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COMET IRA on improved parameterisation of key processes for transfer and dynamic modelling approaches: Results and impact

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Executive Summary

This deliverable report D-N°3.2 gives a short summary from each of the COMET initial research activities (IRA's) adopted during the COMET project period.

The different IRA topics presented are: 1. Marine modelling, 2. Forest modelling, 3. Human food chain modelling, 4. NORM modelling, 5. Particle behaviour and 6. ICRP reference sites.

Within each IRA, the focus has been on improving the parametrisation of key processes that control the transfer of radionuclides in the environment, as well as to include the dynamic aspect of radionuclide transfer in radioecological models.

Even though there is more research to be done, the COMET IRA's have brought the science forward and much has been done to meet the different objectives, both common and specific within each research area.

Each COMET IRA has identified future challenges and priorities of importance for future research within the field of radioecology/environmental radioactivity. These priorities constitute a valuable input to future project cooperation and/or project calls within Radioecology.



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1. Scope of the report

This report presents a high-level summary of the results from the COMET Initial Research Activity (IRA) on improved parametrisation of key processes for transfer and dynamic modelling approaches. A comprehensive report of all WP3 activities, including those of the IRAs, will be presented in D3.7 “Final report of WP3 activities”. This approach was chosen, as several research activities within the COMET IRAs are still ongoing, particularly those relating to marine modelling and particle behaviour. These topics are integrated with the two research projects, FRAME and RATE, which were funded under the COMET Competitive Research and Technological Development Call. These two projects will continue until May 2017 and will be reported at this time. Hence, a detailed report of the COMET IRAs, including a description of the integration of the IRAs with the projects FRAME and RATE will be provided in D3.7.

2. Overall introduction and objectives

The scientific discipline of radioecology aims to provide a quantitative and integrative assessment of radionuclide impacts on humans and wildlife for a wide range of exposure scenarios. This is done through research and modelling to predict transfer of radionuclides in the environment, estimating exposures of and doses to man and wildlife, and by studying effects in wildlife. The radioecological focus central to the proposed Initial Research Activity (IRA) of WP3 is to improve the estimation of exposures and doses to humans and wildlife (whereas the WP4 IRA focuses on studying the actual effects on wildlife).

The first deliverable in COMET WP3, giving a detailed plan for the COMET WP3 Initial Research Activity in the form of a list of research projects and goals, participants and timing, states that the IRA’s should focus on improved parameterisation of key processes controlling the transfer of radionuclides, with a specific emphasis on dynamic modelling approaches. It is also guided by Challenge one of the Strategic Research Agenda of radioecology: “To Predict Human and Wildlife Exposure in a Robust Way by Quantifying Key Processes that Influence Radionuclide Transfers and Exposure”, with its four associated research lines.

The topics and their objectives, in short, are the following:

- 1. Marine modelling** – improving predictions of concentrations in and radiological exposures to marine biota and humans through sophisticated modelling, e.g. trophic transfer modelling and by combining transfer modelling with sediment modelling.
- 2. Forest modelling** - reducing the uncertainties in assessments of short- and long-term impacts of radioactive contamination in forested areas through model development and parameterisation of key processes controlling the transfer of radionuclides.
- 3. Human food chain modelling** - improving human food chain modelling through regional customisation of parameter values, using Bayesian methods and studying the long-term dynamics of soil-to-plant transfers for specific soil types and for long-lived radionuclides.
- 4. NORM modelling** - acquiring the data necessary for the parameterisation of key processes, and improving existing models or developing parametric models linking observed accumulation, mobility, and transfer with environmental parameters and processes.
- 5. Particle behaviour** - improving our ability to describe the processes of hot particle transformation in the environment and radionuclide leaching in various media.
- 6. ICRP reference sites** - providing the data to derive a taxonomically based model of radionuclide transfer for wildlife, independent of site-specific factors.



3. IRA on marine modelling

3.1 Introduction and objectives

The Fukushima accident in 2011 has highlighted the importance of improving our knowledge of marine radioecology. This event constituted the most important accidental release of artificial radionuclides to the marine environment that has ever occurred. Many questions about the behaviour and the future of these released radionuclides in the concerned area appeared. Measurement results in the aftermath of the accident provided realistic data to assess and follow the radiological state of the marine environment, but this knowledge is partial. The purpose of models is to complete this knowledge and to give indications about the future trends of the contamination through the marine environment. The COMET marine group IRA focused on concepts and models that could be applied in an accidental situation such as Fukushima to assess transfers and distribution of radionuclides between the different marine compartments.

3.2 Background and status of the IRA field

An array of dispersion models has been used to determine the radionuclide dispersion over Fukushima coast, with expert teams of modellers dedicated to their improvement and inter-comparisons (IAEA-MODARIA, Science Council of Japan model comparison, UNSCEAR assessment projects). Therefore, the COMET Marine group proposed to focus on the less-explored area of radioecological transfer modelling (transfers to biota in the presence of other interactions, such as with sediments or seawater dispersion). The IRA planned to use existing models as a basis for the work, with a view to improving some of them to achieve more sophisticated models, e.g. trophic transfers modelling or combining transfer modelling with simple sediment transfer to improve not only estimates of concentration in biota, but dose (by considering not only internal dose but also all the pathways of external exposure). This can lead to better understanding of the doses to biota and also the dose to humans due to consumption of seafood. In the aftermath of an accidental situation where radionuclides in the different marine compartments have not equilibrated, time-dependent radioecological models of transfer are required. Whereas the basic concepts of such models were existing, there was no operational tool for this situation and the marine IRA was also posed to address this question.

In the aftermath of the Fukushima accident, contamination of every marine compartment (water, sediment and biota) was observed. Measurements carried out by and the Japanese operators and regulators gave the contamination evolution in these media for the past years, and then a realistic representation of the radiological state and evolution of the environment. These data were bound to be useful to validate and improve our radioecological models.

3.3 Reason for this IRA within COMET

The Fukushima nuclear accident in 2011 constitutes the most important accidental release of artificial radionuclides to the marine environment. This is one of the main reasons why, within the COMET project, a marine group emerged. Furthermore, this situation has refocused marine radioecology research on post-accidental issues. Within the framework of COMET WP3 dedicated to the improvement and the validation of radioecological models, the marine IRA has focused on marine operational modelling tools designed for emergency or post-accidental issues.



3.4 List of participants in the work

The marine IRA had the following participants:

- Céline Duffa, Mokrane Belharet, IRSN - France
- Jordi Vives i Batlle, SCK-CEN, Belgium
- Clare Bradshaw, SU, Sweden
- Mikhail losjpe, Justin Brown, NRPA, Norway

Other participants from FRAME project included:

- Pere Masqué, University of Barcelona, Spain
- Núria Casacuberta, Marcus Christl, ETH Zurich, Switzerland
- Michio Aoyama, IER-Fukushima University, Japan
- Jun Nishikawa, Tokai University, Japan
- Ken Buesseler, WHOI, USA

3.5 How were the key issues addressed within the IRA

The understanding of contamination levels and radionuclide distributions in the environment, along with prediction of their future evolution, requires the use of modelling tools and analyses of extensive monitoring data. Two main radioecological modelling topics were taken into account: Transfers and exchanges between water and sediments, and modelling of transfer to biota.

Some simple models should possibly be included in decision support systems for emergency situations. Others, more sophisticated, should be used in a second step to evaluate and predict more accurately the post accidental situation. The main focus of the marine IRA group centred on emergency and post-accidental issues, as well as on the modelling tools adapted to such situations. It was decided to use existing time-dependent models as a basis for the work with a view to improve some of them to achieve more sophisticated models. The IRA work was therefore divided into three tasks:

- Implementation and use of classical radioecological models based on dynamic transfer equations to evaluate concentrations in marine organisms (fishes, molluscs, crustaceans). This included improvement of radioecological parameters (concentration factors and single or multicomponent biological half-lives) for ^{137}Cs , ^{134}Cs and ^{90}Sr . This task also included comparisons with observed data series from Fukushima.
- Inclusion of sediment processes in dynamic transfer modelling. This work followed ongoing studies adapting an existing dynamic model (the SCK•CEN dynamic transfer model for marine biota D-DAT). The tool was completed and tested with the available data to see if it was capable of reproducing activity of biota and in sediment in the Fukushima coastal area, and it was found to give a more realistic calculation of concentrations in biota and to improve significantly on equilibrium models based on concentration ratios and K_d values, such as the ERICA tool.
- Process oriented modelling for mid and long-term predictions: ecological and environmental processes. This involved modelling trophic transfer to pelagic fishes, including food-web transfers and establishing if there was real potential for biomagnification in the Fukushima food chain.



During the IRA second year (2014), the FRAME project (The impact of recent releases from the Fukushima nuclear Accident on the Marine Environment) supported by COMET after its first call, was integrated within the marine group activities, with complementary objectives (to understand the sources, fate, transport, bioaccumulation and associated impact of radionuclides from the FDNPS). This project included two cruises in the Fukushima coastal area in 2014 and 2015 to collect and analyse many water/sediment and biota samples to give information important for the understanding of the radioecological situation in the years after the accident, and to complete the existing dataset in order to improve the calibration of the D-DAT model.

3.6 Short summary of results so far

Two main kinetic radioecological models were improved during the IRA:

- The IRSN model STERNE, designed to be operated for a first estimation in case of an emergency situation. STERNE calculates both radionuclides transport using advection and diffusion equations offline from hydrodynamic calculation, and radioecological transfers to biota with a simple model based on dynamic transfer equations. The required radioecological parameters (concentration factors and single or multicomponent biological half-lives) were compiled for some important radionuclides and for generic marine species (fish, molluscs, crustaceans). Dispersion and transfer calculations can be carried out simultaneously on a 3D grid (Duffa et al., 2015).
- The aforementioned SCK•CEN D-DAT model, which now includes the dynamics of radionuclide uptake and turnover in biota and sediments, as determined by a balance between the residence time of radionuclides in seawater/sediments and the biological half-life of elimination in the biota. The model calculates activity concentration of ^{131}I , ^{134}Cs , ^{137}Cs and ^{90}Sr in seabed sediment, fish, crustaceans, molluscs and macroalgae from the surrounding activity concentrations in seawater, from which internal and external dose rates are derived (Vives i Batlle et al., 2014). The model was improved by the inclusion of sediment processes in dynamic transfer modelling. As a result of this work, D-DAT is now adapted to include depletion of radionuclides adsorbed onto suspended particulates (particle scavenging), molecular diffusion, pore water mixing and bioturbation (modelled effectively as a diffusive process) represented by a simple set of differential equations that is coupled with the biological uptake/turnover processes (Vives i Batlle et al., 2015). This has had an impact on uncertainty reduction in model predictions compared with equilibrium models. Also, it has impacted the estimation of external dose rate to biota, which now includes the important pathway of external exposure of benthic organisms from radioactivity in sediments (now dynamically calculated), instead of external exposure only from radioactivity in water. With this improvement, the systematic uncertainty margin of the predictions is now reduced by about two orders of magnitude.

More sophisticated modelling tools for simulating transfer to biota were improved (NRPA model) or developed (IRSN trophic transfer model) during this IRA period.

The NRPA model uses time-dependent transfer of radionuclides within the food chain that can be described by first-order differential equations. It assumes that excretion/elimination rates are independent of the uptake route, assimilation efficiency is independent of food type, predators do not assimilate the activity concentration in gut content of their prey, and zooplankton are a homogeneous group.



IRSN developed, with the University of Toulouse (France), a model of radiocaesium transfer to marine biota taking into account large scale organism displacements in the Fukushima area. A trophic level ecosystem model was coupled with an ocean circulation model to take into account the site-specific environmental conditions in the area. The different radioecological parameters of the model were estimated by calibration using published data, and a sensitivity analysis of the parameter uncertainties was carried out, showing a high sensitivity of the model results, especially to the ^{137}Cs concentration in seawater, the rates of accumulation from water and the radionuclide assimilation efficiency.

3.7 Future challenges and priorities

European activities and research on the marine environment are an important challenge for radioecology and radiological protection. Possible accidental inputs in the marine environment and radiological consequences appear now as a real topic to deal with. Modelling tools and monitoring strategies in the emergency phase and in the post-accidental phase need to be improved. Many dynamic processes still need to be studied and understood to be represented. Radionuclide behaviour in the marine environment in non-equilibrium situations is still insufficiently documented, such as radionuclide reactivity and partition between particulate and dissolved phase and transfers through the different trophic chains. Research on these items includes field and experimental measurement data. Investigation of the inputs from contaminated river water and specific processes in estuaries is also an important item.

Moreover, validated dispersion models relevant for normal or accidental discharges from nuclear plants or ship wrecks, usable in a decision support system, are needed, especially for coastal areas.

3.8 List of publications so far

Vives i Batlle, J. (2014). Dynamic modelling of radionuclide uptake by marine biota: application to Fukushima assessment, Oral presentation, ICRER 2014 - 3rd International Conference on Radioecology & Environmental Radioactivity, 7-12 September 2014, Barcelona, Spain. Electronic proceedings 10.1. Lessons learned from the Fukushima accident.

Duffa C. et al. (2015) Development of emergency response tools for accidental radiological contamination of French coastal areas, *Journal of Environmental Radioactivity*, DOI: 10.1016/j.jenvrad.2015.04.019.

Vives i Batlle, J. (2016). Dynamic modelling of radionuclide uptake by marine biota: application to the Fukushima nuclear power plant accident. *Journal of Environmental Radioactivity* 151, 502-11.

Vives Batlle, J. (2016). Impact of the Fukushima Accident on Marine Biota, Five Years Later. *Integrated Environment Assessment and Management* 12, 654-8.

Duffa C., Vives i Batlle J., Belharet M., Bradshaw C., Brown J., Iosjpe (2015) , Modelling radionuclide transfers in Fukushima marine ecosystems - COMET Marine Group (Initial Research Activity), Oral presentation, ICOBTE congress, 12-16 July 2015, Fukuoka-Japan.

Belharet M., Estournel C. and Charmasson S. (2016) Ecosystem model based-approach for the modelling of ^{137}Cs transfer to marine plankton populations: Application to the Western North Pacific populations after the Fukushima nuclear power plant accident, *Biogeosciences*, 13, 499–516.

Iosjpe M., Isaksson M., Thomas R., Halldórsson Ó, Roos P., Logemann K., Jonsson G., Joensen H.P., Suolanen V., 2016. Implementation of a food chain sub-module into a model for radioecological assessments in the coastal waters around Iceland: effects of kinetic modelling of bioaccumulation



processes. Book of abstracts and program, the Second Conference on Radioecological Concentration Processes, November 6-9, 2016, p. 161.

Iosjpe M., Isaksson M., Joensen H.P., Jonsson G., Logemann K., Roos P., Suolanan V., Thomas R. Effects of dynamic behaviour of Nordic marine environment to radioecological assessments, 2016, ISBN: 978-87-7893-442-0, http://www.nks.org/en/nks_reports/view_document.htm?id=111010213400466

4. IRA Forest modelling

4.1 Introduction and objectives for IRA topic

In forest radioecology, the impact of the recent nuclear accidents (Chernobyl and Fukushima) gave evidence of the ability of forests to be at the origin of both significant external and internal exposure by ingestion of forest products (mushrooms, berries and game animals). The Fukushima region is highly forested, with a large proportion of evergreen coniferous trees that are particularly efficient in intercepting radioactivity due to their large leaf area indexes, especially in comparison to agricultural lands.

In such ecosystems, the contamination patterns show a high horizontal variability. In addition, migration of radionuclides in the soil is very low and the existence of a real nutrient cycle that makes available radionuclides leads to a persistence of the radioactive pollution. Whilst modelling work to understand this has been and is being carried out, much effort has to be done to improve the knowledge on these ecosystems and to better predict the behaviour of the radionuclides in such an environment. Moreover, there is a need for validation of models through a variety of scenarios to assess the models' transferability.

In the COMET WP3, IRA on forest modelling, the main objective was to generate guidance on how to reduce the uncertainties in the assessments of short- and long-term impacts of radioactive contamination in forested areas through the production of a guidebook for modellers. The guidebook would include topics such as model sensitivity analysis and parameterisation of key processes controlling the transfer of radionuclides, as well as documentation on model parameter requirements to facilitate the interaction between modellers and experimentalists, as well as examples from ongoing development of current forest transfer models.

The focus was on producing guidance documentation and work on model development dealing with key processes and variables/factors contributing most to the overall uncertainties in forest modelling. Due to the dynamic nature of these processes, most emphasis was placed on time-dependent soil-water-atmosphere interactions (as expressed, for example, in parametric K_d approaches, the evapotranspiration as an engine for water fluxes in trees, etc.) and soil - plant transfers (TF, TC, CR), with forest vegetation selected according to its importance for wildlife intakes and for human dietary intakes.

Starting from existing models as a basis for the guidance, the forest IRA explored the need for innovative modelling approaches. The aim was to discuss process identification and mathematical representation, as well as a parameterisation of key processes controlling the transfer of radionuclides so as to better improve estimated external and internal radiological exposures of biota.



4.2 Background and status of the research field

In forest radioecology, just as in marine, the impact of the recent nuclear accidents (Chernobyl and Fukushima) looms large. The Red (dead) forest in Chernobyl still represents a challenge. Furthermore, the Fukushima accident brought to the forefront several classical problems of forest ecosystems, because the Fukushima region is highly forested and forest trees can trap significant amounts of radioactivity (due to a high interception capacity because of the large leaf area indexes).

4.3 Reason for this IRA within COMET

The activities of the forest modelling WG aimed to:

- Improve knowledge on how to predict radionuclide concentrations in trees and forest products after an accident, on timescales of months to a few years – realisable within the next decade.
- Improve knowledge on how to predict radionuclide concentrations in forest on longer timescales (e.g. for waste disposal) based on future climate projections, inevitably involving larger uncertainties.
- Present this knowledge in the form of guidance documentation, based on modellers' expertise gained in the ongoing development of forest models.

The work was in accordance with Research Lines 1 and 3 of the SRA for Radioecology, namely, (a) to identify and mathematically represent key processes that make significant contributions to the environmental transfers of radionuclides and resultant exposures of humans and wildlife and (b) to develop transfer and exposure models that incorporate physical, chemical and biological interactions, and enable predictions to be made spatially and temporally.

4.4 List of participants in the work

The organisations initially interested in this IRA were: CEH, GIG, IRSN, SCK, UIAR, NRPA, NMBU and STUK.

4.5 How was the issues addressed within the IRA

There is limited research and forest modelling ongoing within radioecology, so brainstorming to share best practices and to publish a common strategy to tackle this topic included the following:

- Necessity to improve our understanding of how forests receive radionuclides from the atmosphere and how realistically this process can be modelled.
- Necessity to understand how forest trees function in an integrated way with soil and atmosphere to bring contaminants from the sub-surface soil to the biosphere.
- Need to strike a balance between the simplest empirical compartment models and the more advanced process-based models.
- Clearly, there is a lot of work on caesium, but rather limited on other radionuclides.

The initial scope and preliminary plans of the forest IRA, which included a significant amount of model development, were considered too ambitious, given the number of model developers (just 2) that had volunteered for this activity. The decision was made to reconsider what is the problem that the IRA intended to study, what forest models the team had, and what can they do in terms of informing the production of guidance for forest modellers and experimentalists. Based on this, we redefined the IRA



deliverable as an expert report giving realistic and practical advice for the modelling of radionuclide cycling in forests. As part of this report, we indicated how our own models can illustrate the importance of several processes/parameters, making the best of what is available. Additionally, we decided on a forest workshop: discussion between modellers and experimentalists of what parameters are needed for models and what data are available to parameterise models, which was integrated with the Workshop 3 of COMET and had as an outcome a report on parameter requirements for forest modelling including a “measurement wish list” for experimentalists.

4.6 Short summary of results so far

This IRA had a successful workshop including both experimentalists and modellers as part of the COMET workshop “Models fit for purpose” in Seville 2016. The results of this workshop are presented as part of the COMET Deliverable 5.5. The IRA also provided the “wish list” of modelling parameters needed for modelling in the forest environment (IRA Forest MS 1 report).

As part of this IRA, SCK•CEN continued the development of a SVAT prototype model (ECOFOR) which had its origins in the Belgian Research Policy Office project ECORISK but continued to be developed as part of the activities of this IRA. This model is currently applied to a Scots pine forest in Mol and is orientated to uptake of radionuclides from soil in chronic situations (such as remobilisation of radionuclides from ground disposal). The ECOFOR model was presented in the Barcelona ICRER conference in 2014. It is currently in advanced prototype stage, meaning that it is fully functional but still needs to be calibrated and validated as new data become available. In the meantime, the model is used in process sensitivity analysis studies, and for determining data requirements for models of this type, as explained in the “wish list” document. The model equations and parameter description were handed over to the German colleagues working on the modelling guide (currently being drafted by BfS) in order to link up with the activity of this guide to review existing forest models and to illustrate the process of selecting processes and associated parameter requirements.

IRSN also developed an approach to help decision-making by rapidly estimating the consequences of an accidental atmospheric fallout, with a special emphasis on the short-term phase (i.e., first few months) and hence focusing on modelling the deposition and plant interception processes.

Presentations and articles from the above work can be found in Section 3.8.

4.7 Future challenges and priorities

The most important future challenge is to secure scientific output within forest ecosystem modelling that can serve as a nexus for the maintenance of expertise and modelling capacity in future projects (e.g. CONCERT).

Potential future activities in forest radioecology could include the following:

- Using conceptual models to help design (a) well targeted field experiments, and (b) better models. Some relatively simple models have been developed in the past, which are fit-for-purpose for regulatory assessments. These can be further compared with measurements and progressively improved to make them more robust by adding or removing processes, though the method of process and parameter sensitivity analysis. This should lead to a robust model, i.e. one that can describe different scenarios with the same basic set of equations, needing only to be re-parameterised according to the scenario to which it is applied.
- Establish a series of tracer experiments with trees in as close to field conditions as possible, exposed to various radionuclides to find out how they process them. Even if not as easily



achievable as direct field measurements, the use of experimental enclosures (e.g. mesocosms) should be investigated.

- Apart from radiocaesium, other radionuclides for which there are poor data for forests and associated wild products should be considered, e.g. ^{36}Cl , ^{79}Se , ^{90}Sr , ^{210}Pb , radioiodine, plutonium, etc. Possibilities to compare data (measurements) and model calculations exist for Cs (and sometimes Sr and Pu), but not for most other radionuclides. The contribution of natural radionuclides to the total exposure should also be assessed where relevant.
- Develop 'instrumented forests' where sap flows in the trees are measured along with rainfall and the water fluxes in the soil (sap flow conductivity measurements, piezometers). In this way, the element fluxes in the forest can be determined and compared/contrasted with the variables affecting evapotranspiration and the processes of interception, translocation and litterfall (the SCK•CEN instrumented forest project at a Belgian NORM site under development as part of the EC project 'TERRITORIES' is an example of this).
- Investigate if there is an influence of climate change on radionuclide cycling in forests and how this can be realistically modelled. Although this is much more challenging than studying the circulation of radionuclides over timescales of just a few years, still this type of research is relevant to waste disposal in order to predict how radionuclides from underground sources will be brought up to the surface in new climate scenarios, and how they behave once they have reached the biosphere, including in response to extreme climate events. Hence, it is a priority to continue the conceptual development of this type of models.
- The plant-animal pathway is currently considered only for specific animals (e.g. wild boar). Kinetic food chain transfer models should be linked to the forest ecosystem (for example transfer to game animals e.g. red deer, moose) to cover this in more detail.
- One of the key issues in forest modelling over the last few years has been model transferability, for example the re-parameterisation of the available dynamic models for specific conditions (permafrost, subtropical, boreal, etc.). The development of regional parameter databases covering these environments is desirable if it can be justified on the basis of there being sources of artificial radionuclides in these environments that may potentially impact populations or the environment.

4.8 List of publications so far

Articles

1. Calmon P., Gonze M-A., Murlon C. (2015) Modeling the early-phase redistribution of radiocaesium fallouts in an evergreen coniferous forest after Chernobyl and Fukushima accidents. *Sci. Total Environ.*, 529, 30-39.

Reports

- IRA Forest MS 1 "[Modelers afternoon in Forest modelling](#)", COMET milestone report IRA Forest
- IRA Forest MS 2 "[COMET workshop for forest modellers and experimentalists](#)" COMET milestone report IRA Forest
- IRA Forest D1 "[Report and wish list to experimentalists on key input data/factors required for forest models](#)", COMET deliverable report IRA Forest



- IRA Forest D2 “Handbook – Guidance on the application of forest models on accidental contaminated sites”, COMET deliverable report IRA Forest – in preparation.

Abstract international conferences

- Vives i Batlle, J., Vandenhove, H., Gielen, S. (2014). Modelling water and ³⁶Cl cycling in a Belgian pine forest. In: Proc. ICRER 2014 - 3rd International Conference on Radioecology & Environmental Radioactivity, 7-12 September 2014, Barcelona, Spain. Electronic proceedings Session: 05.1. Radioactive Waste Management & Disposal, Paper OP-035. Available from: <http://radioactivity2014.pacifico-meetings.com/>.

5. IRA NORM

5.1 Introduction and objectives for IRA topic

Directive 2013/59/Euratom [EC, 2013] enforces the identification of classes or types of activities that involve NORM and which can lead to an increased exposure workers or members of the public to ionising radiation. For this purpose a list of industrial sectors involving NORM is provided including the oil and gas production, coal-fired power plants, the phosphate industry, etc. [Annex VI – EC, 2013]. Member States have to assure that these practices are subject to regulatory control by using a graded approach based on notification, registration and licensing. However, considerable flexibility is given to the Member States as they may also establish exemptions from notification for radioactive materials (including NORM) where the radionuclides activity concentrations do not exceed specified clearance levels laid down either by EC [Annex VII – EC, 2013] or IAEA [2014]. Moreover, notwithstanding these clearance levels, situations where NORM can enter water, significantly contaminating drinking water or affect aquatic life, need to be controlled.

From a radioecology point of view, the new BSS requirements come down to situations when NORM radionuclides can interact with environment. A graded system of authority control requires not only assessment of exposure to humans, but must also demonstrate that environmental criteria for long-term human health protection are met. Actually, the research associated to NORM has been somewhat limited compared to environmental contamination by radionuclides from the nuclear industry, which limits risk assessment. Conservative approaches suit regulators if estimated doses are well below regulatory criteria, i.e. dose limits or constraints. However, NORM often exceeds these criteria and regulatory authorities, industry operators and other stakeholders require more realistic and less uncertain assessment procedures to be used in assessment of risk to humans and wildlife.

The main goals of the IRA NORM group was a better protection of humans and the environment in existing situations (including legacy sites) and prospective analysis of potential risk caused by planned exposures, both related to NORM. Joint research activities to identify scenarios and processes ruling the behaviour of natural radionuclides released from NORM generating industry and to validate/improve assessment procedures or models of interest to all stakeholders were undertaken.

5.2 Background and status of the IRA field

With the new BSS, appropriate risk assessments need to be performed for exposure situations related to NORM, but there is scarce knowledge on the processes that determine NORM mobility and bioavailability and little (site specific) data are available to make assessments. The main reason for the non-existence of a consolidated branch of radioecology dedicated to NORM, is the “intrinsic” complexity associated with the large number of different NORM sources that can be found. While in



the more “traditional” radioecology involving artificial radionuclides the source types are relatively limited: weapon-tests, nuclear reactor accidents, releases from nuclear installations as reprocessing plants etc., mostly with a worldwide or regional impact, in the case of NORM the different sources of natural radionuclides potentially impacting the environment are more variable. There are hundreds of different NORM generating plants worldwide, both uranium production sites and civil activity like oil and gas production, mining, road- and tunnel construction. Most of them potentially have a local impact, depending of the type of activity performed. Usually, the impact is limited to a number of workers that are involved in processing and is mostly observed inside the borders of a plant. However, releases to the environment are often observed and the residues generated in each industrial NORM activity has in each case its own peculiarity depending of their physical and chemical form and the environmental compartment affected. Moreover, notwithstanding the origin, NORM always contains a suite of natural radionuclides subject sequential decay resulting in creation of different isotopes as well as elements differing in chemical and physical properties. Additionally, when doing an impact or risk assessment one has to remember that in contrast to artificial radionuclides, natural ones are ubiquitous in environment and only exposure exceeding natural background is subject to authority control. Additional complexity is introduced by the fact that natural radionuclides enclosed in NORM never occur alone, but other contaminants will usually be present, giving a multiple stressor scenario.

Considering external circumstances, NORM usually consists of residues or by-products from mineral resources processing often classified into a special category of waste that is very abundant comparing to “classical radioactive waste” (e.g. spent radioactive sources or even spent nuclear fuel) and has a completely different form. Solid NORM wastes are usually condensed and accumulated in a relatively small area and the observed contamination of surroundings is limited, even when different erosion processes are taken into account, in comparison to the areas that can be affected due to radioactive fallout (e.g. after a NPP failure). However, some industries generate residues containing natural radionuclides with high solubility in water bodies, while in other industries, a large fraction of the radioactive contamination is dispersed in the environment in the form of particles. Also important is that, except for offshore oil and gas platforms, the overwhelming majority of NORM generating industries are located in industrialised areas. These areas are already significantly altered by human activity and the presence of natural radionuclides is usually a less important source of stress to the environment than chemical pollutants or mechanical disturbances e.g. related to mining or transport and disposal of significant amounts of NORM. Legacy sites, where NORM have been disposed into a landfill without concerns for environmental protection, as no one was aware about natural radioactivity or environmental issues like we are today, are special cases. A significant difference is that secondary biological succession, which is not present on a fresh NORM landfill, is often observed on legacy sites.

5.3 Reason for this IRA within COMET

All the aforementioned issues must to be taken into account when planning an appropriate science-based environmental impact assessment for NORM. However, it will not be feasible to develop specific radioecological studies for all possible NORM activities and sites where the ecosystem can potentially be affected. NORM activities cover broad industrial sectors of a diverse character, where risk assessment methods are needed. As many of these industries have not been regulated in terms of radiation safety, “classical radiation protection” may not work so well when applied towards NORM. While clearance levels for solid NORM are given in EC [2013] or IAEA [2014], situations that lead to the release of NORM in liquid or gaseous form may occur and may require notification of the authorities. In these cases, the probability of an impact on the environment is bigger when compared with solid NORM and the exposure scenario is more complex. Therefore, from practical point of view, a



scientifically based assessment procedure is needed in order to meet authorisation process requirements. The expected solutions should avoid very simplistic and conservative assumptions that can lead to overregulation of sites, affecting in some cases the economical profit and viability of the NORM activity under consideration. The big challenge in NORM radioecology is thus to provide increasingly realistic evaluations, with lower uncertainties.

5.4 List of participants in the work

The institutions involved in the IRA NORM were GIG, NMBU, CIEMAT, SCK-CEN, US and BFS

5.5 How was the issues addressed within the IRA

The objectives of the IRA NORM have been:

- Identification of the sequence of key processes involving NORM impacts, finally leading to the radiological risk to the public and the environment.
- Completing data concerning the behaviour of natural radionuclides released from the NORM industry.

NORM's key safety issues have been analysed in terms of radioecology and environmental radioactivity assessment methods. To develop a systematic method for the identification of relevant NORM cases, various options of NORM presence were considered and assessed as to whether NORM can create an environmental burden.

Based on selected long-term observatory sites (Poland and Norway) new data were collected in order to identify key processes that influence the natural radionuclide behaviour in different environmental compartments such as soil and water/bottom sediments in a fresh water lake ecosystem.

5.6 Short summary of results so far

After extensive available literature review the *sine qua non* conditions when NORM may influence the environment, were identified. Namely, existence of big amounts of waste deposited and release of contaminated water or air. Based on this, the conclusions was drawn that three scenarios cover the majority of NORM situations: landfills, discharges of water and stack emissions. These basic exposure scenarios were analysed in terms of secondary processes determined by interaction of releases with the receiving environment. To define relevant scenarios for safety assessment studies the approach based on features, events and processes (FEPs) was used for defining the behaviour of a radionuclide in environment. The proposed approach was discussed in article [1] based on the examples of phosphogypsum heaps, stack emission and release of brines from coal mine and process water from oil and gas industry.

In NORM generating industries, the technology determines properties of residues and its way of release. Hence, identification of key processes ruling environmental behaviour of radionuclides must start at the plant. However, it is quite easy to identify the sequence of technological processes typical for the industry involved in mineral resources exploitation, recycling and processing, which is the main source of NORM. The three defined exposure scenarios were combined with these processes, and a structured approach to identification NORM industries able to affect environment seriously was developed (article [2]).

Studies at the NORM observatory sites have been performed, focusing on:

- contamination of terrestrial environment affected by discharge of mine water :



- 5 soil bulk samples (about 15 kg each) were characterised in terms of chemical, mineralogical composition and radionuclide concentrations; then they were prepared as a substrate for further experiments planned (leaching, TF) and sent to SCK-CEN,
 - rules of identification of NORM contaminated area based on measurement of radionuclides activity concentration in a soil profile (article [3]),
- characterisation of the current state of Rontok lake contaminated due to discharge of radium rich water from coal mines (abstract submitted to ICRER conference) considering:
 - comparison of radionuclide (^{226}Ra and ^{210}Pb) distribution in bottom sediments (1999-2016),
 - assessing the K_d for ^{226}Ra
 - ^{226}Ra and ^{210}Pb TF to selected biota and dose modelling using ERICA tool ,
 - application of FEPs for identification main processes determining radionuclide behaviour in contaminated fresh water lake ecosystem
- completion of Polish observatory site characterisation with qualitative and quantitative analysis of mine effluents discharged to the inland waters (abstract submitted to ICRER conference),
- testing of the conceptual model parameterised notification process proposed in article [1] and [2] on an example of coal mining industry, especially on aquatic environment affected by discharge of radium rich mine waters,
- studies of leachate from tunnel- and road construction in a Norwegian observatory site show increased metal and NORM concentrations in the surrounding environment and uptake in fish and benthic organisms. No effects are observed on fish, but there is effect on the benthic community. Modelling (ERICA and Cumulative Risk Assessment) possible chemical and radiological effects, showed risk towards crustaceans and the benthic community. This predicted risk includes both radiotoxicity (radionuclides) and chemical toxicity (metals), i.e. a multiple stressor scenario.

5.7 Future challenges and priorities

Obtained results are sufficient to apply a recommended graded approach in regulatory oversight of the coal mining industry, based on the concepts of exemption and clearance in the case of terrestrial environments and verified prospective analysis of long term effects in the case of fresh water lake environments. However, parameterisation of key processes that influence natural radionuclides speciation/fractionation and then transfer of this knowledge into a mechanistic model sufficiently complex to describe the radionuclide behaviour in the environment in each defined exposure scenario is needed. In the case of solid NORM leaching is the most important process. However, the diversity in contaminated NORM sites causes differences in NORM transfer due to different chemical, physical and physical-chemical properties. Therefore, there is a need to look into specific parameters influencing leaching of NORM.

In spite of generic processes responsible for radionuclides transfer from biotope to biocenosis having been identified in detail, ecosystem transfer of NORM is still not clearly understood and studies of bioavailability and possible effects of NORM together with other stressors (multiple stressor approach) is lacking.

There are still NORM sites and scenarios that need to be included into the research. Discharge of process water from oil and gas industry seems to be the most urgent NORM case to scrutinise.

In general, considering that natural radioactivity is ubiquitous in environment and clearance levels set does not cover all possible media (e.g. liquids or biota), NORM also needs to be put into a multiple stressor context, because NORM is never alone in the environment.



5.8 List of publications so far

Articles:

1. Boguslaw Michalik, Nathalie Vanhoudt, Rafael Garcia-Tenorio, Lindis Skipperud, An unified approach to environmental impact assessment caused by releases from NORM generating industry. (prepared for submission)
2. Boguslaw Michalik, Juan Carlos Mora Canadas, Identification and classification of key processes ruling environmental behaviour of radionuclides released from NORM industry. ((prepared for submission)
3. Boguslaw Michalik. NORM contaminated area identification using radionuclides activity concentration pattern in a soil profile. *Journal of Environmental Radioactivity* 169-170 (2017)p. 9-18

Conference presentations:

- B. Michalik, *NORM contaminated area identification using radionuclides activity concentration pattern in a soil profile*. V. Terrestrial Radionuclides in Environment International Conference on Environmental Protection, 17 -20th May 2016, Veszprém, Hungary
- B. Michalik, *Identification of key processes ruling environmental behaviour of naturally occurring radionuclides on an example of Polish Observatory Site*. Third International Conference on Radioecology and Environmental Radioactivity (ICRER 2014) Barcelona 7-12 September 2014
- B. Michalik, *Unified approach to environmental impact assessment caused by releases from NORM industry* presentation, Eighth International Symposium on Naturally Occurring Radioactive Material –NORM VIII, Rio de Janeiro, Brasil, October 18-21, 2016
- L. Skipperud et al. *Dispersion and transfer of nor & metals due to construction in u-bearing minerals*. Presentation, Eighth International Symposium on Naturally Occurring Radioactive Material –NORM VIII, Rio de Janeiro, Brasil, October 18-21, 2016
- B. Michalik, *Identification and classification of key processes ruling environmental behaviour of radionuclides released from NORM industry*, presentation , Second International Conference on Radioecological Concentration Processes (50 years later), Sevilla, Spain, November 6-9, 2016 (article under preparation)
- L. Skipperud et. al., *Dispersion, Transfer and Risk of NORM and Metals due to Construction in U-bearing Minerals*. presentation, Second International Conference on Radioecological Concentration Processes (50 years later), Sevilla, Spain, November 6-9, 2016 (article under preparation)
- L. Skipperud et al. *Challenges with naturally occurring radioactive material (norm) in road- and tunnel construction*. presentation, NETS, October 2016.
- *Natural radioactivity in Polish mines - an assessment of radium activity in formation water and mine affluents*. M.Wysocka, S. Chałupnik, I.Chmielewska, E.Janson. Abstract submitted to ICRER 2017
- B. Michalik, K. Samolej, M. Bonczyk, M. Wysocka, I. Chmielewska. *Long term behaviour of radium rich deposits in a lake ecosystem*. Abstract submitted to ICRER 2017



6. IRA Human food chain modelling

6.1 Introduction and objectives for IRA topic

This IRA focused on general agricultural systems. There is a close link with the “Terrestrial Food Chain and Dose Module” (FDMT) used in the two standard European Decision Support Systems (DSS) ARGOS and JRODOS (Müller et al., 2004), and with the SYMBIOSE simulation platform for performing radiological risk assessments used by IRSN (Gonze et al., 2011; Simon-Cornu et al., 2015). The overall objectives of this IRA were to improve human food chain modelling through regional customization of model parameters, by using novel and advanced statistical methods, and through studies of long-term dynamics of soil-to-plant transfer of radionuclides for different types of soil.

6.2 Background and status of the IRA field

Region-specific parameters are lacking for many countries, leading to uncertain predictions of doses from the human food chain following radioactive fallout. ARGOS and JRODOS use German default parameters in their integrated FDMT, whereas the default parameter values of SYMBIOSE are largely based on French conditions. Both FDMT and SYMBIOSE, however, can be adapted to other regions. Generally, the Mediterranean area has been understudied so far, and the derivation of region-specific values would greatly improve the model prediction capability. Other regions, such as the Nordic Countries, have climatic conditions and agricultural/grazing practices that differ significantly from those in central European countries. Specific soil properties can also have a considerable impact on the transfer of radionuclides. For instance, very high transfer of radiocaesium from soil to plants, followed by slow decrease in contamination over time, is typical for European wet peat ecosystems and needs to be specifically addressed. Regarding element- or nuclide-specific model parameters, most empirical data concern isotopes of Cs, I, and Sr. Thus, it is interesting to provide new transfer data for other radionuclides that have been less studied, but that could make a contribution to doses in the long run, such as isotopes of Pu, Am, and Tc. Finally, in recent years there has been substantial interest in the use of probabilistic modelling for deriving more robust parameter values for modelling purposes. In particular, Bayesian methods offer modellers and decision-makers options when faced with a lack of knowledge and data. The Bayesian Theorem provides a method for modification of probability in the light of new evidence. It allows for both prior knowledge (e.g. generic data) and site- or study- specific empirical data to be used. In a food dose assessment model, the use of Bayesian networks could aid the separation of uncertainty and variability in model parameters.

6.3 Reason for this IRA within COMET

The research undertaken aims at improving parameter values for human food chain modelling in line with challenge 1 of the strategic research agenda of radioecology (Hinton et al., 2013) – research line (RL) 2, and to some extent RL1 and RL3.

6.4 Participating organisations and research approach

The participating organisations in this IRA were CIEMAT, IRSN, NUBIP, NRPA, and STUK, and four tasks were specified for the work. Partners included in each task are also shown:

- **Task 1:** Derivation of human food chain parameter values that are appropriate for Nordic and Mediterranean ecosystems (STUK, NRPA, CIEMAT, IRSN).



- **Task 2:** Bayesian methods to derive more robust values where data are lacking or have a large variability (*IRSN, NRPA*).
- **Task 3:** Long-term dynamics of radiocaesium mobility and plant availability for peat soils with unusually high transfers (*NUBIP*).
- **Task 4:** Long-term dynamic soil-to-plant transfers for Tc-99, Pu, and Am (*NUBIP, NERC, IRSN*).

6.5 Short summary of results so far

In the first task, important parameters related to regional update were identified based on recommendations from the JRODOS or ARGOS communities – and to some extent on available sensitivity analyses (see Milestone COMET IRA-Human-M1). Generally, the most relevant parameters could be divided into four broad groups: Contamination of plants due to direct deposition, animal parameters, human consumption habits, and uptake from soil. The focus of D3 was on: (1) parameters of relevance to growing season and harvest periods of crops and grass – including seasonal development of leaf area indices (LAI), (2) animal feeding practice, and (3) human consumption of foodstuffs. JRODOS (STUK, CIEMAT), ARGOS (NRPA), and SYMBIOSE (IRSN) model runs using dry and wet deposition test scenarios were used to study the effect of parameter updates on e.g. time development for activity concentrations (Cs-134/137, Sr-90, and I-131) in selected foodstuffs and intake doses for different age groups (see Milestone COMET IRA-Human-M3).

The second task explored the use of Bayesian statistics through a study in two parts. In the first part (A), Hierarchical Bayesian models were developed for the parameter “weathering loss” – i.e. the time-dependent reduction of radionuclide concentrations in plant foliage due to wind, rain, plant growth, etc. This approach was applied on three data sets. The models developed in task 2 demonstrated the influence of plant species and/or type of radionuclide on weathering, depending on type of data set (i.e. iodine field studies, other radionuclides – laboratory or field studies). Part (B) was a test case (same as the dry case for task 1) using SYMBIOSE – incorporating results from (A) and from earlier studies on dry deposition (see Milestone COMET IRA-Human-M2). An article is under development regarding part (A). The work within task 2 is also part of a PhD thesis (Sy, 2016).

In the third task, a two-year laboratory experiment using peat-bog soils of Rokytné district (Ukraine) was performed. Soils from this area show very high radiocaesium bioavailability – resulting e.g. in high levels in milk from cows grazing in these areas. The present study compared root uptake of ‘freshly added’ and ‘old’ Chernobyl radiocaesium by herbaceous plants (*Juncus acutus* L.). Equilibrium was reached after 1–2 years (see Deliverable COMET IRA-Human-D1). A paper has been published in Ukrainian (Maloshtan et al., 2015).

The fourth task was a continuation of earlier experiments in the Chernobyl exclusion zone (CEZ). The aim of these studies was to reveal changes of transfer with time due to the radionuclide “ageing”, and thus improve the dynamic modelling of soil-to-plant transfers for Tc-99 and trans-uranic elements (Am-241, Pu-238,239,240). Soil-to-plant transfer data for several agricultural plants (lettuce, radish, wheat, potato) from four types of soil were provided. In addition, vertical migration / soil retention of Tc-99 was considered (see Deliverable COMET IRA-Human-D2). A paper has been produced concerning the Tc-99 part of this work and is currently in preparation for resubmission.

The use of results from the experimental work in tasks 3 and 4 to improve input parameters in FDMT and SYMBIOSE still needs to be evaluated.



6.6 Future challenges and priorities

Future challenges and priorities

Within COMET, the first priority under this IRA will be to finalise publications beyond deliverables and milestone reports (as mentioned above).

New foodstuffs have been difficult to implement in FDMT (as mentioned in Milestone COMET IRA-Human-M3). This issue needs to be sorted out in the near future, since local foodstuffs may play an important part in relation to regional adaptation. Moreover, regional updates in soil-plant transfer parameter values was not considered in our first task due to time constraints (see Deliverable COMET IRA-Human-D3). Since such parameters may significantly affect the long-term trends of e.g. Cs-137 they should be properly addressed. This also applies to the 'special case' of peat (or organic) soils used as pastures in task 3 (see Deliverable COMET IRA-Human-D1), where the perspective should be broadened by inclusion of data from other countries for the derivation of regionally appropriate transfer parameters – in line with suggestions in task 2 of the Human Food Chain Roadmap of the European Radioecology Alliance.

In parallel with the work presented here, the topic of regional adaptation is also considered within the on-going EURATOM-project HARMONE (funded by the Second call of OPERRA). Outputs from our work and HARMONE constitute the basis for further work within the Alliance – where 'radioecological regions' are defined and relevant model parameters derived, as described in task 2 of the Roadmap. All European areas will be considered, and partners of the roadmap not involved in HARMONE and/or COMET are also invited to contribute to the provision of regional parameter values. This work is expected to improve predictions on a regional basis and an enhanced ability to plan and predict the effect of remediation.

Other future challenges/priorities relevant for Human Food chain modelling could be validation of regional model updates by using real cases (i.e. deposition data and measured concentrations in relevant products) and handling uncertainty in FDMT – since this is presently not possible. Improvement of DSS through probability modelling is also addressed in the Human Food Chain Roadmap (task 4) – including the use of Bayesian statistics.

6.7 List of publications so far

Articles or other reports:

1. The PhD thesis of Moustapha M. Sy (2016) (partly financed by COMET) is found at: http://www.irsn.fr/fr/larecherche/formation_recherche/theses/theses-soutenues/dei/documents/2016-these-sy.pdf
2. Maloshtan, I. Polischuk, S., Khomutinin, Yu., Kashparov, V. (2015). Dynamics of Cs-137 uptake to herbaceous plants at the peat bog soils with abnormally high radiocaesium availability. Nuclear Physics and Atomic Energy v.16, No.3, p 263-272. The whole article in Ukrainian is available from: http://jnuae.kinr.kiev.ua/16.3/Articles_PDF/jnuae-2015-16-0263-Maloshtan.pdf.

Reports

- Thørring, H. Liland, A., Calmon, P., Dyve, J.E., Peltonen, T., Real, A., Simon-Cornu, M., Sy, M.M. (2015). [Derivation of human foodchain parameter values appropriate for Nordic and Mediterranean ecosystems – Sensitivity analysis of parameters in FDMT and SYMBIOSE](#). Milestone COMET IRA-Human-M1, 28 p.



- Sy, M.M., Hosseini, A., Liland, A., Simon-Cornu, M., Thørring, H. (2016). [Uncertainty and variability in food chain exposure by radioactive fallout. What can Hierarchical Bayesian modelling bring to the radioecology community?](#) Milestone COMET IRA-Human-M2, 59 p.
- Thørring, H., Dyve, J.E., Lahtinen, J., Montero, M., Simon-Cornu, M., Trueba, C. (2016). [Human foodchain modelling. Summary of results from the FDMT and SYMBIOSE model runs.](#) Milestone COMET IRA-Human-M3, 11 p.
- Kashparov, V., Maloshtan, I., Levchuk, S., Polichuk, S. (2015). [Analysis of long-term dynamics of radiocaesium mobility and plant availability in peat systems.](#) Deliverable COMET IRA-Human-D1, 14 p.
- Levchuk, S., Kashparov, Howard, B. (2015). [Long-term dynamic soil-to-plant transfers for Tc-99, Pu and Am.](#) Deliverable COMET IRA-Human-D2, 17 p.
- Thørring, H., Dyve, J.E., Hevrøy, T.H., Lahtinen, J., Liland, A., Montero, M., Real, A., Simon-Cornu, M., Trueba, C. (2016). [Set of improved parameter values for Nordic and Mediterranean ecosystems for Cs-134/137, Sr-90, I-131 with justification text.](#) Deliverable COMET IRA-Human-D3, 52 p.

7. IRA Particle behaviour

7.1 Introduction and objectives for IRA topic

For areas contaminated with radioactive particles, information on particle characteristics and weathering dynamics essential for ecosystem transfer is highly needed. The parameters of the models of dissolution for different radioactive “hot” particles (HP) in soils, deposition/disposal of radioactive waste (including NORM) and characteristics of contaminated soil/sediments (and at the drained areas) describe important aspects of the mobility and bioavailability of radionuclides. Refinement of these parameters by systematisation of existing data and by obtaining novel data on the long-term environmental behaviour of HP, as well as characterisation of hot particles and their matrix (composition and size distributions, crystalline structure and oxidation state, solubility in the soil and solutions, transfer of radionuclides to plants and into organisms) are very important for radiation protection of humans and the environment.

7.2 Background and status of the IRA field

Chernobyl radioactive fallouts are represented by a significant fuel component, i.e., particles of fine-dispersed nuclear fuel with different composition, and by a condensed component, formed during release or transport as a result of condensation of volatile fission products on surface of different particles. The presence of fuel particles (FP) in radioactive fallouts is the key feature of the Chernobyl accident. The near 30-km zone of the accident (about 2000 km²) was contaminated mainly by different fuel particles that contained the main part released from the reactor (3-4 tons of fuel). In addition to refractory (non-volatile) radionuclides such as uranium and plutonium, the particles also contained a significant amount of biologically relevant radionuclides such as ⁹⁰Sr. Fuel particles were also found at long distances from the Chernobyl Nuclear Power Plant (ChNPP) in many European countries including Scandinavia, about 2000 km from the source. Over time, fuel particles deposited in the environment were exposed to weathering processes and many particles (especially oxidised uranium fuel particles) have been dissolving, and this process still determines the long-term environmental impact and risks for ecosystems contaminated with radioactive particles. In contrast, fuel particles in the sediments of the Chernobyl NPP cooling pond have not dissolved during 30 years after the deposition due to low oxygen level and neutral pH of the media in the pond. At the end of 2014, after the start of the COMET project, the ChNPP cooling pond draining was initiated. This will most probably increase the rate of dissolution of fuel particles within the drained areas and the released particle associated radionuclides



will be included in biogeochemical fluxes and ecosystem transfer. Therefore, it was very important to increase our understanding of the mechanisms determining behaviour of radioactive fuel particles in the environment, and to enhance our ability to estimate the potential impacts of this on man and the environment.

7.3 Reason for this IRA within COMET

Refinement of model parameters for dissolution of fuel particles deposited in soils and sediments (and at the drained areas), which determine the mobility and bioavailability of associated radionuclides needed to be undertaken. This refinement was based on systematisation of existing data, as well as on the production of novel data on particle characteristics and on long-term environmental behaviour of Chernobyl fuel radioactive particles (solubility in the soil and sediments, transfer of radionuclides to plants and animals). This information was to be utilised to describe the function of the source term in different dynamic models of radionuclides behaviour in the environment and to estimate the exposure doses to humans and biota.

7.4 List of participants in this work

Participating organisations and research approach

- Ukrainian Institute of Agricultural Radiology (UIAR) of National University of Life and Environmental Sciences of Ukraine (UA -NUBiP of Ukraine)
- Norwegian University of Life Sciences (NO-NMBU).
NMBU is also partner in RATE and work in RATE concerning the Chernobyl particles will also be reported here.
- Institut de Radioprotection et de Surete Nucleaire (IRSN)

7.5 How was the issues addressed within the IRA

The objective of this IRA was to obtain parameters related to weathering and dissolution of radioactive particles and the subsequent leaching of particle associated radionuclides, depending on the properties of the environmental medium (pH, oxygen concentration, etc.) and on the physico-chemical properties of the contaminating particles (composition, size, structure, etc).

The IRA focused on:

- Chernobyl Fuel Particle behaviour in soil.
- Characterisation of fuel particles (FP) and associated dissolution kinetics, in particular on fuel particles in the Red Forest radioactive waste trench
- Identification of parameters essential for fuel particle dissolution in the Cooling Pond drained bottom sediments, including the characterisation of FP matrix
- Characterisation of radioactive particles containing NORM

The determination of the fuel particle dissolution rates in soil and sediments in natural conditions was based on autoradiography and on the comparison of the level of exchangeable forms of ⁹⁰Sr in soils (from Chernobyl fallout) and ⁸⁵Sr injected in water-soluble form into the soils. Following sampling of soil, grain and sediment, measurement of the total activities of radionuclides and ⁹⁰Sr exchangeable fractions, and autoradiography was performed. Sequential extraction procedures were applied for



assessing leachability/mobility, while a simplified procedure suggested in RATE, will be utilised for comparison purposes (comparing properties of particles released from different sources).

In 2015, the efficiency of the models on Chernobyl fuel particles dissolution, initiated 15 years ago, and the reliability of the predictions of the dynamics of radionuclides leaching, were verified in the experimental site situated in the Red Forest within the Chernobyl exclusion zone. At this site, radioactive waste was disposed into shallow sub-surface storages i.e., waste trenches overlaid with clean soils, covering an area about 2.5 km west from the ChNPP.

Assessments of the chemical forms of the radionuclides ^{90}Sr , ^{137}Cs , ^{238}Pu , $^{239,240}\text{Pu}$, ^{241}Am as well as the composition of fuel particles in bottom sediments of the Cooling pond of the Chernobyl nuclear power plant (CP ChNPP) were also carried out. An experimental field plot for the simulation of drainage of the sediment and their exposure under natural conditions was created. For this purpose, samples of the bottom sediments were collected in the northern-western part of the Cooling pond at a depth of 5–7 m by a bottom sampler. On the drained sites of the ChNPP cooling pond, six research points were chosen. Thus, the model experiments should elucidate the dynamics of radionuclides species retained in the bottom sediments of the Cooling pond, mimicking the dissolution processes taking place during draining when exposed to the natural conditions.

The water level in the ChNPP cooling pond started to decrease in September 2014. Today, the water level has decreased by 4.5 m. In this regard, monitoring the dynamics of chemical forms of radionuclides on drained sites was started.

Radioactive particles containing NORM focuses on the characterisation of residues of uranium ore processing localised in the tailings of Pridneprovsky Chemical Plant (PChP). Samples were collected at various depths of the three tailings during the “dry” auger drilling of the boreholes. The physical-chemical characteristics (pH, moisture content, dispersal composition) and the hot particles properties (small, discrete, high α -active particle or cluster) in sludge sampled in the PChP tailings, as well as the chemical composition and the total contents of the uranium series radionuclides in the waste samples were studied. Similarly, NORM particles from a thorium rich area in Norway, were characterised using advanced 2D and 3D synchrotron microscopic techniques. Analyses showed that a series of metals are included in the NORM particles, and that radioactive elements exist as small inclusions within the entities.

7.6 Short summary of results so far

In the years 2013-2014, the efficiency of the models on Chernobyl fuel particles dissolution, elaborated 15 years ago, and reliability of their predictions for the dynamics of plants contamination with ^{90}Sr , were verified. The theoretical/modelling estimates of 1997-2000 of the exchangeable fraction of ^{90}Sr activity in soils with different pH within the Southern trace of fallout coincide with the experimental results obtained in 2013-2014. The fraction of ^{90}Sr activity present in the biologically available form has reached its maximum values for the post-accidental period due to fuel particles dissolution. The monotonic increase of the concentration ratio (CR) of ^{90}Sr observed in the grain grown in the survey area was due to dissolution of the fuel particles, which coincides well with theoretical forecasts.

The main results include identification of the radionuclide source term: the description of the physical and chemical properties of the fuel particles encountered in the waste trench, and a model of fuel particles dissolution and subsequent radionuclide leaching into the soil solution inside the waste storage site under natural conditions. The theoretical/modelling estimations of 2000-2001 of the



exchangeable fraction of radionuclide activity in soils coincide with the experimental results obtained in 2015. The data show that the mobile form of ^{90}Sr in the trench has reached the maximal value.

According to the obtained results, the predominate fraction (> 98 %) of ^{137}Cs , $^{239+240}\text{Pu}$ and ^{241}Am in sediments from the Cooling Pond (CP ChNPP) after drying and 26 months exposure under natural conditions on the experimental site is still in a non-exchangeable forms. The fraction of ^{90}Sr as exchangeable species may have reached 10 %. The low rate of radionuclides leaching from modified sediment from the Cooling Pond is caused by a prolonged slightly alkaline pH of the medium. In turn, this is due to the presence of a large number of remnant shells of zebra mussels in the bottom sediments of the pond. Hence, a sharp increase of the radionuclide mobility should not be expected for the newly contaminated sediments. Probably, this fact indicates that parts of the radionuclides are kept in a chemically very stable form within particles, and for this reason these radionuclides will not be transferred into the environment for many years.

By autoradiography using track detectors the homogeneity of the distribution of α -emitting radionuclides in the U containing sludge of PChP tailings has been investigated. Results showed a homogeneous background with the distribution of α -activity single spots with significantly higher activity concentration of α -active radionuclides. Analysis of the obtained information indicates the presence of small, discrete and high α -activity particles (or cluster) in the sludge of the tailing PChP. The size of detected particle ranged from 2 to 20 μm .

7.7 Future challenges and priorities

The study of the physical and chemical properties of the chemically extra stable U-Zr-O fuel particles encountered in the waste trench will be performed using advanced technologies, in order to improve the long-term predictions of radionuclides behaviour in the environment. Previous investigations have showed that the weathering rate would depend on the crystalline structure and the oxidation state of the carrying U matrix of the particle. Thus, such information will be important for understanding the underlying mechanisms of particle weathering and dissolution.

The obtained results indicate very slow dissolution rate for fuel particles in drained sediments under natural conditions and that the radionuclides are heterogeneously distributed in the sediments of the ChNPP cooling pond. In this regard, to obtain reliable parameters of the FP dissolution models, more detailed studies and regular monitoring observations over a longer period of time should be performed.

The main hazards associated with α -active hot particles of NORM are inhalation and ingestion routes of entry. Thus, the study of the characteristics of the α -active hot particles in tailing dumps as well as the weathering kinetics should be continued (AMAD, solubility class, desolation in environment, etc.).

7.8 List of publications so far

Articles or other reports:

- Otreshko, L.N., Levchuk, S.E., Yoschenko, L.V. Concentration of ^{90}Sr in grain on fuel traces of the Chernobyl radioactive fallout //Nuclear Physics and Atomic Energy, 2014, V.15, N.2, p.171-177 (Ukr.)
- Otreshko L. M., Zhurba M. A., Bilous A. M., Yoschenko L. V. ^{90}Sr and ^{137}Cs content in wood along the southern fuel trace of Chernobyl radioactive fallout //Nuclear Physics and Atomic Energy, 2015, V.16, N.2, p.183-192(Ukr.)



- Protsak V. P., Odintsov O. O. Assessment of forms finding of Chernobyl radionuclides in bottom sediments of cooling pond of the ChNPP //Nuclear Physics and Atomic Energy, 2014, V.15, N.3.

8. ICRP reference sites - developing alternative transfer models for wildlife

8.1 Introduction and objectives for IRA topic

A large number of organisms and radionuclides have to be considered within assessments of the potential impact of radioactivity on the environment. It should, therefore, not be surprising that empirical data to derive transfer parameters for many organism-radionuclide combinations are lacking. This lack of data is especially the case for the Reference Animals and Plants (RAPs) which form the basis of the ICRP framework (ICRP 2008, 2009) that is likely to be used by many EC member states in the future. To compensate for the lack of data, in ICRP Publication 114 (2009), it was suggested that a series of sites be selected and sampled to determine transfer parameters for the various RAPs. At the start of the COMET project one such study had been conducted in a forest ecosystem in the UK (Barnett et al. 2014) by COMET partner NERC-CEH and NRPA were beginning a similar study at a Norwegian site.

Whilst studies such as Barnett et al. (2014) provide a considerable amount of data for poorly studied organisms and elements/radionuclides the resultant transfer parameters are likely to be highly site specific. However, if data were available from a number of such sites it should enable the parameterisation of an alternative modelling approach, in part developed within the STAR Network of Excellence (Beresford et al. 2013, 2016). The approach uses a Residual Maximum Likelihood (REML) mixed-model regression and in-effect removes the effect of site; 'site' contributing to the considerable variability observed in existing concentration ratio (CR_{wo}) databases. The output of the REML model is the relative concentration values for organisms taking into account inter-site variation. This to enables predictions to be made for organisms for which no data are available from those for which data are available.

Another criticism of the existing data for the transfer of radionuclides to wildlife is their considerable geographical bias, most data originating from Europe or North America. A bias is true even within Europe, with virtually no, if any, data originating from Mediterranean ecosystems in the international database (Copplestone et al. 2013) used by ICRP and IAEA to derive their recommendations.

8.2 Objectives and hypothesis

The overall objective of the IRA was to establish taxonomic models for the estimation of the activity concentration of radionuclides in terrestrial wildlife. This was to be accomplished through sharing of datasets and targeted sampling and analyses. In addition, the IRA had the objective of significantly improving the empirical transfer parameter data available for the ICRPs RAP.

The hypothesis the IRA aimed to test was: *radionuclides activity concentrations in (terrestrial) wildlife can be predicted using a taxonomic model which takes out the effect of site.*

8.3 Why this IRA within COMET

The IRA contributed to COMETs aim to reduce model uncertainty through fostering the sharing of infrastructure (Chernobyl Observatory), on-going studies (UK funded TREE project and study by NRPA in Norway), and datasets (Barnett et al. 2013), together with targeting additional activities to address the issues highlighted above. The results will contribute to the Radioecology SRA's research lines 1 to



3 by providing the data to derive a taxonomically based model of radionuclide transfer independent of site specific factors.

8.4 List of participants in the work

NERC-CEH, Chernobyl Center, NRPB, Fukushima University, CIEMAT; in collaboration with the TREE project (<http://tree.ceh.ac.uk/>) and the universities of Extremadura (Spain) and Salford (UK)

8.5 How was the issues addressed within the IRA

The sampling approach as described by Barnett et al. (2014) has been adopted for this work. This involves the targeted collection of species falling under the definitions of the ICRPs RAPs (which are defined at the taxonomic level of family) from a single sampling site; representative soil samples (0-10 cm layer) were also collected from the sampling sites. Radionuclide and stable element concentrations were subsequently analysed in the collected samples. Where species falling outside the definition of the RAPs were caught these were also retained for analyses from some of the sampling sites.

As already noted above the work was conducted in collaboration with some nationally funded project. The sites for which data are available are:

- Chernobyl Exclusion Zone (Ukraine)
- Extremadura (Spain)
- Northumbria Site K (United Kingdom)
- Northumbria Site H (United Kingdom)
- Tjøtta (Norway)
- Yamakiya (Japan)

The Chernobyl Exclusion Zone (CEZ) site is the location of a former village on the edge of the Red Forest (trees were not killed in 1986); the CEZ was selected as a sampling area on the basis of the ability to measure a range of radionuclides in wildlife. The site in Extremadura was selected on the basis of the lack of data for Mediterranean ecosystems in international databases. The site is a semi-natural grass land (dehesa) which is managed for the hunting of red deer (*Cervus elaphus*) and wild boar (*Sus scrofa*). The two Northumbrian sites are managed coniferous plantations; Site K is dominated by Sitka spruce (*Picea sitchensis*) whilst Site H comprised mixed coniferous species. Tjøtta is a 11.3 km² island off the coast of central Norway. The Yamakiya site is a 7 ha area of (formerly managed) Japanese cedar forestry located 35 km north-west of the Fukushima Daiichi nuclear power plant complex.

Reference Animals and Plants and other sample types collected from the three sites are presented in Table 1.

8.6 Short summary of results so far

Results are available for ¹³⁷Cs and ⁹⁰Sr in all samples collected in the CEZ; ²⁴¹Am, ^{239,240}Pu and ²³⁸Pu activity concentrations are available for selected samples. Samples from Norway, Spain and the two UK sites have all now been analysed for stable element concentrations using ICP-MS; some gamma-emitting radionuclide results are also available for the UK and Norwegian sites. Results for the Norwegian site have recently been published (Thørring et al. 2016). For the Japanese site only ¹³⁷Cs activity concentrations have been determined.



8.7 Future challenges and priorities

Data from most of the studies has only become available in the last few months and that from some of them is still being processed and quality controlled. Once this is completed then the data will be subjected to REML analyses to define taxonomic models initially at the family level (given the ICRP define the RAPs at the level of family. We will perform the REML modelling for Sr and Cs first as the REML model outputs derived from this structured sampling can be compared to those coming from REML analyses of data from the international wildlife transfer database (Copplestone et al. 2013). We will then aim to produce REML models for some of the less studied radionuclides.

The work will also input to the activities of ICRP Task Group 99 which is currently preparing a number of reports which will include transfer parameters for the ICRP RAPs. The studies outlined here (together with Barnett et al. 2014) significantly improve the availability of RAP specific data as previously compiled in ICRP (2009).

Table 1 Representative RAP samples collected (underlined); other sample types are also listed.

Spain	Northumbria ¹	Japan	Norway	Chernobyl
<u>Pine Tree</u>	<u>Pine Tree</u>	Other tree species	<u>Pine Tree</u>	<u>Pine Tree</u>
<u>Wild Grass</u>	<u>Wild Grass</u> & other vegetation	<u>Wild Grass</u> & other vegetation	<u>Wild Grass</u>	<u>Wild Grass</u>
<u>Earthworm</u>	<u>Earthworm</u>	<u>Earthworm</u> & other earthworms spp. ²	<u>Earthworm</u>	<u>Earthworm</u>
<u>Deer</u>	<u>Deer</u>	-	<u>Deer</u>	-
<u>Rat</u>	<u>Rat</u> & vole species	-	<u>Rat</u>	<u>Rat</u> (2 species) & other small mammals
<u>Frog</u>	<u>Frog</u> & toad	<u>Frog</u>	<u>Frog</u>	<u>Frog</u> & other amphibians
<u>Bee</u>	<u>Bee</u> & other insects	<u>Bee</u> & other bees ²	<u>Bee</u>	<u>Bee</u> & other insects
<u>Duck</u>	-	-	Duck	-
-	-	Liverworts, mosses, fungi	-	-

¹The same RAPs and other samples were collected from both Northumbrian sites; ²not all earthworms or bees fall within the ICRPs family level definition of the respective RAPs.

8.8 List of publications so far

Articles

We have been invited to submit refereed papers on the CEZ and Spanish sites to two special issues of the *Journal of Environmental Radioactivity*; submission dates are spring 2017 for both issues.



Conference papers in-press:

J. Guillén, N. A. Beresford, A. Baeza, M. Izquierdo, M.D. Wood, A. Salas, A. Muñoz-Serranoa, J.M. Corrales-Vázquez and J.G. Muñoz-Muñoz. Submitted. Transfer parameters for ICRP's Reference Animal and Plants in terrestrial Mediterranean ecosystems. International Conference On Radioecological Concentration Processes (50 years later); Seville November 2016. Extended abstract.

J. Guillén, N. A. Beresford, A. Baeza, M. Izquierdo, M.D. Wood, A. Salas, A. Muñoz-Serranoa, J.M. Corrales-Vázquez and J.G. Muñoz-Muñoz. Submitted. Seasonal variation of concentration ratios for ICRP's Reference Animal and Plants in terrestrial Mediterranean ecosystems. International Conference On Radioecological Concentration Processes (50 years later); Seville November 2016. Extended abstract.

9. Conclusions

The COMET IRAs have contributed to improved parameterisation of key processes controlling the transfer of radionuclides, with a specific focus on dynamic modelling approaches, within each chosen research area. In this sense, the project has contributed a higher predictability of the phenomena observed, and it has contributed significantly to quantify and constrain uncertainties. The issue of whether uncertainties really have been reduced and the degree to which predictability has been improved, will be addressed in D3.7 (Final report of WP3 activities), both for each IRA separately and for the WP as a whole. Further work is however needed with respect to validation of the resulting models, as well as quantification of the degree of uncertainty reduction provided by these model improvements.

The COMET IRAs have also brought people not working together earlier into close collaboration, working together towards common goals. Furthermore, the IRAs made an important contribution to the preparation of COMET roadmaps, bringing the researchers even further together in the future. In this respect, the important goal of further integration of radioecology at the European level has been substantially advanced.

Each COMET IRA has identified future challenges and priorities of importance for future research within the field of radioecology/environmental radioactivity. These priorities should be the focus in future projects and calls.



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