the history of environmental radiation research in Finland. The major starting point was the era of atmospheric nuclear tests in the 1950's and 60's when prof. Jorma Miettinen and scientists from the Meteorological Institute started the environmental follow-up studies of fallout nuclides in the Finnish environment, in particular in Lapland that was closer to the Soviet test site in Novaja Zemlja. In the 1960´s and 70´s, the Finnish scientists had a key role in the discovery of radon as major contributor to radiation exposure of citizens via drinking water and indoor air. Chernobyl accident in 1986 boosted research programs and the fallout in Finland was studied in detail. Finland is the first country in the world starting the final deposition of nuclear waste. This process has been accompanied by a national research programme considering potential long-term consequences and carefully studying physico-chemical processes in the bedrock and the environment. Dr. Maarit Muikku gave an introduction to the Strategic Research Agenda of the European Radioecology Alliance, encouraging universities to participate in the joint research activities. The three main research challenges of the ALLIANCE are: 1) to predict human and wildlife exposure in a robust way by quantifying key processes that influence radionuclide transfers and exposure; 2) to determine ecological consequences under realistic exposure conditions, and 3) to improve human and environmental protection by integrating radioecology. Prof. Sisko Salomaa then gave introduction to the Cores consortium that was formed after the Government decided to introduce a comprehensive reform of the Finnish research and innovation system in 2013. At the moment, the Cores consortium has 11 members, STUK as well as ten universities. The first version of a National Programme for Radiation Safety Research

was published in 2015. The research areas for the national programme include health (low dose risk as well as medical use of radiation), environment (radioecology) and emergencies (emergency preparedness and response, security of sources). As cross cutting themes, risk assessment, risk management as well as technological development (metrology and dosimetry) are addressed. Dr. Roy Pöllänen told about the metrology for processing materials with high natural radioactivity, such as oil and gas production, mining activities, rare earth extraction, water treatment and phosphate fertilizer industry, where Naturally Occurring Radioactive Materials (NORMs) present in raw materials may result in enhanced radiation exposure especially for workers but also for public in the surrounding environment. To ensure safety and protection against NORM, the regulation of NORM practices and NORM waste management is necessary. In addition, up-to-date measurement methods are of importance for controlling the radiation exposure to the workers and public. A very promising example is the development of an in-situ alpha spectrometer at STUK by Dr. Pöllänen.

The second session on Radioactive substances in geosphere was chaired by Dr. Nina Huittinen.

Transfer of radionuclides in porous bedrock was discussed by Dr. Marja Siitari-Kauppi. Deep geological repositories are being considered for long-term disposal of spent nuclear fuel in multiple countries. Safe disposal of spent nuclear fuel requires information about the radionuclide transport and retention properties within the porous and water-containing rock matrix and water flowing fractures. Dr. Siitari-Kauppi's group has investigated the transport properties of safety-relevant radionuclides in the geosphere and engineered barriers to provide parameters for the safety analysis of the repository. The research topics included e.g. sorption, diffusion and structure characterization in laboratory and in situ experiments. The studies involved long-term in situ experiments as well as laboratory studies and interpretation of the results through modelling. Mobilization of radionuclides and trace metals in tailings from Rautuvaara mine was described by Mila Pelkonen. The old Rautuvaara mining area is located in the municipality of Kolari. Currently there are no mining activities at the Rautuvaara mining area, but it has become an object of interest, since an on-going mining prospect in Hannukainen is planning on transporting their produced tailings to Rautuvaara and depositing them above the existing ones. Sulphide-bearing ores and mine waste are known to form acid mine drainage (AMD) when sulphuric acid forms in the oxidative dissolution of sulphide minerals. The decreased pH in the area affected by AMD can further dissolve and mobilize harmful elements from the surroundings minerals, which may contaminate the natural water systems. Therefore reference data were obtained for possible future need. Dr. Richard Andrew Klos reviewed

the evolution of the regulatory framework for use in the radiological assessment of potential releases of radionuclides to the biosphere from waste disposal facilities on timescale beyond a few millennia. Finland and Sweden lead the world with their advanced programmes for geologic disposal. The detailed site investigation programmes, coupled with global information system techniques allow details of the site to be processed and understood in unprecedented detail. As the capabilities of the disposal programmes evolve this feeds into the model description.

The third session on Transfer of radionuclides in the environment was chaired by prof. Sisko Salomaa. Dr. Jari Turunen presented sensitivity analysis of radionuclide transport in biosphere analysis. Sensitivity analysis is an important tool dealing with uncertainties in modelling. Sensitivity analysis can give insights for understanding the dependence between input and output parameters and checking errors of the model. They used a farm scenario where the radionuclide transportation to humans is analyzed using Ecolego simulation tool and analyzed the model parameters using five different sensitivity analysis methods. He concluded that the tested sensitivity analysis methods are capable to order the largest influence parameters in correct order. Dr. Jarmo Ala-Heikkilä told about Finnish technologies for monitoring The Comprehensive Nuclear-Test-Ban Treaty, CTBT. Even though not still in force, preparations for the CTBT and its verification are ongoing. A Preparatory Commission for the CTBT Organization has been established in Vienna and a global verification regime has been built. Finland has contributed to the technological development by the high-volume samplers for airborne radionuclide detection by Senya Oy and the analysis software Sampo and Shaman for gamma-ray spectrometry developed at TKK. Prof. Jaakko Leppänen gave an introduction to Serpent Monte Carlo code for radiation transport code applications. Serpent is a three-dimensional continuous-energy Monte Carlo transport calculation code, developed at VTT Technical Research Centre of Finland. During the past few years the field of applications for the Serpent code have been broadened from reactor physics to fusion neutronics, radiation transport and shielding, and the code is becoming a practical calculation tool for the radiation safety community. Ari Ikonen addressed biosphere modelling in safety assessments for various purposes. In general, such assessments comprise of relatively similar components, and here the focus is on assessments for radioactive waste disposal, even though the same principles can well be, and have been, applied for example to normal-operational and accidental releases from nuclear power plants and other nuclear facilities or to evaluate situations with naturally occurring radioactive materials (NORM) posing a concern.

The fourth session on Uptake of radionuclides in biosphere was chaired by Dr. Kaisa Vaaramaa. Prof. Jukka Juutilainen presented observations on non-linear transfer of elements into organisms, challenging the concept of element- and organism-specific concentration ratios (CR) that are generally used in radioecological models to describe the transfer of radionuclides into organisms. The CR approach is based on the assumption that uptake into an organism is linear with respect to concentration in the medium (soil or water). They carried out a series of studies on the uptake of elements into terrestrial and aquatic organisms using samples collected from the nature and experimental meso- and microcosms. The initial aim was to determine CR values relevant to boreal ecosystems, but it was soon found that the CR approach was not adequate, and studying non-linearity of uptake became the main focus of the studies. Although the studies aimed at understanding uptake of radionuclides into plants and animals, total element concentrations were measured. This approach is often used, based on the assumption that radioactive and stable isotopes of the same element are taken up similarly. Dr. Nina Huittinen reported on Cm complexation with aqueous phosphates at elevated temperatures. Orthophosphate ions may originate from the natural decomposition of rocks and minerals, agricultural runoff, or from wastewater treatment plants. Phosphates are strong complexants and can be expected to influence the speciation of dissolved radioactive contaminants. Very little data is available on the complexation of actinides with aqueous phosphates. In addition, the existing data suffers from systematic absence of independent spectroscopic validation of the stoichiometry of the proposed complexes. In this study, laser-induced luminescence spectroscopy has been used to study the complexation of Cm as a function of phosphate concentrations in the different temperatures. The new thermodynamic data derived in this fundamental study will contribute to a fundamental process understanding necessary to critically assess the environmental fate of actinides in the environment. Dr. Merja Lusa reported the effects of bacterial selenium oxyanion reduction on selenium transfer on plants. Microorganisms in the environment can modify soil characteristics through e.g. oxidation and reduction reactions and secretion of metabolites, altering the availability of chemical elements for plant-uptake. Soil bacteria can form favourable symbiotic associations with plants (like nitrogen fixing root nodule bacteria), but they can also enhance mobilization of hazardous substances, such as radionuclides or heavy metals in soil, increasing their accessibility to plants and further accumulation into the food chains. Translocation and accumulation of radionuclides and heavy metals to plant biomass and to edible crops is extremely undesirable, but as bacteria and plants can be utilized as phyto-bioremediation agents, these bacteria-plant interactions can be harnessed to benefit the environment and mankind. The

chemical compounds of interest (e.g. radionuclides, heavy metals) can be made more accessible for microbe-co-cultivated plants, by treating the contaminated areas with appropriate bacteria. The undesirable compounds can thereafter be removed from the soil, by harvesting the plant material. Dr. Antti Kallio gave an overview on an investigation on radioactivity in ashes from bioenergy production in Finland that was carried out during 2015-2017 by STUK in co-operation with the bioenergy producing industry. Ash samples from 11 different bioenergy producing facilities were analyzed at STUK for Cs-137 and selected natural radionuclides using gamma-spectrometry. The measured activity concentrations were used to estimate the level of potential radiation exposure from the handling and reuse of ash. STUK guide ST 12.2. contains activity indices which can used to estimate whether the use of a material in certain predetermined ways can lead to exceeding the reference levels for public or occupational radiation doses. The results show that there would be no restrictions to the handling of the ashes from the radioactivity point of view, as all the samples have activity indices of <1 for handling of ash. The activity indices for building materials, road construction, landfill and landscaping were exceeded for some of the fly ash and grate ash samples. If these materials were to be used for such purposes, a more detailed dose calculation would need to be made by the applicant to demonstrate compliance with the reference levels.

The final session on Effects on humans and biota was chaired by Prof. Jukka Juutilainen. Prof. Anssi Auvinen reported on the results of the Finnish register-based case-control study of childhood leukemia for residential background gamma radiation exposure and risk of childhood leukemia. The results provide some support for involvement of low doses of ionizing radiation from natural background radiation in the etiology childhood leukemia, particularly at the ages of the highest leukemia incidence. However, background radiation may account only for a small fraction of all cases, as the doses and, accordingly, the risks are low. A significant effect was found only in the age group 2-7 years, but the risk estimate was substantially larger than expected and the findings of this sub-group analysis may be partly explained by random error. The findings suggest some potential differences by leukemia subtype (in particular possibly larger risk associated with high hyperdiploidy), but further confirmation is needed. Kati Kivisaari reported on effects of ionizing radiation on wild rodent populations in Chernobyl exclusion zone, studying whether low levels of chronic radiation can cause selection pressure in wild populations. Studies on the bank vole (Myodes glareolus) showed that there was similar organ related damage related to brain size and cataracts in eyes as observed from barn swallows. A negative correlation between radiation level and sperm size and quality was found and radiation also had a significant negative effect on litter sizes. In

addition, there was evidence of increased oxidative stress both in genetic side and fenotypic properties. Dr. Jarkko Akkanen introduced the concept of microcosms for testing of individual stressors and environmental samples. While standardized ecotoxicity testing is based on single species assessment using individual level end-points, microcosms allow the assessment of interactions of species that are selected to represent ecologically important trophic levels.

Overall, the presentations provided an excellent overview of the scientific achievements and societal challenges related to the environmental radiation research in Finland. The organisers wish to thank the participants for the high quality of the presentations and the lively discussion.

Session 1: Analytics and metrology

Chair: Dr. Merja Lusa

Environmental radioactivity research – where to go?

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Nuclear weapons test fallout studies – radioecology competence developed

Environmental radioactivity in Finland is an established research field and has long traditions. Research started at the University of Helsinki and at the predecessor of the Finnish Authority for Radiation and Nuclear Safety (STUK) in the last years of 1950's. The most important task at the time was to clarify the effects of radioactive fallout from the atmospheric nuclear weapons tests in the environment and food chains. A major achievement from that period was the study on a critical radiocesium-enriching food chain from lichen via reindeer into reindeer-herding Sami people. Fallout studies remained the largest field in environmental radioactivity studies in the 1960's to 1970's and were extended to environmental behavior of transuranium elements in the early 1970's.

Importance of indoor radon becomes evident

Early 1980's the researchers at STUK revealed the importance of indoor radon to radiation doses to humans and in the coming years radon research of this became more and more important. Today we know that indoor radon is responsible for more than half of the average radiation dose to Finnish population and measures to decrease its intrusion into dwellings have been developed at STUK.

Fallout of the Chernobyl accident requires good competence

The competence developed in the weapons tests fallout studies became very important in 1986 when Finland was exposed to a radioactive fallout from the

Chernobyl accident. In fact Finland received the largest fallout excluding the three closest countries, Ukraine, Belorussia and Russia. The environmental surveillance and research department of STUK had considerably increased to about 60 persons of which 20 were researchers. Thus they had good resources to react to the accident which they very well did. Department of Radiochemistry, University of Helsinki, focused on the behavior of transuranium elements in the environment and, for example, together with STUK and the Finnish Meteorological Institute unique spatial distribution maps of Pu, Np, Am and Cm in Finland were developed. It is no exaggeration to say that no other country has such a deep knowledge on distribution and behavior of radionuclides from the Chernobyl fallout on their own territory.

National needs to study naturally occurring radionuclides

Through intensive fallout studies we have, however, learned to know that the radioactive pollution has only a minor effect on the radiation doses to humans and also in the most critical groups, such as reindeer herders in 1960's and most exposed groups after the Chernobyl accident, the doses from pollution radionuclides have been below that received from indoor radon on average. During the last twenty years or so, the importance of naturally occurring radionuclides has gained more attention in research. Studies like uranium series radionuclides U, Ra, Po, Pb in the drinking water and in forest environment have tackled important national problems and considerably increased our knowledge in these areas. Latest achievements are studies on the behavior of naturally occurring radionuclides in the mining processes. Due to increasing mining activities and potential this field definitely needs further attention.

Radiochemistry at the University of Helsinki in key role in educating experts

From the very beginning the environmental radioactivity studies have been mainly implemented by physicists and chemists. The chemists working in the field have been mainly educated at the Radiochemisty Department/Laboratory/Unit of the University of Helsinki. The unit has almost 60 years' experience in working in this field, so it has required traditions and knowledge to educate new experts to the field. It has relevant facilities, including laboratories and instruments, to do work in this field. Radiochemistry education at the University of Helsinki has internationally a unique, extensive teaching program covering handling and measurement of radionuclides and behavior of radionuclides in the environment.

Where are we now? – Where to go?

The environmental radioactivity research in Finland is a well-established discipline. The resources have, however, considerably decreased during the last twenty years. There are only a few institutes carrying out research in the field. Including those already mentioned, research is being done also in the University of Eastern Finland, for example. In all of them except STUK the personnel working on environmental radioactivity consists of only a few members funded mainly by short-term projects. The resources of STUK's research have considerably decreased due to great funding cuts by the government and thus STUK mainly focuses on environmental surveillance.

Even though it would be unrealistic to think that the greatest days of environmental radioactivity studies could be regained there are still important reasons to continue the research and not only continue but deepen it. The reason to continue the research is not because completely new radiation sources would be discoverable or because uncertainty of presently known sources would be large. We need environmental radioactivity research first of all because it is of utmost importance to keep up knowledge and competence for radiological preparedness. This is required to adequately react to potential nuclear accidents, attacks and other unexpected events. For this, Finland needs not only highly qualified researchers but also competent authorities to advice decision makers in such situations. So far, the quality of researchers and authorities has been on a top level internationally. To keep the level high, high quality research is needed; short courses or only master's level education is not enough to guarantee the competence.

Second reason to continue the research is that there still are some areas what we know a lot too little about. This applies particularly to environmental effects of mining industry which has a great potential to increase in Finland. A great number of mines and potential mining sites are located in Lapland where the nature is more vulnerable than in other parts of Finland. General knowledge on the effects of mining already exists but each mining site and process has its own characteristic features that need to be taken into consideration when assessing their potential environmental and health effects.

Third reason to continue the research in this field is the deepening of our knowledge on various processes, such as various transfer processes in the environment. We know already most important processes at macro scale but not, or at least not well, the processes and mechanisms at atomic and molecular scale. Deepening the knowledge using old tools is not possible. Traditionally environmental radioactivity research has limited to measurement of radionu-

clide concentrations in various compartments (soil, vegetation, animal etc.) and modelled the results using simplified compartment models. This might still be a right approach for environmental monitoring for authority purposes but for modern research it is not that. Research at atomic and molecular scale requires new, modern methodology, all not even available in Finland. The competition of research funding from national sources, such as from Academy of Finland, is strict and gets even stricter. To find funding for environmental radioactivity research, it of course needs to be socially important but also scientifically at a high level. This cannot be guaranteed only by traditional, conventional tools.

Furthermore, new insight into processes can be gained from frontiers and interfaces of disciplines. The environmental processes are very complicated and cover areas traditionally studied by various disciplines at their own compartments without contacts to others. Microbes, for example, play an important role in most environmental processes. Combining traditional or more advanced radiochemical methods with those of microbiology has increased our knowledge on processes taking place in the environment.

Since the processes occurring in the environment are complicated and cover a number of components and variables, the processes and mechanisms may not necessarily be solvable at natural conditions. Model experiments using stripped systems in laboratory are needed. Here the traditional environmental radioactivity research could follow the example of research carried to study geological behavior of radionuclides from nuclear waste. In that field laboratory-scale model experiments in controlled conditions are everyday life. With these techniques parameters, such as pH or ionic strength for example, can be varied systematically and large number of data can be attained in relatively short time.

At last, modern research requires high level mathematical modelling. The models used by authorities are simple and robust and it understandable that they also should be that. But what we need more are models for scientific purpose, especially for various transfer processes. They should combine various physical and chemical processes and try to help not only understanding better the processes but also predicting them in varying conditions.

The European Radioecology ALLIANCE – the Strategic Research Agenda and associated topical roadmaps

Maarit Muikku
Radiation and Nuclear Safety Authority (STUK)

Abstract

Members of the European Radioecology ALLIANCE bring together parts of their respective research and development programmes into an integrated programme that maintains and enhances radioecological competences and experimental infrastructures, and addresses scientific and educational challenges in assessing the impact of radioactive substances on humans and the environment. The ALLIANCE Strategic Research Agenda created under EU-funded STAR Network of Excellence in 2012 provides a long-term vision of radioecological research needs. It has been complemented later on by the first transitional roadmap and the topical roadmaps dealing with specific scientific research areas.

The European Radioecology ALLIANCE

The directors of eight European organisations signed a Memorandum of Understanding, stating their commitment to the long-term integration of radioecology within Europe in 2009. The final aim was to maintain and enhance radioecological competences and experimental infrastructures, and to address scientific and educational challenges in assessing the impact of radioactive substances on humans and the environment. The first step was to create the European Radioecology ALLIANCE [1], which was officially formed as an association in September 2012. The ALLIANCE has expanded from the initial eight founding members to 27 members from 14 countries (Belgium, Croatia, Finland, France, Germany, Greece, Ireland, Kazakhstan, Norway, Poland, Portugal, Spain, Sweden and United Kingdom) in 2017.

The ALLIANCE Strategic Research Agenda (SRA) and roadmaps

The first ALLIANCE strategic research agenda (SRA) provided a long-term vision (20 years) of radioecological research needs and was created under the

STAR (Strategic Network for Integrating Radioecology) Network of Excellence [2] in 2012. The strategic research agenda has been updated several times, taking into account comments from stakeholders and experts and by adding a strategic part focusing on education and training challenges as well as infrastructure-related issues [3, 4].

The SRA highlights the required scientific knowledge as well as methodological and technical know-how for the main components of any environmental risk assessment related to ionising radiation. It identifies three scientific challenges and fifteen associated research lines, consistent with a strategic vision of what radioecology can achieve in the future via a prioritisation of efforts. These challenges are:

- Challenge 1 – To Predict Human and Wildlife Exposure in a Robust Way by Quantifying Key Processes that Influence Radionuclide Transfers and Exposure
  - Approach: Improve human and environmental dose and impact assessments by mechanistic/process-based modelling of environmental transfer and exposure in the biosphere.
  - Outcome: Fit-for-purpose environmental models to support human and wildlife impact assessment and risk management.
- Challenge 2 – To Determine Ecological Consequences under Realistic Exposure Conditions
  - Approach: Unravel causes and mechanisms of radiation-induced effects in wildlife from molecular to individual and population scales.
  - Outcome: Knowledge of the causes of biological effects to detect early damage and to protect wildlife populations.
- Challenge 3 • To Improve Human and Environmental Protection by Integrating Radioecology
  - Approach: Improve risk characterisation by better quantification of uncertainty and variability of exposure and effects.
  - Outcome: An integrated approach to enhanced risk characterisation and communication (connecting science, economy & society).

An initial short-term (5-year) scientific roadmap for radioecology was developed by EC funded project COMET (COordination and iMplementation of a pan-European instrumenT for radioecology) [5]. It was a transitional implementation plan to structure and enhance interactions between the ALLIANCE and two of the other European radiation protection research platforms, namely the European platforms on nuclear and radiological emergency response (NERIS [6]) and low dose risk research (MELODI [7]). Many of the research areas for

radioecology are also relevant to post-emergency management and low-dose effect research.

Under the COMET project, with the help and endorsement of the ALLIANCE, several topical roadmap working groups (WG) have been initiated to make progress in addressing the challenges and priorities described the SRA in a structured way. Each WG has built a 5-year topical roadmap dealing with specific scientific areas to complement the SRA and the transitional roadmap [8]. The selected scientific areas are:

- atmospheric transfer processes;
- marine radioecology;
- human food-chain modelling
- environmental issues associated with naturally-occurring radioactive materials (NORM);
- inter- and intra-species radiation sensitivity and transgenerational effects.

Research proposed by the WGs is intended to interlink the three different challenges presented in the SRA. Several criteria were considered to prioritise the research in the roadmaps: impact, achievability, relevance and public perception, and good science. The topical roadmaps are primary elements to develop the global roadmap for radioecology.

The strategic research agenda, the first transitional roadmap and the topical roadmaps are the essential elements for preparing the radioecological part of the Joint Roadmap for Radiation Protection Research under the European Joint Programming project EJP-CONCERT [1, 2], [9]. The Joint Roadmap will be based on the roadmaps produced by MELODI, NERIS, EURADOS (the European dosimetry group [10]) and EURAMED (The European alliance for medical radiation protection research [11]). The Joint Roadmap for Radiation

[1] The European Joint Programme ('EJP') under Horizon 2020 is a co-fund action designed to support coordinated na-tional research and innovation programmes. The EJP aims at attracting and pooling a critical mass of national resources on objectives and challenges of Horizon 2020 and at achieving significant economies of scales by adding related Horizon 2020 resources to a joint effort.

[2] The 'CONCERT-European Joint Programme for the Integration of Radiation Protection Research' under Horizon 2020 is operating as an umbrella structure for the research initiatives jointly launched by the radiation protection re-search platforms MELODI, ALLIANCE, NERIS, EURADOS and EURAMED (The European alliance for medical radiation protection research). CONCERT is a co-fund action that aims at attracting and pooling national research efforts with European ones in order to make better use of public R&D resources and to tackle common European chal-lenges in radiation protection more effectively by joint research efforts in key areas.

Protection Research is intended as a guide to plan radiation protection research over the next decades. It will promote long-term research to assess the effects of ionising radiation on humans and the environment, and to develop tools to improve practical radiation protection related to different situations resulting in exposure to ionising radiation, with the aim to improve the radiation protection system, to answer priority radiation protection questions and to support decision making. The first steps and current ideas to build a joint roadmap for radiation protection research, a set of exposure scenarios to identify potential radiation protection needs as well as a first set of radiation protection research challenges has been proposed under EJP-CONCERT [12]. The ALLIANCE has produced three annual SRA statements (2015, 2016 and 2017) for EJP-CONCERT to provide and justify research priorities for radioecology in the short- and medium-term. The ALLIANCE will continue to contribute to the Joint Roadmap.

Mechanisms of sustainable cooperation

The activities of the ALLIANCE are organized within working groups, in which any interested member can participate [13]. These working groups provide input to promote ALLIANCE functioning, strengthen integration and sustainability.

The ALLIANCE working groups

The ALLIANCE has formed four working groups to develop and promote the issues related to their topics in the future:

- Strategic Research Agenda & Roadmaps; The working group is now mainly working with EJP-CONCERT project.
- Infrastructures & Sustainability; The focus of the working group to date has been on the future of the Radioecology Exchange website [14] , which is the gateway to access on-line radioecological resources and news items. The infrastructure-related work is at the moment being further developed under the EJP-CONCERT Work Package 6.
- Education & Training; for more information see Bradshaw et. al. 2018 [15]
- Stakeholder Involvement; The stakeholder involvement has been mainly limited to key stakeholders of the wider research community concerned with radiation protection of humans and wildlife and regulatory authorities. As a

partner of the EJP-CONCERT stakeholder working group, the ALLIANCE has made sure that the radiation protection goals will not only target at protecting humans but also at conserving the biodiversity of wildlife and that environmentalists will be part of the future CONCERT stakeholder group.

The Observatory working groups

Radioecological Observatories are contaminated field sites that provide a focus on long-term joint field investigations. The Observatories provide also excellent training and educational sites. Four contaminated sites have been selected under the STAR Network of Excellence and COMET project as the most promising options for Radioecological Observatories [16]:

- the Chernobyl Exclusion Zone
- the aquatic environment of a previous coal mining and processing site in Poland
- a forest in the Fukushima prefecture in Japan
- the waste landfill "Kepkensberg" from the Belgian phosphate industry.

Research at the Radioecological Observatories will primarily focus on radioecological topics outlined within the SRA.

Conclusions

The European Radioecology ALLIANCE has now a solid basis, having expanded from the initial eight founding members to 27 members from 14 countries in 2017. Whilst focussed on Europe, the ALLIANCE is also open to organisations throughout the world with interests in supporting research in radioecology. In the short-term the work to promote radioecology in Europe will continue under the EJP-CONCERT (2015 – 2020) which aims to develop a sustainable structure for promoting and administering joint programming and open research calls in the field of radiation protection research for Europe. In the longer term, radioecological research will be facilitated by the ALLIANCE. Under EJP-CONCERT the ALLIANCE will contribute to the Joint Roadmap based on the roadmaps produced by each of the platforms (i.e. ALLIANCE, MELODI, NERIS, EURADOS, EURAMED). The ALLIANCE contribution to this Joint Roadmap reinforces the crosscutting features of radioecology.

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National Programme and consortium for radiation safety research

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Introduction

While there has been long-standing co-operation between STUK and universities, such ties were further tightened and formalized since the Government decided to introduce a comprehensive reform of the Finnish research and innovation system in 2013. This subsequently led to the setting up of the national Consortium for Radiation Safety Research (Cores) and formulation of national program on radiation safety research in Finland. The Finnish Government Resolution on Comprehensive Reform of State Research Institutes and Research Funding took place in September 2013. The main goal of the reform was to strengthen multidisciplinary, high-level research of social significance. One line of action was to deepen cooperation between research institutes and universities. To achieve this goal, the Resolution envisaged a step-by-step integration process leading to centers of competence (agreement-based consortiums). According to the government policy, such agreement-based consortia must have common research equipment, laboratories and information resources (eg. follow-up material, sample material, statistical and register material) as well as engage in close co-operation in research and education (eg. sharing of mutually complementary competencies, joint professorships and duties, and shared staff). Furthermore, it was envisaged that, within the consortiums, the research institutes and universities form joint campus areas with common functions on a regional basis.

Cores – Consortium for Radiation Safety Research

National strategy

A roadmap for the integration during the period 2013-2017 was prepared. As a first step, a national strategy was formed during 2013-2014, evaluating the impact of the Government Resolution and setting the objectives for the national

programme and consortium. STUK surveyed potential national co-operators, made initial contacts and organized seminars. Stakeholder consultations were carried out by STUK, in particular with the universities and at the ministerial level. Radiation safety research was identified as one of the priority programs in the national research strategy of nuclear sector (Työ- ja elinkeinoministeriö, 2014), along with current research programs for nuclear safety and waste management and the research program for future nuclear energy (fission and fusion). It was envisaged that the national radiation safety program will ensure the national competence and scientific base in radiation safety, secure the needs for knowledge of the national authorities and integrates research with education and innovation. The strategy envisaged the setting-up of a national research consortium along the lines of the Government Resolution. Furthermore, the national program would be linked with the European radiation protection research area (Euratom/Fission).

Establishment of the Consortium

Based on the Government Resolution, a process was initiated to strengthen the co-operation between STUK and universities and to create a national research consortium that would carry out research on various aspects of ionizing and non-ionizing radiation safety. This process has involved an analysis of scientific disciplines required for radiation protection and surveying the profiles of Finnish universities. Existing collaborations were formalized and additional competencies were identified. As part of Euratom OPERRA project (Open Project for European Radiation Research Area), a research seminar was organized in STUK in June 2014, highlighting European research agendas and STUK research activities. Another workshop presenting relevant research activities in the universities and discussing the national program was organized in November 2014. To state the joint intent for forming a long-term Consortium, Letters on Intent were signed between STUK and universities during 2013–2014.

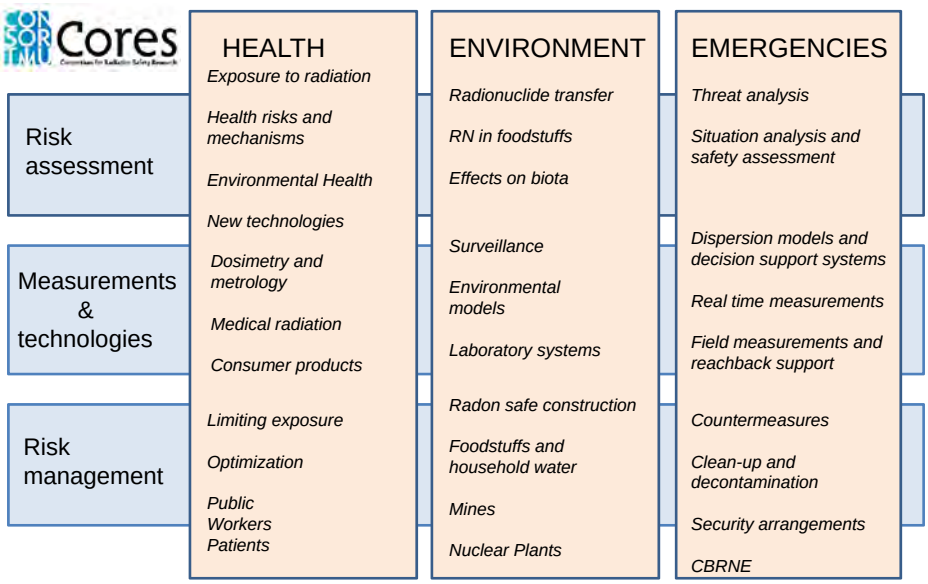
The actual Agreement to set up the Consortium was signed between STUK and nine universities by 2015. In addition of STUK, the following universities signed the Consortium Agreement forming the Finnish Consortium for Radiation Safety Research (Cores), and contributed to the national program: Aalto University, Lappeenranta University of Technology, Tampere University of Technology, University of Helsinki, University of Eastern Finland, University of Jyväskylä, University of Oulu, University of Tampere and University of Turku. The first version of a National Programme for Radiation Safety Research was published in parallel (Salomaa et al, 2015). More detailed plans for the use of infrastructures and information resources were made for

specific areas and bilateral agreements have been prepared between STUK and University of Eastern Finland, between STUK and University of Tampere, and between STUK and University of Jyväskylä. The first meeting of the Board was held 3.12.2015 and the rules for operation of the Consortium were approved. Cores was adopted as acronym for the **Co**nsortium of Radiation Safety **Res**earch.

Implementing the programme

Research areas for the national programme include health (low dose risk as well as medical use of radiation), environment (radioecology) and emergencies (emergency preparedness and response, security of sources), see the figure below illustrating the framework for the radiation safety research program. As cross cutting themes, risk assessment, risk management as well as technological development (metrology and dosimetry) are addressed. Overall, the programme is well aligned with the objectives of European radiation protection research platforms (MELODI, ALLIANCE, NERIS, EURADOS and EURAMED), with additional elements relevant for non-ionizing radiation safety, security research and metrology research. While the European SRAs also include a research agenda for Social Sciences and Humanities (SSH), this area is so far not covered in the national programme.

Framework for Radiation Safety Research Programme



During the preparation of the Euratom Joint Program Co-fund action (CONCERT) in 2014, the Ministry of Social Affairs and Health (Program Owner) mandated STUK to participate as Program Manager and participant in the EJP Consortium. Since then, the role of Finnish universities in radiation safety research has been increasing. This development is in line with the National KOTUMO Roadmap for Co-operation between universities and research institutes (KOTUMO), outlined by the Ministry of Education and Culture. In 2016, the Ministry of Education and Culture mandated University of Eastern Finland as a Programme Manager and University of Eastern Finland joined CONCERT as of June 2016. Furthermore, University of Tampere joined CONCERT as Linked Third Party to STUK.

Stakeholder feedback

After publishing the national programme (Salomaa et al. 2015), a stakeholder consultation on the national program was carried out in 2016. The report was sent to almost 80 stakeholders and statements were received from about half of them. The feedback from stakeholders was analysed by the Cores Board and taken into account in the new version of the program (to be published in 2018).

Overall, the stakeholders considered that the program was well structured and necessary in order to maintain and develop national competence in radiation safety. However, more concrete plans were requested in many statements. Furthermore, the importance of international research was emphasized, to complement the national programme. The first version of the national program was seen as a well justified list of research needs and what each one is doing. The roadmap describing the development of activities was taken positively. The statements also emphasized that arrangements for long-term stable funding were necessary. However, there were no practical solution to point out possible funding sources. Many stakeholders also noted that the programme is focused on collaboration between STUK and universities only and the research carried out by other institutes should also be mentioned. A major issue was raised by stakeholders: who is taking the general responsibility for the maintenance of knowledge and competence in Finland? This question is of cause very much linked to the funding of research.

Future plans

The stakeholder feedback as well as emerging research needs are analysed. A new strategy and a roadmap for years 2018–2022 is set up and the national programme is updated accordingly. This involves engaging new partners, universities as well as research institutes, and organizing activities fostering the co-operation, such as joint symposia and working groups. Dissemination of

Cores aims and achievements is supported by newletters and the Cores website. Fostering the education and training and promoting the joint use of infrastructures and databases (open science) are important lines of action. Cores also promotes national and international funding for radiation sciences and links with the European radiation protection research platforms. In the longer run, the objective is to actively participate in international collaboration, in particular the 9th framework programme and European research activities. A long-term funding plan is among the key objectives for the next strategy period.

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MetroNORM - Metrology for processing materials with high natural radioactivity

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1 Introduction

In the case of industrial activities, such as oil and gas production, mining activities, rare earth extraction, water treatment and phosphate fertilizer industry, Naturally Occurring Radioactive Materials (NORMs) present in raw materials may result in enhanced radiation exposure especially for workers but also for public in the surrounding environment. In addition to raw materials, NORMs may be present in the products, by-products and waste causing economic and ecological burden if not appropriately disposed or re-used. Typical NORM radionuclides considered here are, for example, isotopes of U and Th and their progeny.

In general, radionuclide metrology is focused on artificial radionuclides, and NORMs are often considered as normal background in the measurements regardless of their concentration. To ensure safety and protection against NORM, the regulation of NORM practices and NORM waste management is necessary. In addition, up–to–date measurement methods are of importance for controlling the radiation exposure to the workers and public.

2 MetroNORM project

New traceable, accurate and standardized measurement capabilities for NORM industries were developed in the European metrological joint research project known as MetroNORM in order to improve the industrial processing of NORM resources and waste (Maringer et al., 2017; http://www.metronorm-emrp.eu). The developed capabilities included novel NORM measurement systems/procedures to be used in a laboratory or on-site as well as production of reference and calibration materials for validation of the measurements. Development of *in-situ* methods were of particular consideration in MetroNORM.

Scientific and technical objectives of the MetroNORM project are following (Maringer et al., 2017):

- Elaboration of measurement methods and techniques including in-situ systems.

- Development of traceable procedures for measuring industrial NORM raw materials, products, by-products, residues and waste.
- Establishment of traceable standard sources and reference materials needed for detector calibration.
- Decay data improvement for selected natural radionuclides of the ^{238}U and ^{235}U decay series and ^{138}La.
- Testing the developed methods, standards and reference materials.

The MetroNORM consortium consists of twelve National Metrology Institutes or Designated Laboratories and five other partners. Duration of the project was three years.

3 Measurement methods, techniques and procedures developed in the MetroNORM project

During the project novel hand-held prototype instruments have been developed for *in-situ* alpha-particle (Pöllänen et al., 2014, see text later) and gamma-ray spectrometry. A hybrid Si pixel detector (MEDIPIX/TIMEPIX) were designed, developed and tested for *in-situ* gamma-ray measurements. In addition, a production chain for ^{220}Rn in a vacuum chamber has been designed and tested. Measurement technique for the on-line activity determination of the ^{220}Rn activity was developed and tested. Elaboration of the NORM measurement procedures were developed (and are under evaluation) according to the questionnaire sent to a set of stakeholders. A number of on-site tests were performed to verify the developed methods, instruments and procedures.

4 Development of traceable reference materials and standard sources and improved decay data

During the project, ten NORM sources and standard reference materials in the activity range of 0.1 – 100 kBq/kg were prepared for calibrating alpha and gamma-ray spectrometers. The sources contained the nuclides ^{238}U, ^{235}U, ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th, ^{208}Tl, ^{228}Ac, ^{214}Bi, ^{214}Pb, and ^{40}K (Maringer et al., 2016). Interlaboratory comparisons were carried out successfully by using the developed sources. Special sources containing the radionuclides ^{235}U, ^{227}Ac, ^{226}Ra, ^{210}Pb and ^{138}La were prepared to improve the decay data.

5 Impact of the MetroNORM project and dissemination of the results

Results of the MetroNORM project will be (have been) published in open scientific literature. During the project the partners participated in 40 scientific conferences, and 18 peer-reviewed publications have been done so far

(Maringer et al., 2017). An open-access e-learning course (http://www.npl.co.uk/commercial-services/products-and-services/training/e-learning/naturally-occurring-radioactive-materials/) was established during the project for training the end-users, regulators and the public. In addition, NORM course material for post-graduate university students was developed.

6 Development of an in-situ alpha spectrometer at STUK

Alpha particle spectrometry has been applied for different type of samples but the chain of sampling, sample processing and spectrum analysis is usually time consuming and requires appropriate laboratory infrastructure and special expertise. However, in-situ alpha spectrometry with good energy resolution at ambient air pressure is feasible by using sophisticated sampling procedures and techniques in the measurements (Pöllänen et al. 2012).

During the MetroNORM project a novel hand-held equipment known as ADONIS was developed. It allows to measure thin samples/objects, such as air filters, swipes or smooth surfaces, at ambient air pressure with good energy resolution. This was carried out by using a collimation technique (Fig. 1). Since collimation causes reduction of the detection efficiency approximately by a factor of 10 the equipment can also be used without the collimator for fast detection of the alpha particles.

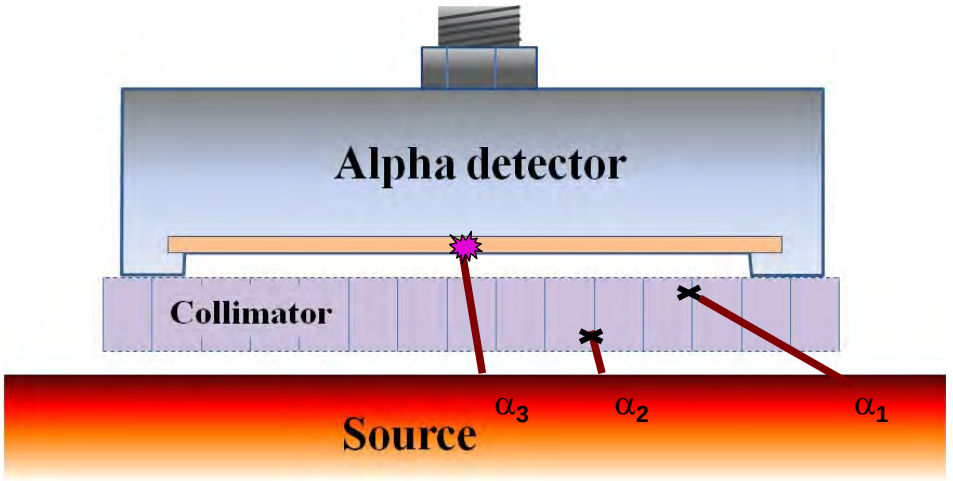

Fig. 1. A honeycomb collimator between the source and the detector reject alpha particles entering the detector on the slant (such as alphas α1 and α2) but allows to detect those alphas (such as α3) arriving the detector perpendicularly. This means that the path length of alpha particles entering the detector is almost constant and, thus, the energy loss in air is also almost constant. By this way it is possible to obtain an alpha spectrum with good energy resolution.

The main components of ADONIS consists of the detector head, the multichannel analyzer and a laptop computer for the data acquisition and real-time data transfer. All the instruments were mounted in a backpack (Fig. 2).

Using the collimator facilitates radionuclide identification in the case of thin sources (Fig. 3). The collimator removes alpha particles that do not carry relevant information for the radionuclide identification. With collimation, the values of the full width at half maximum were between 130–170 keV depending on the emission energy of the alpha particles. Although thin sources are optimal for the alpha-particle detection, the equipment can also be used for thick homogeneous sources. However, in that case the activity determination is not possible. In general, appropriate spectrum analysis tools are necessary for analyzing the results.

Determination of the detection efficiency is needed to estimate the relation between the detected count rate and the activity per unit area. Following numbers were obtained with and without collimation:

- No collimation: 1 Bq cm^{-2} refers to 4.2 cps
- No collimation: 1 cps refers to 0.23 Bq cm^{-2}
- Collimation: 1 Bq cm^{-2} refers to 0.47 cps
- Collimation: 1 cps refers to 2.1 Bq cm^{-2}.

Since beta particles may deteriorate the spectrum unfolding at small energies (Fig. 3), only channel numbers >150 were considered for determining the sum activities per unit area of the long-lived alpha-particle emitting radionuclides. The user-interface of ADONIS reports cps-values for different regions of inter-

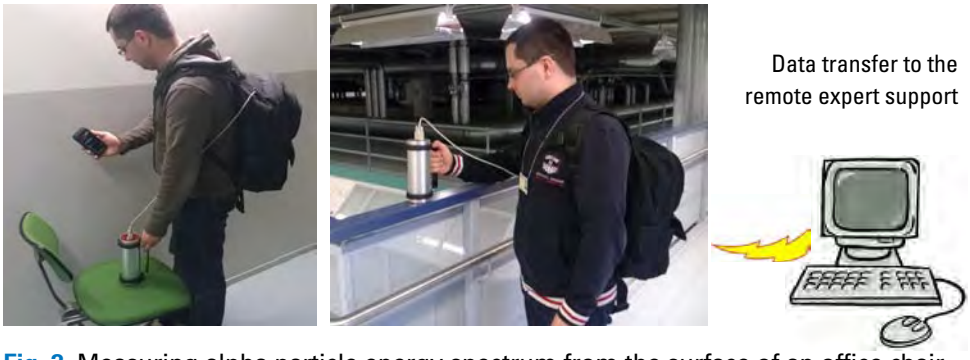

Fig. 2. Measuring alpha particle energy spectrum from the surface of an office chair (left) and from a balustrade (right). The measurement data (as well as other data) can be followed on-site by using a mobile phone (left). The data can also be sent in real-time to the reachback center (right).

est. Respective activities can be easily obtained on-site or in the remote expert support. Nuclide-specific activities can be obtained by using the ADAM program (Ihantola et al., 2011).

As a summary, the ADONIS equipment is viable and during the MetroNORM project thorough tests were done to verify its performance. ADONIS can send the measurement data in real time to the remote expert support. This allows performing the in-field measurements even by a personnel, which has no deep understanding on radiation measurements issues.

Acknowledgement

The MetroNORM research project EMRP/IND57 were performed in the frame of the European Metrology Research Programme EMRP, and is jointly funded by the EMRP participating countries within EURAMET and the European Union.

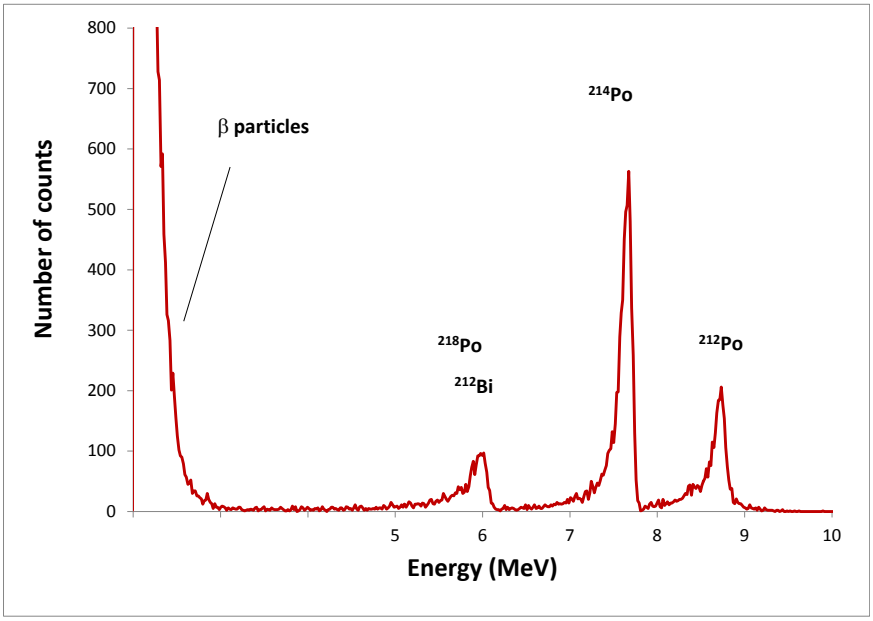

Fig. 3. Alpha spectrum measured from a membrane air filter. The sample was from indoor air containing radon progeny (^{218}Po, ^{212}Bi, ^{214}Po and ^{212}Po). In addition to alpha particles, the ADONIS equipment can also detect beta particles, which may be advantageous in some cases. In the spectrum, the beta particles are located at channels <150 referring to the alpha particle nominal emission energy of 3.9 MeV (Pöllänen et al., 2014). Total number of channels in the spectrum was 102 4.

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Session 2: Radioactive substances in geosphere

Chair: Dr. Nina Huittinen

Radionuclides in porous bedrock-deposition of spent nuclear fuel

Marja Siitari-Kauppi
University of Helsinki

Deep geological repositories are being considered for long-term disposal of spent nuclear fuel in multiple countries. We investigate the transport properties of safety-relevant radionuclides in the geosphere and engineered barriers to provide parameters for the safety analysis of the repository. Our research topics include e.g. sorption, diffusion and structure characterization in laboratory and in situ experiments.

Safe disposal of spent nuclear fuel requires information about the radionuclide transport and retention properties within the porous and water-containing rock matrix and water flowing fractures. Objective of this study is to support the implementation of geological disposal of spent nuclear fuel by improving knowledge base for the safety case. To this end, the RIP team have been involved in several long-term in situ experiments in Olkiluoto, Finland and Grimsel, Switzerland. In addition, it has been conducted supporting laboratory studies and interpretation of the results through modelling.

One main focus has been the work in ONKALO, the underground rock characterization facility in Olkiluoto, as part of the project ”rock matrix REtention PROperties” (REPRO). The research site is located at a depth of 420 meters close to the repository site and the aim is to study the diffusion and sorption properties of radionuclides in the rock matrix in real in situ conditions (Fig. 1).

The participation in the **Grimsel** *in situ* project which has been funded by the Ministry of Economic Affairs and Employment under the KYT cluster, has lasted more than 10 years. The first in situ diffusion experiment in Grimsel (Monopole 1), started in 2007 and was stopped in 2013. The experiment was overcored and the diffusion profiles of the radionuclides were obtained. The sec-

ond in situ diffusion experiment (Monopole 2) started in 2013 and was stopped in 2017.

When assessing the safety of nuclear waste management, sorption onto mineral surfaces and diffusion into the pore network of rock are the most significant processes that retard the transport of radionuclides from the repository in the water conducting fractures of the rock. Sorption of safety relevant radionuclides has been investigated from molecular scale up to in situ scale. Distribution coefficients are commonly determined using batch sorption experiments and experimental results are interpreted with hydrogeochemical modelling tools to determine the sorption mechanisms. In addition, sorption is studied in intact rocks with laboratory diffusion methods in drillcore samples and larger rock blocks to combine sorption with diffusion.

Investigations related to the material properties of the different geological formations that serve as host rocks for nuclear waste repositories call for thorough evaluation of the transport and retardation properties of geomaterials. The relevant rock properties to be quantified are the accessible internal pore volume that determines the diffusive transport/retardation of non-sorbing and slightly sorbing radionuclides and the accessible internal surface area that controls sorption/fixation of radionuclides and chemical interactions. These properties are linked to the spatial porosity distribution of the geomaterials, which have been studied over the last decades with the C-14-PMMA autoradiography technique.

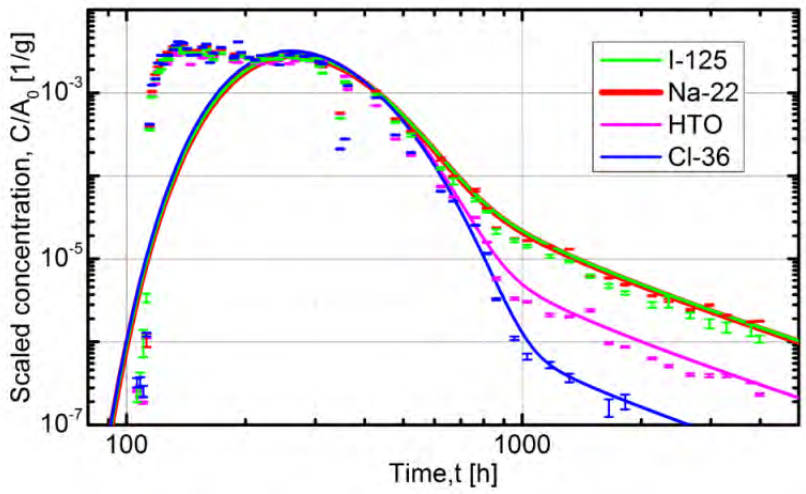

Fig. 1. Late time behavior of the breakthrough curves of radioactive tracers show power-law behavior by the matrix diffusion and sorption in rock matrix.

The C-14-PMMA technique involves the impregnation of centimeter-scale samples with C-14-labeled methylmethacrylate, a low molecular weight and low-viscosity monomer that mimics water in its behaviour in the pore space. The labeled MMA is then polymerized with gamma radiation or heating, which results in a solid radiolabeled polymer, PMMA, within the pore network. The impregnated rock samples are analyzed with autoradiographic methods and digital image analysis of autoradiographs provides spatial porosity distribution of the geomaterials.

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Mobilization of radionuclides and trace metals in tailings from Rautuvaara mine

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The old Rautuvaara mining area is located in North-West Finland, in the municipality of Kolari. Rautuvaara mine was operational between the years of 1962 to 1988 and the tailings field, located in the Niesajoki valley, was used to dispose of mine tailings from several different mines until 1995. Currently there are no mining activities at the Rautuvaara mining area, but it has become an object of interest, since an on-going mining prospect in Hannukainen is planning on transporting their produced tailings to Rautuvaara and depositing them above the existing ones. Sulphide-bearing ores and mine waste are known to form acid mine drainage (AMD) when sulphuric acid forms in the oxidative dissolution of sulphide minerals. The decreased pH in the area affected by AMD can further dissolve and mobilize harmful elements from the surroundings minerals, which may contaminate the natural water systems.

The aim of this study was to evaluate the occurrence and possible mobilization of the base metals Ni, Zn, Cu, Co, Fe, and Mn, potentially toxic metals As, Pb, Cr, and Cd, as well as the radioactive elements 238U and 232Th, in samples collected from the Rautuvaara mining area. The collected samples included water samples from the mining area and its surrounding and four types of solid samples: enrichment sand samples from the tailings, sediment samples from the old settling pond, waste rock samples from the old mining area and acidic pond sand samples from the area of the tailings affected by AMD. A six-step sequential extraction procedure, with progressively increasing leaching reagents, was applied for all the solid samples in order to access the geochemical association of metals and radionuclides in the solid samples. The studied fractions were the exchangeable fraction I (pH 7,8), exchangeable fraction II (pH 5,0), mild acid-soluble fraction (pH 4,1), reducible fraction (pH 2,0), oxidizable fraction (pH 1,5) and strong acid-soluble fraction (pH 0). The metal concentrations in the leaching supernatants and water samples from the mining area were determined by Inductively Coupled Plasma Mass Spectrometry. Furthermore,

the behaviour of the mineral composition in the samples during extraction was studied by using Synchrotron Radiation X-ray Powder Diffraction (SR-XRPD) and by applying the Rietveld method for the acquired data.

Water analysis showed the metal concentrations in the settling pond and downstream river samples are comparable to the typical levels of the area, aside from slightly elevated base metal and uranium concentrations. The highest metal and radionuclide concentrations were seen in the acidic pond waters. In the sequential extractions the total amounts of leached metals were in order of: enrichment sand, sediment, acidic pond sand and waste rock. The overall trend is that the highest amounts of metals are leached out in the oxidizable and strong acid-soluble fractions, indicating that the elements are tightly bound to the sample matrix. The dissolution of thorium and uranium was different: thorium mainly dissolved in the strong acid-soluble fraction, whereas uranium started dissolving in the mild acid-soluble fraction as the pH of the solution decreased. The SR-XRPD data showed that the samples mainly consisted of Albite, Dolomite and Quartz. A complete dissolution of dolomite, bassanite and calcite was seen, as well as a partial attack on pyrrhotite and pyrite, during the extractions. Also gypsum formation happened in the mild acid-soluble leaching step as the pH decreased during the extractions. The results can be used to evaluate the possible hazards the occurring metals and radionuclides pose to the surrounding environment and they also give a reference point of contamination for the future, if the use of the old tailings area continues.

Progress and implications: 20 years of increased system understanding

Ryk Kłos
Aleksandria Sciences Limited

Introduction

In 2001 STUK published a list of release constraint limits (RCLs) for use in the radiological assessment of potential releases of radionuclides to the biosphere from waste disposal facilities on timescale beyond a few millennia (STUK, 2001). The RCLs are used to determine acceptability of calculated radionuclide fluxes from the geosphere to the biosphere in terms of the risk posed to human populations at the time of the release.

The RCLs are based on dose calculations that work backward from the STUK (2001; 2013) dose constraint limits. To determine the RCL requires a dose conversion factor (DCF) calculated assuming constant unit release of each radionuclide across the geosphere–biosphere interface to selected biosphere objects from which exposure can reasonably be expected to arise.

The biosphere objects identified for the 2001 RCLs were based on the best understanding of Nordic biospheres available at the time. In turn, the models for dose assessment used the best available techniques for dose assessment; the BIOMASS Reference Biosphere Methodology (IAEA, 2003).

In 2016 STUK commissioned a review of the RCLs based on updated site descriptive modelling. This incorporated enhanced system understanding that had become possible as a result of the POSIVA site characterisation programme around Olkiluoto, where a deep geological repository for spent nuclear fuel is presently under construction (POSIVA, 2012). The results (Kłos, 2017) show some notable differences compared to the STUK (2001) values.

This paper looks at what has changed, how this impacts the assessment of potential radiological risk and indicates what influence the increased depth of knowledge might have for future assessments and the potential regulatory response.

Site description and modelling

Dose assessment: regulatory background

STUK (2013) provides the background for the *dose assessment* of waste facilities. The requirement is that for *the radiation impacts arising as a consequence of expected evolution:*

a. the annual dose to the most exposed individuals remains below the value of 0.1 mSv; and

b. the average annual doses to other individuals remain insignificantly low.

Strictly, this constraint applies to an assessment period from closure of the repository over a few millennia, when the radiation exposure of humans can be assessed with sufficient reliability, taking into account changes in the living environment that arise from changes in ground and sea level.

The RCLs are derived release values, corresponding to the dose constraint, that are to be used over longer timescales. In practice they are numerical values for release fluxes across the geosphere–biosphere interface determined by STUK using simple reference-biosphere level dose models.

The key is to fit patterns of human behaviour into the potential for exposure expressed by the biosphere system around the potential geosphere release locations. Human behaviour, in terms of exposure, means ingestion, inhalation and external irradiation. Dose models – for a few millennia or beyond – need to be placed in the context of what its understood and anticipated about the evolving landscape and potential radionuclide accumulations that can arise following inputs from the geosphere.

Turn of the century interpretation

The models on which the RCL calculations were based are typical of the *reference biospheres approach*. POSIVA (Karlsson & Bergström, 2000) used models developed for SKB (Bergström *et al.*, 1999). Figure 1 illustrates sub-models for irrigation and lakes. There were modules for different dose object: coastal waters, lakes, running waters, groundwater wells, agricultural land and peat bogs. Description of landscape features was in the form of a qualitative description, building on earlier Nordic biosphere modelling.

Given the nature of Fennoscandian biospheres the objects included a number of features not found elsewhere. Where the BIOMASS example reference biospheres included well and groundwater discharge scenarios, the range in the SKB/POSIVA models reflects the coastal nature of the sites and the fact of landrise due to crustal rebound following the loss of the Weichselian ice sheet

around 10 kyear before present. While agricultural land and wells are common in dose assessments that support early-stage siting assessments the state of site understanding at the turn of the century recognised that system change was an important feature. Landrise would see coastal water turn to lakes, which would, in turn, terrestrialise as they infilled with decaying vegetation, forming peat bogs. Only minor differences between the Swedish and Finnish implementations were necessary in respect of the national regulatory frameworks in the respective countries.

2016 implementation

By the TURVA-2012 assessment, POSIVA had developed a detailed site investigation programme that allowed the evolution of the landscape around Olkiluoto island to be described quantitatively. Of particular utility are the terrain and ecosystem development models and the hydrological interpretation. POSIVA (2013) used this material in the biosphere safety assessment (BSA-2012) to assess doses over the period from 2020 CE to 12020 CE, in accordance with the regulatory requirement.

Additionally, a database of radionuclides specific properties related to the types of media in the sedimentary overburden had been produced. Rather than the individual sub-models in the 2000 interpretation, a standardised structure of bedrock-deep overburden, mid- and upper overburden was possible, allowing for the thick-ness of the different layers to be included in the models.

Table 1 compares the interpretation of landscape objects in the Kłos (2017) and for the STUK (2001) RCL calculations. Where the state of site understanding at 2001 identified five distinct objects types, the improved site descriptive database in POSIVA (2014) recognised more distinct types, broadly corresponding to the original set, albeit with more refinement. Statistical anal-

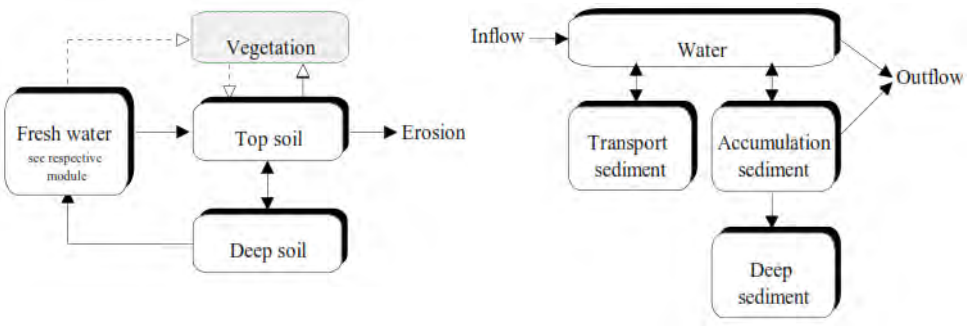

Figure 1. Typical models from the 2000 interpretation: lake and freshwater irrigation, taken from Bergström et al. (1999).

Table 1. Comparison of release scenarios between the 2016 and 2001 determinations of RCL values. The scenarios are grouped to show corresponding situations. The 2001 derivation used calculated doses at the end of a 10 kyear period. The 2016 review included explicit persistence of the release objects in the landscape. Scenarios for which doses were evaluated in the Karlsson & Bergström (2000) modelling but which were not used in the determination of the RCLs are shaded.

2016 RCL calculations (Kłos, 2017)			**2001 RCL calculations (STUK, 2001) – Doses at 10 kyear**	
Release scenario	**variant**	**persistence kyear**	**Release scenario**	**Status at 2000**
Accumulation / Exposure	Wetland → Cultivation	10	Agricultural land	discounted
			Peat Bog	discounted – high doses
Wells	Bedrock wells	~ 5×10^{-2}	Garden plot (well)	included
	Overburden Wells	10		
Lake	Small, forest	3	Lake	included
	Medium, forest	8		
	Large, forest	10		
	Small, agriculture	3		
	Medium, agriculture	8		
	Large, agriculture	10		
Rivers	Tributary	10	Running waters	discounted
	Local	10		
	Regional	10		
Coast	Bay	8	Coast	included – low doses
	Open sea	2		

ysis of the landscape quantitatively describes different sizes of lakes and rivers and their sub-catchments.

Together with the terrain and ecosystem development modelling the *persistence* of the object can be addressed. This is linked to the accumulation time that is appropriate for the dose calculation for each object type. Small objects retain their initial character for a short time, larger objects for longer. The largest lakes are deep enough to persist for several thousand years and, on the timescale for the derivation of RCLs – up to 10 kyear – they are effectively permanent. The retreat of the coastline with land uplift means that there are no open sea areas around the island after around 2 kyear, some bays can exist for up to around 8 kyear.

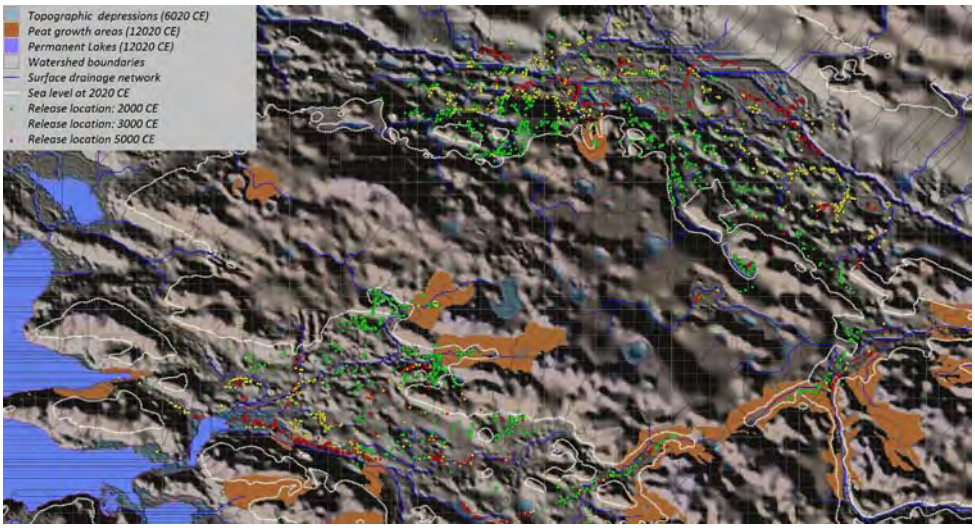

Figure 2. Details from the POSIVA (2016) digital terrain model shown as a relief map of the anticipated landscape around Olkiluoto island at 12020 CE. The landscape at this time shows peat areas, terrestrialised lakes, and the location of the main drainage features. To the west are large persistent lakes. Release locations correspond to the TURVA-2012 estimates for the deep repository are at topographic low-points. The release locations are used to inform understanding and are not taken to be definitive.

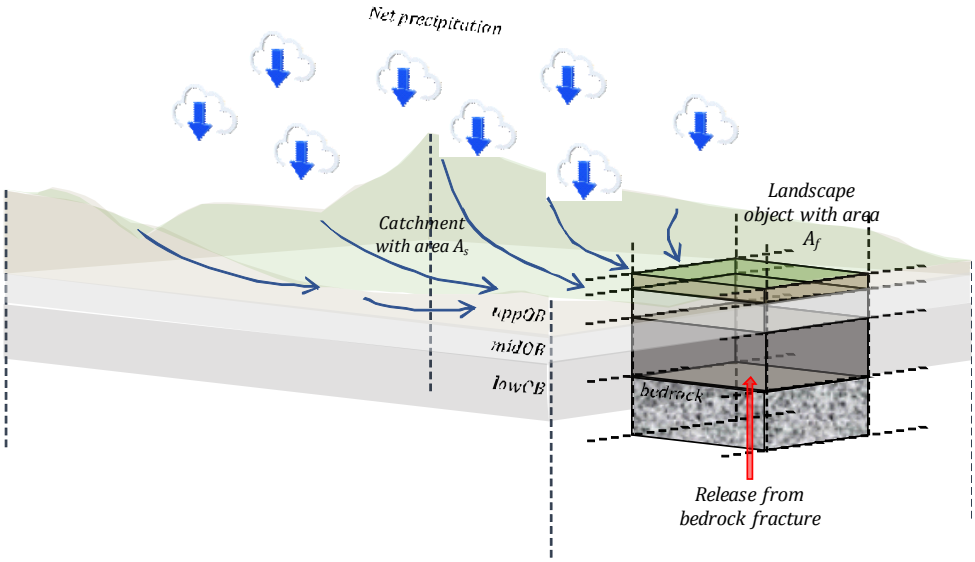

Figure 3. Landscape dose object in the landscape context. Water fluxes in the overburden are determined by the net precipitation, topography of the catchment and the properties of the overburden material. Water fluxes from the bedrock are small in comparison. The compartment models use similar concepts to the Bergström *et al.* models illustrated in Figure 1.

Three object types were discounted in the original RCL derivation: *running waters, agricultural land* and *peat bogs*. It was known that peat bog objects were possible but the site descriptions at the time did not allow for sufficiently robust models to be defined and the assumptions necessary to allow calculations implied overly cautious and unrealistic doses. The corresponding objects in the 2016 implementation are identified as *accumulation / exposure* objects (Kłos & Wörman, 2013). In these objects there is a period of accumulation (up to 10 kyear) in their natural state (wetland with limited drainage) at the end of which measures are taken (installation of drainage) to allow cultivation, as described by Biebighauser (2007).

This leads to a cautious radiological assessment assumption, whereby activity is conserved and doses are evaluated over the subsequent 50 year period, with the average value of dose over this time used, in accordance with the guidance in STUK (2013). These objects are foreseen to be amongst the most radiologically sensitive in the future Olkiluoto landscape. In contrast with the earlier models they are much better motivated in the landscape context.

An axiom of the simple–and–robust reference biosphere modelling approach has always been that wells provide the highest dose. The key feature of wells is dilution. Two types of well are considered in the 2016 RCL derivation: bedrock and overburden. In the former, it is conservatively assumed that the deep well is located in a fracture in the bedrock that can supply the needs of the community. Abstracted water has radionuclide concentrations determined by the release rate from the geosphere and the dilution in the well. The *persistence* of well objects is acknowledge to be short, typically not more than 50 years (Bergström *et al.* 2008). As there is no sorption in the bedrock fracture, therefore, doses are calculated for the irrigation water in the first year of the constant release to the geosphere.

On the other hand, as dilution in the overburden well is determined by the collecting power of the landscape's topography, there can be sorption and accumulation in the deep overburden. Doses are calculated at the end of the 10 kyear accumulation period. Dilution in other landscape objects is important. Doses from marine and bay objects are low in both the 2001 and 2016 models. Similarly the larger the lake, the greater the dilution and it is small lakes that give higher doses from aquatic exposure pathways. These are exceeded by doses from accumulation–exposure objects, where lakes implicitly form wetlands before drainage and cultivation.

Overall then, bedrock and overburden wells and the accumulation/exposure objects dominate the doses used to calculate RCLs in the 2016 modelling. This is broadly consistent with the results from the original models, where wells and lakes featured. In the new RCL calculations wells are also important

Table 2: Recommended geo-bio constraints (RCLs) based on the 2016 probabilistic modelling of key landscape features of the future Olkiluoto landscape. Arithmetic mean of the calculated DCFs (1000 samples, LHS) are used to generate the RCL values (taken from Kłos, 2017). Results ordered according to the ratio of 2016 to 2001 values.

nuclide	RCL (2016) GBq a^{-1}	RCL(2016) / RCL(2001)	nuclide	RCL (2016) GBq a^{-1}	RCL(2016) / RCL(2001)
Se-79	0.3	300%	U-238	0.01	10%
C-14	0.3	100%	Pu-239	0.001	3%
Cs-135	0.1	33%	Pa-231	0.001	3%
Cl-36	0.1	33%	Th-229	0.001	3%
Tc-99	1	33%	Ra-226	0.001	3%
Sm-151	10	10%	Pd-107	3	3%
Ni-59	3	10%	I-129	0.003	3%
Zr-93	1	10%	Sn-126	0.003	0.30%
Cm-245	0.003	10%	Nb-94	0.003	0.30%
Am-243	0.003	10%	Am-241	0.01	–
Np-237	0.003	10%	U-235	0.01	–

and the cultivated land objects are better justified by the landscape context provided by the POSIVA (2016) dataset. Most significantly, the peat bog objects of the Karlsson and Bergström (2000) modelling are underpinned by a more sophisticated interpretation of the conditions whereby accumulations might arise provided by the POSIVA material.

The POSIVA (2014) biosphere data description concentrates on radionuclides relevant to the assessment over the few millennia period specified by the regulatory requirement. Olkiluoto specific data for radionuclide distribution coefficients in the overburden media and for the concentration ratios used to model uptake in biota. Data in 2001 were taken from a variety of sources. The most recent region-specific database for a range of radionuclides concerned was therefore taken from the SKB SR-PSU assessment (Tröjbom *et al.*, 2013).

Results – RCLs 2016

Having calculated the dose per unit release (1 Bq $year^{-1}$) for each object type (DCF – dose conversion factor, Sv Bq 1), taking media properties and object size into account the RCL is calculated, for a dose limit of D_{lim} = 0.1 mSv $year^{-1}$, as

$$RCL = \frac{D_{lim}}{1000}.\frac{10^{-9}}{\max(DCF)} \text{ GBq year}^{-1}.$$

The maximum DCF from the ensemble of objects is used and, the RCLs are rounded to either 1×10^{n} or 3×10^{n}. The recommended values from Kłos (2017) are given in Table 2, with the ratio of the new values to the original indicated as a percentage. In all but two cases the recalculated RCL is more restrictive. In part this reflects the increased site understanding and the way in which the objects that were ruled out in the original calculations have now been included. Of the release scenarios in Table 1, the objects that dominate are bedrock wells, overburden wells and the accumulation exposure cases.

The results in Table 2 use the arithmetic mean of the calculated doses. These are somewhat cautious since they are strongly influenced by the smaller objects in the distribution of landscape sizes. In practical terms the results emphasise the role of smaller objects with lower capture of net precipitation and with lower in dilution.

Conclusions

Finland and Sweden lead the world with their advanced programmes for geologic disposal. The detailed site investigation programmes, coupled with global information system techniques allow details of the site to be processed and understood in unprecedented detail. As the capabilities of the disposal programmes evolve this feeds into the model description. In practical terms the mathematical representation of features, events and processes (FEPs) remains closely similar to the methods employed at the turn of the century. What has changed is the detail and confidence with which the FEPs can be expressed mathematically.

The implications of the site descriptive material are that the restriction of detailed evolving models of the biosphere to a few millennia may be overly cautious. While RCLs might be used for very long timescales, say beyond 100 kyear (e.g. STUK, 2013), it is possible to construct well-justified representative models of radiologically significant landscape objects up to this time, if only for illustrative purposes. This would mean that detailed datasets for a broader range of radionuclides would be needed for the specific sites under consideration.

How to use the RCL concept remains an open question. The 2016 derivation has included a lot of site-specific material so that the RCLs are then site-specific and inherently subjective, rather than being more generically applicable. The approach developed in this project is to be used to update the reference biospheres methodology as part of the IAEA's MODARIA II project for BIOMASS 2020.

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Session 3: Transfer of radionuclides in the environment

Chair: Prof. Sisko Salomaa

Sensitivity analysis of radionuclide transport in biosphere

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Introduction

Uncertainty is a common denominator in biosphere analysis, especially when the, usually non-deterministic, computational models are based on measured values. Measurements are always related to measurement accuracy and the accuracy may vary significantly, i.e., orders of magnitude depending on whether it is a historical measurement or is based on the latest modern laboratory technology.

The aim of a biosphere model is to mimic real-world environment as realistically as possible. However, technology limitations (computer processing power and memory consumption) are still defining the size and realism of biosphere models. The essential questions in biosphere modelling are: what aspects should be included and what should be left out? and what aspects can be simplified?

The safest way to simplify a model is to compare the simplified model to the original one changing one factor at a time (OAT). For example, in (Pohjola et al. 2016a) the effect of grouping dietary features into bigger sets on the dose conversion factors of various radionuclides is examined. It was found that grouping and re-arranging food items could simplify the model significantly while the total dose conversion factor estimate remained within an order of magnitude. Also, in (Pohjola et al. 2016b) it was found that lake volume was not significant compared to lake outflow rate in a model where household water was taken from a contaminated lake.

One approach to assess uncertainties is to use two different types of modelling methods, as in (Erichsen et al. 2013). They model a coastal ecosystem using compartment based model as well as dynamic driven calibrated hydrodynamic model. Although the models are different, the results are quite similar.

In most cases, uncertainties in parameter values are dealt by means of probability distributions. Probability distribution gives the range of the parameter value and clues for the most probable value of the parameter. When parameters are presented as probability distributions, a well-known method called Monte Carlo simulation, can be used to pick random values from distributions and feed them into the non-deterministic model. In one of the earliest papers on ecosystem modelling (Tiwari and Hobbie 1976), simple aquatic ecosystem is modelled using random differential equations and the parameters are randomly picked from Gaussian distributions of predefined means and standard deviations. By repeating this procedure hundreds or thousands of times, the distribution of the model output can be obtained.

An essential question is: what parameters affect most the modelling results? Sensitivity analysis is an important tool dealing with uncertainties in modelling. Sensitivity analysis can give insights for understanding the dependence between input and output parameters and checking errors of the model. Sensitivity analysis is useful for parameter reduction and model simplification. (Saltelli et al. 2006) is a good review of the process and methods of sensitivity analysis.

In this study we present farm scenario where the radionuclide transportation to humans is analyzed using Ecolego simulation tool. The model parameters are analyzed using five different sensitivity analysis methods.

Data and methods

Sensitivity analysis presented in this study is based on a self-sustained Finnish farm (Pohjola et al. 2016b), with cattle breed and cultivation. The household water of the farm is taken from the nearby lake. As a lake model we use Lake Liponjärvi, which will be formed near nuclear waste repository at Olkiluoto as the result of land uplift. Contamination from a leaking waste canister is assumed to be carried by the groundwater to the surface and released to the lake as a constant unit flux of 1 Bq/year. It is known that under Lake Liponjärvi there are two major lineaments that can carry radionuclides to the lake. On the other hand, the bottom sediments, till and clay, will form effective barrier to the pathway. In this case the bottom sediments will act as an inhibitor layer and the transport trough sediments is not taken into account.

Radionuclide transport model and parameters

In Figure 1 the radionuclide transport model is presented in the form of a flow-chart. The model has originally been presented in (Pohjola et al. 2016b) and since then several model parameters have been updated with the introduction of uncertainties. Sensitivity analysis of the model was done for 34 parameters in total. The parameters can be further divided into the following groups: K_d-values (solid-liquid distribution coefficients) for soil and lake sediment, 14 food intake parameters, 4 parameters concerning lake properties, 7 root uptake parameters and 7 parameters related to translocation of intake for animal products.

Five most common variance-based sensitivity analysis methods, FAST, EFAST, RBD, EASI and SOBOL, which are used in this paper are presented below.

FAST and EFAST

Fourier Amplitude Sensitivity Test (FAST) is a variance based sensitivity analysis method by (Cukier et al. 1973; Schaibly and Schuler 1973). Outputs of the model are converted into coefficients using multiple Fourier Transforms that represent conditional variance. After that the multidimensional Fourier coefficient integral is converted into one-dimensional integral using the theory developed by (Weyl 1938). The frequencies are usually irregular non-integer frequencies and to reduce computation, a set of integer (multiples of) frequencies are selected. Integer frequencies are selected in a way that enables the error to be controlled. By using integers, the resulting function of one-dimen-

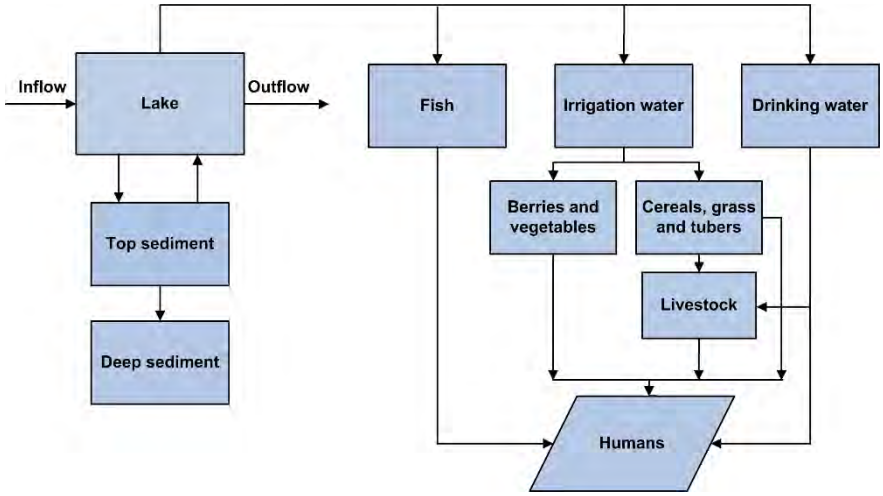

Figure 1. Flowchart of the radionuclide transport paths in the model.

sional integral is periodic and only a single period is needed for evaluation. The continuous integral function can be sampled using Nyquist-Shannon sampling theorem and the resulting sampled function is evaluated from the summation of function values. FAST method is so-called "first-order sensitivity index" method.

Extended Fourier Amplitude Sensitivity Test (FAST) is a modification of FAST proposed by (Saltelli et al. 1999). The EFAST method estimates total effect indices by computing the variance of a complementary set to that introduced in FAST. Also, more flexible sampling scheme is introduced when compared to the FAST algorithm. EFAST algorithm can produce both "first-order" and "total-order" sensitivity indices.

RBD

Random Balance Design (RBD) was first proposed for regression problems by (Satterthwaite 1959). (Tarantola et al. 2006) modified the RBD method by combining it with the FAST method. While FAST algorithm uses integer multiples of frequencies, RBD uses random permutations of the selected frequencies to generate scrambled set that covers the input space. RBD is a "first-order" sensitivity method.

EASI

Effective Algorithm for Sensitivity Indices (EASI) was developed by Plischke (2010). EASI algorithm is basically similar to FAST and RBD but the frequencies are obtained by sorting and shuffling the input samples and not the frequencies of the Fourier transform. Input samples are sorted and output samples are re-arranged based on sorted input samples. Sorted data is then analyzed using power spectrum. EASI is a "first-order" sensitivity method, but it can be extended to higher order method by implementing multidimensional search curve to the search algorithm.

SOBOL

In Sobol method, the output variance of the model is decomposed into summands of input parameter variances for increasing dimensionality (Sobol 1993; Zhang et al. 2015). Sobol sensitivity analysis determines the contribution of each input parameter and their interactions related to the output parameter variance. While the FAST method uses sinusoidal components of the Fourier method for pattern search, Sobol uses Monte Carlo simulation for that purpose. Sobol algorithm can produce both "first-order" and "total-order" sensitivity indices.

ECOLEGO simulation tool

The modelling platform used for the radionuclide transport modelling and the sensitivity analysis is the Ecolego software product (Facilia AB, version 6.5.39) (Avila et al. 2003). Ecolego can be used for creating dynamic models and running simulations deterministically or probabilistically. Radiological risk assessment has been emphasized in the software, for example, by using a built-in database of radionuclides so their half-lives are automatically taken care of in the dose assessment calculations. The sensitivity analysis toolbox includes various methods and the emphasis in this study is on variance-based methods described above.

Results

In Tables 1 and 2 the sensitivity analysis results for ^{36}Cl are presented. Respectively, similar results are presented in Tables 3 and 4 for ^{135}Cs. The sample size was set to 1000 Monte Carlo simulations in order to ensure convergence in the results. The simulations were done for the 34 parameters mentioned above.

Table 1. First-order sensitivity indices for ^{36}Cl (%).

	EASI	EFAST	FAST	RBD	SOBOL
K_d value for soil	69.4	75.3	75.9	54.8	83.8
Soil to plant CR ('other vegetables')	13.0	10.3	10.3	6.9	6.7
K_d value for lake sediment	5.8	5.6	6.7	4.3	5.2
Outflow rate of the lake	4.1	4.5	3.3	1.8	4.2
Other parameters	7.7	4.3	3.8	32.2	0.1

Table 2. Total-order sensitivity indices for ^{36}Cl (%).

	EFAST	SOBOL
K_d value for soil	28.3	71.1
Soil to plant CR ('berries')	7.9	0.1
Soil to plant CR ('other vegetables')	6.6	5.8
K_d value for lake sediment	4.5	9.3
Outflow rate of the lake	3.0	5.6
Other parameters	49.7	8.1

Table 3. First-order sensitivity indices for ^{135}Cs (%).

	EASI	EFAST	FAST	RBD	SOBOL
K_d value for lake sediment	62.6	64.9	66.5	48.0	80.6
Water to fish CR	22.1	24.3	21.2	14.2	10.6
Volume of the lake	6.8	5.7	7.6	6.1	8.7
Other parameters	8.5	5.1	4.7	31.7	0.1

Table 4. Total-order sensitivity indices for ^{135}Cs (%).

	EFAST	SOBOL
K_d value for lake sediment	45.1	58.5
Water to fish CR	19.0	28.2
Volume of the lake	5.2	2.6
Intake rate of fish	2.3	3.9
Other parameters	28.4	6.8

Discussion and conclusions

When looking at the first order results of ^{36}Cl and ^{135}Cs in Tables 1 and 3, EASI, EFAST and FAST methods produce somewhat similar results but SOBOL overestimates and RBD underestimates the influence of the first parameter when compared with the others. However, the order of the parameters in terms of their influence remains the same in all sensitivity tests. Similar phenomenon can be seen in total order results in Tables 2 and 4. However, the "Soil to plant CR (Berries)" parameter in SOBOL case is not in order when compared to the EFAST case in Table 2. Also, for the ^{135}Cs (see Table 4), the "Volume of the lake" parameter explains less of the variability in model outcome in the SOBOL case compared to the EFAST case. However, order of the largest influence parameters remains the same regardless of the analysis methods.

Overestimation of the first parameter in SOBOL method might be explained by the computation routine. In SOBOL the different occurrences of parameters to be fed into the model are decomposed into summands using the Fourier-Haar transform (Sobol 1993). The summands are orthogonal to each other but the numerical integration of summands (with respect to the output) may increase the influence of large components. It is stated in (Sobol 1993) that some functions may cause numerical integration to behave poorly and may cause unexpected results.

The underestimation problem when using RBD is a direct consequence of using random permutation of selected frequencies. According to (Tarantola

et al. 2006), the analysis covers a subset of input space that is permuted. This permutation operation is done to generate scrambled set of points that cover the whole input space. It seems that the whole subset analysis process acts like a lowpass filter for the variable variances smoothening the spikes out while keeping the order of magnitude in correct places.

To conclude the analysis in this paper, the tested sensitivity analysis methods are capable to order the largest influence parameters in correct order.

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Finnish technologies for monitoring of the Comprehensive Nuclear-Test-Ban Treaty

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Introduction

The Comprehensive Nuclear-Test-Ban Treaty, CTBT, was a result of decades of political and technical negotiations. It was adopted by the UN General Assembly on 10 September 1996 and opened for signature two weeks later. 71 states, including the five official nuclear-weapon states, signed the treaty then and the number of signatures has grown to 183 during the past twenty years.

The CTBT is not as well known as the Non-Proliferation Treaty, NPT, that was opened for signature in 1968 and entered into force two years later. One explanation is that the CTBT has not entered into force yet. There are still 30 states that have not ratified the treaty and seven of them belong to so-called "annex 2"-states. Namely, the CTBT was written so that 44 states with nuclear technology, as listed in the treaty annex, need to ratify it before entry into force. Not surprisingly, states hindering the CTBT are the unofficial nuclear-weapon states DPRK, India, Israel and Pakistan, but also China and the USA have not yet ratified the treaty although they were two of the original signatories.

The enthusiasm in 1996 has changed to a less wishful atmosphere. Nevertheless, preparations for the CTBT and its verification are ongoing. A Preparatory Commission for the CTBT Organization has been established in Vienna and a global verification regime has been built. International conferences are held every two years to facilitate the entry into force of the CTBT.

Verification regime

Already in the treaty text, a verification regime was defined for the CTBT. Besides confidence-building measures, consultations and clarifications, two very concrete elements were envisioned: an International Monitoring System (IMS) and On-Site Inspections (OSI). The latter has been politically controversial, which is easy to understand: an OSI means that the international community requests access to some state territory with suspicions of illegal activities.

On the other hand, building of the IMS has made remarkable progress reaching soon 90% certification level.

In the early phases of CTBT negotiations, monitoring of seismic signals was considered the key monitoring technique. Practically all nuclear detonations are observable in seismic signals, like the DPRK nuclear tests of recent years. Scenarios can, however, be postulated where the seismic signal is not so clear, so needs for monitoring of acoustic waves under water and in the air were clear. Arrays of sensors for seismic, hydroacoustic and infrasound signals have been set up around the globe, with 170, 11 and 60 stations, respectively, as the ultimate goal.

Seismic and sound waves can be produced also by other means but nuclear devices. In addition to the three so-called waveform techniques, an indisputable indication of a large number of fission or fusion reactions is needed: the smoking gun. A fourth network – actually consisting of three separate components – has been built for this purpose.

The primary radionuclide monitoring part of the IMS consists of 80 stations world-wide where ambient air is pulled through a filter medium at high flow rates. After a sampling period of 12 or 24 hours, the filter medium with attached particulate matter is measured with a gamma-ray spectrometer. The measurement and subsequent analysis reveal radionuclide concentrations of the order of 10 uBq/m3, well sufficient for detecting even small nuclear events.

With meticulous planning, nuclear test sites may manage to contain all particulate releases, but confining noble gases has proven to be the most challenging task. Therefore, radionuclide monitoring of particulate matter is complemented with systems that filter noble gases from air at 40 of the 80 IMS stations. A lower air flow rate is sufficient for these samplers since they are able to produce very pure samples of xenon gas. Isotope proportions of four xenon isotopes produced in fission and activation reactions readily reveal fresh anthropogenic releases. As with particulate samples, the natural variations in the background have to be known.

Whenever interesting content is observed in a particulate or xenon sample, it will be shipped to a CTBT radionuclide laboratory. There are 16 laboratories, one of them at STUK's premises in Helsinki, doing more thorough analyses. They complement the three-component radionuclide monitoring system of the IMS.

Finnish technologies

Finland was an active participant in the CTBT negotiations. In addition to people from the Foreign Ministry, experts from the Institute of Seismology of the University of Helsinki and STUK, the Radiation and Nuclear Safety Authority,

participated in the scientific negotiations. Domestic support was provided by the Finnish Defence Research Agency and Aalto University known back then as TKK, Helsinki University of Technology.

The seismic station FINES in Sysmä that has been operated by the Institute of Seismology since 1985 was one of the first certified stations of the IMS. There is very little seismic activity near FINES and it is easily accessible from Helsinki for eventual services, so the station has belonged to the backbone of the IMS. For example, seismic waves from DPRK are always observed within ten minutes at FINES.

Air sampling techniques were developed at STUK in the 1980's. A spin-off company called Senya Oy was established to commercialize the knowhow. The company now has a range of air samplers for radiation detection from portable battery-operated models to the heavy-duty automated models utilized in the IMS. A significant share of the IMS air samplers have been manufactured by Senya and similar samplers are naturally used also in national surveillance networks.

Analysis software for gamma-ray spectrometry have been developed at TKK since the 1960's. Sampo and later Shaman have found wide use for various applications involving high-resolution gamma-ray spectra. In the 1990's the development effort was concentrated on air filter spectra of the IMS network and a spin-off company named Baryon Oy was established.

Results obtained with the Sampo-Shaman system in a completely automated analysis system have consistently been better than with the alternative systems. During the early post-accident weeks after the Fukushima Daiichi accident, it is fair to say that the CTBT Organization was very much depending on the results from the Finnish-made analysis system. The system is also being used by several National Data Centers, processing IMS data in parallel with the International Data Center of the CTBTO, and CTBT laboratories.

Conclusion

It was a lengthy political and diplomatic endeavor to finally obtain a treaty text that could be signed by nuclear-weapon states and most others. When the treaty is still after twenty years not in force, one may ask whether the effort was worth it?

As a positive result, the international community has built four global networks of sensors. These networks can be utilized for secondary purposes that may be considered even more important than the primary purpose of treaty monitoring. For example, the seismic and hydroacoustic data of the IMS network can provide real-time data for a tsunami warning system. The infrasounds picked by the IMS network can be utilized for predicting volcano erup-

tions and, naturally, the radionuclide network complements the picture given by national radioactivity surveillance networks.

These and other non-verification uses of the IMS data are discussed at the CTBT Science and Technology conferences that are held every few years. It is clear that when these comprehensive collections of data are made available for scientists, the development of various analysis methods is boosted.

It is difficult to foresee whether the CTBT will ever be ratified. For the future of the human kind, it would be advisable, but it all depends on the wisdom of future decision-makers.

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Serpent Monte Carlo code for radiation transport applications

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1 Introduction

Serpent [1] is a three-dimensional continuous-energy Monte Carlo transport calculation code, developed at VTT Technical Research Centre of Finland.[1] The development started in 2004, originally for the purpose of generating input data for reduced-order (nodal diffusion) calculation codes used for nuclear reactor analysis [2]. Serpent has been distributed free of charge for research and educational use by the OECD/NEA Data Bank and RSICC since 2009. The user community has grown to almost 800 users in 200 organizations in 40 countries around the world. The most typical Serpent user is a university student, applying the code for academic research and thesis work. This is also seen in the large number of publications and theses on Serpent development and applications. More than 500 peer-reviewed journal articles and conference papers and 140 B.Sc., M.Sc. and Ph.D.-level theses have been published on Serpent-related topics worldwide since 2005. In Finland the project has contributed to six doctoral and several Master's theses at Aalto University and Lappeenranta University of Technology.

During the past few years the field of applications for the Serpent code have been broadened from reactor physics to fusion neutronics, radiation transport and shielding, and the code is becoming a practical calculation tool for the radiation safety community. The background and motivation for this work is introduced below, followed by an overview of calculation methods and potential future applications.

2 Background

One of the major research topics in Serpent development for the past six years has been multi-physics reactor modeling [3], which requires solving the coupled problem between neutronics, heat transfer and coolant flow. High-fidelity meth-

[1] For more information, see the Serpent website - http://montecarlo.vtt.fi

ods, such as Monte Carlo simulation, are not widely used for this type of applications because of their high computational cost, which is why the traditional approach relies on reduced-order methods instead. Implementing the coupling framework required developing new interfaces for passing the power distribution, temperature and density fields between Serpent and thermal hydraulics, CFD and thermal mechanics (fuel performance) solvers.

As a result, the calculation routines implemented for handling complicated 3D data structures were also available for developing new geometry types to complement the traditional constructive solid geometry (CSG) model. Two new geometry types based on CAD and unstructured polyhedral mesh were consequently implemented in 2014–2015 [4,5]. With these capabilities it became possible to model complicated irregular structures, often encountered in fusion neutronics and radiation shielding applications.

High-fidelity multi-physics calculations also require accurate heat deposition models, which in addition to fission heating also account for the effect of prompt fission and capture gammas. This, in turn, required implementing a photon physics model, which was completed in 2016 [6]. With this capability it became possible to perform radiation transport calculations for dose rate evaluation and shielding purposes as well. Since Serpent was originally developed as a reactor physics code, the neutronics model is internally coupled to a Bateman depletion solver [7] to account for the changes in the isotopic composition in nuclear fuel. The same routine is also available for producing radiation source terms for spent fuel and activated materials.

3 Methods

The transport simulation in Serpent is based on the continuous-energy Monte Carlo method. A brief overview on the methodology is provided in the following.

3.1 Transport physics

Serpent reads neutron and photon transport cross sections from ACE format library files. Since the data format was originally developed for the MCNP code [8], there are some considerable similarities in the transport physics as well. The neutron physics model follows the ENDF reaction laws as they are represented in the data libraries, including unresolved resonance probability tables and special scattering laws for important moderator materials. Because the two codes share the same “laws of physics”, neutron transport calculations carried out using Serpent can be expected to match reference MCNP results to within statistics.

The photon physics model, however, is not an exact match to MCNP. Cross sections determining the reaction probabilities of photon interactions are

obtained from ACE format libraries, but the models used for interaction physics requires additional data provided in auxiliary files. The photon transport mode can be used for elements from hydrogen to californium over the energy range from 1 keV to 100 MeV. The four main photon interactions are included: Rayleigh scattering, Compton scattering, photoelectric effect, and electron-positron pair production. A thick-target bremsstrahlung (TTB) approximation is used for taking into account the bremsstrahlung emitted by electrons and positrons. Secondary particles are also created through atomic relaxation in the form of fluorescence photons and Auger electrons, but their transport is not explicitly modeled. A more detailed description of the photon physics model can be found in Ref. [6].

Production of prompt gammas emitted in neutron reactions, such as fission, capture and inelastic scattering, can be accounted for by running coupled neutron-photon transport calculations [9]. Implementation of photonuclear reactions in photon transport mode (i.e. neutrons produced in photon reactions) is currently under way.

User-defined detectors (tallies) can be set up for calculating various volume- and surface-integrated reaction rates. Built-in or user-provided response functions can be used for calculating photon dose rates, but Serpent also has several analog detector types for evaluating the absorbed gamma energy explicitly for different purposes.

3.2 Radioactive decay source mode

Serpent applies a matrix exponential solver based on the Chebyshev Rational Approximation Method [10] for solving the Bateman depletion equations. The methodology relies on an automated calculation routine that also forms the transmutation and decay paths and calculates the neutron cross sections for the coefficient matrix. Serpent is capable of handling more than 100,000 irradiated material zones, each comprised of more than 1500 nuclides. The burnup calculation capability has been available almost from the beginning of the project, and it has been extensively used for reactor physics applications.

Burnup or neutron activation calculation can also be combined with a radiation transport simulation. Irradiated material compositions are written in a binary restart file, and can be read with another input to form the source distribution for neutrons or photons produced in radioactive decay. The calculation combines the radionuclide compositions to source spectra read from ENDF format decay data files. The calculation requires no additional input from the user, since the normalized source distributions are formed automatically.

The radioactive decay source includes discrete emission lines and continuous spectra for neutrons and photons. A separate model to account for

bremsstrahlung associated with beta-decay was also recently implemented. The methodology has been put to practice, for example, in shut-down dose rate calculations performed for the ITER fusion reactor, in which a neutron activation calculation performed using a realistic fusion plasma source was followed by a photon transport simulation in which the source distribution was formed by the activated components [11,12].

3.3 Geometry models

The standard CSG geometry type in Serpent relies on combinations of elementary and derived surface types to construct the geometry from homogeneous material cells. This type of approach can be considered sufficient for most reactor applications, in which the geometries are regular and most structures can be modeled using cylinders and planes. This, however, is often not the case in radiation transport and shielding calculations, in which the particles are transported through complicated and irregular structures.

The unstructured mesh based geometry type [4] applies a polyhedral mesh to describe complicated structures. The mesh is read in OpenFOAM data format, widely used, for example, for computational fluid dynamics. For geometries without internal structure Serpent also provides a CAD based geometry type [5], in which the solid bodies are described by their triangulated outer surface. The geometry is read in the stereolithography (STL) data format. Since this data type is also used for 3D printing, it is supported by virtually all CAD tools. The CAD based geometry type has been successfully used, for example, for modeling complicated fusion reactor geometries [11,12].

Serpent applies a combination of conventional surface-tracking and the rejection-sampling based Woodcock delta-tracking method [13] for transporting particles through the geometry. Delta-tracking was originally implemented because of its good performance in reactor geometries, in which the neutron mean-free-path is long compared to the dimensions. The method has been found to be well suited for complicated CAD and mesh based geometries as well, not only because of its performance, but also because it can easily overcome problems related to small geometry errors that are a common nuisance when CAD based models are used in transport calculation.

3.4 Variance reduction

Monte Carlo transport simulations in reactor physics applications are typically carried out to calculate reaction rates inside or near the reactor core. Since the region of interest is close to the source, these calculations can usually be carried out without applying any variance reduction techniques. This is not always the case for radiation transport, in particular for shielding applications,

in which dose rates are calculated in a region where the particles are intentionally kept away from by physical barriers.

Development of efficient variance reduction methods has been one of the main new topics in Serpent development during the past few years [14]. The methodology relies on the conventional weight windows technique, in which an importance mesh is superimposed on the geometry, and the particles are encouraged to flow in the direction of higher importance by splitting and Russian roulette. The importance mesh can be read in the MCNP WWINP format [15], which is supported by a wide range of deterministic variance reduction tools. As an alternative approach, Serpent provides a built-in light-weight importance solver based on the response matrix method.

4 Future work and potential applications

Even though Serpent is widely used for fission reactor applications, the code is still relatively unknown outside the reactor physics community. There is some considerable interest in adopting the code as a standard calculation tool for fusion research, but it is believed that Serpent could also be used for a wide range of applications in other fields involving radiation transport problems.

The advantages of Serpent include its versatile geometry types, capable of handling complex structures that can be imported from CAD based models without extensive user effort. Another favorable feature is the built-in Bateman solver, which when combined with the radioactive decay source mode enables very detailed modeling of complicated source terms without additional solvers or processing steps. Such source types include spent fuel and activated materials, but also radioactive emissions from reactor accidents and nuclear tests. Practical experience with Serpent users has shown that working with a code that is actively developed and maintained is a considerable advantage, since the developers are capable of providing support and expertise that originates from source-code level understanding of the underlying calculation methods.

The on-going work on radiation transport capabilities is currently focused on several topics, including photon physics and variance reduction. One interesting possibility is coupling Serpent with the Attila radiation transport software [16], which is widely used for shielding design, radiation transport and medical physics applications. The most important goal for future work, however, is expanding the Serpent user community from reactor physics to other fields involving radiation transport problems. During the past ten years Serpent users have actively participated in the design and implementation of new features and capabilities, and it is believed that similar collaboration between code developers and users would considerably benefit the radiation transport community as well.

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Biosphere modelling in safety assessments

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This contribution addresses biosphere modelling in safety assessments for various purposes. In general, such assessments comprise of relatively similar components, and here the focus is on assessments for radioactive waste disposal, even though the same principles can well be, and have been, applied for example to normal-operational and accidental releases from nuclear power plants and other nuclear facilities or to evaluate situations with naturally occurring radioactive materials (NORM) posing a concern. Even if there is some difference in the assessment endpoints (e.g., doses to the most exposed group of humans (or reference persons) and/or other exposed people, and exposure of 'non-human biota') and other assessment context, formulation of assessment scenarios, and the associated demonstration of compliance, the mathematical models for radionuclide transport and dose calculations that are applied in the safety analysis part of the assessments are very similar especially for their structure. It seems that largest differences lie in the rigour and style of addressing the remaining uncertainties and their significance. However, also there general methodologies can and, to a large extent, have been applied to all the assessment types.

The oral presentation addresses a general workflow of a 'biosphere assessment' (although recognising it being a part of the overall whole-system safety case) largely following the IAEA BIOMASS-6 approach developed for geological disposal of solid radioactive waste that is currently under revision through collaboration of the BIOPROTA Forum and Working Group 6 of the IAEA MODARIA II programme. However, to provide the general audience background information, first basics of the compartment modelling methods generally applied are summarised. This encompasses the general assumptions of an approximate equilibrium or steady-state situation in respect of the size of the compartments (both in the model and in the 'reality') and dynamics of the modelled system, and examples of compartment structures, transfer equations and dose calculation schemes.

The further presentation then addresses the application of the models and rationales to satisfy with the state of the model (assessment) development. In addition to the broader assessment context, a key issue is the appropriate representation of the biosphere system being assessed in the models. This often requires not only general knowledge and support from literature, but also understanding of the site and its behaviour. In more elaborate assessments, a number of 'site-interpretive models' (e.g., hydrological or landscape development modelling) may be required to inform the identification, justification and specification of the necessarily more stylised radionuclide transport models and dose calculations to be used. In some cases, though, a static 'snapshot' of the system, or series of them, may be well sufficient. Short-term assessments such as those for normal operation of facilities most often fall into this category. In addition to the representativeness of the site in question, the development and application of the models needs to consider also the exposure groups to be assessed. These may include most exposed humans or representative persons, other exposed people (or a collective dose or a similar quantity) and/or non-human biota at their various levels of biological organisation (individuals, populations, communities, ecosystems).

To demonstrate a sufficient safety assessment, required in addition to demonstrating compliance with the numerical (or qualitative) safety criteria set for the endpoints, sensitivity and uncertainty analyses and verification and validation of the models and modelling procedures are rather well established practices, but especially for the (very) long-term assessments, validation is often challenging. For a more complete picture, approaches such as pedigree analysis (e.g., scoring of the knowledge level underpinning different aspects of model structures and input data) and systematic mapping of the multiple categorical sources of uncertainties and their location across the degree of potential impact on the assessment outcome may be applied.

Session 4: Uptake of radionuclides in biosphere

Chair: Dr. Kaisa Vaaramaa

Non-linear transfer of elements into organisms

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Element- and organism-specific concentration ratios (CR) are generally used in radioecological models to describe the transfer of radionuclides into organisms. This approach is based on the assumption that uptake into an organism is linear with respect to concentration in the medium (soil or water). We have carried out a series of studies on the uptake of elements into terrestrial and aquatic organisms using samples collected from the nature and experimental meso- and microcosms. The initial aim was to determine CR values relevant to boreal ecosystems, but we soon found that the CR approach was not adequate, and studying non-linearity of uptake became the main focus of our studies.

Although the studies aimed at understanding uptake of radionuclides into plants and animals, total element concentrations were measured. This approach is often used, based on the assumption that radioactive and stable isotopes of the same element are taken up similarly.

In a study at two forest sites in North Savo, we studied transfer of essential (Co, Mo, Ni, Zn) and non-essential elements (U, Pb) from soil into three understory species and two tree species (Tuovinen et al., 2011; Roivainen et al., 2012). The radionuclides of these elements are potentially important in the risk assessment of radioactive waste disposal. An important finding was that, for all species and elements studied, the CR was not constant but decreased with increasing soil concentrations. A non-linear equation was found to fit with the observations.

In another study, we used experimental mesocosms and microcosms to investigate the transfer of U, Co, Mo, Ni, Pb, Th and Zn into three plant species, snails and earthworms (Tuovinen et al., 2016a). Non-linear transfer into

plants was observed also in this experimental study, confirming the findings of the natural forest study. Transfer from soil or food (plants) to animals was also found to be non-linear for many of the elements studied. The uptake of U was nearly linear, indicating that different modelling approaches may be needed for individual elements. However, the results did not support the previously proposed simple hypothesis that transfer would be non-linear only for essential elements.

An important finding was that, in conditions found in natural forests, non-linear transfer leads to a practically constant total element concentration in plant tissues (Fig 1). Based on this observation, we developed a novel approach to radioecological modelling. The impact of non-linear transfer on the predictions of radioecological models was studied by comparing the linear model, a non-linear model based on equations derived from our own data, and our novel "constant concentration" model (Tuovinen et al., 2016b). The three models were used to predict transfer of 234U, 59Ni and 210Pb into spruce needles. The predictions of the non-linear model and the novel model were essentially the same, but the linear model underestimated the uptake of radionuclides when the total element concentration in soil was low, but within the range commonly observed in nature. The proposed new modelling approach could potentially reduce uncertainty in model predictions. It was concluded that linear modelling could easily be replaced by a new approach that more realistically reflects the

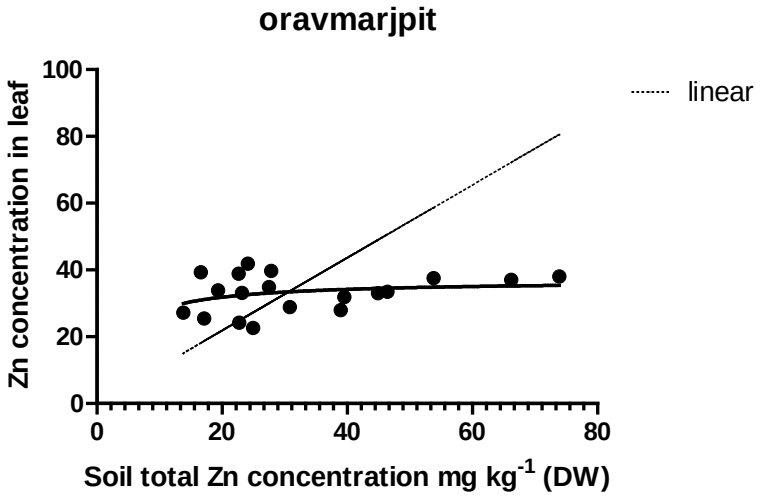

Figure 1. Zinc concentration in the leaves of may lily (Maianthemum bifolium): comparison of observed plant concentrations, conventional assumption of linearly increasing concentration, and a nonlinear function fitted with the data.

true processes involved in the uptake of elements into plants. The new modeling approach is simple, and does not increase the complexity of modelling in comparison to CR-based linear models. Data needed for model parameters are also readily available from different types of ecosystems.

In ongoing studies, we are investigating uptake of elements into aquatic species (chironomid larvae, fish). Preliminary data indicates nonlinear transfer for many elements, but there may be variation between elements.

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Cm complexation with aqueous phosphates at elevated temperatures

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Orthophosphate ions ($H_2PO_4^-$, HPO_4^{2-}, and PO_4^{3-}) are ubiquitous in the environment and may originate from the natural decomposition of rocks and minerals (e.g. monazite or apatite), agricultural runoff, or from wastewater treatment plants. Furthermore, the potential use of monazite ($LnPO_4$) ceramics for the immobilization of specific actinide-containing waste streams may become an important source of phosphates in the future [1–2]. Among the inorganic ligands, phosphates are strong complexants and can be expected to influence the speciation of dissolved radioactive contaminants when present in solution. However, very little data is available on the complexation of especially actinides with aqueous phosphates, even though these complexation reactions precede any aqueous synthesis of monazite ceramics and are expected to occur in natural waters as well as in the proximity of monazite-containing high-level waste repositories. The existing data also suffers from an almost systematic absence of independent spectroscopic validation of the stoichiometry of the proposed complexes.

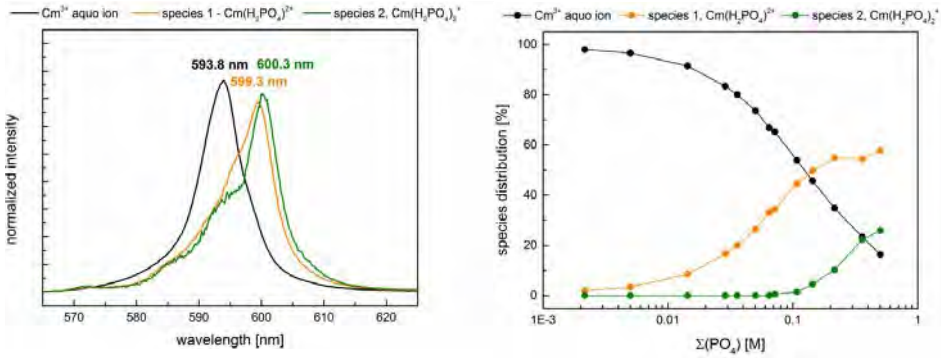

Figure 1. Left – Emission spectra of the Cm^{3+} aquo ion and the two identified Cm^{3+} -phosphate complexes, i.e. $CmH_2PO_4^{2+}$ and $Cm(H_2PO_4)^{2+}$. Right – The extracted species distribution at T = 25°C, I = 1.1 M, and -log[H^+] = 1.

In the present work, laser-induced luminescence spectroscopy has been employed to study the complexation of the actinide Cm^{3+} (5×10^{-7} M) as a function of total phosphate concentration (0–0.5 M $\Sigma(PO_4)$) in the temperature regime 25–80°C, using $NaClO_4$ as a background electrolyte (0.5–2.1 M). The studies have been conducted in the acidic pH-range ($-\log[H^+]$ = 1–2.5) to avoid precipitation of solid Cm rhabdophane ($CmPO_4\times nH_2O$). Under these experimental conditions, the trivalent actinide cation was found to form a complex with the anionic H_2PO_4 species, i.e. $CmH_2PO_4^{2+}$ and $Cm(H_2PO_4)_2^{+}$, depending on the solution pH and the total phosphate concentration, Figure 1.

The complexation reaction occurs at lower total phosphate concentration when increasing the ionic strength or the temperature. Using the specific ion interaction theory (SIT) and the Van't Hoff equation, obtained conditional constants at varying ionic strengths and temperatures have been extrapolated to infinite dilution ($\log\beta^0$) and values for the enthalpy $\Delta_R H°$ (assumed constant between 25 to 80 °C) and entropy $\Delta_R S°$ of reaction have been acquired. The results of the extrapolations are shown exemplarily for the $CmH_2PO_4^{2+}$ species in Figure 2.

The new thermodynamic data derived in this fundamental study will contribute to a fundamental process understanding necessary to critically assess the environmental fate of actinides in the environment.

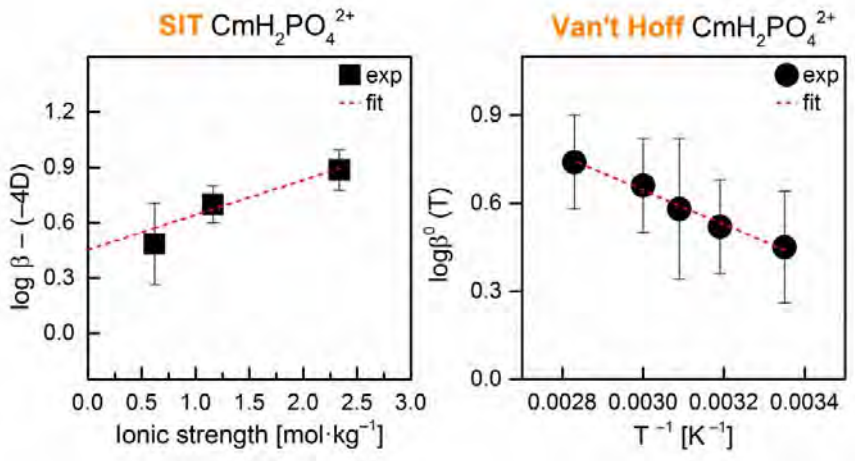

Figure 2. The extrapolation of obtained complexation constants to infinite dilution was done using SIT (left). $\Delta_R H°$ and $\Delta_R S°$ were derived using the linear Van't Hoff equation (right)[3].

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Bacterial selenium oxyanion reduction – effects on bacteria-plant interactions

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Microorganisms in the environment can modify soil characteristics through e.g. oxidation and reduction reactions and secretion of metabolites, altering the availability of chemical elements for plant-uptake. Soil bacteria can form favourable symbiotic associations with plants (like nitrogen fixing root nodule bacteria), but they can also enhance mobilization of hazardous substances, such as radionuclides or heavy metals in soil, increasing their accessibility to plants and further accumulation into the food chains. Translocation and accumulation of radionuclides and heavy metals to plant biomass and to edible crops is extremely undesirable, but as bacteria and plants can be utilized as phytobioremediation agents, these bacteria-plant interactions can be harnessed to benefit the environment and mankind. The chemical compounds of interest (e.g. radionuclides, heavy metals) can be made more accessible for microbe-co-cultivated plants, by treating the contaminated areas with appropriate bacteria. The undesirable compounds can thereafter be removed from the soil, by harvesting the plant material [e.g. 1].

Considerable amounts of selenium (Se) enter the environment through anthropogenic activities, which include coal combustion, mining, refining of sour crude oils and agricultural irrigation of seleniferous soils [2–5]. The biological effects of Se make its releases particularly hazardous in the environment, where the mobility and biological effects of Se are mainly controlled by its chemical speciation [6–8]. In animals, Se acts as an antioxidant and helps in immune responses and thyroid hormone metabolism [9]. However, especially

the higher valence states (selenate, Se(VI)O_4^{2-} and selenite, Se(IV)O_3^{2-}) are toxic at elevated concentrations. Epidemiologic studies have shown that chronic exposure to raised Se intake, especially on regions with elevated soil Se concentrations, is associated with several adverse health effects [10]. These include harmful effects on synthesis of thyroid hormones as well as the metabolism of growth hormone. Other negative effects include impairment of natural killer cells activity, hepatotoxicity and gastrointestinal disturbances [10]. In addition, high levels of environmental Se induces nail and hair loss and dermatitis (skin inflammation) [10]. Also degeneration of motor neurons after chronic exposure is possible. As a result of Se related health hazards, it has been questioned whether the current environmental Se exposure limits are sufficient to prevent these harmful health effects [10]. Plants are the main source of dietary Se, but the essentiality of Se to plants is still debatable [9]. At low doses, Se may protect the plants from variety of abiotic stresses like cold, drought, desiccation and metal stress [9]. However, at higher doses Se toxicity causes oxidative stress and distorted protein structure and function in plants, which may impact the crops through impaired plant growth, development and metabolism.

From the radiation protection point of view, the radioactive long-lived isotope of selenium, ^{79}Se, is one of the high priority radionuclides in the long term biosphere safety assessments of spent nuclear fuel. While bacterial Se reduction is an environmentally important process, only a few SeO_3^{2-} respiring bacteria have been isolated, including *Pseudomonas* sp. strains T5-6-I and PS-0-L [11]. We have found, that these *Pseudomonas* sp. strains form intracellular aggregates of reduced Se0 when incubated in SeO_3^{2-} containing solutions, which were previously verified using TEM and EDX [11]. Nitrate, nitrite and sulfate additions enhanced Se(IV) uptake in the *Pseudomonas* sp. strains, but uptake sustained also under sulphur and nitrogen starvation. This indicates two dis-

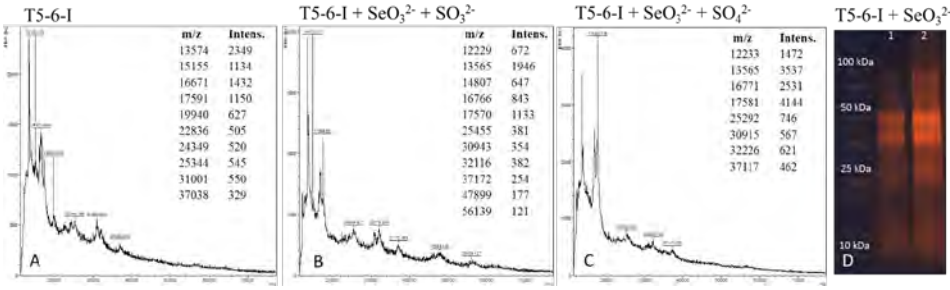

Figure 1. MALDI-TOF spectra and m/z (Da) peaks of total soluble protein fractions of Pseudomonas T5-6-I expressed in response to Se(IV) and other oxyanion treatments at 4 °C (A, B and C). In (D) 4–20% gradient SDS-page gel with T5-6-I control (1) and T5-6-I + SeO_3^{2-} (2). Equal amounts of protein (ca. 30 µg) were loaded onto each well on SDS-page.

tinct Se(IV) transport mechanisms; a low affinity transport system regulated by nitrate, nitrite or sulfate and a distinct Se(IV) regulated transport system [11]. The following proteome analysis of Se(IV) supplement and temperature responses by SDS-PAGE and MALDI-TOF (Figure 1) showed several variations in the protein expression on the 10–60 kDa regions following these stress factors, probably through enzymes associated to oxidative stress, uptake or temperature adaptation.

We cultivated *Arabidopsis thaliana* and *Brassica oleracea* (kale) with previously isolated heterotrophic aerobic Pseudomonas sp. strains to better understand the phenomena of bacteria mediated selenium oxyanion reduction on selenium availability and transfer of Se(IV) to plants. A. thaliana and B. oleracea grown on MS-salt agar were labelled using 100 Bq of $^{75}[Se(IV)O_3^{2-}]$ (with 2.7×10^{-8} M stable Se(IV) carrier) per 5 mL of agar suspension with (Figure 2A) and without (Figure 2B) addition of $5{\cdot}10^7$ CFU *Pseudomonas* sp. amendments. In addition, similarly labeled MS-growth agar with additional 125 µM Se(IV) with (Figure 2C) and without (Figure 2D) the addition of $5{\cdot}10^7$ CFU *Pseudomonas* sp. was prepared. Negative control samples were used in all experiments to ensure no contaminations occurred. Both *A. thaliana* (data not shown) and *B. oleracea* (Figure 2) showed transfer of Se to their roots, leaves and stems. Se accumulation was verified both with autoradiography (Figure 2) and x-ray microtomography (µCT) (Figure 3).

Interestingly, *Pseudomonas* sp. (especially T5-6-I) seemed to enhance Se(IV) plant-uptake both in *A. thaliana* (data not shown) and *B. oleracea* (Figure 2), seen as an increase in the intensity of autoradiograms in Figure 2. In preliminary tests using $^{75}[Se(IV)O_3^{2-}]$-label and gamma spectroscopy on

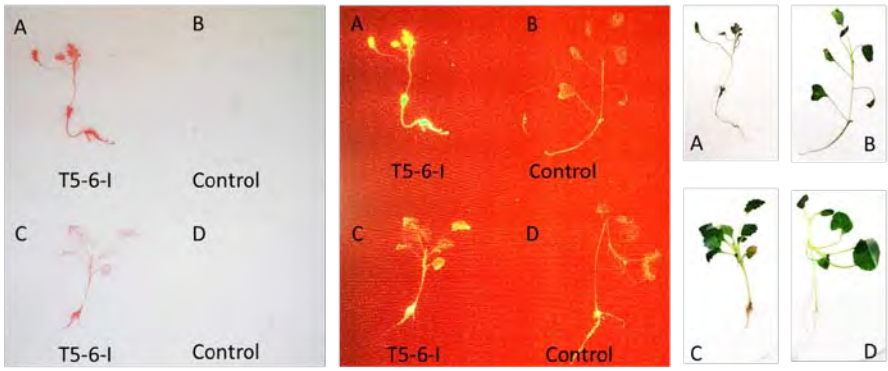

Figure 2. On left and middle Brassica oleracea (kale) grown on MS-salt agar labelled with $^{75}[Se(IV)O_3^{2-}]$-label with (A) and without (B) Pseudomonas sp. addition. In (C) and (D) 75[Se(IV)O32–]-labeled MS-growth agar with additional 125 µM Se(IV) with (C) and without (D) Pseudomonas sp. addition. On right photographs of the plants used for autoradiograms.

average 34% more activity was translocated to *A. thaliana* when $5{\cdot}10^7$ CFU *Pseudomonas* sp. T5-6-I or $5{\cdot}10^7$ CFU *Pseudomonas* sp. PS-0-L or a mixture of these bacteria (total of $5{\cdot}10^7$ CFU) was added to the growth MS-salt agar, compared to the situation when no bacteria were present on growth agar. In addition, in these preliminary tests, Se(IV) concentrations above 125 µM (625 µM and 1250 µM) were observed to impair or prevent growth in *A. thaliana*. However, when using only somewhat increased concentrations of Se(IV) (250 µM), 5·107 CFU *Pseudomonas* sp. amendment seemed to enhance growth in *B. oleracea*.

It is still however unknown, whether *A. thaliana* and *B. oleracea* form intracellular selenium granules, similar to *Pseudomonas* sp. strains seen in

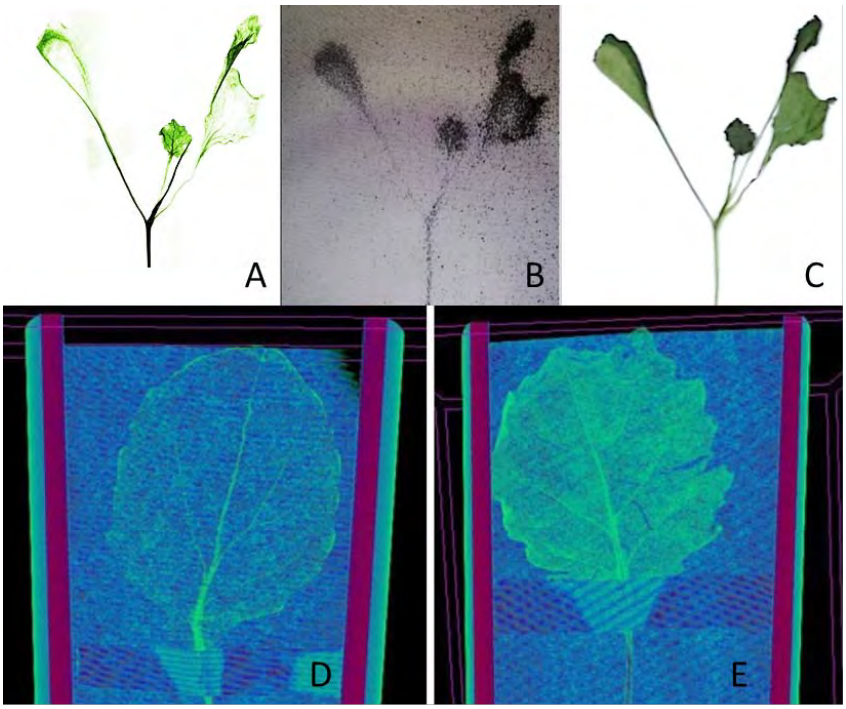

Figure 3. Leaves of Brassica oleracea visualized using autoradiography and x-ray microtomography (µCT). In (A) and (B) the same plant (B. oleracea) on which Se has been accumulated on was visualized using µCT (A) and autoradiography (B). In (C) a photograph of the same plant is shown. In (D) and (E) mature leaves of B. oleracea were freeze dried in a cold vacuum. (D) = B. oleracea without Se(IV), (E) = B. oleracea with 125 µM Se(IV) amendment. In (E), Se accumulation is seen as an increase in signal (green color). The colors in (D) and (E) reflect different absorption densities in increasing order from green (plant tissue) through blue (the filter paper on which the sample was attached to) to red (the test tube).

our previous studies [1] or deposit selenium elsewhere in their tissues (e.g. cell walls). To better understand this phenomenon, we have planned synchrotron experiments targeted for imaging Se accumulation in plant roots in 3D in-vitro in cryo format. Both whole root sample electron densities (with x-ray ptychography) and presence of Se (x-ray fluorescence) would be measured. In addition, experiments using X-ray absorption near edge structure spectroscopy (Xanes) are conducted to verify the chemical form of Se taken up to the plants as well as further microscopic experiments (e.g. confocal microscopy).

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Radioactivity in the ashes from biomass-fired bioenergy production in Finland, 2016 case study

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Radiation and Nuclear Safety Authority of Finland (STUK) performs surveillance of environmental radiation in Finland every year. In addition to the regular annual surveillance program, smaller investigations on specific chosen topics are made from time to time. During 2015-2017 STUK made an investigation "Radioactivity in ashes from bioenergy production in Finland", in co-operation with the bioenergy producing industry. This presentation gives an overview of the results from the investigation. The full report will be published as a separate STUK report, and will be publicly available.

Ash samples from 11 different bioenergy producing facilities were analyzed at STUK for Cs-137 and selected natural radionuclides using gamma-spectrometry. The total number of samples was 157, out of which 66 samples were fly ash, 74 bottom ash and 17 samples were grate ash. 61 of the fly ash and bottom ash samples were collected pairwise from the same facilities at the same time.

The measured activity concentrations were used to estimate the level of potential radiation exposure from the handling and reuse of ash. STUK guide ST 12.2. contains activity indices which can used to estimate whether the use of a material in certain predetermined ways can lead to exceeding the reference levels for public or occupational radiation doses. The results show that there would be no restrictions to the handling of the ashes from the radioactivity point of view, as all the samples have activity indices of <1 for handling of ash. The activity indices for building materials, road construction, landfill and landscaping were exceeded for some of the fly ash and grate ash samples. If these materials were to be used for such purposes, a more detailed dose calculation would need to be made by the applicant to demonstrate compliance with the reference levels.

The activity indices exceeding the reference level were mainly caused by elevated Cs-137 activity concentrations. The grate ash at facilities where fuel collection was solely from the highest Chernobyl fallout area contained Cs-137 with a median of 3100 Bq/kg dw, and a maximum of 6100 Bq/kg dw. It is therefore necessary to make measurements and activity index calculations if planning to reuse ash when the biomass fuel comes from the highest Chernobyl fallout area. Grate ash from the second highest and lowest Chernobyl fallout areas contained Cs-137 with median activity concentrations of 470 Bq/kg dw and 130 Bq/kg dw, respectively. There is an added complication for the grate ash data from these two areas, namely that in these cases the fuel also contained peat, which contributes to the variations in Cs-137 concentrations.

In some of the fly-ash samples Pb-210 is significantly enriched compared to the other uranium-series nuclides, which must be taken into account from the radiation protection perspective. The activity indices in STUK guide ST 12.2. do not take into account elevated Pb-210 concentrations because Ra-226 is representing the whole uranium series in the index calculation. Thus the activity indices do not cover all possible situations, especially if one or a few specific nuclides are concentrated in processed materials. Based on this investigation, it is recommended that the possible high activity concentration of Pb-210 in fly ash is considered when estimating radiation doses from fly ash handling and reuse, and guidance thereof.

The results of this investigation will benefit bioenergy producers and regulators alike, when estimating radiation doses from the handling and reuse of ash from bioenergy production. In the future, follow-up investigations are recommended in the surroundings of biomass-fired bioenergy production facilities to map out possible impacts of radioactivity in the ashes.

Session 5: Effects on humans and biota

Chair: Prof. Jukka Juutilainen

Residential background gamma radiation exposure and risk of childhood leukemia: Results of the Finnish Register-based Case-control study of Childhood Leukemia (FRECCLE)

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High doses of ionizing radiation are an established risk factor for childhood leukemia. However, substantial uncertainty remains about the effect of low doses of radiation. Natural background radiation has a substantial contribution to radiation exposure of the population, for instance in Finland half of the average gamma ray dose is from natural background radiation (mean 1.1 mSv out of 1.6 mSv annually). Radiation-induced cancer risk is larger for exposures during childhood than later in life. Some studies have suggested an association between the level of natural background radiation (Kendall et al. 2013, Spycher et al. 2015) and risk of childhood leukemia, though the results are not entirely consistent (UKCCS 2002, Demoury e al. 2017). We investigated the effect of the indoor and outdoor gamma radiation on childhood leukemia using a nationwide register-based case-control study (Nikkilä et al. 2016).

Material and methods

Our study population consisted of all cases of childhood leukemia diagnosed in Finland in 1990–2011 identified from the Finnish Cancer Registry. Further details of the diagnosis were abstracted from medical records. Three age- and gender-matched controls were selected for each case, and the analysis involved

1093 cases and 3279 controls. Information on pregnancy and birth characteristics were obtained from the Medical Birth Register maintained by the Institute for Health and Welfare. Complete residential histories were reconstructed for the study subjects through the Population Registry with moving dates, addresses and coordinates of each dwelling, as well as information on building characteristics such as type of house, building material etc. Dose rate outdoors and indoors from terrestrial gamma radiation both from natural radionuclides and Chernobyl fallout at each dwelling were reconstructed for each subject from birth to diagnosis of the case using a map of 8x8 km grids provided by STUK. The average dose rate in houses was 41 nSv/h and in flats 70 nSv/h. Doses to the red bone marrow were estimated using a conversion coefficient of 0.7 Sv/Gy. Cosmic radiation was ignored due to it small contribution and minimal variation. Conditional logistic regression was used in the data analysis with adjustment for birth weight (large for gestational age) and maternal smoking.

Results

Of the cases, 81% were acute lymphoblast leukemia (ALL) and 13% acute myeloblast leukemia (AML). the most common genetic abnormality was high hyperdiplody (26%), followed by Tel/AML1 (9%). Other abnormalities were seen in 40% and 27% showed no aberrations.

The mean dose rate from environmental gamma radiation to red bone marrow was 67.2 nSv/h for the cases and 66.4 nSv/h for the controls (Figure 1). The corresponding median cumulative effective dose to RBM for cases was 1.96 mSv and for controls 1.90 mSv.

In the main analysis, no strong association was found for background gamma radiation: the odds ratio of childhood leukemia was 1.01, 95% CI 0.97, 1.05 for 10 nSv/h increase in average equivalent dose rate to red bone marrow. The dose-response analysis involved considerable uncertainty (Figure 2). In subgroup analyses, significant differences by age at diagnosis were observed (p for interaction 0.01). The age group 2–7 years showed an odds ratio of 1.27, 95% CI 1.01, 1.60 for 1 mSv increase in equivalent cumulative dose to red bone marrow. Even higher but non-significant odds ratio was seen in the youngest age group (<2 years). Analyses by genetic subtype were also conducted (high hyperdiploidy showing the highest odds ratio), but the findings were limited by a small sample size (p for interaction 0.14).

Discussion

Our results provide some support for involvement of low doses of ionizing radiation from natural background radiation in the etiology childhood leukemia, particularly at the ages of the highest leukemia incidence. However, back-

ground radiation may account only for a small fraction of all cases, as the doses and, accordingly, the risks are low. A significant effect was found only in the age group 2-7 years, but the risk estimate was substantially larger than expected and the findings of this sub-group analysis may be partly explained by random error. Our findings are suggest some potential differences by leukemia subtype (in particular possibly larger risk associated with high hyperdiploidy), but further confirmation is needed.

The strengths of our study include a comprehensive residential history, which allowed us to assess exposure at each dwelling inhabited by the subjects. This has not been possible in previous studies. On the other hand, some uncertainty stems from the fact that we were unable to make any direct

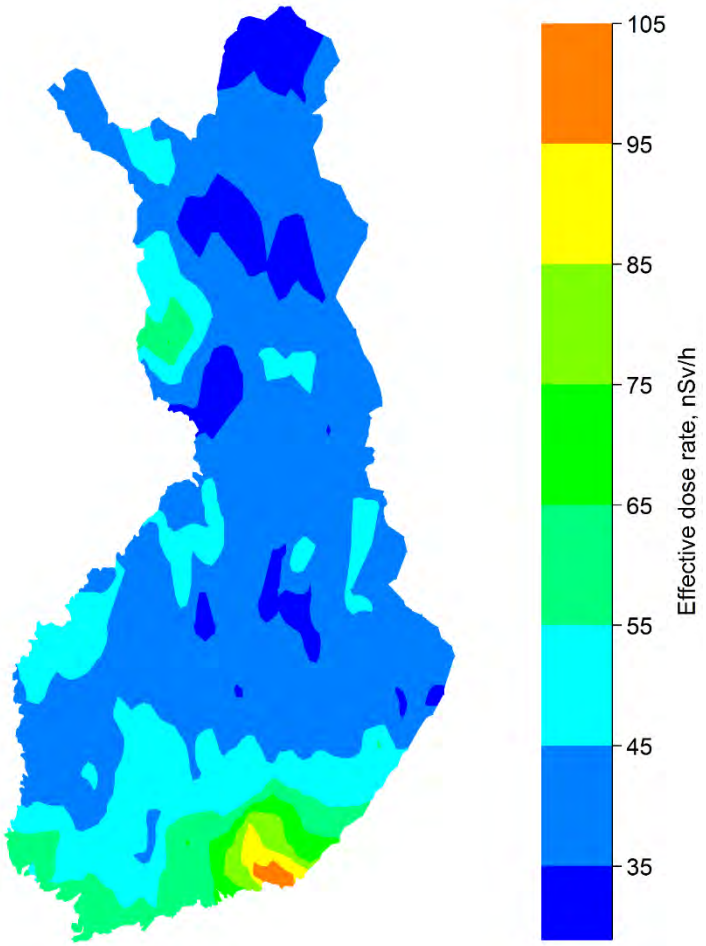

Figure 1. Distribution of background gamma radiation dose rate in Finland.

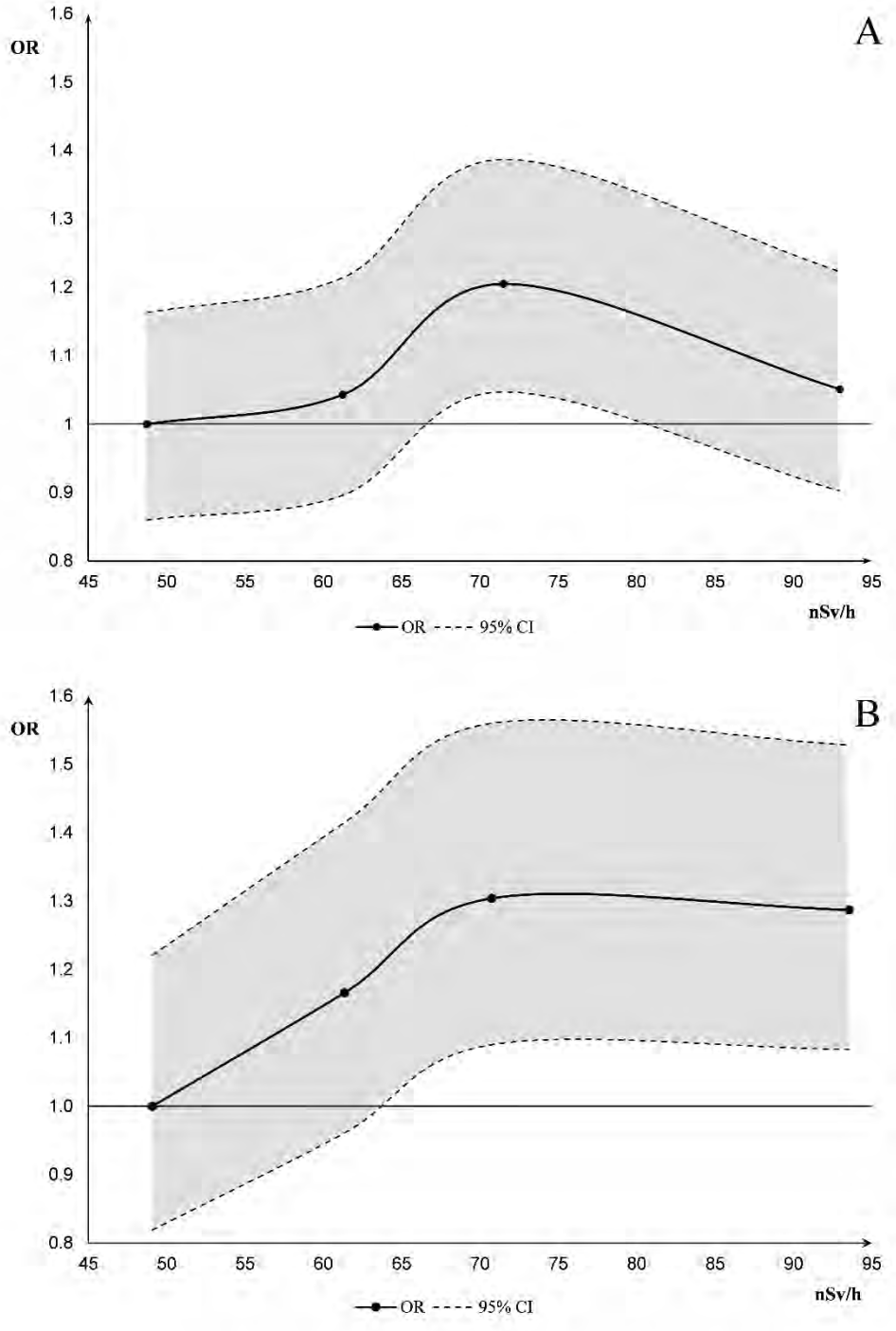

Figure 2. Dose-response analysis of background gamma radiation dose rate and childhood leukemia.

measurements of dose rates, but exposure assessment was based on earlier measurements of ambient dose rate levels. The resolution of our exposure grid was larger than in the previous Swiss and French studies, but our study is the only one taking into consideration also building characteristics (shielding effect and radiation from building materials). Further, radiation exposure outside the home was not considered i.e. background radiation elsewhere. Only one study on the topic has used actual radiation measurements (UKCCS 2002), and despite the more accurate measurement results, major drawbacks involved low participation with potential selection bias and coverage of only current residence. We also had more comprehensive information on the risk factors of childhood leukemia than other studies (with possible exception of the Swiss study).

Our study was limited in its precision to estimate the effects of low doses of radiation from background gamma radiation. The number of cases was smaller than in the UK and French studies, but larger than in the Swiss cohort study. However, the findings were consistent with previous studies and suggest that even low doses may have some effect on risk of childhood leukemia.

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The effects of low-dose ionizing radiation on wild populations – Chernobyl field studies

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Abstract

Nuclear disasters can spread radioactive contamination to large areas with highly variable dose-ranges. Medical studies have shown that ionizing radiation in high doses can have strong negative effects and fatal doses have been established partly as consequences to large nuclear accidents. However, quantifying the long term and cumulative effects of low-dose radiation is quite challenging. In addition, the question about the effects of low-dose ionizing radiation in wild populations remains unclear, and there is contradicting evidence whether the effects in nature are positive or negative.

Recent advances in ecological studies of wild animal populations in Chernobyl (Ukraine) and Fukushima (Japan) have shown significant negative physiological, developmental, and fitness effects due to radioactive contamination. Several studies report lower abundance of several species in contaminated areas in Chernobyl and Fukushima. Studies in Chernobyl have covered multiple species from soil organisms to large mammals, but studies involving small mammals have mainly focused on genetic variation. Our studies focus on small rodents and whether low levels of chronic radiation can cause selection pressure in wild populations. Being in key position in forest ecosystem and one of the most abundant rodent species in Chernobyl exclusion zone, bank vole (Myodes glareolus) is an ideal study species to test the effects of ionizing radiation.

Studies in birds and insects show for example decreased abundances, lower fertility and organ damage such as radiation cataracts in eyes and lowered brain mass. Our aim was to investigate whether similar effects would also be visible in mammalian populations. In our yearly trappings, we have seen a negative correlation between bank vole population size and radiation level. Lower population size is clear indicator of selection pressure in wild populations. We also have investigated bank vole sperm properties and female litter

sizes in Chernobyl to see how radiation in the environment translates into growth, reproduction and survival in radioactive environment. Our results with Chernobyl bank voles were strikingly similar with previous results with birds and insects. There was similar organ related damage related to brain size and cataracts in eyes as observed from barn swallows. We found negative correlation between radiation level and sperm size and quality. Radiation also had a significant negative effect on litter sizes. In addition we have found evidence of increased oxidative stress both in genetic side and fenotypic properties.

Radiation related research aims mainly on human safety and the importance of ecosystem level studies are less valued. Censuses of numbers of individuals cannot necessarily tell how healthy the population is and misinterpretation of the results might lead to underestimating the risks that populations face in contaminated environment. In our studies we have found the effects of radiation from cellular level to population level. Moreover, the results are not species specific and not even location specific. Our results show the importance of the ecosystem studies in contaminated areas. We suggest that more ecosystem level studies are needed, and these should be taken more into account in radiation risk assessment.

Microcosms for testing of individual stressors and environmental samples

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Ecotoxicology is a field of science, which is providing data and tools for evaluation of environmental effects of stressors, principally different types of chemicals. Ecotoxicity testing, especially standardized, is based on single species assessment using individual level end-points. The species have been selected to represent ecologically important trophic levels. For example for the aquatic environment the chosen ones are often green algae (producer), daphnia (1st level consumer) and a fish (2nd level consumer), but due to nature of the tests possible changes at trophic level interactions, which may be then reflected at the population and ecosystem level, cannot be detected. In addition, same tests are many times used to assess toxicity of environmental samples (water, sediment, soil), also without precise comprehension what exactly is causing the detected effects. On the other hand, these tests are usually quite easy to conduct, replicate and repeat. The causal relationships are also clear.

To overcome the shortcomings of the regular testing set ups, it is possible to use more complex settings such as mesocosms or simply field monitoring. However, usually these do not enable studies with single stressors and cause-effect relationships are uncertain. For that laboratory scale microcosms offer more controllable platform than the previously mentioned set ups, but cover also trophic level interactions and ecosystem level measures. Microcosm tests include always more than trophic level, but can be replicated and repeated.

Nordic Cosm Pilot Study (NORCO)-project compiled a pilot study for the use of microcosms in radioecological research. The consortia included participants from following organizations: Norwegian Centre of Excellence for Environmental Radioactivity (CERAD), Norwegian Radiation Protection Authority (NRPA), Stockholm University (SU), Finnish Radiation and Nuclear Safety Authority (STUK), University of Eastern Finland (UEF), Norwegian University of Life Sciences (NMBU).

In this study, the effects of different levels of external ionizing radiation on aquatic ecosystem were studied to reveal not only direct effects of the radiation on individual species but also secondary effects that arise from the interactions within the model ecosystem (Hevroy, TH and Golz A-L et al 2018, submitted). The microcosms (volume 4 L) consisted of artificial freshwater and sediment as well as two phytoplankton, one zooplankton, three plant and one gastropod species. In addition, litterbag with tree leaves was used to monitor microbial degradation. General observation was that the producers appeared to be more sensitive to the gamma radiation than the consumers in the microcosms. Both individual and ecosystem level responses have to be monitored to reveal the stressor effects in these kind of test systems.

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Hevroy TH, Golz A-L et al. 2018, submitted.

POSTERS, Session 1 and 2

Radioactivity on peatlands of various uses and development stages – an overview

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Introduction

Peatlands are ecosystems maintained by a moist local climate and characterised by high groundwater levels, where only partially decomposing organic matter accumulates as peat. Peatlands are often also habitats that are dominated by peat-forming vegetation. Variation in hydrology, peat thickness and nutrition is caused by different development stages and topography resulting from the peat accumulation that may span thousands of years. Naturally, the composition of vegetation and occupying fauna vary as well, and peatlands and particularly their pool areas often offer a suitable environment for many acidmire species and rare and endangered species. (E.g., Laine & Vasander 1996, Rydin & Jeglum 2006, Wieder & Vitt 2006, Päivänen & Hånell 2012, Beadle et al. 2015).

Regarding radioactivity, peatlands not only effectively retain atmospheric deposition falling on them, but they also often are more or less direct recipients and buffers or storages of radioactivity carried by surface and ground waters due to their inherent location in the wetter areas of the landscape and due to the relatively high capacity of the peat to retain many contaminants. In this contribution, sources and storages of radioactivity and its behaviour in peatland environments are outlined, largely based on the earlier work of the authors (in particular, Kaunisto et al. 2002, Aro et al. 2009, Ikonen et al. 2015, Kangasniemi et al. 2016, Aro et al. 2017, Ikonen 2018). Here, after summarising the sources, the subsequent migration and distribution patterns of radionuclides and their changes with the mire succession and land use modifications are discussed, supplemented with a brief account of modes and pathways of exposure of people and non-human biota to the radioactivity and radiation.

Sources and origins of the radioactivity

Naturally occurring radionuclides in peatlands typically originate from Earth's crust and the subsoil (both through erosion/leaching and as gases such as radon or other noble gases) or from the atmosphere (products of cosmic radiation). Further inputs mediated, directly or secondarily through runoff from upstream areas, by atmospheric deposition, include radioactive releases from nuclear weapons tests, nuclear accidents, operational releases from nuclear facilities and use of naturally occurring radioactive materials (NORM; e.g., combustion of coal, shale or peat). Radionuclide inputs carried along by surface and groundwater flow may result also from disposal of radioactive waste, liquid effluents from nuclear, research or medical facilities or, for example, uranium or thorium mining. Coastal peatlands may also receive radionuclides in seawater through high-water events or sea spray. (E.g., Eisenbud & Gesell 1997, Bréchignac & Howard 2001).

Migration, retardation and distribution of the radioactivity

Even though there is large variation in peatland ecosystems, there are also some common characteristics over the types (e.g., Clymo 1984, Rydin & Jeglum 2006, Limpens et al. 2008, Päivänen & Hånell 2012): the vegetation typically consists of a moss carpet and a field layer, density of which varies by the relatively small-scale pattern of differing moisture conditions ('mire microforms', e.g., hummocks, lawns, carpets, hollows and pools). In drier areas, also shrubs and trees can be found. Open surface-water areas and ditches typically support aquatic mosses and macrophytes. The upper peat layer (about 5–60 cm; 'acrotelm') is unsaturated with water and oxic during the growing season, supporting most of the biological activity and soil organic matter turnover, whereas the peat layer below ('catotelm'; Figure 1) is waterlogged, anoxic and characteristically slow in matter turnover. However, the oxic–anoxic boundary shifts with water table fluctuations, and the associated change in the redox potential is decisive for many biogeochemical processes that exhibit a strongly vertically structured environment (e.g., aerobic and anaerobic processes). The peat layer grows, though, due to the input of fresh plant residues. The newly formed layer collapses under its weight, which results in a rather sharp decrease in the hydraulic conductivity, and the acrotelm–catotelm interface rises relative to the bottom of the peat. The transport of water, solutes and gases is very slow at depth, compared to the surface, due to decrease in diffusion coefficients and hydraulic conductivity of the peat with increasing water saturation and compaction. In some conditions, a hydraulically active and relatively nutrient-poor sand layer overlies the tighter and more fertile bottom clay, but the sub-soil can also be hydraulically poorly conductive throughout.

In crystalline bedrock environments (such as in Fennoscandinavia), radionuclides transported by groundwater solutes and gases arrive in the surface system along bedrock fractures and travel through the water-saturated mineral sub-soil rather similarly to other ecosystems. In the catotelm, the advective transport is the slower the thicker the peat column is, and the radionuclide transport may be dominated by diffusion until reaching more surficial conditions conductive enough. Peat is also an effective sorbent for many contaminants and radionuclides. However, even in areas with thick peat layers, open-water pools may considerably aid the radionuclide transport from the depth to the surface, the more the deeper they are, as well as cracks and pipes in the peat layer (e.g., Limpens et al. 2008). The organic matter in the peat decays also in the catotelm, albeit slowly, gaseous results of which may also aid the transport of the radionuclides. Lateral transport in the catotelm is typically minor (except for cracks and pipes). In the acrotelm, however, lateral transport dominates over the vertical for the water flow. For radionuclide inputs through atmospheric release and surface water flows, movement along and sorption in the acrotelm dominate this 'abiotic' part of transport (microbes may play a considerable role in the sorption to the 'inert' peat, though). Most of the releases from peatlands occur with the surface runoff along the acrotelm and open-water areas, or as gas.

The uptake into the macro-biological cycling occurs from the canopy atmosphere or from the substrate; mainly from the acrotelm, but particularly some plants specialised into growing in waterlogged conditions take nutrients and other substances also from the catotelm. When the peat layer is thin enough, roots of trees and other plants may reach also the mineral sub-soil. The plants specialised to waterlogged conditions provide oxygen to their root system via their stems, and also provide an escape route (aerenchymal transport) from the catotelm for gaseous species (e.g., ^{14}C-methane). Gases may also escape from the catotelm or the gas-trapped space in the acrotelm through ebullition ('bubbling'). Some gaseous radionuclides released to the canopy atmosphere may become taken up by the plant foliage, and some radionuclides may pass relatively freely. In any case, the ventilation of the canopy air is a key process for the release of gaseous contaminants to the wider atmosphere.

Radionuclides captured by the vegetation through root or foliar uptake are mainly transferred to the acrotelm by leaching by the rain (throughfall) and by litterfall, possibly occurring in the various vertical layers of the vegetation (Figure 1). The plant litter must be decomposed by microorganisms before the radionuclides are available for uptake again, and the degree and rate of decomposition, leaching and bioavailability, and further fate of the radionuclides, vary considerably with the litter quality and environmental conditions. For the

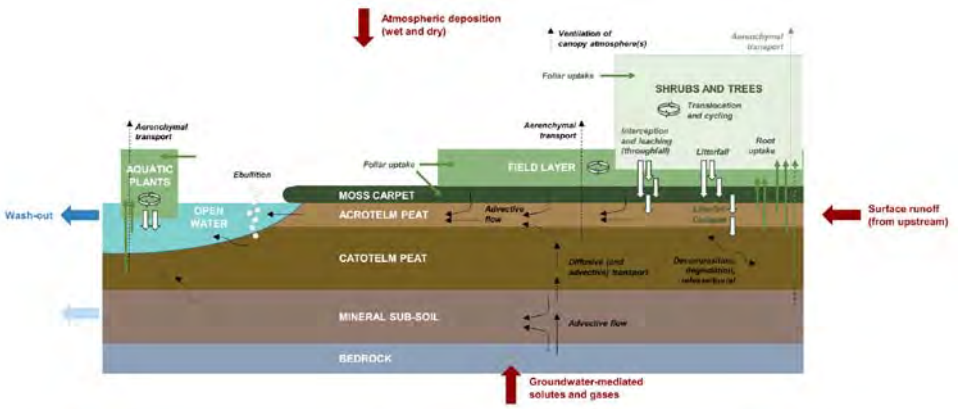

Figure 1. A schematic illustration of typical inputs and characteristics governing radionuclide distributions in peatlands (not to scale; the figure on courtesy of Enviro-Case, Ltd.).

direct atmospheric deposition or throughfall, some nuclides may even be taken up directly through the leaves. Once captured by plants, internal nutrient cycling transfers radionuclides between different plant parts (translocation). Radionuclides in certain plant parts, or on them, may become consumed by animals and transported further in the food web.

Impacts of mire succession

The biogeochemistry, and thus the migration and uptake of radionuclides, is intrinsically coupled with the hydro-geomorphological development of the mire. Hydrology drives both the initiation and development of the mire and the biogeochemistry, in addition to the diffusion that is the key transport process in the typically hydraulically poorly-conductive deep peat layers. Both the formation of new peat, that is, the growth of the peat deposit, and the lateral extension of the mire are regulated by the shape and size of the water 'bubble' held by the peat, its size growing with the gradual increase in the peat thickness (as explained e.g. by Clymo 1984). Lateral expansion through paludification into areas where not prohibited by higher ground or other factors maintaining relative dryness.

The link with the groundwater-mediated releases from a repository to the surface ecosystem depends on, and changes with, the mire succession: In littoral and overgrown areas of ponds and lakes, there is a strong link also with the aquatic system, but there, as well as in minerotrophic mires with a thin peat layer (fen), the releases are readily available for the biological circulation. With the development towards an ombrotrophic bog the link, in the broader picture, weakens but accumulation of the released radionuclides in deeper peat

layers and their re-release due to subsequent change in land use (see below) becomes more important. Whereas in old bogs the connection to the groundwater has ceased, the laggs (peripheral areas) may remain minerotrophic (Figure 2 and the text below) and thus offer the groundwater-carried radionuclides a direct access to the rooting zone.

In areas of post-glacial land uplift, there is a relatively straightforward link between the age of the peatland and its geographical position (elevation; 'spatio-temporal distance from the sea'): In a coastal marsh, open-water pools and/or thin peat cover prevail, and water chemistry and movement is affected by the sea (e.g., groundwater intrusion, flooding, sea spray). With increasing peat thickness and distance from the sea, the link of plant roots to the groundwater gradually ceases. With further development into a 'middle-aged' mire, different morphological sectors of the cross form have to be considered (e.g., lagg, central area, and marginal slope in between; Figure 2). In the laggs, runoff is possibly from both the central area of the mire (nutrient-poor) and upland mineral sites (nutrient-rich). Groundwater typically is available in the root zone, and the laggs are prone to flooding. In the central area, larger areas are losing the connection to groundwater with time. Small-scale microforms, reflected also in the root systems, are typical. Furthermore, forested and open peatlands support different microbial and vegetation communities, and tree cover has a strong influence on peat physical and chemical properties (Ratcliffe et al. 2017). The ombrotrophic central parts are only fed by precipitation. As a consequence, mineral nutrient status is poor and the cycling of organic matter and nutrients, and thus of also radionuclides, will mostly take place in the most superficial peat layers of the bogs.

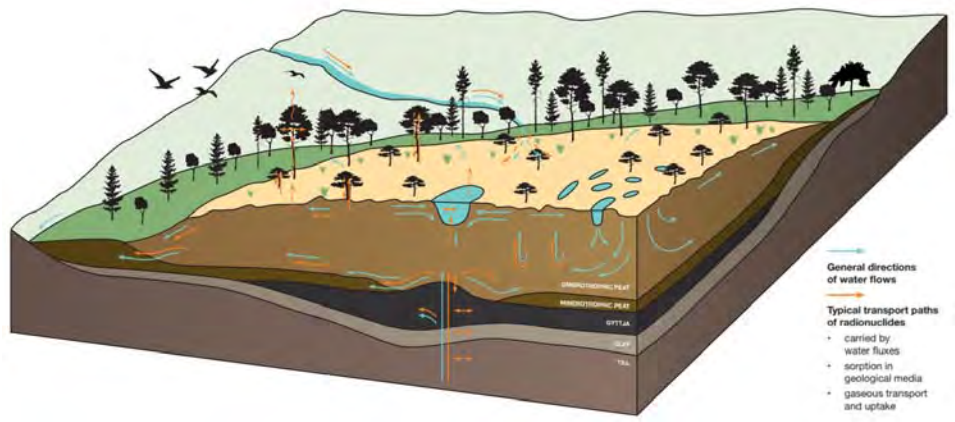

Figure 2. A schematic illustration of the overall pattern of ground and surface water flow and radionuclide transport paths in a 'middle-aged' peat bog (reproduced from the poster of Ikonen et al. 2015 on courtesy of EnviroCase, Ltd.).

For the contaminants brought into the peatland with surface water or atmospheric deposition, the overall picture tends to remain similar throughout the mire succession, even though the flow paths usually become somewhat altered with the growth of the 'peat cake' and also the details of the vegetation-mediated transport change along with the changes in the plant communities.

Impacts of land use changes

Natural processes may be disturbed by humans or natural disasters. The former encompasses a variety of land uses, of which drainage for forestry or agriculture and harvest of peat for fuel or for horticultural substrate are the most common. After such uses, a former peatland may be rewetted for a wetland habitat, or the area may be used for forestry or agriculture (or other purposes) more or less as such after the removal of the peat layers of value. Some constructed or natural wetlands are also used for nutrients and contaminant removal and/or for surface-water discharge management.

In drained mire peat layer dries and compresses which results in the bottom layer becoming root-available or washed out. Better aeration of the rooting zone increases microbial activity and thus availability of nutrients to vegetation, and drainage may enable re-established (at least partial) connection to groundwater. Mixtures of peat and mineral subsoil (soil texture and properties) in the rooting zone are possible in certain situation (e.g., cultivation, peat harvesting).

Radiation exposure of people and non-human biota

For the exposure of biota to the radionuclides and radiation emitted by them, food webs and intake are typically the most important factors. Also the occupancy affect the exposure; migrating or visiting organisms may get exposed as well, although typically to a smaller extent than the resident ones. For wetlands, particularly, the environment is transitory between terrestrial and aquatic types of habitats, and the open-water areas (e.g., bog pools) may be of importance to identify biota species and/or populations of interest. Radiation exposure of humans may arise from various activities, most typical of which are berry and mushroom picking and hunting, as well as various modes of recreation (from hiking and skiing to swimming in mire lakes or pools). Various materials available from peatlands may be used to a smaller extent as well. These include, for example, medical and other herbs, decorative materials, cloth fibres and use of peat and peat-derived products for healthcare, peat therapy or art.

Conclusions

Peatlands and particularly mires are inherently complex systems, but a relatively few basic principles seem to characterise them and their development. A conceptual model of their basic functioning, radionuclide transport and accumulation, and exposure of people and biota to the radioactivity has been outlined here. However, due to the diversity characteristic to peatlands, care needs to be taken in application to particular situations; the authors foresee further work to provide further illustrations and empirical support on further intricacies in later contributions.

Acknowledgements

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Speciation and analysis of soil-plant transfer of I-129 and Tc-99 for dose estimation in crop cultivation

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Motivation

This project aims at obtaining detailed knowledge on the radionuclide transfer to the biosphere following unintentional release from a potential repository for radioactive waste. The impact of expected climatic changes over relevant periods and subsequent changes of soil characteristics and agricultural use will be taken into account for different scenarios. Within the scope of this work, the chemical speciation in soils and the radionuclide transfer to plants will be investigated.

The current radioecological models for long-term safety of potential repositories assume an input of radionuclides into the biosphere via the water path in accident scenarios [1–3]. In addition to irrigation and natural rainfall, near surface groundwater is particularly relevant for the contamination of soil. Groundwater can reach the soil surface naturally via springs or capillary rise. Furthermore, groundwater can be extracted by man via wells for drinking or irrigation purposes (Fig. 1).

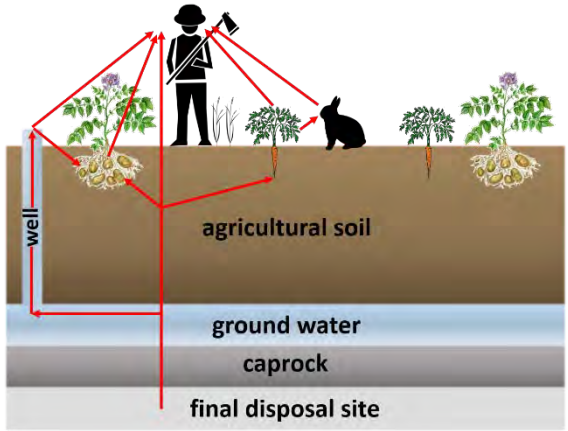

Figure 1. Possible pathways of radionuclides from final disposal site into the biosphere.

According to the different contamination scenarios outlined above, radionuclides can accumulate in agricultural soils over long periods of time. Many aspects of the transport processes involved, the adsorption to soil components, speciation of long-lived radionuclides, and the transfer between different environmental compartments are still unknown.

Radionuclides

This work considers two of the most important fission products from nuclear power plants, I-129 and Tc-99. These are long-lived fission products of U-235 with half-lifes of $T_{1/2} = 1.57 \cdot 10^7$ and $T_{1/2} = 2.1 \cdot 10^5$ years. Due to the potentially high mobility, which will be discussed below, and the long half-lifes, these two radionuclides may dominate radiotoxicity of release from a repository after long time scales (Fig. 2) [4].

Under environmentally relevant conditions, iodine can exist as solid, liquid, or gas, and can occur in a variety of oxidation states (-I, 0, +I, +V, and +VII). It is available as iodide (I^-), iodate (IO_3^-), and readily reacts with organic

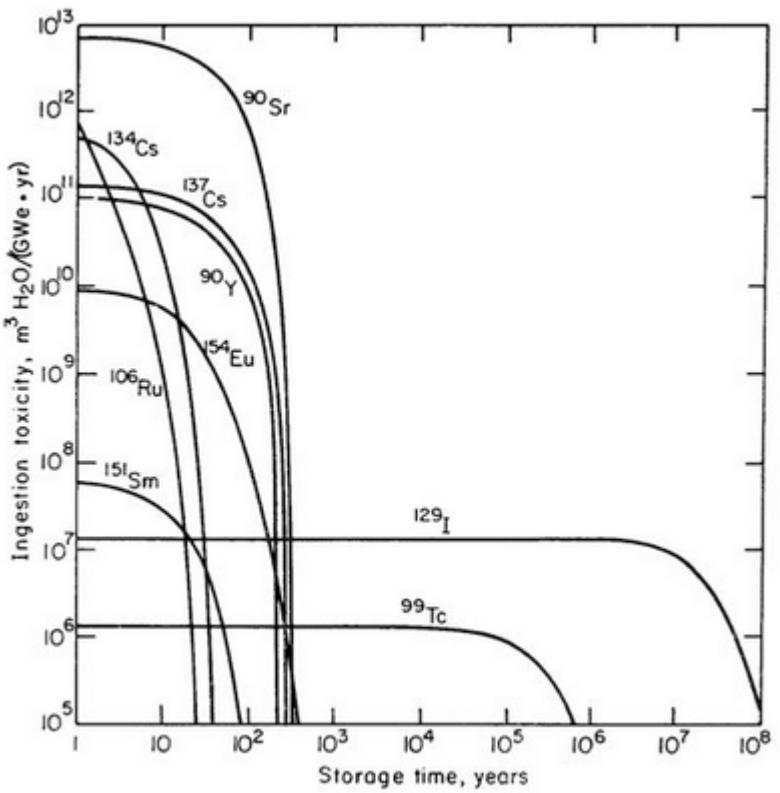

Figure 2. Radiotoxicity of selected radionuclides.[4]

compounds. However, there is still considerable uncertainty regarding the role of microorganisms on iodine adsorption and the bonding mechanisms deriving from the heterogeneous nature of organic matter. In organic-poor soils, iodine adsorption is primarily influenced by soil minerals, especially iron and aluminum oxides, and pH [5, 6].

The mobility of technetium depends on the current oxidation state. In principle, technetium can occur in all oxidation states from +VII to -I. In spent nuclear fuel, technetium mainly occurs in oxidation state +VII as pertechnetate-ion (TcO_4^-). Depending on soil conditions, Tc(+VII) can be reduced to Tc(+IV). Depending on the species, the sorption capacity of the soil differs and subsequently the mobility of the compound. TcO_4^- has a high mobility and can easily enter the biosphere while the mobility of TcO2 (+IV) is very limited [7–10].

Planned Experiments

In column experiments, the migration process of iodine and technetium in four different soils will be investigated. Therefore, lab lysimeters for in situ measurements of pH and pE will be used (Fig. 3). From the measurements of the pH- and pE-values, the chemical species of iodine and technetium can be determined by modelling with PhreeqC.

Figure 3. Lab lysimeter that will be used in this work. [ecoTech]

Figure 4. Growth chamber with carrots (top) and potatoes (bottom).

Two different reference soils have been chosen representing typical soils types in Germany, one of them being a dystric cambisol (RefeSol-01A) mainly consisting of sand (ca. 73%) with a low content of Corg (0.9% in average), the other one an eutric cambisol (RefeSol-03G), consisting of silt-loam, having a Corg content of around 3% [11]. The other two soils, representing future developments of the chosen RefeSols resulting from climate change, will be identified in the course of this project.

For a general dose estimation, also the uptake of radionuclides into plants has to be considered. For this purpose, various crops such as carrots, peas, potatoes, and wheat are studied. The plants are growing under controlled conditions in growth chambers (Fig. 4).

The crops are cultivated in pots, which are placed in glass bowls filled with sand for irrigation and radionuclide contamination from the bottom to simulate uptake from groundwater. A first test with Brilliant Blue FCF (E133) was done to confirm the tracer uptake after irrigating from the bottom (Fig. 5).

Figure 5. Tracer test with Brilliant Blue FCF shows the uptake via roots when irrigating from the bottom.

The radionuclide concentration in different plant parts will be determined by means of radiometric methods (γ- and LSC-Measurements). Furthermore, the speciation in the individual parts of the plants will be studied by coupling of separation techniques like (IC-)ICP-MS. From these measurements, transfer factors for the soil-plant system can be determined from which dose calculations can be carried out.

In the experiments, I-125 (half-life of $T_{1/2}$ = 60 d) is used as homologue to I-129 . Tc-99 will be used in form of a NH^4TcO^4 solution in 0.1 mol·l^{-1} NH_3. The project also includes experiments with the long-lived radionuclides plutonium and americium.

This work is part of the joint project *"Transport and Transfer Behaviour of Long-lived Radionuclides along the Pathway Groundwater – Soil – Surface – Plant in Consideration of Long-term Climatic Changes (TRANS-LARA)"*.

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Vertical Migration of Radionuclides in Soil Samples from Pripyat

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Motivation

The accident of Chernobyl led to significant contamination in large regions of Europe, particularly in Ukraine, Belarus and Russia. In the Chernobyl exclusion zone (CEZ), which includes a 2200 km² large area around the Chernobyl nuclear power plant, the highest amount of radioactivity was distributed. The decrease of surface activity concentrations is governed not only by physical decay but also by vertical migration. In this study the vertical distribution of ^{137}Cs, ^{90}Sr, ^{241}Am and plutonium in upper soil layers from the exclusion zone was investigated to get information on the retention mechanisms of these nuclides. Therefore, several drill cores of 30 cm length were collected in the city of Pripyat, which is about 4 km away from the nuclear power plant. Each drill core was cut into 10 layers of different thicknesses. Due to their half-lifes ^{137}Cs ($T_{1/2}$=30,17 a) and ^{90}Sr ($T_{1/2}$=28,78 a) still dominate the contamination in this region. Because of their relevance to long-term considerations the different isotopes of plutonium (^{238}Pu, ^{239}Pu, ^{240}Pu) as well as ^{241}Am being the daughter of ^{241}Pu were also investigated.

Methods and Measurements

In the first step, each sample was analyzed by γ-spectrometry to determine the amount of ^{137}Cs and ^{241}Am by their characteristic γ-lines (^{137}Cs: Eγ=661,7 keV; ^{241}Am: Eγ=59,54 keV). In the next steps, the soil samples have been ashed and plutonium and strontium were chemically separated. For the detection of plutonium α-spectrometry was used. The ^{90}Sr measurements were performed by liquid scintillation counting (LSC).

Results

Figure 1 shows the specific activity of ^{241}Am, ^{137}Cs, ^{90}Sr and the three plutonium isotopes in different depths of one drill core. While the absolute specific activities in layers of equal depth differ considerably, the decrease follows similar slopes in all cores and all nuclide below the 15 cm. Each nuclide is detected down to 30 cm depth.

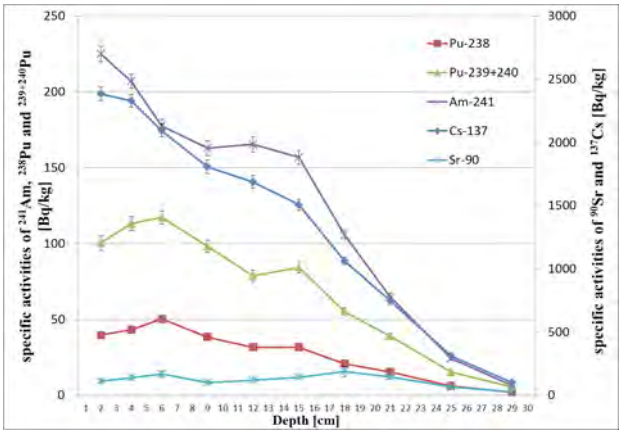

Figure 1. Specific activities in soil layers.

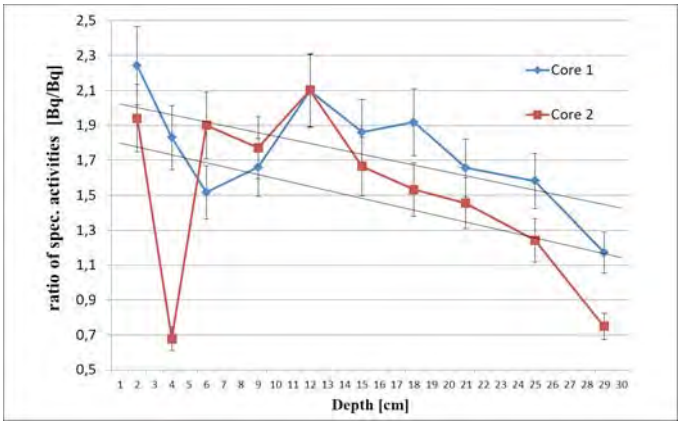

Figure 2. Ratio of ^{241}Am and $^{239+240}$Pu activities for two cores.

Important information on the different migration behavior is obtained from the specific activity ratios of different nuclides (Figure 2).

From the decrease in the ratio of ^{241}Am and $^{239+240}$Pu activities migration rates of plutonium and americium are deduced by a simple model.

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Radon concentrations in Finnish houses built in 2013–2015

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Abstract

Indoor radon concentrations were measured in 1332 randomly chosen dwellings that received building permission between November 2012 and October 2013. The data included 48% of the dwellings in the original sample. The average radon concentration was 71 Bq/m^3 and the median 42 Bq/m^3. These were roughly 25 % lower values than that measured in dwellings constructed in 2006–2008. Six percent of the measurements were higher than the reference level of 200 Bq/m^3. Correlation between air tightness and radon concentration were also studied. Within those dwelling, where the radon prevention was not used, radon concentration increased with the improving air tightness. This observation emphasizes the importance of radon prevention in new energy efficient houses.

Introduction

Elevated indoor radon levels are common problem in Finnish dwellings. Radon concentrations vary between different areas in Finland. The highest average radon concentrations have been found in the following regions: Kymenlaakso, Päijänne Tavastia, Pirkanmaa, South Karelia and Tavastia Proper. Later in this paper, these are called *high radon regions*.

The negative pressure of the dwellings causes air flow from ground to indoors through the gaps in the base floor. However, it is easy to prevent radon ingress from the ground by sealing the base floor and installing a radon piping [1]. The radon piping consists of a perforated drainage pipe installed in the drainage layer below the floor slab, Figure 1. The radon piping is connected to exhaust duct which is led to the roof. The radon piping is naturally ventilated due the temperature differences and wind.

The energy efficiency requirements in the Finnish national building code were considerable tightened in 2010 and 2012. This has led into improving of the air tightness of houses. If the radon prevention is neglected, theoretical

calculations suggest that improved air tightness can cause increased radon concentrations [2].

In this study, radon concentrations and prevalence of radon prevention in Finnish houses were investigated. In addition, the correlation between air tightness and radon concentration was studied.

The previous *2009 survey* focused on houses constructed during 2006–2008 [3]. The average radon concentration in that survey was 95 Bq/m^3 which was 30% lower than that measured in houses constructed during 2000–2005 [4].

Materials and Methods

The study was based on a random sample provided by Population Register Centre. The sample size was 2800 houses, including detached, semi-detached and row houses [5]. The sampling consists of new houses, for which a building permission was given between November 2012 and October 2013 and that were in use by the October 2015.

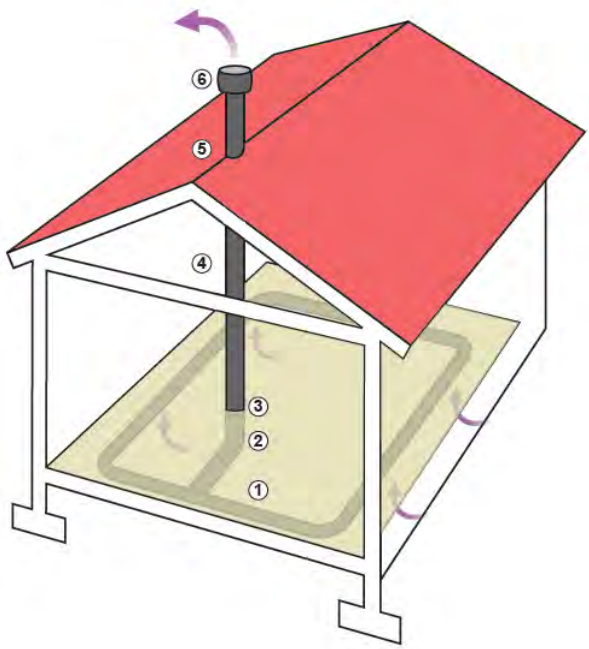

Figure 1. An example of an effective radon prevention in new house. Radon piping is installed below the floor slab. The joint between the floor and wall is also sealed as well as pipe penetrations.

Table 1. The average and median radon concentrations in the high radon regions and elsewhere in the country.

	Average radon concentration Bq/m³			Median radon concentration Bq/m³		
	Detached houses	Row and semi-detached houses	All	Detached houses	Row and semi-detached houses	All
High radon regions	76	89	78	59	55	58
Elsewhere in the country	70	59	69	41	37	41
Whole country	72	65	71	44	39	42
Change in radon concentration (%) compared with the 2009 survey						
	Detached houses	Row and semi-detached houses	All	Detached houses	Row and semi-detached houses	All
High radon regions	–31	–42	–38	–18	–30	–22
Elsewhere in the country	–19	–23	–17	–25	–21	–23
Whole country	–22	–35	–25	–25	–28	–28

The radon measurements were carried out between December 2nd, 2015 and June 8th, 2016. One radon detector per house was used. Typical measurement time was 2–3 months and most of the measurements were started in December 2015. With the radon detector a questionnaire was send to the inhabitant in order to obtain information about the house features and radon prevention.

Since the areal variation of radon concentration is caused by varying radon emanation from the soil, it causes variation to the results. Therefore, *relative radon levels* were also calculated in the analysis of air tightness vs. radon concentration. The relative radon level was calculated for each house by dividing the radon concentration by a reference value, which was median radon concentration of postal code area of the measured house. The reference values were obtained from the STUK's radon measurement data base containing data from 120 000 houses. If there was less than 10 measurements in a postal code area, the median value of the municipality was used.

Results and discussion

The average radon concentration of houses was 71 Bq/m³, Table 1. That is 25% lower value than that in the 2009 survey (95 Bq/m³). Median was 42 Bq/m³ and it was reduced by 26% compared to 2009 survey (58 Bq/m³). In the high radon

regions, radon concentration have reduced 22% and 23% elsewhere in the country compared to 2009 survey.

The reference level of 200 Bq/m^3 exceeded in 6% of houses. This value has reduced 8,1 percentage units in high radon regions and 3,5 percentage units in rest of the country.

In houses with crawling space, radon concentrations were the lowest and all the measured radon concentrations were below 200 Bq/m^3. In houses with basement or semi-basement radon concentrations were the highest. In houses with floor slab on ground, 6% of the measured values were above 200 Bq/m^3 and 11% in houses with basement or semi-basement.

Radon prevention methods had been used in 81% of detached houses that is 27 percent units more than in the 2009 survey. In the high radon regions, radon prevention had been used in almost all the houses both in this survey (98%) and the 2009 survey (92%). Elsewhere in the country, radon prevention had been used in 61% of houses that is 23 percent units more than in the 2009 survey.

The air tightness value was reported for 216 (18%) houses, Figure 2. The average air tightness value was 0.96 h-1 and the median 0.83 h^{-1}.

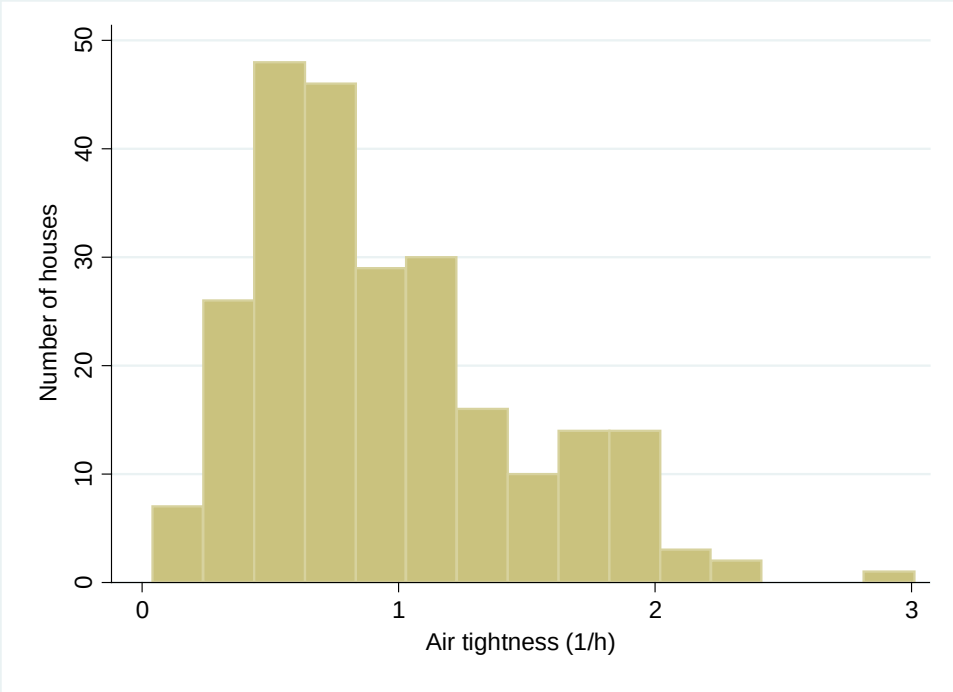

Figure 2. Distribution of air tightness value q50.

Table 2. Radon concentrations by the air tightness classes. Only houses without radon prevention were included.

	Air tightness q_{50}			
	0,04–0,59	**0,60–0,79**	**0,80–1,19**	**1,20–3,01**
Number	14	14	12	17
Average radon concentration (Bq/m³)	45	64	29	31
Median radon concentration (Bq/m³)	32	52	29	20
Median of relative radon level (Bq/m³)	0,53	0,96	0,27	0,17

The data were divided into four categories by the air tightness, Table 2. If all cases (N=216) were included there was no linear correlation between air tightness and measured radon concentration. Similarly there was no linear correlation between air tightness and measured radon concentration if houses with radon prevention were excluded. However, there was a small negative correlation ($r=-0.39$, $p<0.05$) between the air tightness and relative radon level when houses with radon prevention were excluded from the analysis. Figure 3 shows relative radon level as a function of air tightness.

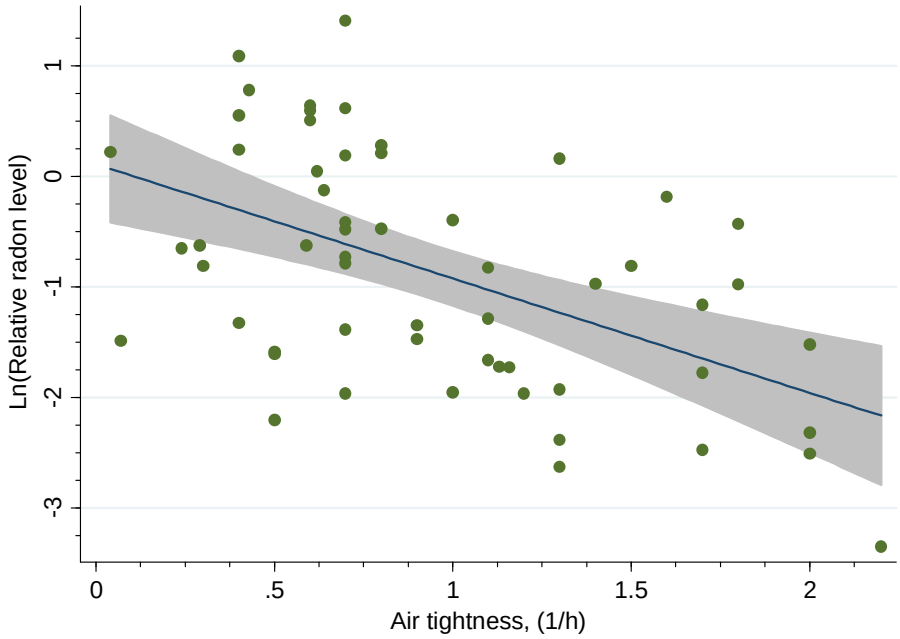

Figure 3. Relative radon level vs. air tightness in the houses without radon prevention (N=57).

Conclusions

The current study shows clearly that radon prevention in new houses is more common than earlier. Therefore, indoor radon concentrations in new houses have reduced. Radon prevention was used in 81% of houses build during 2013–2015. Especially, in the high radon regions, radon prevention was widely used but also elsewhere in the country the radon prevention was used more often than earlier. This has lead to lower radon concentrations: average and median radon concentration in the measured new houses were 71 Bq/m^3 and 42 Bq/m^3, respectively. The concentrations were clearly lower than in the previous survey. The reduction of the radon concentrations was greater in the high radon regions than elsewhere in the country.

The structure of the base floor affects to indoor radon concentrations. The highest radon concentrations were measured in houses with basement or semi-basement. This could indicate that sealing of basement walls in contact with soil was still inadequate. Good results require good design and application of radon prevention. On the other hand, houses with basement or semi-basement may have been build more often on an esker or rock and in these cases, the radon emanation could be higher than on average. The effectiveness of the passive radon piping could also be reduced in the case of blasted rock below the footings.

The air tightness of houses has been improved in recent years due to new energy saving requirements. This could lead to higher negative pressure in houses and therefore higher indoor radon concentrations. In general, indoor radon concentrations have reduced in new houses in Finland. This positive progress are due to several reasons: radon prevention is effective enough to overcome the effect of increased negative pressure; sealing of the base floor has been done more often and more carefully than before; and ventilation has been adjusted better than earlier. However, in the houses without radon prevention, relative radon levels were increased with improving air tightness. This observation suggest that it is increasingly important to use radon prevention in the new low energy houses. It would be worthwhile for the house owner to require thorough radon prevention from designers and workmen and to supervise radon prevention installations carefully.

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On conceptual and modelling uncertainties in biota dose assessments of releases from nuclear waste repositories and other nuclear facilities

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Exposure of also "non-human biota" may result from releases of radioactivity from nuclear facilities. There have been a number of approaches been developed for assessing such exposures, and also the International Commission of Radiological Protection (ICRP) has reasonably recently provided their own (e.g., ICRP 2008, 2014) and incorporated the use of that in their general recommendations applying also to nuclear facilities. Practical implications are currently being studied for example by Working Group 3 of the IAEA MODARIA II programme, as also the International Atomic Energy Agency (IAEA) follows the ICRP recommendations in their safety requirements and guides.

Whilst off-the-shelf assessment procedures, particularly when packaged in a convenient tool (*e.g.*, the ERICA Assessment Tool, Brown *et al.* 2016), are useful for guidance and communication of assessments of the degree of exposures of biota from radioactive releases and to ease the workflow, there are also unavoidably a number of conceptual and modelling uncertainties involved, and those may not be so readily recognised than with customised assessments. Such uncertainties may bear a considerable role in interpretation of the results and compliance with requirements. In this contribution to the symposium, most prominent of these are mapped for their location and type by applying evaluation tools earlier collated by the authors (Ikonen & Kangasniemi 2017).

In general, most of the approaches to assess exposure of biota follow the similar structure: selection of the target organisms, simplification of the geometry and exposure configuration for each target organism, radionuclide transport modelling, dose rate calculations, and effects analysis and/or comparison with benchmark values.

As it is practically impossible to assess every single kind of an organism possibly being exposed, some representatives must be chosen. The ICRP recommends (ICRP 2008, 2014) a system of Reference Animals and Plants (RAPs), being comparable to that of the Reference Man, for the general frame of reference that should be complemented with more specific representative organisms where appropriate. These twelve RAPs bear colloquial names (e.g., "duck") and characteristics derived from a few species, even though they are intended to cover a larger group of biota (close to the taxonomic concept of a Family).

Clearly, such generalisations and selections of the more context-specific representatives involve a considerable degree of scenario and conceptual uncertainties that are largely unquantifiable.

For the dosimetric calculations to translate the radioactivity amount in the environment and within the organisms, model simplifications are needed for practical reasons: the Monte Carlo simulations of the radiation interactions are computationally demanding and the combination of configurations of the (external) radiation source, medium and the receiving organism are practically infinite. Therefore, it is customary to simplify the organisms into an ellipsoidal form and assume a homogeneous distribution of radionuclides within them, as well as the organisms having the same major-elemental composition and density as the medium around. Here, there are many modelling uncertainties arising from various details, but also conceptual uncertainties and issues of communication what comes to the simplified shapes particularly for plants (*e.g.*, Aro & Ikonen 2011); it may be difficult to perceive that an ellipsoidal solid mass would be an appropriate approximation of a complex-shaped grass with roots, culms, leaves and inflorescence even if the attention would be only in the exposure of the reproductive parts (*e.g.*, meristem and buds) and numerically the estimated error would be insignificant. Also, the exposure configuration is often stylised to overestimate the dose (*e.g.*, birds are assumed to constantly lie on the ground). Some information on the potential of alternative assumptions seems to be available from earlier development stages of the assessment methodologies, but such discussion could still be worthwhile at least for some of the details, in benefit of an overall view of the multifaceted uncertainties involved in this kind of assessments.

After the conceptual setting of the stage in terms of the geometries and material properties, the actual calculation of the absorbed dose (rate) – that is presently used to characterise the degree of the exposure almost without an exception – is rather straightforward. Same goes also for the radionuclide transport calculations (*i.e.*, estimating the radionuclide concentrations in the environmental media and in the organisms), that anyway would preferably be undertaken in the framework of the overall assessment (*i.e.*, same modelling for dose assessments for humans and biota). The dosimetry itself aside, the media-to-organism concentration ratios seem to dominate in the significance to the calculation results (*e.g.*, Ikonen et al. 2011), and due to the notoriously large variability in concentration ratios, they do so also for the uncertainties. Of course, these steps in the assessment process involve a number of model and data uncertainties, in addition to those related to correctly identifying and representing scenarios or calculation cases of relevance.

The effects analysis, whether in form of comparing the calculated doses or dose rates directly to empirical radiation effects data or to a benchmark

value, seems to bear the most fundamental conceptual uncertainties, however (in addition to numerous data and model uncertainties). This is since there is a consensus of the level of individuals of biota being too constrictive to be protected, but the higher levels of biological organisation (populations, communities, ecosystems) being more appropriate protection targets (*e.g.*, ICRP 2008, 2014). However, nearly all solid-enough empirical evidence of radiation effects addresses the level of individuals. Thus, methodologies such as species sensitivity distributions or plain expert judgement needs to be used in setting the comparison values, were they common benchmarks (for which there are presently a number of 'competing' candidates) or specific to the assessment. In addition, alternative choices could be made also regarding the effect endpoints (reproduction, mortality, genetic effects, etc.). Even if the recommendations by the ICRP and others help in the choices, these remain important aspects for communication of the assessments and their results to various audiences.

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Uptake of Nickel in boreal heterotrophic bog Pseudomonas strains

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Toxic concentrations of nickel (Ni) can be released into the environment as a result of mining and industrial activities (Nriagu 1990). Various industrial processes can increase Ni mobility resulting in potentially large releases to various environmental compartments through surface and groundwater flows. In addition, the potentially high mobility of the long-lived nickel radioisotope, ^{59}Ni, can present concerns for long-term safety of spent nuclear fuel disposal (Hjerpe et al. 2010). Many environmental microbes have an ability to bind nickel, and various Ni uptake mechanisms, including biosorption on cell wall structures and active bioaccumulation, have been presented (Ledin and Pedersen 1996; Lopez et al. 2000; Mulrooney and Hausinger 2003). In the boreal regions, nutrient-poor bogs provide unique growth environments for distinct microbial populations, but only limited knowledge about their metabolism is available.

In this study, a new *Pseudomonas* sp. strain V4-5-SB was isolated from the boreal Lastensuo bog, identified using 16S rRNA gene sequencing and characterized using e.g. catalase and oxidase activity tests and RapID™ system. The newly isolated *Pseudomonas* V4-5-SB and previously isolated *Pseudomonas* strains PS-0-L and T5-6-I originating from the same bog (Lusa et al. 2016A and 2016B), were used in the following Ni removal tests. The ability and efficiency of these bacteria to remove Ni from solution was examined using batch experiments (Ni $6{\cdot}10^{-10}$ mol/L; in 1% Tryptone and 1% Yeast extract; at 4°C and 20°C; with incubation time 7 or 14 days; at pH range 6–8) and sorption isotherms (Ni 10^{-10}…10^{-3} mol/L; in 0.1 mol/L NaCl + 0.01 mol/L MOPS or TRIS buffer; at pH 6 and 8). In addition, the morphology of the isolated bacteria before and after incubation with $8{\cdot}10^{-4}$ mol/L Ni was examined using transmission electron microscopy (TEM).

All studied *Pseudomonas* strains removed Ni from the 1% Tryptone and 1% Yeast extract broths and the uptake efficiency was affected by nutrient source, incubation temperature and time (Figure 1). The highest Ni uptake ability was shown by *Pseudomonas* PS-0-L in 1 % Tryptone (20°C, 7 d), with a

maximum K_d value of 1 890 L/kg DW. Compared to other *Pseudomonas* V4-5-SB (max K_d 640 L/kg DW) and T5-6-I (max K_d 420 L/kg DW), Ni uptake was three times higher in *Pseudomonas* PS-0-L. All used *Pseudomonas* strains differed in their substrate utilization patterns examined using RapID™ tests. This presumably affects also their ability to remove Ni under variable nutritional conditions. Generally, the Pseudomonas sp. strains used in this study preferred 1% Tryptone as their carbon source, which was reflected as stronger bacterial growth and higher nickel removal, compared to 1% Yeast extract. In addition, Ni uptake was observed to increase as incubation temperature increased from +4°C to +20°C, which may be explained by weaker growth and uptake ability of these strains under lower temperature. Incubation time had a minor role in Ni removal by all studied Pseudomonas strains, especially in 1% Tryptone, in which a fast initial uptake was observed (Figure 1 A). In 1 % Yeast extract, the uptake rate was somewhat decreased, compared to 1 % Tryptone solution, especially under lower temperature (+4°C) and in V4-5-SB and T5-6-I strains (Figure 1 B).

Sorption isotherms of Pseudomonas V4-5-SB and PS-0-L showed a slight deflection at both pH 6 and 8, resulting either from A) the saturation of the exchange sites or B) from the retention of Ni on multiple sorption sites or C) from competition between different ions (e.g. Na^+) and Ni in the solution (Figure 2 B). It has previously been shown that no Ni sorption on TRIS or MOPS compounds occurred (Perrin and Dempsey 1974). On the other hand, variation in the solution pH did not significantly influence Ni removal by Pseudomonas V4-5-SB, PS-0-L or T5-6-I (Figure 2 A). As energy-independent interaction mechanisms (principally straight biosorption on cell wall functional groups) would be

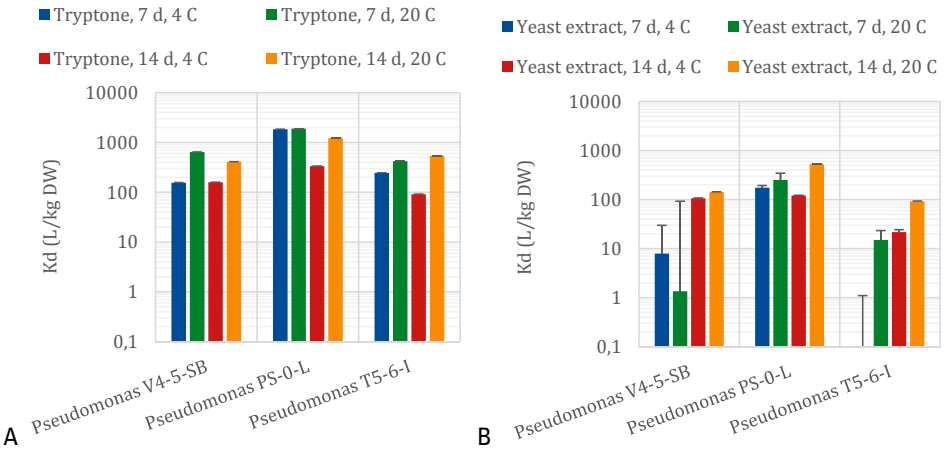

Figure 1. Uptake of nickel (6·10-10 mol/l) by Pseudomonas PS-0-L, V4-5-SB and T5-6-I in 1% Tryptone and 1% Yeast extract at +4°C and +20°C with 7 and 14 d incubation time.

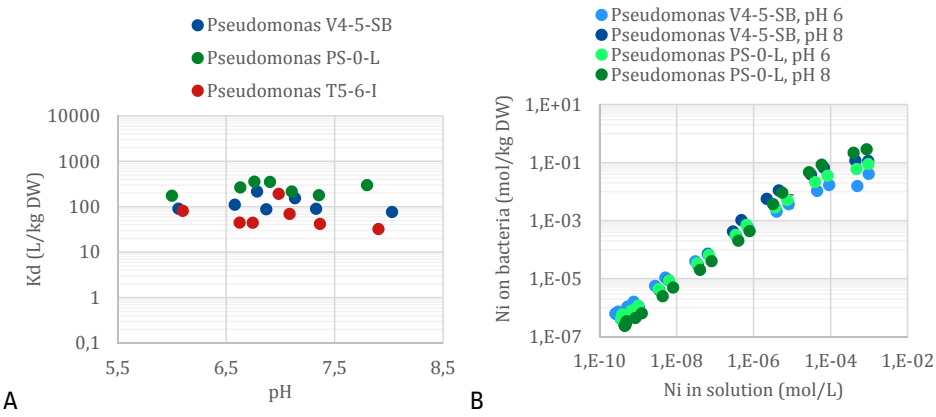

Figure 2. A: Uptake of nickel (6·10-10 mol/L) by Pseudomonas V4-5-SB, PS-0-L and T5-6-I in 1% Tryptone at +20°C with 7 d incubation time at pH range 6–8. **B:** Sorption isotherms of Ni (10^{-10}…10^{-3} mol/L) by Pseudomonas V4-5-SB and PS-0-L in 0.1 mol/L NaCl +0.01 mol/L MOPS or TRIS buffer at pH 6 and 8.

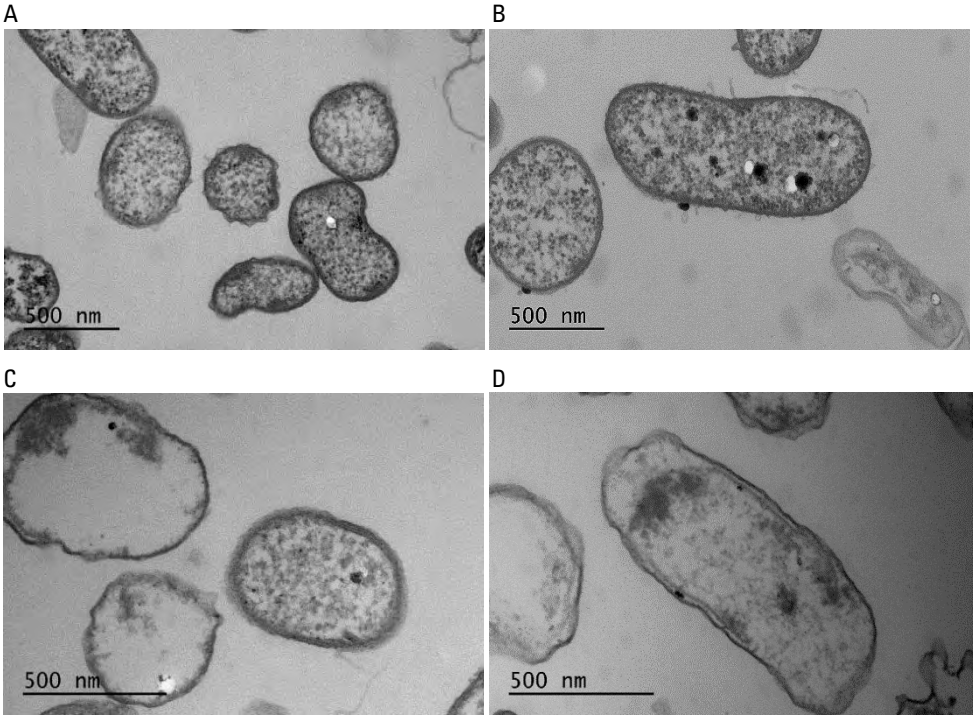

Figure 3. TEM images (10000x) of Pseudomonas PS-0-L incubated **A** without nickel and **B** with nickel ($8·10^{-4}$ mol/L). Respectively, TEM images (15000x) of Pseudomonas V4-5-SB **C** without and **D** with Ni. In B dense probable Ni accumulations are seen.

affected by pH, this observation possibly results from Ni cellular bioaccumulation or from the complexing metabolites secreted by these bacterial strains.

In the morphological TEM images of Pseudomonas PS-0-L after incubation with $8{\cdot}10^{-4}$ mol/L Ni solution, dense crystalline accumulates, suggesting that Ni affects the cellular processes and is probably accumulated into the cell, were observed (Figure 3 A and B). A similar nickel uptake to the bacterial cell was not detected by Pseudomonas V4-5-SB (Figure 3 C and D). However, further analysis is still needed for more detailed elemental analysis of these accumulates.

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Environmental radiation survey prior to FiR1 research reactor decommissioning

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Introduction

FiR1 TRIGA Mark II type research reactor by General Atomics (US) in Otaniemi, Espoo, was the first nuclear reactor in Finland. Its initial power in 1962 was 100 kW until it was raised to 250 kW in 1967. The early operations included intensive neutron beam research, activation analyses and isotope production. In the 1990s, a facility for Boron Neutron Capture Therapy (BNCT) was constructed to the reactor and between 1997 and 2012 over 200 cancer patients were treated. During the over 50 years of operation, FiR1 has had a key role in Finland's nuclear energy program and in training of nuclear energy and technology professionals. After the final shutdown in 30th of June 2015, FiR1 will also be the first reactor to be decommissioned in Finland.

Environmental aspects are an important part of the FiR1 decommissioning project. The project was started with an Environmental Impact Assessment in 2013–2015. The documentation is publicly available at the web pages of Ministry of economic affairs and employment [1]. The conclusions showed that decommissioning of FiR1 will not cause a significant impact on the environment. However, in accident situations there is potential for release of radionuclides, and the effects have been discussed in reference 2. In addition to the general description of the measures to restrict the burden caused by the nuclear facility on the environment (Appendix 6 of VTT's decommissioning license application [3]), an environmental radiation survey will be given to STUK as a supporting material. Also, a plan for the environmental monitoring during the decommissioning will be prepared and the environmental radiation survey will be repeated after the decommissioning is finished.

Environmental sampling during operation

During the years of operation, there were two possible pathways for contaminants to migrate from the reactor building to the environment. First pathway was via waste water originating from hand washing. The waste water was collected into holding tanks and it was always measured for radioactivity prior to

release into Ossinlampi pond (Picture 1). The waste waters released during the last 20 years did not contain measurable amounts of artificial radionuclides such as Mn-54, Co-60, Cs-134 and Cs-137. In order to verify the environmental impact via the waste water, water and sediment samples were regularly collected from Ossinlampi pond [4]. The results showed that the water samples did not contain artificial radionuclides, but sediment samples contained Cs-137 originating from Chernobyl nuclear power plant accident.

Second pathway for the contaminants to migrate from the reactor building to the environment was via air ventilation. However, the air was always monitored and only gaseous Ar-41 (activation product of air) was released during the operation hours [5]. Therefore, it can be concluded that the environmental impact of FiR1 regarding release of radionuclides has been negligible.

Selection of sampling sites and sample types

The planning of the environmental radiation survey was closely linked with the unique surroundings of FiR1 (Picture 1) [6]. Otaniemi is a peninsula extending into the shallow bay of Laajalahti, including a nature reserve, a university campus, student residences, small businesses, companies, greeneries, and small

Picture 1. Map of Otaniemi. FiR1 reactor is marked as a red dot. Green line shows the nature reserve and blue lines planned areas with significant changes in the infrastructure.

Table 1. Environmental radiation survey plan.

Target	Sampling site*	Number of samples	Analysis
Aerosols	4	Four air filters – during the survey the air filters will be changed every 2 weeks during 2 months period	Gamma nuclides (especially ^{60}Co and ^{137}Cs)
Soil	2, 3, 5, 6	Four top soils	Gamma nuclides (especially ^{60}Co and ^{137}Cs)
Plant indicator: moss	4	One moss sample from the roof of nearby building	Gamma nuclides (especially ^{60}Co and ^{137}Cs)
Plant indicator: fern	2, 5, 6	Three fern samples	Gamma nuclides (especially ^{60}Co and ^{137}Cs)
Water	1, 3, 4	Three water samples • Laajalahti sea water • Ossinlampi pond water • Water from the roof of the reactor building	Gamma nuclides (especially ^{60}Co and ^{137}Cs) and ^{3}H
Sediment	3	One top sediment sample from Ossinlampi	Gamma nuclides (especially ^{60}Co and ^{137}Cs)

* See Picture 2

forests. Additionally, large area of Otaniemi was under construction during the sampling since new infrastructure was being built. City planning maps also showed that new construction sites will change the landscape significantly in the near future [7]. All this information was considered in order to select sampling sites, which would be accessible also after several years when FiR1 has been decommissioned and a final environmental radiation survey needs to be made.

Second step in the planning phase was the selection of sample types. As a continuation to historical environmental sampling, sediment and water samples from Ossinlampi pond were selected. Water samples were also planned to be collected from Laajalahti and reactor roof. An air filter sampler (Hunter JL-150 air sampler by Senya) was purchased and placed on the roof of the nearby building. A plant survey was needed for the selection of plant indicators and sampling sites for plants and soil. Therefore, a plant survey was carried out during spring and summer of 2017. The survey included mapping of existing plants, especially moss and fern, which are good plant indicators as recommended by STUK. Main focus in the survey was given to the 0.5 km radius from the reactor. The results showed that the small forests contained large areas of lily of the valleys and wood anemones and patches of fern, fireweed, cowberry, and nettle. Also some small patches of moss was growing on top of big rocks.

After the examination of the map and the plant survey, a plan for the sample types, sampling sites and analyses was suggested to STUK. Main concepts of the plan are given in Table 1 and Picture 2. Due to the activation of the reactor components, one of the main gamma radionuclide, which could be released to the environment during the dismantling, is ^{60}Co. On the other hand, a representative fission product emitting energetic gamma is ^{137}Cs, the presence of which would indicate leaking of the reactor fuel. Therefore, these two gamma nuclides were chosen to be the main artificial radionuclides of interest. Additionally, ^{60}Co forms stable complexes especially with organic matter and does not transfer to plants effectively [8]. ^{137}Cs forms stable complexes with organic matter, but especially strong complexes with clay [8]. ^{137}Cs also transfers to plants [8]. Therefore, focus on top soil was chosen. ^{3}H was chosen as an indicator for betas because it is one of the main activation radionuclides and it easily migrates together with water in the biosphere [8].

Picture 2. Planned sampling sites. 1) Water sample, 2) Plant and soil samples, 3) Sediment, water, and soil samples, 4) Plant, air filter and water samples, 5) Plant and soil samples, 6) Plant and soil samples. Green line shows the nature reserve and blue lines planned areas with significant changes in the infrastructure (compare with picture 1). FiR1 is marked as a red dot.

Table 2. Results of the environmental radiation survey.

Target	Sampling site*	ID number	Gamma nuclides	Beta nuclides
Air filters	4	13	$^{60}Co < 8{,}1 \cdot 10^{-6}$ Bq/m³ $^{137}Cs < 6{,}7 \cdot 10^{-6}$ Bq/m³	–
		14	$^{60}Co < 8{,}0 \cdot 10^{-6}$ Bq/m³ $^{137}Cs < 6{,}6 \cdot 10^{-6}$ Bq/m³	–
		15	$^{60}Co < 8{,}1 \cdot 10^{-6}$ Bq/m³ $^{137}Cs < 6{,}6 \cdot 10^{-7}$ Bq/m³	–
		16	$^{60}Co < 9{,}3 \cdot 10^{-6}$ Bq/m³ $^{137}Cs < 6{,}7 \cdot 10^{-7}$ Bq/m³	–
Soil (top soil)	2	6	$^{60}Co < 0{,}007$ Bq/g $^{137}Cs = 0{,}08 \pm 0{,}01$ Bq/g	–
	3	9	$^{60}Co < 0{,}01$ Bq/g $^{137}Cs = 0{,}015 \pm 0{,}002$ Bq/g	–
	5	10	$^{60}Co < 0{,}007$ Bq/g $^{137}Cs = 0{,}020 \pm 0{,}002$ Bq/g	–
	6	11	$^{60}Co < 0{,}01$ Bq/g $^{137}Cs = 0{,}033 \pm 0{,}004$ Bq/g	–
Plant: Moss	4	4	$^{60}Co < 0{,}03$ Bq/g $^{137}Cs = 0{,}05 \pm 0{,}01$ Bq/g	–
Plant: Fern	2	1	$^{60}Co < 0{,}04$ Bq/g $^{137}Cs < 0{,}03$ Bq/g	–
	5	2	$^{60}Co < 0{,}04$ Bq/g $^{137}Cs < 0{,}03$ Bq/g	–
	6	3	$^{60}Co < 0{,}04$ Bq/g $^{137}Cs < 0{,}03$ Bq/g	–
Water	1	5	$^{60}Co < 0{,}009$ Bq/g $^{137}Cs < 0{,}008$ Bq/g	$^{3}H < 0{,}03$ Bq/ml
	3	8	$^{60}Co < 0{,}009$ Bq/g $^{137}Cs < 0{,}007$ Bq/g	$^{3}H < 0{,}03$ Bq/ml
	4	12	$^{60}Co < 0{,}009$ Bq/g $^{137}Cs < 0{,}008$ Bq/g	$^{3}H < 0{,}03$ Bq/ml
Sediment	3	7	$^{60}Co < 0{,}013$ Bq/g $^{137}Cs = 0{,}040 \pm 0{,}004$ Bq/g	–

*See Picture 2

Sampling campaigns and measurements

After the approval of the sampling plan by STUK, sampling campaigns were executed during September–December 2017. A spade was used for the collection of soil samples, shears were used for the collection of plants, water samples were poured into plastic vials, and sediment samples were collected by hand. Air filters, soil, plant and sediment samples were let to dry at room temperature before packing into measurement vials. 250 ml Nalgene vials were used for water, soil and plant samples. Air filters were carefully folded into petri dishes in order to obtain consistent and efficient geometry.

Gamma activities were determined using ISOCS gamma spectrometer with an HPGe detector (BE2020). Geometry calibrations were carried out with Canberra's Geometry Composer program version 4.4.

Beta activity concentrations in the water samples were also measured using Wallac 1415 liquid scintillation counter.

Results and conclusions

The environmental radiation survey results are shown in Table 2. Based on the results, it can be concluded that the measured samples did not contain significant amounts of artificial radionuclides ^{3}H, ^{60}Co and ^{137}Cs, activity concentrations being mainly below the detection limits. However, soil, sediment and moss samples contained small amounts of ^{137}Cs (15–80 mBq/g), which most likely originates from Chernobyl fallout.

The obtained results give a base line for the current status of artificial radionuclides in the FiR1 surroundings and it can be concluded that FiR1 has not caused radioactive contamination in the environment. However, the risk of contamination will be increase during the dismantling phase but the risk will be minimised by efficient radiation protection. For example, all outgoing air will be filtered through HEPA filters, which will be changed regularly and measured for gamma radiation. Also the air filter sampler will be in operation during the dismantling phase in order to have an additional verification point. When the decommissioning will be finished in 2020s, final environmental radiation survey will be carried out.

Acknowledgements

Authors would like to thank colleagues at VTT, namely Jori Helin, Perttu Kivelä and Antti Räty, for their assistance in execution of the environmental radiation survey.

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Fate of radionuclide ^{14}C in soil-plant-atmosphere continuum: on the potential uptake of soil ^{14}C into plants

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Radioactive waste is generated at all stages of the nuclear fuel cycle. Finland and many other countries support deep geologic disposal as the best method for isolating highly radioactive, long-lived waste. However, there is still risk that release might take place and pose a threat to health or the environment. Detailed waste management studies and knowledge about the long-term environmental behavior and migration of radionuclides are needed to guarantee the safety of geological disposal of radioactive waste. One of the radionuclides that have been identified as being potentially significant in terms of release from deep geological disposal is radiocarbon (^{14}C). It has a high potential to escape as carbon dioxide (CO_2) from radioactive waste repositories, and there are risks that it is taken up by plants and thus incorporated into the food chain. However, the exact amount of ^{14}C possibly taken up through plant roots or assimilated within the sub-canopy is largely unknown.

We investigated the potential transfer of soil-derived C into two plant species (reed canary grass, *Phalaris arundinacea* and Scots pine, *Pinus sylvestris*) by using a new approach to separate soil-derived carbon from that originating from the atmosphere. The principal idea of this approach is to study uptake of soil carbon into plants in cultivated cutaway peatlands, where a distinct natural ^{14}C pattern exists (very large difference in ^{14}C content between modern plants with "normal" ^{14}C levels and the up to 8000-year-old leftover peat strongly depleted in ^{14}C). Samples of soil, plants (roots and leaves) and CO_2 in the canopy were collected and analyzed for ^{14}C content by accelerator mass spectrometer (AMS). Two-pool isotope mixing model was applied to calculate the relative contribution of atmospheric C and soil-derived C in the plants. Both field and laboratory studies under controlled conditions were conducted.

Although majority of plant C was obtained from atmosphere by photosynthesis, our study suggested significant uptake of soil-derived C into roots, particularly pine roots. The proportion of soil-derived C was higher in pine roots than in RCG roots (4.67% vs 2.01%), which might be related to the high mycorrhizal colonization rate in pine roots. Carbon transfer from the mycorrhizal fungus to the host has been suggested by others, though soil sources have not yet been identified. Likely, dark CO_2 fixation by heterotrophs plays a role. Furthermore, the results showed that, despite of abundant soil-derived C being available in plant canopy air for assimilation, no trace of soil-derived carbon was found in the aboveground parts of the plants. This may have been affected by the fact that the plants canopy was not fully closed during assimilation of C from the atmosphere. Further work is needed in other ecosystems with different plants and soil types to investigate the transfer of 14C and contribution of soil-derived C to ascertain to what extent these results are generalizable.

Determination of ^{90}Sr and Plutonium in environmental samples from drinking water abstraction areas in Northern Germany

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Water as an essential resource for life is exposed to different environmental pollutants originating from human activities. Liquid and gaseous releases from nuclear facilities, especially from nuclear fuel reprocessing plants, in permissible amounts result in a continuous accumulation of radionuclides in the environment. Radionuclides washed out from the atmosphere by precipitation may infiltrate the soil and enter the groundwater. Within the framework of the project 'TransAqua', which was supported by the German Federal Ministry of Education and Research (contract number 02NUK030), the sensitivity of a drinking water reservoir was investigated with regard to input of man-made radionuclides. Environmental samples (ground water, surface water, soil) were collected in the "Fuhrberger Field" catchment, situated about 30 km northeast of the city of Hannover. The catchment covers 300 km2 and its unconfined aquifer extends within 20–30 m thick Quaternary sediments. Gleysols, gleyic Podzols, and Podzols are the dominant soil types [1]. Soils are highly permeable with low cation exchange capacity.

Main part of this work was the development of a new chemical sample preparation method for very low strontium- and plutonium concentrations, which allows the determination of both elements from the same sample volume. The method was adapted to the chosen analytical techniques of Liquid Scintillation Counting (*LSC*) for ^{90}Sr and Accelerator Mass Spectrometry (*AMS*) for the plutonium isotopes ^{239}Pu and ^{240}Pu. Figure 1 shows the developed preparation method from the raw water sample to the quantification of radionuclides.

Although the techniques have low limits of detection, the sample must be pre-concentrated from 40L to 1L, which causes high salinities. Subsequently, well-defined amounts of the yield tracers ^{85}Sr and ^{242}Pu are added to the solution, to monitor process losses. Separating plutonium from strontium is based on $Fe(OH)_3$-coprecipitation, while 99% of Sr^{2+} remain in solution. Due to the similar behaviour of Sr^{2+} and Ca^{2+} as chemical homologues, problems with the

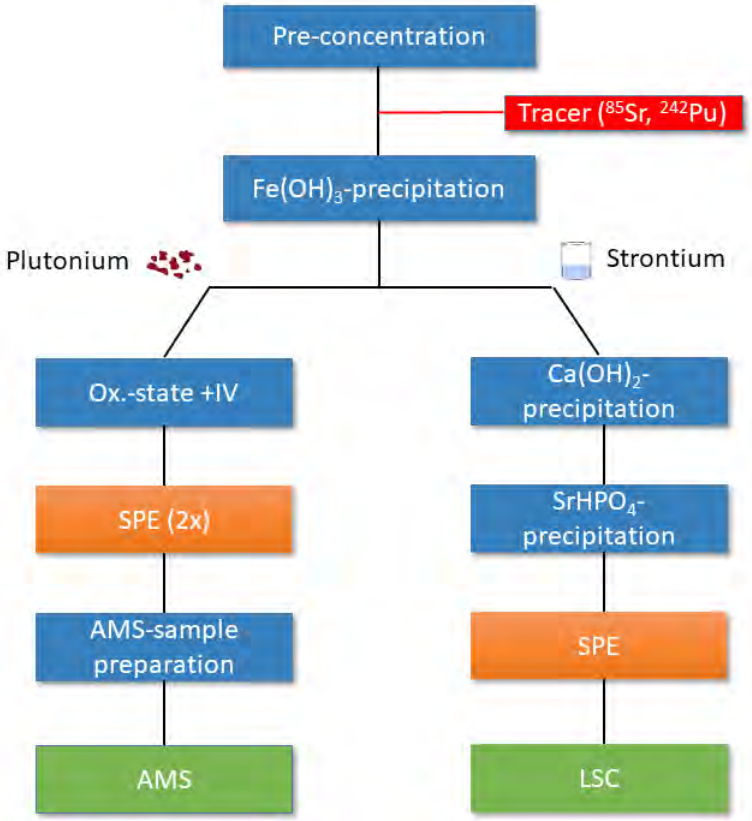

Figure 1. Flowchart of the sample preparation method.

solid-phase-extraction (*SPE*) appear (TRISKEM Sr-resin). The cavity of the crown ether used as extraction agent also selects Ca^{2+} in competition to Sr^{2+}, which results in decrease of Sr-yields to nearly 0%, depending on Ca^{2+} concentrations [2]. Unfortunately, Ca/Sr separation methods are very rare. Patti and Hernandez precipitated Ca^{2+} selective as $Ca(OH)_2$ in basic aqueous solution [3]. This method is also used here, with the side effect that the amount of Ca^{2+} remaining dissolved in solution is sufficient for co-precipitation of Sr^{2+} with Ca^{2+} as Ca/$SrHPO_4$. Residual matrix components in the precipitate can be removed by Sr-solid-phase extraction. The purified Sr-solution is prepared for LSC and stored for three weeks to reach equilibrium of ^{90}Sr and ^{90}Y. After 24h of measurement, chemical yield and ^{90}Sr activities can be calculated from one spectrum simultaneously by the 2-window-method with ^{85}Sr (window 1) and ^{90}Sr+^{90}Y (in window 2), like shown in Figure 2.

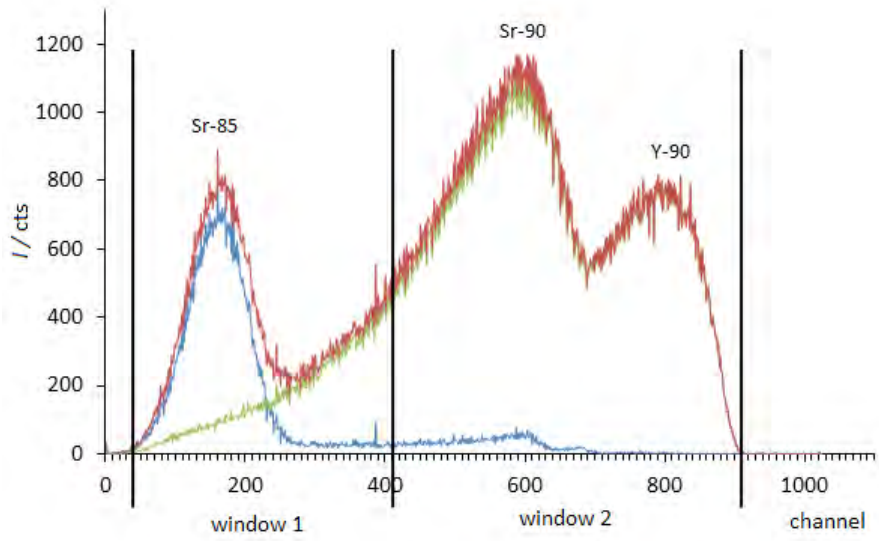

Figure 2. Measurements of yield tracer ^{85}Sr (blue), analyte ^{90}Sr in equilibrium with ^{90}Y (green) and both measured simultaneously (red) [4].

The plutonium co-precipitated with $Fe(OH)_3$ is prepared for solid-phase-extraction by adjusting the oxidation state to +IV, which is necessary to form stable complexes with the extraction agent (Triskem TEVA resin). To avoid mass-spectrometric interferences of ^{239}Pu and $^{238}U^{1}H$, uranium is eluted from the column during extraction. The purified Pu-fraction is co-precipitated with $Fe(OH)_3$ again, and subsequently converted into iron oxide in the muffle furnace at high temperatures. The oxide is mixed with elemental Niob powder and pressed into a titanium holder. AMS measurements are performed by LIP (Laboratory of Ion Beam Physics, ETH Zurich). The method can also be used for soil samples. Due to the lower Ca-content, the calcium-removing reaction steps are skipped. After spiking with yield tracers and digesting the soil by hydrofluoric acid, plutonium-SPE is performed. The Sr-containing fraction is then prepared for the Sr extraction. The development of the method was monitored by yield tracers, which could be recovered satisfactorily. γ-spectrometric measurements of ^{85}Sr showed 87% process yield, α-spectrometric measurements of ^{242}Pu showed 65% process yield. The method outlined above was also tested with water samples from the drinking water abstraction area and resulted in

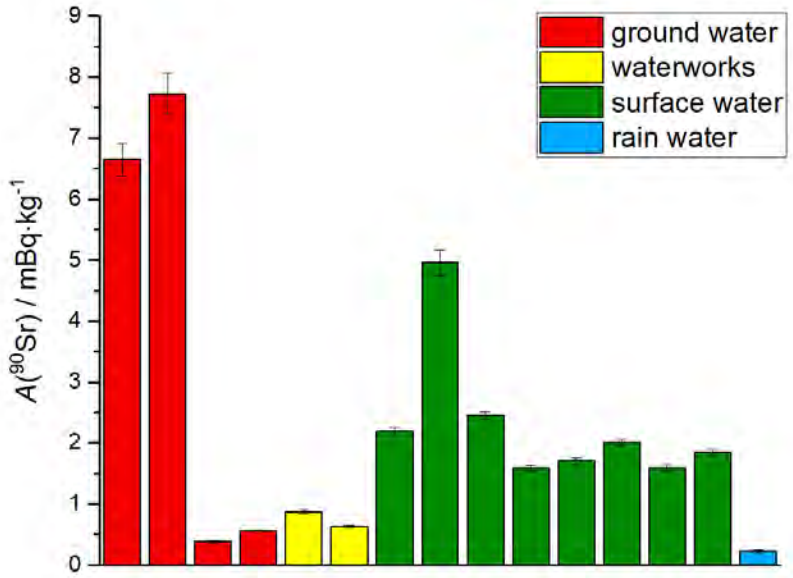

Figure 3. Mass related ^{90}Sr activities of the water samples.

good chemical yields. Figure 3 categorizes the water samples according to their origin and shows the calculated mass related ^{90}Sr-activities.

The highest ^{90}Sr content (7,7 mBq/kg) was found in one of the ground water samples, the lowest content in the rainwater sample. Due to the inhomogeneity of the aquifer, the 90Sr activities of the groundwater samples vary by an order of magnitude. Both water samples from the waterworks (before and after the raw water treatment) show similar activities below 1 mBq/kg. The surface water samples span a range between 1,5 mBq/kg and 5 mBq/kg. As expected, results of the measured soil samples of the upper soil layer covering the groundwater exhibited values for mass related activities ranging two orders of magnitude higher than those of the water samples. They were in the range of 370 mBq/kg to 925 mBq/kg.

Despite the low detection limits of the AMS, plutonium could only be detected in a few samples. The isotopic ratios, also measured by AMS, of $^{240}Pu/^{239}Pu$ = 0,18 suggest plutonium originating from weapons fallout. The highest Pu-activities (1–5 Bq/kg) were found in the organic litter layers. Below this layer, nearly no plutonium could be detected, which suggests the conclusion, that plutonium primarily is sorbed to the organic matter in the litter layer and does not reach the aquifer water for this reason.

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European joint programme for radiation protection research

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The "European Joint Programme for the Integration of Radiation Protection Research – CONCERT" under Horizon2020 is operating as an umbrella structure for the research initiatives jointly launched by the radiation protection research platforms MELODI, ALLIANCE, NERIS and EURADOS. CONCERT is a co-fund action that aims at attracting and pooling national research efforts with European ones in order to make better use of public R&D resources and to tackle common European challenges in key areas of radiation protection research.

CONCERT strives for a better integration of the radiation protection scientific community at the EU level, leading to a better coordination of research efforts and the provision of more consolidated and robust science based policy recommendations to decision makers in this area. In the long-term, these efforts will translate into additional or improved practical measures in view of the effective protection of people and the environment. In view of the effective protection of people and the environment, CONCERT aims at establishing a joint programme for the radiation protection research in Europe and thus to create synergy effects based on the strategic programmes of the European research platforms. Currently MELODI (radiation effects and risks in the low dose range), ALLIANCE (radioecology), NERIS (nuclear and radiological emergency response), EURADOS (dosimetry) as well as EURAMED (radiation protection in medicine) belong to the platforms committed to the radiation protection research in Europe. The aim is to improve radiation protection in Europe, answer open questions, reduce scientific uncertainties and provide scientific support for the implementation of the Euratom Basic Safety Standards in national regulations.

The CONCERT Co-fund action was granted five years and started its work in June 2015. The Federal Office of Radiation Protection (BfS) in Germany coordinates CONCERT. CONCERT interlinks Europe-wide research

in all fields of application of ionizing radiation. 69 partner institutions from almost all EU countries plus Norway and Switzerland have joined forces to combine their expertise and research activities in order to improve radiation protection. In Finland, STUK and University of Eastern Finland (UEF) are partners in CONCERT. UEF is leading the work package on Strategic Research Agenda development and integration.

CONCERT was running two open research calls to strengthen the scientific research in the strategic priority research areas of radiation protection. CONCERT committed 17.1 MEUR funding for the open research calls. The EU contribution is limited to 70% and the remaining 30% of the project costs are covered either by in-kind contributions or by cash funding provided by the CONCERT partners. Among the 37 proposals that were submitted to the two open calls in 2016 and 2017, nine have been selected for funding by the international peer review panel.

Molecular modelling approach to the separation of rare earth elements using phyllosilicates

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Rare-earth elements (REE) consist of the fifteen chemical elements of lanthanide series together with scandium and yttrium. Scandium and yttrium are considered rare-earth elements because they tend to occur in the same ore deposits as the lanthanides and exhibit similar chemical properties. For REEs it is typical, that they consist of several isotopes, the most of which are radioactive. Despite their name, REEs (except the radioactive prometheum) are rather plentiful in the Earth's crust and they are significantly more abundant than for example silver or gold. However, their separation from each other is difficult, because they tend to occur together in nature. On the other hand, REEs are needed in many modern innovations such as lasers, hybrid engines and laptop hard drives [8]. Their demand is continuously increasing, while growing the production is difficult. Thus, there is demand for new resources and recycling of REE's.

Generally, the production of REEs can be divided into five phases: 1) extracting the REE containing material (for example an ore), 2) concentration of the REE content from low levels to about 60-70 %, 3) purifying the material to generate a REE containing mixture, 4) separating REEs or their fractions from the purified mixture and 5) refining into REE compounds or metals. [9] However, this mining and processing produces significant amounts of wastes which can be used as a secondary source for REE extraction. This work considers the use of molecular modelling methods in the separation of REEs using phyllosilicates (sheet silicates).

Most REEs have good sorption properties, and they tend to adsorb onto the alumino-silicate clays, from which they can be more easily processed than by traditional mining of REEs from for example Bastnaesite and Monazite minerals. The alumino-silicate clays are typically 2:1 -layered smectites where the layers are formed of two tetrahedral (T) silicate sheets sandwiching an octahedral (O) aluminium sheet. The resulting negatively charged TOT layers

stack together with a space for water and charge compensating cations between the layers, the interlayer. Smectites have the ability to adsorb water into the interlayer and to change their cationic composition. [1, 2] The capability for cation exchange reactions makes it possible to enrich REEs from smectites. The properties of smectites are under extensive study in connection to mining industry, oil drilling industry, fluid filtration, catalytic processes [3] and the disposal of spent nuclear fuel [4].

An example of the smectites is montmorillonite which is a fine grained mineral: approximately 100 nm wide TOT layers form stacks which pack together to form grains of which the bulk material is composed. The structure from nanoscale to microscale is illustrated in Figure 1. Montmorillonite's key properties are to swell when water is transported into the interlayer (swelling can be to several times the original volume) and to act as a sorbent material e.g. binding a possibly released radionuclide. In addition to interlayer cation exchange, sorption can occur in the basal and edge surfaces of the mineral stacks. These properties are expected to derive from the mineral's nano level structure like chemical composition, type and number of defect sites, layer charge and the type of interlayer cations. The surrounding material (e.g. water) and its chemical composition, pressure and temperature also have an effect on the mineral's properties. An example of the effect of interlayer cation species is shown in Figure 2 where the two dimensional self-diffusion coefficients for interlayer water and cations of Na- and Ca-montmorillonites are plotted (results from our previous work [5]). In the presence of monovalent Na the diffusion coefficients are larger than in the presence of divalent Ca meaning that transport is faster in the Na-montmorillonite interlayer.

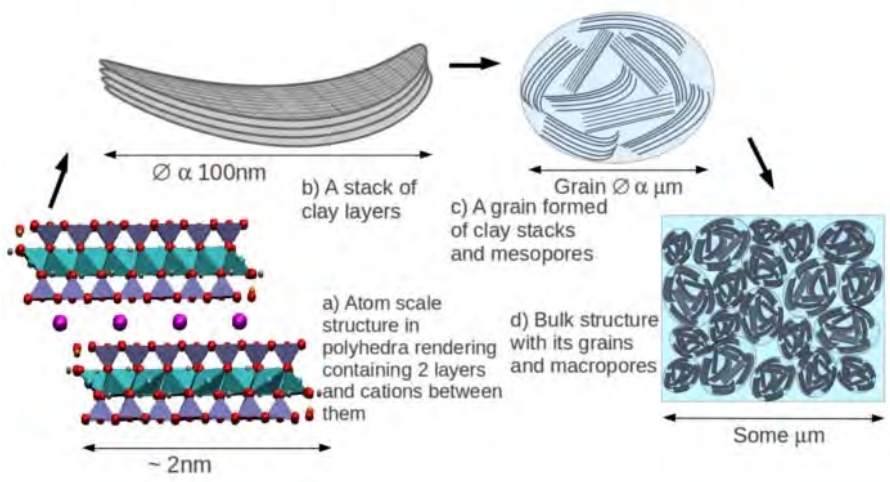

Figure 1. The structure of montmorillonite from nanoscale to microscale.

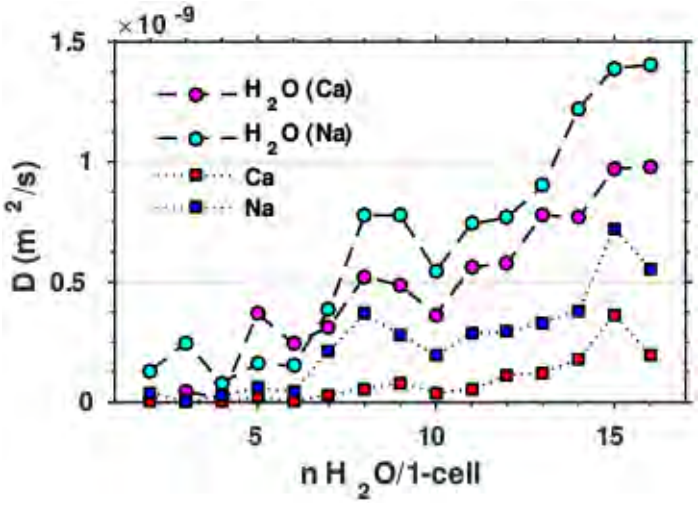

Figure 2. Two dimensional self diffusion coefficients of water and cations in the interlayers of Na- and Ca-montmorillonites.

Molecular dynamics simulations produce the time evolution of a system by solving Newton's equations of motion. This method can be applied to investigate the transport behaviour of different REE species in the montmorillonite interlayers. Additionally, it is possible to perform these simulations using different compositions of the mineral (layer charge, water content, cationic composition) to further the understanding of the REE behaviour in different environments.

In addition to the molecular dynamics studies, we have utilized quantum mechanics to study specific interactions between montmorillonite and cations. In Figure 3, there is an example of the sorption of the Eu^{3+} ion onto the TOT

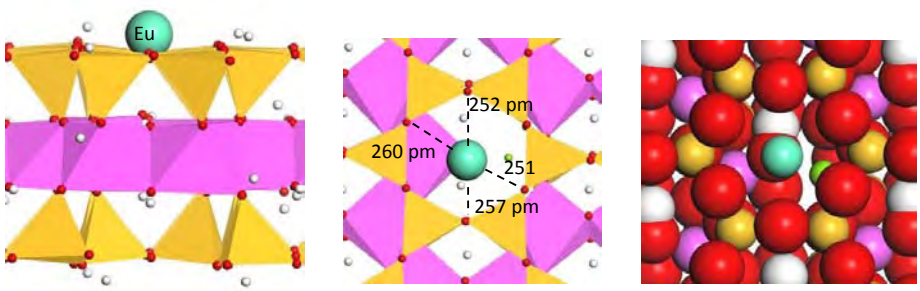

Figure 3. Sorption of Eu^{3+} ion onto the TOT layer of the montmorillonite. A side view in the left, a top view in the middle, and a top view with van der Waals spheres in the right.

layer of montmorillonite. The calculations were performed using quantum mechanics DMol3 code included into Materials Studio version 8.0 [6]. In the calculations, the total electronic energy and overall electronic density distribution are solved in order to define the energetically stable [7] montmorillonite-Eu^{3+} structures. The study indicated that Eu^{3+} ion coordinates in the middle of Si-O ring on the tetrahedral sheet of montmorillonite. Corresponding sorption studies are also possible to other REEs.

Conclusions

The impact of this research is to assist in the development of new technologies to extract and recycle REE elements from mining and other waste streams by exploiting clays. Processes are assumed to decrease the usage of harmful chemicals and energy consumption while also increasing the recovery efficiency. This is an excellent example of a sustainable approach to the world's resources. Together, these represent growth in production with a smaller price and lower environmental impact.

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En route towards mass spectrometric imaging on plant parts: First DESI MS measurements

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Motivation and introduction to DESI MS

As daughter of ^{241}Pu ($T_{1/2}$ = 14.35 a), ^{241}Am is currently the dominating α-emitter in the Chernobyl exclusion zone due to the nuclear accident. On the one hand, this nuclide can end up in human food via contaminated soil and the transfer into plant material. On the other hand, plants can be used for phytoextraction to clean contaminated soil areas. In both cases understanding the transfer from radionuclides into plants is crucial for a better understanding of the environmental behaviour of this radionuclide [1,2].

An important function is the determination of the transfer factor for the radionuclide uptake into plants. The major impact factors are the physicochemical properties of the radionuclide, the plant species, the soil properties, the plant growing technology and the time after contamination. For the determination of the transfer factor, it is important to know the exact concentration of the radionuclide in soil and in plants afterwards. The analytical method of choice is usually ICP MS after an extended sample preparation [3,4].

Furthermore, the speciation in plant parts could give important indications on the environmental behaviour of radionuclides. To this end, TRLFS (Time-resolved laser fluorescence spectroscopy) is applied for fluorescence active elements like europium, americium and curium [5]. The Desorption Electrospray Ionization Mass Spectrometry (DESI MS) used here, shall give a spatially resolved mass spectrum of a sample with easy sample preparation in order to realize a quick radionuclide speciation. This technique was developed for quick and easy qualitative analysis of several samples, especially liquid and solid materials. It is based on the electrospray ionization process to generate an ionized solvent spray. With gaseous nitrogen and a pressure up to 15 bar, the solvent is led to the sample (Figure 1 A). Further processes desorb the analyte: droplet pickup, charge transfer followed by a so-called sputter process and charge transfer in the gaseous phase after evaporation from sample. A

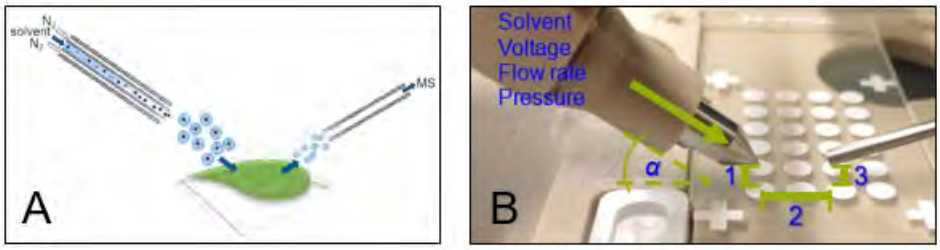

Figure 1. Schematic illustration of the desorption electrospray ionization process (A) and adjustable parameters.

small and heated ion transfer tube collects the desorbed analyte molecules and leads into the mass spectrometer. In the end for a successful desorption and a high ion formation efficiency the adjustable parameters have to be optimized for each analyte and sample material. The main important parameters are the voltage of spray capillary, the solvent flow rate, the angle α, the pressure of gaseous nitrogen, and the distances 1, 2 and 3 (Figure 1 B) [6,7].

Material and methods

The uptake of europium as a homologue for the trivalent actinides in plants was investigated for first indication of americium behaviour. First experiments were made with stable europium (47.8% ^{151}Eu and 52.2% ^{153}Eu) in *Secale cereale L.* from Kiev as described above. Plants were grown for two weeks in Hoagland medium [8] in a growth camber with about 22°C and a 16 h/8 h day/night cycle at 120 µE/m²s. Afterwards, the medium was contaminated with an aqueous europium nitrate solution. Two different europium concentrations of 1 mmol/L and 10 mmol/L were used in Hoagland medium.

Results obtained with nano ESI MS, DESI MS, TRLFS and ICP MS. Europium species in Hoagland medium were determined by nano ESI MS (nano Electrospray Ionization Mass Spectrometry) before treatment. These species were used for searching m/z signals in the DESI MS spectra after five and seven days of incubation. DESI MS measurements were recorded from background (sample plate), leave, stem and root of control group and contaminated plant parts. Afterwards, TRLFS gives indications of the contained europium species for supporting the MS evaluation. At least with ICP MS the element concentration of europium was determined.

Results and discussion

Due to the two stable europium isotopes ^{151}Eu and ^{153}Eu, the isotope ratio was a useful indicator for data evaluation. After five days of contamination, there

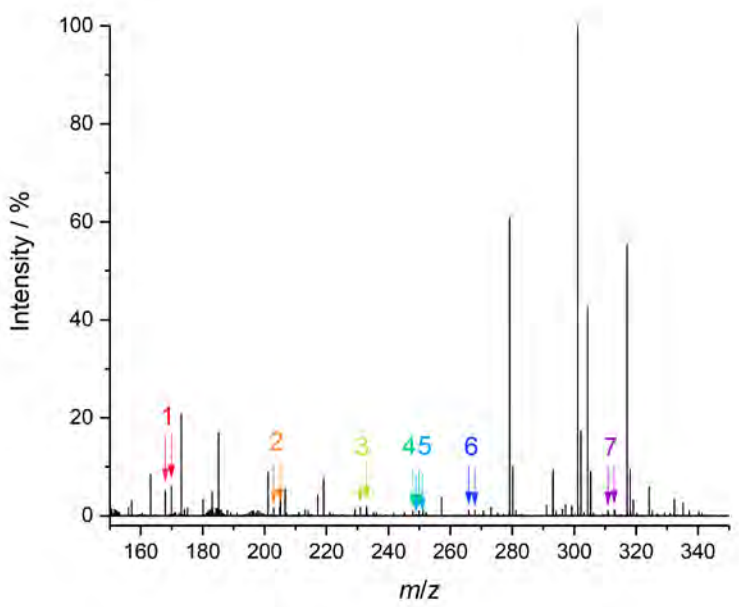

Figure 2. DESI MS spectrum of a contaminated root of Secale cereale L. after five days with marked europium species.

Table 1. Determined europium species in the DESI MS spectrum of contaminated Secale cereale L. root after five days of treatment.

No.	*m/z*		Species	Relative intensity / %	
1	167.92	169.92	$[Eu(OH)]^+$	5.11	5.86
2	202.94	204.94	$[Eu(OH) + H_2O]^+$	1.41	1.65
3	230.92	232.92	$[Eu(NO_3) + H_2O]^+$	1.67	1.84
4	247.92	249.92	$[Eu(OH)(NO_3) + H_2O]^+$	0.87	0.97
5	248.93	250.93	$[Eu(NO_3) + 2\ H_2O]^+$	0.25	0.30
6	265.93	267.93	$[Eu(OH)(NO_3) + 2\ H_2O]^+$	1.21	1.22
7	310.92	312.92	$[Eu(NO_3)_2 + H_2O]^+$	0.97	1.15

are two to three isolated m/z signals per isotope above 0.1% relative intensity of determined europium species in each samples. In the root sample with a contamination of 10 mmol/L europium seven signals above 0.1% relative intensity were determined (Figure 2, Table 1). A very few of the m/z signals are caused by Eu^{2+} species, which do not exist under the current pH and redox conditions. The reason is a measurement artefact due to the ionization and included charge transfer via collisions with residual gaseous nitrogen in the mass spectrometer. Also the europium hydroxide species are caused by measurement artefacts due to reaction with water molecules [9].

Mainly the solvent, species present in ambient air and the plant itself cause the additional m/z signals in the MS spectrum. Further europium species could also be included and are currently evaluated.

Furthermore, DESI MS spectra were recorded after seven days of contamination with less signals of the europium Hoagland species. Therefore, TRLFS measurements were performed on a root. They show a strong complexation of europium based on the $^7F_1$–$^7F_2$ band ratio (Figure 3). Calculations with bi-exponential fits evaluated by use of Horrocks equation [10] showed longer lifetimes of the europium-contaminated root. According to that, a stronger complexation with less water molecules around the element ion is possible. Possible candidates are carbonate and phosphate complexes with similar lifetimes. This is also in accordance with the HSAB concept (Hard and Soft Acid and Bases) because europium as a hard acid preferably binds to carboxylates and phosphates being hard bases. Therefore, more experiments with europium and carboxylate or phosphate ligands will be performed for determining the exact lifetimes.

With the support of ICP MS analysis, the europium concentrations in plant parts were determined. Each plant part incorporated europium and the results show a decreasing trend with root > leave > stem due to the uptake mechanism. Europium is translocated from the roots in the leaves via the xylem of the stem, which do not store Eu^{3+} ions.

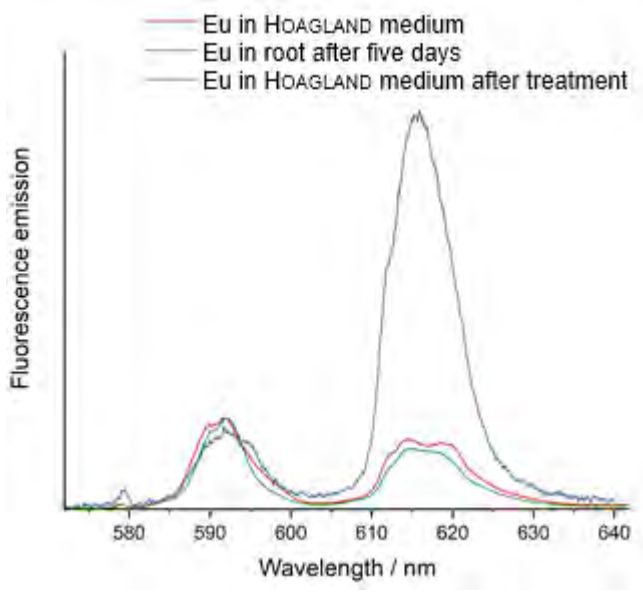

Figure 3. Fluorescence emission spectra of europium in HOAGLAND medium before, after contamination, and of a Secale cereale L. root after contamination recorded with TRLFS.

Conclusion and future experiments

First results of europium contaminated plant parts were obtained by DESI MS analysis. The technique is able to analyse radionuclide species in plants with a quick and easy sample preparation and complex evaluation. Future experiments with other crop plants and americium will give more experience and information about the environmental behaviour of this radionuclide. Additionally, an analysis of curium with TRLFS can give indications about the species formation in plants. Curium has only one fluorescence emission band, which is very sensitive to environmental shift and easy to identify. Furthermore, experiments with different ligands added to the Hoagland medium, like carboxylates and phosphates, will show the influence of these naturally occurring small organic compounds on radionuclides and their plant uptake.

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What is the optimum season and length for radon measurement in Finnish homes?

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Background

Radon (Rn-222) in indoor air is a well-know carcinogen which causes 200–300 lung cancers every year in Finland. Home-owners are encouraged to test their homes for radon and use radon mitigation methods if levels above 200 Bq/m^3 are found.

Seasonal variation of indoor radon concentration is a well known phenomenon in homes in cold climate. The extent of variation is dwelling-specific and the difference in radon concentration can be one order of magnitude between summer and winter months. Temperature gradient between indoor and outdoor air creates a pressure gradient that generates convective air flow from soil to indoor air through gaps in the building foundation. Since year-long measurements are impractical, the radon concentration is normally measured by a two-month sampling during the cold season. The annual average concentration is then assessed by multiplying the result by a correction factor. The season and the length of sampling are important factors that attribute to the uncertainty of the assessment.

In this work, we investigated whether the present recommendations on the season and the length of sampling are justifiable. Another aim was to establish a seasonal correction factor that can be used for assessing radon exposure in homes and to find out uncertainties related to the use of the correction factor.

Methods

We analyzed data from 329 homes where radon was measured monthly for a period of one year. The data was collected in the national radon survey conducted in 1996. We calculated correction factors for each home assuming 1) different starting month of sampling and 2) three lengths (one, two or three months) of sampling. Hence, for each home we obtained 36 seasonal correction factors representing 36 different sampling seasons and lengths. Then the distribution of correction factors for each sampling season and length were assessed and the related descriptive statistics were computed.

Results

The correction factors were log-normally distributed and therefore the geometric mean and geometric coefficient of variation were selected as descriptive quantities. As expected, the variation decreased with the increasing sampling length (Fig. 1).

The geometric mean of correction factor computed for measurements starting between September and March remained relatively constant. The lowest coefficients of variation were obtained for three-month measurements and those which started between September and April (Figure 2).

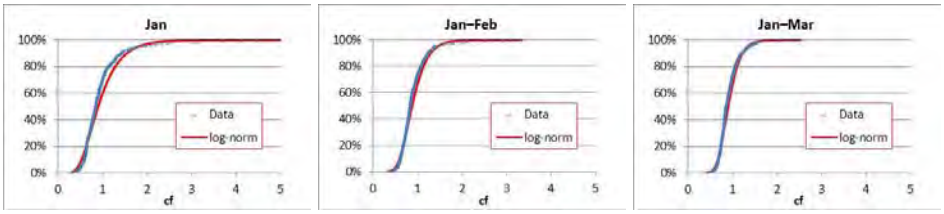

Figure 1. Cumulative distribution of seasonal correction factor (cf) for one-, two- and tree-month measurements starting in January.

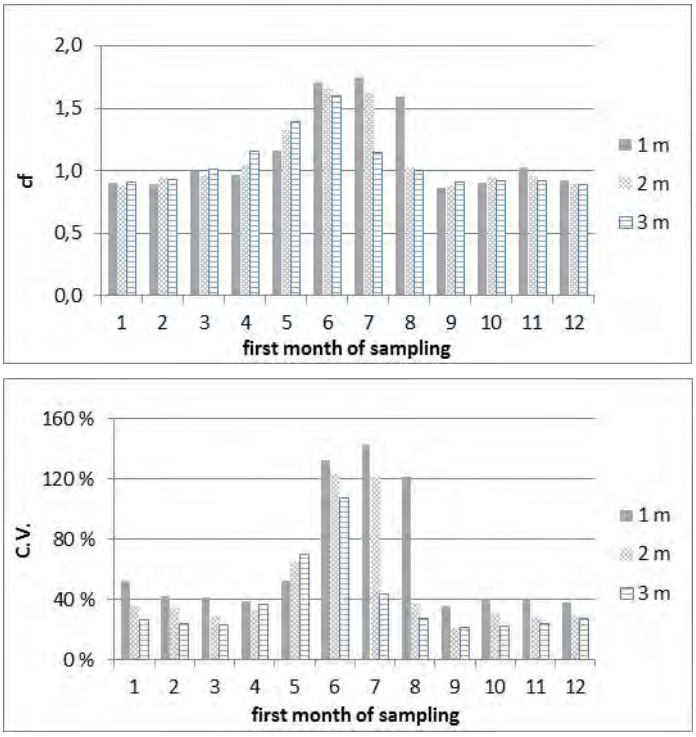

Figure 2. Geometric mean of seasonal correction factor (cf) and related coefficient of variation (C.V.) for one-, two- and three-month measurements starting in different months (Number 1 refers to January).

Table 1. Comparison of three different sampling strategies. cf stands for seasonal correction factor and C.V. for coefficient of variation.

	two-month sampling	three-month sampling	three-month sampling
Months of sampling	9–5	8–5	9–5
cf, geometric mean	0.93	0.93	0.92
cf, 90 % confidence interval	0.63–1.46	0.68–1.35	0.67–1.34
cf, C.V.	30%	24%	24%

Table 2. Seasonal correction factor for houses where annual mean radon concentration is <100 Bq/m3 and >100 Bq/m^3.

	geometric mean (and coefficient of variation) of seasonal correction factor		
Sampling length (months)	two	three	three
Months of sampling	9–5	8–5	9–5
Rn conc. All	0.93 (30%)	0.93 (24%)	0.92 (24%)
Rn conc. <100 Bq/m^3	0.98 (38%)	0.97 (29%)	0.97 (30%)
Rn conc. >100 Bq/m^3	0.90 (25%)	0.90 (22%)	0.89 (21%)

It is obvious that sampling carried out during summer months has generally a poor predictive value in assessing the annual mean concentration. Based on this preliminary analysis, we compared three measurement strategies: two-month sampling between September and May, three-month sampling between August and May and three-month sampling between September and May (Table 1). The three-month sampling results in lower uncertainties when assessing the annual mean concentrations.

Finally, we investigated the influence of annual average radon concentration on correction factors. It is hypothesized that with low radon concentrations the proportion of radon produced by building materials is significant. Radon released from building materials does not exhibit seasonal variation unlike convective flow from the soil (Table 2). Our data confirms this hypothesis.

Discussion and Recommendations

When assessing the annual mean radon concentration the three-month sampling is preferable, since the related uncertainty is smaller especially when the annual mean radon concentration is below 100 Bq/m^3. Many residents, however, feel that even the present recommended minimum of two-month sampling is too long. Some companies have realized this and have started to supply "express measurements" that take only one week. The results of these measurements will in many cases lead to false positives or false negatives when compared to the reference value of 200 Bq/m^3. Keeping this in mind, we recommend

that the minimum length of indoor radon measurement remain two months in order to curb the popularity of the ”express measurements”.

The present recommendation for sampling months (between November and April) is too conservative and the data supports extending the period to the end of May. The results suggest that residents could start measuring radon already in August if three-month sampling is applied. In August, however, the mean correction coefficient and related coefficient of variation differs significantly from those computed for September and October. Therefore, August should not be included in the sampling months. Since we consider three-month sampling not an applicable strategy, our new recommendation for sampling period is between September and May.

The seasonal correction factor obtained in this analysis is somewhat higher than our previous estimate (0.85 for >100 Bq/m^3 annual mean concentrations and 0.89 for all concentrations). It is likely that in the future the seasonal variation in radon concentration in indoor air will be even smaller. New houses are generally built radon safe and have sophisticated mechanical supply and exhaust ventilation. Hence the pressure gradient between indoor and outdoor air remains more stable. Many new houses also have air conditioning reducing the need to ventilate through windows during the warm season, which would dilute the radon concentration in indoor air.

^{14}C content in vegetation in Finland

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Introduction

^{14}C is a beta emitter (E_{max} 156 keV) and it has a relatively long half-life (5730 years). The main sources of radiocarbon in the environment are natural production in the upper atmosphere, the atmospheric nuclear weapons testing carried out during the 1950s and 1960s, and discharges from nuclear power plants and fuel reprocessing plants. On the other hand, ^{14}C concentrations decrease in the atmosphere due to the increased burning of ^{14}C-free fossil fuel.

Due to fallout from the atmospheric nuclear weapons testing (in the mid-1960s), the ^{14}C specific activities in the terrestrial environment reached the maximum values (higher than 400 Bq/kg C). Before atmospheric testing, the specific activity of ^{14}C was 226 Bq/kg C in 1950.(Roussel-Debet S, *et al*. 2006) In the present day the background value of ^{14}C is higher than in 1950 in terrestrial environment.

^{14}C is released during normal operation of nuclear power plants. In the gaseous effluents of boiling-water reactors (PWR) most of ^{14}C is in carbon dioxide form (95%) and rest in carbon monoxide and in hydrocarbon. Correspondingly in the effluents of pressurized-water reactors (BWR), 80% of ^{14}C is in organic form (CH_4) and 20% is in form of CO_2. The carbon species in the liquid releases are various organic compounds and carbonates. In terrestrial environment, most of the carbon is taken up by leaves as CO_2 form during photosynthesis. Root absorption by plants is negligible (Garnier-Laplace J, Roussel-Depet S, 2010).

In Finland there are four nuclear power plants units, two in the Southern Finland (Loviisa) and two in the Western Finland (Olkiluoto). Two pressurized-water reactors (BWR) are in Loviisa and two boiling-water reactors (PWR) in Olkiluoto. In addition, commissioning of the third unit will be near future in Olkiluoto NPP. Studies of ^{14}C activity concentrations in the environment of Finnish nuclear power plants are one of the monitoring program in STUK. In order to evaluate ^{14}C releases from Finnish NPPs, the background level of the ^{14}C activity concentrations should be known. The objective of this study was to investigate the ^{14}C background level in vegetation in Finland. In addition, we

also look for possible regional differences, e.g. by dilution from industrial combustion of fossil fuel.

In this paper the specific activities of ^{14}C equal to the activity per kg of carbon (Bq/kg C) are presented in the samples of pine needle, grazing grass, grain and birch leaves.

Materials and methods

The samples were collected in August 2008. The sampling places were selected considering the vicinity of coal-fired and peat-fired power stations. The influences of coal-fired power stations to the contents of ^{14}C were supposed to be minor in the Eastern Finland (Fig.1, Heinävesi, Juankoski). Both coal-fired and peat-fired power stations are in the Pietarsaari area, for example. Pudasjärvi was the Northernmost sampling place, which locates below 100 km from the peat-fired station of Oulu. The peat-fired station of Kuusamo is located in the Northeast Finland, 120 km from the Pudasjärvi area (Fig. 1). In Southern Finland in Helsinki, the samples were taken from near the coal-fired power station of Hanasaari (pine needle samples about 400 meters from the station).

Samples of pine needles were taken from ten different pines in the sampling site (one branch/pine), except in Lohja (one branch from five pines). Samples of birch leaves were taken from eight to ten different trees. The sample size of grain and grazing grass samples was 50–100 g fresh weight. In the laboratory, the samples were dried overnight at 105°C and homogenized.

The samples were prepared for the low background liquid scintillation counting (1220 Quantulus, PerkinElmer) with a 307 Sample Oxidizer (Vartti V-P. 2009). Determination of the

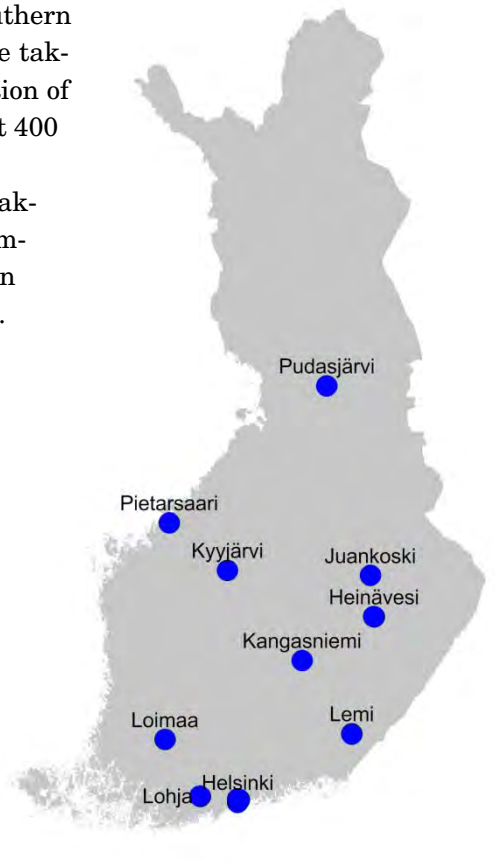

Figure 1. Location of the sampling sites in Finland.

total carbon from the samples was performed by CHN-analyzer (LECO CHN-1000 elemental analyzer).

Results

The results of ^{14}C specific activities (Bq/kg C) in the vegetation samples are presented in Table 1. The values of percent Modern Carbon (pMC) were calculated by the equation 1 (Stenström et al.2011):

$$A = \frac{pMC}{100\%} \cdot \left(\frac{\left(1 + \frac{\delta^{13}C}{1000}\right)}{0.975} \right)^2 \cdot e^{(1950-y)/8267} \cdot 226 \text{ Bq/kg C} \quad (1)$$

Correction for sample isotope fractionation was performed by using a value of $\delta^{13}C = -25‰$. pMC values were calculated because these are often used for environmental samples.

Table 1. ^{14}C activity of pine needle, grazing grass, grain and birch leaves samples (Bq/kg C and pMC %).

Location	Sample	C-14 Bq/kg d.w.a ± uncertainty [b]	TC (m-%) ± uncertainty [b]	C-14 Bq/kg C ± uncertainty [b]	pMC %
Pudasjärvi 65°23.42N 27°15.22E	Pine needles	98 ± 7	50 ± 8	194 ± 17	100 ± 11
	Birch leaves	162 ± 24	48 ± 10	335 ± 60	159 ± 18
	Grain (barley)	128 ± 11	43 ± 10	298 ± 40	112 ± 13
	Grazing grass	105 ± 13	45 ± 10	232 ± 37	96 ± 16
Juankoski 63°01.26N 28°25.37E	Pine needles	114 ± 21	49 ± 8	231 ± 47	114 ± 20
	Birch leaves	154 ± 20	48 ± 10	318 ± 52	151 ± 16
	Grain (barley)	120 ± 7	43 ± 10	277 ± 32	106 ± 12
	Grazing grass	127 ± 9	45 ± 10	284 ± 35	115 ± 12
Heinävesi 62°30.26N (Pn, Bl,Gb) 28°30.15E 62°30.57N (Gg) 28°29.04N	Pine needles	116 ± 21	50 ± 8	234 ± 47	117 ± 20
	Birch leaves	150 ± 21	50 ± 10	307 ± 53	149 ± 17
	Grain (barley)	94 ± 6	43 ± 10	221 ± 26	81 ± 12
	Grazing grass	134 ± 11	45 ± 10	298 ± 38	122 ± 13
Lemi 61°03.10N (Pn, Bl,Gb) 27°51.57E 61°03.00N (Gg) 27°51.32E	Pine needles	110 ± 10	50 ± 8	220 ± 27	112 ± 12
	Birch leaves	149 ± 21	50 ± 10	297 ± 52	152 ± 17
	Grain (barley)	106 ± 6	43 ± 10	248 ± 29	92 ± 12
	Grazing grass	131 ± 11	46 ± 10	288 ± 37	121 ± 13

Location	Sample	C-14 Bq/kg d.w.a ± uncertainty [b]	TC (m-%) ± uncertainty [b]	C-14 Bq/kg C ± uncertainty [b]	pMC %
Helsinki, Hanasaari and Viikki 60°11.17N (Pn) 24°58.95E 60°10.97N (Bl) 24°58.73E 60°13.53N (Gb) 24°58.56E 60°13.27N (Gg) 25°01.37E	Pine needles	130 ± 20	49 ± 8	264 ± 45	130 ± 17
	Birch leaves	113 ± 7	49 ± 10	232 ± 27	112 ± 12
	Grain (barley)	77 ± 5	42 ± 10	185 ± 22	65 ± 12
	Grazing grass	97 ± 15	44 ± 10	219 ± 41	87 ± 19
Lohja 60°14.37N (Pn) 24°02.25E 60°14.61N (Bl) 24°01.62E	Pine needles	95 ± 17	50 ± 8	192 ± 37	96 ± 20
	Birch leaves	111 ± 18	48 ± 10	229 ± 43	109 ± 19
Loimaa 60°55.53N 23°04.21E	Pine needles	129 ± 19	51 ± 8	255 ± 43	132 ± 17
	Birch leaves	141 ± 6	48 ± 10	291 ± 32	139 ± 11
	Grain (barley)	110 ± 7	43 ± 10	259 ± 30	95 ± 12
	Grazing grass	133 ± 6	44 ± 10	302 ± 34	119 ± 11
Kangasniemi 61°58.07N 26°34.13E	Pine needles	146 ± 22	50 ± 8	294 ± 50	148 ± 17
	Birch leaves	147 ± 24	48 ± 10	307 ± 58	143 ± 19
	Grain (barley)	99 ± 6	42 ± 10	233 ± 27	85 ± 12
	Grazing grass	128 ± 21	45 ± 10	287 ± 54	116 ± 19
Kyyjärvi 63°04.18N 24°30.13E	Pine needles	139 ± 19	50 ± 8	277 ± 45	141 ± 16
	Birch leaves	181 ± 37	49 ± 10	368 ± 84	181 ± 23
	Grain (barley)	83 ± 5	43 ± 10	192 ± 22	73 ± 12
	Grazing grass	121 ± 16	44 ± 10	274 ± 44	109 ± 16
Pietarsaari 63°37.36N 22°49.42E	Pine needles	139 ± 15	50 ± 8	276 ± 37	142 ± 13
	Birch leaves	160 ± 21	49 ± 10	325 ± 54	160 ± 17
	Grain (barley)	81 ± 5	43 ± 10	190 ± 22	70 ± 11
	Grazing grass	133 ± 14	43 ± 10	307 ± 45	117 ± 15

The ^{14}C specific activities of pine needle samples varied from 192 to 294 Bq/kg C, birch leaves from 229 to 368 Bq/kg C, barley grain from 185 to 298 Bq/kg C and grazing grass from 219 to 307 Bq/kg C. The results of different samples are at same level considering also the uncertainties of the results. The ^{14}C specific activities of the birch leave samples were highest mainly in all sample sites (Table 1). With this analyzing method, any differences in ^{14}C levels between the sample locations did not observed. The average values of ^{14}C activities in pine needles, in birch leaves, in grains and in grass were 244 Bq/kg C,

Table 2. The average value of the ^{14}C activities (Bq/kg C, pMC%) of four sample types in Finland.

Sample type	^{14}C activity Bq/kg C ±%	pMC %
Pine needle	244 ± 16%	123 ± 16%
Birch leave	301 ± 17%	145 ± 17%
Grain (barley)	234 ± 12%	87 ± 12%
Grazing grass	277 ± 15%	111 ± 15%

a coverage factor of 95% (k = 2)

301 Bq/kg C, 234 Bq/kg C and 277 Bq/kg C, respectively (Table 2). The average value of the ^{14}C specific activities taken all together is 264 ± 40 Bq/kg C (2008).

The background level of ^{14}C is a dynamic parameter (Otlet R.C. et al 1997) i.e. ^{14}C background level changes over time. According to Garnier-Laplace J and Roussel-Depet S, the ^{14}C specific activities of terrestrial samples were around 238 Bq ^{14}C/kg C (2009 measurements).

Conclusions

The analysis of pine needle, birch leave, grain and grass samples gives a mean value for ^{14}C background levels for the year 2008 of 244 Bq/kg C, 301 Bq/kg C, 234 Bq/kg C and 277 Bq/kg C in Finland, respectively.

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