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Executive Summary

The overall aim of work package 3 of COMET was to strengthen the pan-European links between the radioecology and the emergency and post-accident communities and to undertake joint research activities to improve and validate radioecological models of interest to both communities. The research in WP3 was centred on improving parametrisation of key processes controlling the transfer of radionuclides, with a specific focus on dynamic and mechanistic modelling approaches. The research was initially conducted within topical working groups on marine, forest, human food chain, NORM, and wildlife transfer modelling, as well as a group on particle behaviour. After an open call, these groups were complemented by two projects, FRAME and RATE, in which additional investigations on the mechanisms radionuclide transfer in the marine environment and on particle behaviour, respectively, would further reduce the uncertainties associated with key transfer processes.

In work package 3 of COMET, significant contributions have been made to improve radioecological modelling capabilities, both through model development and through improvements in model parametrization. For example, kinetic transfer models were improved to better capture the dynamics of post-accident situations, and more sophisticated trophic transfer models were developed in the marine modelling group, while an advanced hierarchical Bayesian statistical model taking parametric uncertainty was developed in the human food chain modelling group. In forest modelling, where less comprehensive existing models were available, a handbook on forest modelling was prepared. New modelling approaches were also explored, such as an alternative transfer model for wildlife.

Both experiments and fieldwork contributed towards improved model parametrization. For example, the scientific cruises conducted under the FRAME project made data available for both marine model improvement and validation, while experimental work in the human food chain modelling and particle behaviour groups provided data on dynamic parameters for their respective models. Furthermore, data from ICRP reference sites contributed to the alternative wildlife transfer model and data from the Polish Observatory site was used to evaluate a framework for structured impact assessment from NORM generating industries.

Work package 3 of COMET also fostered integration, within radioecology in Europe and internationally, as well as between radioecology and other radiation protection communities and other scientific disciplines. We covered topics highly relevant for emergency and post-accident communities, such as updating decision support systems with regional parameters. We engaged in collaboration with researchers from countries where major nuclear accidents had occurred, such as Japanese researchers in relation to the Fukushima accident in the FRAME project and Ukrainian researchers in relation to the Chernobyl accident in the particle behaviour and human food chain working groups. Furthermore, we conducted interdisciplinary research, such as a very fruitful collaboration with oceanographers in the FRAME project.

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Objectives and Scope

In the present deliverable, we report on the scientific output from work package (WP) 3 in the COMET project. Hence, the focus of this report will be the Initial Research Activity (Task 3.1) and the research projects FRAME (Task 3.3) and RATE (Task 3.4), as well as the work on integration, validation and implementation of RTD activities (Task 3.5). The output from task 3.2 (Identification of common research needs for radioecology and the emergency and post-accident communities) has been reported previously in COMET deliverables D2.1 "Towards a first phase Radioecology Alliance RTD roadmap and implementation plan as input for the preparation of the Competitive Call organised in collaboration with OPERRA " and D1.2 "Flex Fund Report and Amended Workplan" and is thus not discussed further in this document.

Introduction

The overall objectives of work package 3 were to strengthen the pan-European links between the radioecology and the emergency and post-accident communities and to undertake joint research activities to improve and validate radioecological models of interest to both communities, to better protect humans and the environment in existing, planned and emergency exposure situations.

The radioecological focus of WP 3 was to improve the estimation of exposures and doses to humans and wildlife. Hence, the work in WP 3 of COMET relates to Challenge One of the Strategic Research Agenda of Radioecology (Hinton et al, 2013): "*To Predict Human and Wildlife Exposure in a Robust Way by Quantifying Key Processes that Influence Radionuclide Transfers and Exposure"* and its four associated research lines:

1. Identify and mathematically represent key processes that make significant contributions to the environmental transfers of radionuclides and resultant exposures of humans and wildlife.
2. Acquire the data necessary for parameterisation of the key processes controlling the radionuclides.
3. Develop transfer and exposure models that incorporate physical, chemical and biological interactions, and enable predictions to be made spatially and temporally.
4. Represent radionuclide transfer and exposure at a landscape or global environmental level with an indication of the associated uncertainty.

The research work in WP 3 consisted of two components. An Initial Research Activity (IRA) was initiated by the original partners of COMET from the start of the project and two research projects were later added after an open call, bringing several new partners into the consortium.

The Initial Research Activity aimed to improve parameterization of key processes that control the transfer of radionuclides in the environment, with a specific focus on dynamic modelling approaches. The work was structured in six topical working groups: marine modelling, forest modelling, human food chain modelling, NORM modelling, particle behaviour and ICRP reference sites for alternative wildlife transfer modelling. A detailed plan for the goals of these groups and their associated research projects, including participants and timing, was elaborated and published as Deliverable D3.1. As the project progressed, this plan was revised at a meeting for all WP 3 participants in Oslo, Norway in January 2015, and synergies with the two projects from the open calls were taken into consideration.

The two research projects added to WP3 in 2014 after winning the COMET Competitive Research and Technological Development Call, were FRAME (The impact of recent releases from the Fukushima nucleaR Accident on the Marine Environment) and RATE (RAdioactive particle Transformation processes). These two projects addressed the marine modelling and particle behaviour topics of the

call, respectively. Hence, where appropriate, efforts were made to integrate the work done in these projects with the existing work in the relevant IRAs.

In the present report, we present the scientific achievements of WP 3 by topic. Furthermore, for each topic, we describe how these results have improved radioecological modelling capabilities and, where appropriate, compare earlier modelling approaches with our new ones. Model validation, as well as data requirements for further validation where this is needed, is also addressed. Finally, the impact of the results, both for the radioecology and emergency and post-accident communities is also discussed.

In addition to integrative activities described above, scientifically oriented COMET position papers based on the research in WP3 are also under production for the Marine modelling, NORM modelling and Particle behaviour topics. These position papers are aimed at publication in international peer reviewed journals and will thus not be further elaborated in this report. The particle position paper will be published in Journal of Environmental Radioactivity (*In press*) in 2017.

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Marine Modelling

1.1 Initial Research Activity

1.1.1 Background

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 a marine group has emerged within the COMET project. The main lines of research from Challenge One of the ALLIANCE SRA directly relevant to Fukushima were (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, (b) acquire the data necessary for parameterisation of the key processes controlling the transfer of radionuclides and (c) develop transfer and exposure models that incorporate physical, chemical and biological interactions, and enable predictions of radionuclide concentration to biota to be made, both spatially and temporally. This provided the reference point for the activities of the Initial Research Activity for Marine Radioecology (IRA). Such activities focussed on how modelling tools could be adapted to the highly dynamic post-accident situations, improving the existing models by making them more dynamic and process-based, and trying to make more realistic estimates of marine dispersion and transfer to biota with these models in order to assess radiological consequences, using improved data for parameterisation, calibration and validation of models.

Many questions about the behaviour and the fate of radionuclides released after Fukushima accident in the marine environment appeared unsolved. There has been an ongoing need to obtain reliable data to assess and follow this radiological state. The objective of the IRA was to improve predictions of concentrations in, and exposures of, marine biota and humans through dynamic modelling, trophic transfer modelling and by combining transfer to biota with sediment modelling. The philosophy adopted in the Marine Radioecology IRA was to consider as complementary operational models based on simple accumulation/depuration process and more realistic models taking into account trophic transfers through food webs.

Therefore, the marine group focused on concepts and models that could be used in an accidental situation to assess transfers and distribution of radionuclides between the different marine compartments. Radioecological transfer modelling (transfers to biota and sediments) was highlighted as the key topic, with plans to improve estimates of concentrations in biota.

1.1.2 Methods and materials

The understanding of contamination levels and radionuclide distributions in the environment, along with prediction of their future evolution requires the use of modeling tools and analyses of detailed monitoring data. Two main radioecological modelling topics are to take into account transfers and exchanges between water and sediments, and ii) transfer to biota.

Decision support systems for emergency situations usually include fairly simple models. Other, more sophisticated, models should be used in a second step to evaluate and predict more accurately the post accidental situation. The main focus of the Initial Research Activities of this group has been centered on emergency and post-accidental issues, and on the modelling tools adapted to such situations. Existing time-dependent models were chosen as a basis for the work, with a view to improving some of them to achieve more sophisticated models. The IRA work was divided in 3 tasks:

- Implementation and use of classical radioecological models based on dynamic transfer equations to evaluate concentrations in marine organisms (macroalgae, fish, molluscs, crustaceans) and improvement of radioecological parameters (concentration factors and

single or multicomponent biological half-lives) for ^{137}Cs, ^{134}Cs and ^{90}Sr. This task included comparisons with observed data series from Fukushima.

- Inclusion of sediment processes in dynamic transfer modelling. This work has followed ongoing studies adapting an existing dynamic model (D-DAT,). The tool has been completed and tested with the available data to see if it is capable of reproducing activity in sediment to give a more realistic calculation of concentrations in biota and improving the dose models for humans and biota based on Kd values.
- Process oriented modelling for mid- and long-term predictions: ecological and environmental processes. This included modelling trophic transfer to pelagic fishes, including food-web transfers and establishing if there is real potential for biomagnification in the Fukushima food chain.

1.1.3 Results

Two operational kinetic radioecological models were improved during the Initial Research Activity:
The first one is the IRSN model STERNE, which is now optimised to obtain fast results for the first analysis of any marine radiological situation. STERNE calculates both radionuclide transport using advection and diffusion equations offline from hydrodynamic calculation, and radioecological transfers to biota with a simple model based on dynamic transfer equation. Required radioecological parameters (concentration factors and single or multicomponent biological half-lives) have been compiled for some important radionuclides and for generic marine species (macroalgae, fish, molluscs, crustaceans). Dispersion and transfer calculations are carried out simultaneously on a 3D grid (Duffa et al., 2015). STERNE is designed to be operated for a first estimation in case of emergency release to the sea.

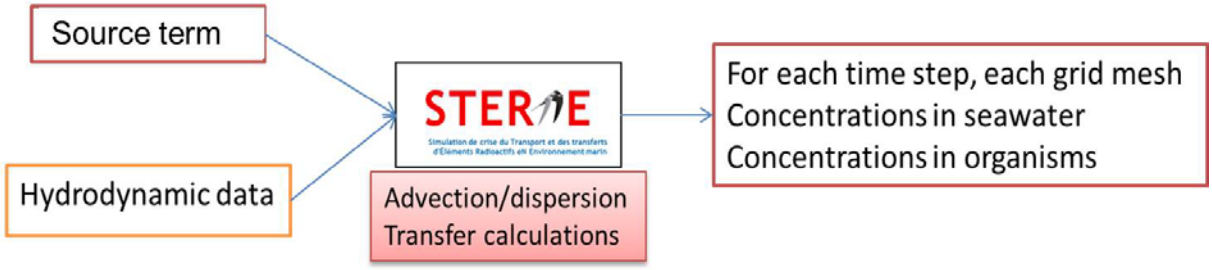

Figure 1.1.1: Schematic diagram of STERNE implementation principle

Concentrations in living organisms are calculated at each time step of the dispersion calculation as follow:

$$\frac{dC_o(t)}{dt} = k_i \cdot C_w(t) - (\lambda_p + k_o) \cdot C_o(t)$$

EQUATION 1.1

where $k_i = (k_o + \lambda_p).CF$

C_o is the activity concentration in the organism (Bq.kg^{-1} fresh weight)
C_w is the activity concentration in seawater (Bq.l^{-1})
k_i is the uptake or accumulation rate constant (d^{-1})
k_o is the elimination or depuration rate constant (d^{-1})
λ_p is the physical decay constant (d^{-1})
CF is the concentration factor (l.kg^{-1} f.w.)

In STERNE code, this equation is computed as following for a time step i (see Fievet and Plet, 2003 for details):

$$C_{o(i)} = a \times C_{o(i-1)} + FC \times (1 - a) \times C_{w(i)}$$

EQUATION 1.2

Where *i* is the time increment.
With $a = e^{-T(k_o+\lambda_p)}$
and $T = t(i) - t(i-1)$ the constant time step duration.

In order to refine this one compartment dynamic model and adapt it to post-accident conditions (for which various studies report multiple depuration rate constants), the STERNE simulation tool allows users to combine two independent compartments as A.Co1+B.Co2. A and B values are between 0 and 1 and A+B=1. Each compartment Co1 and Co2 has its own transfer parameters (CF and Tb).
All required radioecological parameters (concentration factors and single or multi-component biological half-lives) are compiled from literature (IAEA, 2004, Gomez *et al.,* 1991 and Vives i Battle *et al.*, 2007) for main radionuclides and generic marine species (fish, molluscs, crustaceans, algae). These default values can be change by the user if necessary.

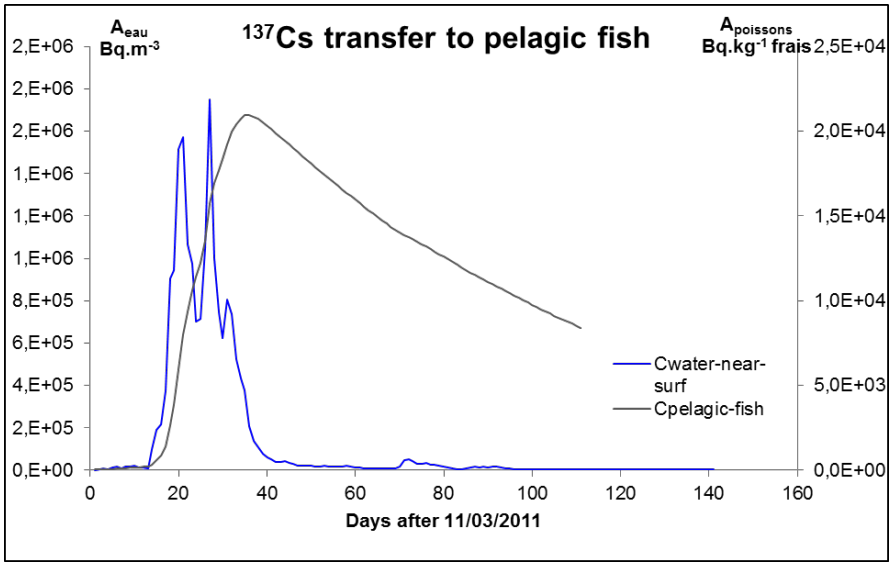

Figure 1.1.2: Example of STERNE calculation results

The second model was the SCK•CEN D-DAT (Dynamic Dose Assessment and Transfer), which 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 and sediments and the $T_{B½}$ (biological half-life) of radionuclides in the biota. As for STERNE, a dual $T_{B1/2}$ approach can be calibrated to predict acute and long-term levels. The model calculates activity concentrations of ^{131}I, ^{134}Cs, ^{137}Cs and ^{90}Sr in seabed sediment, fish, crustaceans, molluscs and macroalgae from surrounding activity concentrations in seawater, with which the organism exchanges radionuclides. Although the model does not explicitly include the incorporation of radionuclides via food intake, this process is captured indirectly by imposing the condition that the concentration ratio at equilibrium equals the concentration factor (CF), which incorporates all processes including ingestion. D-DAT can also be used to calculate internal and external dose rates (Vives i Batlle, 2014, Vives i Batlle, 2016).

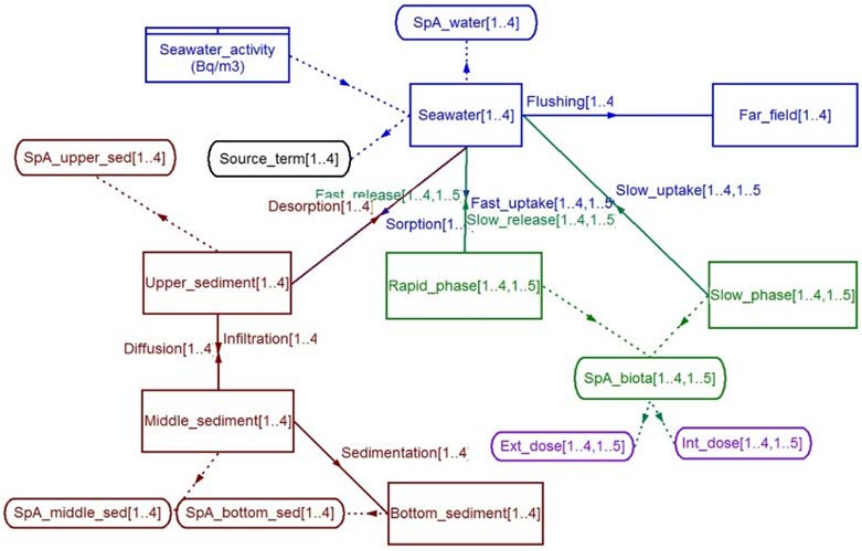

Figure 1.1.3: Schematic diagram of D-DAT model

Model radioecological parameters were calibrated using Fukushima ^{137}Cs data. Results obtained then were compared with radionuclide measurements for macroalgae and molluscs. ^{137}Cs activities prediction are consistent with UNSCEAR data for the 50 km from the FDNPP. ^{137}Cs prediction matches fish data near FDNPP from 450 days after the accident (Figure 1.1.4).

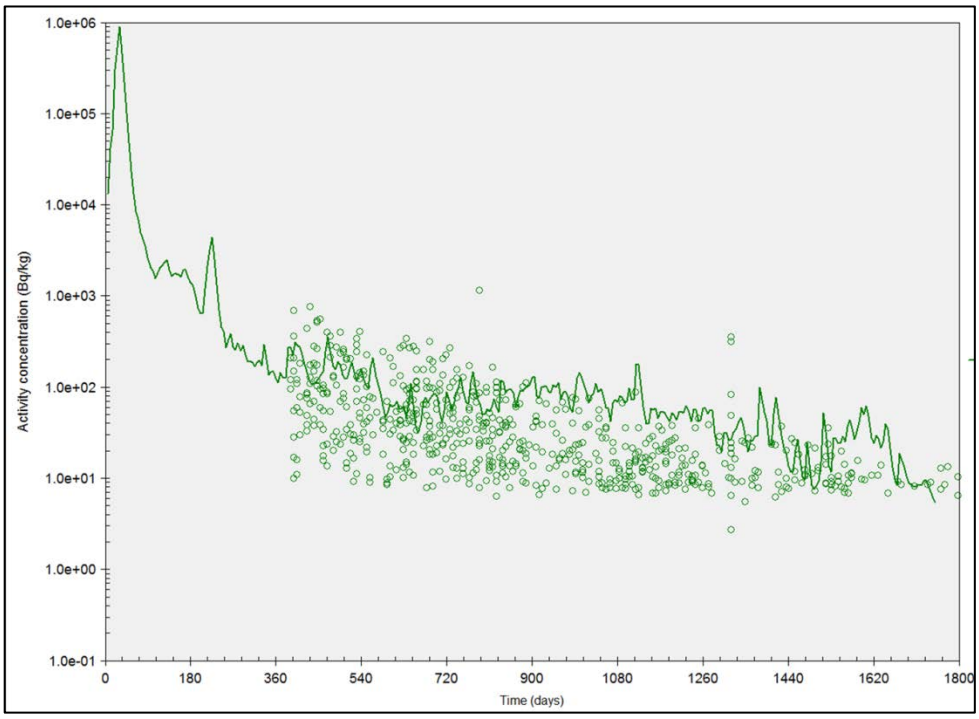

Figure 1.1.4: Long-term comparison between D-DAT and model-predicted ^{137}Cs in greenling from Fukushima (MAFF data)

The production of an advanced version of D-DAT with the inclusion of sediment processes in dynamic transfer modelling was performed. The model is now adapted to include depletion of radionuclides adsorbed on to 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). In its current implementation, D-DAT comprises a sub-model of three sediment layers (Fig 1.1.5).

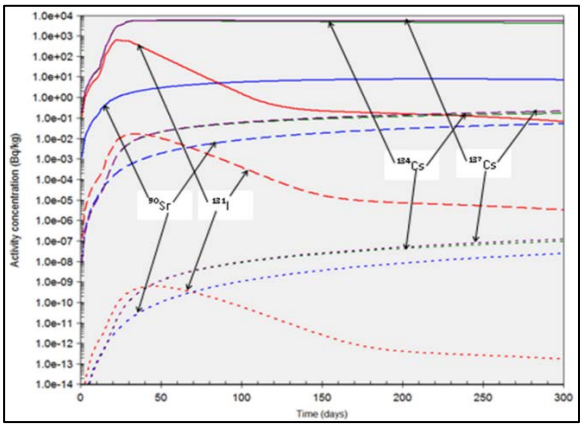

Figure 1.1.5: Activity in 3 sediment layers: ^{90}Sr (blue), ^{131}I (red), ^{134}Cs (green) and ^{137}Cs (purple) in surface (——), middle (– – –) and deep (·····) sediment at <2 km from the FDNPP

Calculations provide good results: ^{134}Cs activity in top layer in the vicinity of the FDNPP (an average for all sediments within a distance of 2 km) is 5600 Bq kg^{-1} 50 days after accident. This matches with measurement data.

The third IRA task was established on process oriented modelling for mid and long-term predictions, factorising ecological and environmental processes. This involved modelling trophic transfer to pelagic fishes and establishing if there is real potential for biomagnification in the Fukushima food web. Modelling tools for simulation of transfer to biota were improved, such as the pre-existing NRPA model (Iosjpe et al., 2016a) or the IRSN trophic transfer model (Belharet et al., 2016) developed during this IRA period.

The NRPA model, based on improved version of the Thomann (1981) model, uses a time-dependent approach to represent transfer of radionuclides within the food chain that can be described by first-order differential equations (Iosjpe et al., 2016b). The food chain considered involves phytoplankton, zooplankton, small fish (preying on the previous two categories) and large fish (preying on zooplankton and small fish). The NRPA model assumes that: excretion/elimination rates are independent of the uptake route; assimilation efficiency is independent of food type; predators do not assimilate the radionuclides in the gut content of their prey; phyto and zooplankton are a homogeneous group. The model can also be linked to exposures to humans consuming the biota, an output that is important for decision makers seeking better harmonisation with doses to humans in accidental situations.

IRSN, together with the University of Toulouse (France), has developed a model of radiocaesium transfer to marine biota taking into account large scale organism displacements in Fukushima area (Belharet et al., 2016). It simulates dynamically the uptake and turnover of a radiocaesium by marine organisms. This model is based on different exchanges, either (1) between the organism and its surrounding environment (marine water) by direct uptake of radionuclide from water and elimination of part of it by egestion and excretion, or, (2) between different organisms living in the same environment, essentially by grazing or predation (predator-prey relationship). Radiocaesium activity in water was deduced from dispersion modelling using a coastal circulation model SYMPHONIE (Estournel et al., 2012), while biomasses of the different marine populations, grazing, excretion and egestion fluxes were obtained through application of a 3-dimensional ecosystem model composed of a bio-geochemical model NEMURO (Kishi et al., 2007) coupled with the bioenergetic model NEMURO.FISH (Ito et al., 2004; Megrey et al., 2007). In the NEMURO biogeochemical model, phytoplankton is categorized into two groups: diatoms and non-diatoms small phytoplankton, and the zooplankton into three groups: small zooplankton, large zooplankton, and predatory zooplankton Transfer to

zooplankton has been calibrated with in situ data (Fig 1.1.6).This model showed that the maximum values of the 137Cs concentrations in phytoplankton and zooplankton populations were mainly reached 1 month after the accident and were about 2 to 4 orders of magnitude higher than those observed before the accident depending on the distance from FNPP.

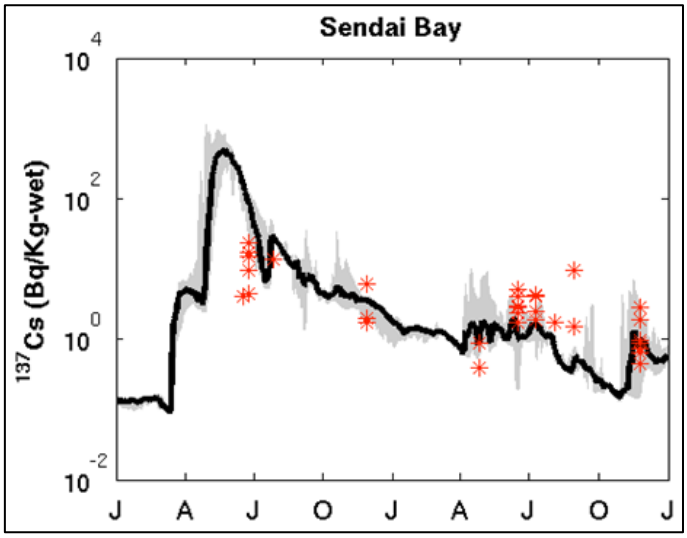

Figure 1.1.6: Results of the model calibration represented as the spatial median of the weighted average of 137Cs concentration in the three zooplankton groups situated in Sendai Bay. The red stars represent the field data of 137Cs activity in zooplankton in the same location (Kaeriyama et al., 2014)

Model results for fish are also significant. For example, they show the importance of taking into account fish migration in their contamination history (Fig. 1.1.7).

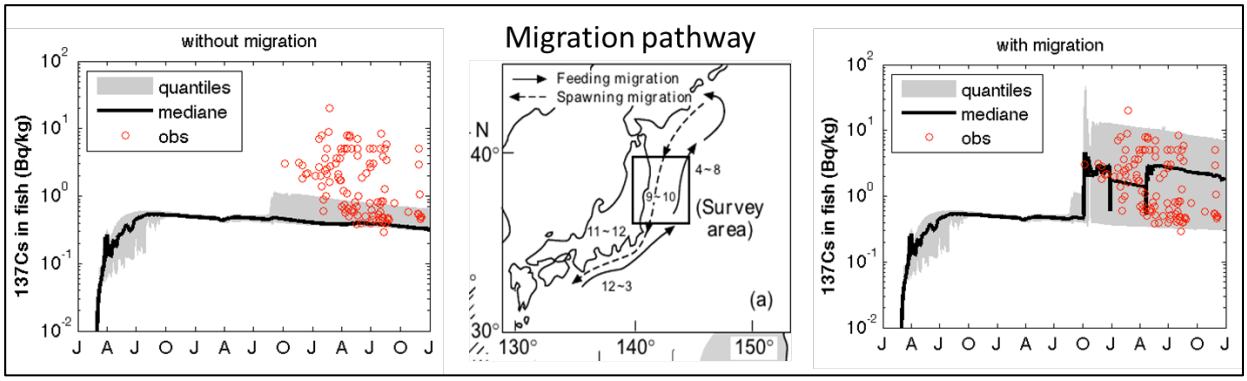

Figure 1.1.7: ^{137}Cs activities in sardine: model/measurement comparison with or without taking into account fish migration given by Tameishi et al. (1996)

1.1.4Discussion

The marine modelling performed under COMET has demonstrated that biokinetic models can better represent radionuclide transfer to biota in non-equilibrium situations, bringing more realism to predictions, and has also enabled a better understanding of important marine processes, such as the interaction of Fukushima radionuclides with seawater, particulate matter and biota. Thus, it can truly be said that the main achievement during COMET has been to directly address temporal dynamics in the assessment process. There has also been an attempt to involve more biological factors in the modelling process, by including (for example) biological half-lives ($T_{B½}$'s), bioturbation, fish migration, trophic transfer and uptake/elimination. We are readier now than we were before Fukushima in terms of having models that can be applied to dynamic situations. Although there were initially few partners

in the Marine IRA at the start of the project, since there was no dedicated marine network in the Alliance framework before the Fukushima accident, the objectives of the marine working group were met. It provided six publications, and presented marine activities and results in three different international congress (ICOBTE, ICRER, and ASLO).

The development and practical application of marine models, especially D-DAT, was enhanced substantially by interaction with the project FRAME (COMET Deliverable D3.6 Impact of recent releases from the Fukushima nuclear accident on the marine environment). Field data collected in FRAME, as well as those provided by Japanese authorities and universities, were used to improve model parameterisation and calibrate the models, which were then used to interpret current activity levels and doses to marine biota. For example, the complex time evolution of ^{137}Cs activity concentrations in different types of marine biota from 2011 to present is now modelled by D-DAT as a multicomponent exponential decrease influenced by a bi-phasic depuration mechanism and the radionuclide hold-up processes by seabed sediments. This model optimisation was possible due to the implementation of local parameters derived from the FRAME marine cruises.

The original approach of coupling a biogeochemical model and a dispersion model with a radioecological model gave significant results that could represent a good way for future research to better represent very complex behaviour that involves many processes.

1.1.5 Impact and further work

European activities and research on the marine environment are an important challenge for radioecology and radiological protection. Modelling tools and monitoring strategies in the emergency phase and in the post-accidental phase need to be improved. Many dynamic processes, such as those involved in the transfer of the radioactivity still accumulated on land, in groundwater and in bottom sediments to the water column and to marine organisms, still need to be studied and understood in order to be adequately represented in models. Radionuclides' behaviour in the marine environment in non-equilibrium situations is still poorly documented, such as radionuclides' reactivity and partitioning between particulate and dissolved phases and transfers through different trophic chains. Research on these items will need further field and experimental measurement data. Investigation of the inputs from contaminated river water and specific processes in estuaries is also an important issue.

Although simple biokinetic models that consider accumulation and depuration terms seem to be efficient as operational tools to evaluate the initial trends after any accidental release, a second generation of trophic transfer models that could better represent variability of transfers depending on considered organisms are to be developed, especially for mid to long term post-accidental contamination.

Another area that requires further effort is gathering new measurement results and consolidating existing data from Japanese authorities and universities, in order to further improve and calibrate modelling tools, especially for radionuclides other than radiocaesium. On this depends the future development of properly validated dispersion models relevant for normal or accidental discharges from nuclear plants or ship wreckage, usable in decision support systems for different release scenarios, especially for coastal areas. The scope of marine radioecology is very large, and the behaviour and fate of radionuclides is complicated to investigate due to the difficulty and the cost of getting measurement data. At the same time, marine radioecology is of cross-border importance and requires having a harmonised approach. Hence, there is a strong need to have more funded collaborative projects with the wider research community dealing with how other contaminants disperse and transfer to different parts of the environment.

Lastly, research in marine radioecology is not only purely scientific but also has societal relevance. However, currently there is little interaction with the socio-economic research community, something that would improve both the science and decision-making processes in the future.

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1.2 FRAME – The impact of recent releases from the Fukushima nuclear accident on the marine environment

1.2.1 Background

The earthquake and subsequent tsunami on 11 March 2011 led to the most important accidental release of artificial radionuclides to the marine environment from the Fukushima Dai-ichi nuclear power plants (FDNPP) since peacetime exploitation of nuclear energy. Initially studies carried out on

the impact in the ocean were limited, although it was estimated that about 80 % of all radioactivity was released to the marine environment and contamination of every marine compartment (water, sediment and biota) had been observed. Almost 3 years after the accident the contamination of the waters and biota remained elevated and leakages continued.

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 detailed monitoring data. There was an opportunity to validate and improve models that aim to be included in decision support systems for emergency situations. The overall goal of this project was to improve predictions of concentrations in the marine environment and exposures of marine biota (and, indirectly, humans) through fieldwork and radionuclide transfer modelling from seawater and sediments to marine species near the FDNPP. We planned to evaluate and model the radioactivity concentrations in waters and marine life off Japan, including new data to be obtained during this project, to better understand the sources, fate, transport and accumulation of radionuclides in the water column, the seafloor and the marine food web. We aimed to address why levels of contamination in marine biota remain high near shore and make predictions of long-term impacts of Fukushima on the ocean.

Concept and Objectives

This project was built on the collaboration of scientists from Spain, US, Belgium, Switzerland and Japan to better understand the sources, fate, transport, bioaccumulation and associated impact of radionuclides from the Fukushima Dai-ichi NPP accident in Japan. The investigation encompassed the main compartments of a complex marine environment, namely the water column, the seafloor and marine biota living within them. To that end, we measured a variety of radionuclides on samples collected during various oceanographic. This allowed addressing relevant questions such as:

i) What is the fate of the contamination in the ocean, what fraction of the total releases is stored in marine sediments and what is the aerial extent of the contamination in the seafloor.
ii) How much radioactivity is still leaking from the Fukushima Dai-ichi site and which are the mechanisms that govern the releases to the ocean, such as the role of submarine groundwater discharge.
iii) To which extent have the concentrations of contaminants in the ocean changed since 2011, and
iv) What are the current impacts on marine biota.

1.2.2 Methods and materials

Several oceanographic cruises were carried out in 2014 and 2015 to obtain samples for this project. Also, samples were collected from beaches and specific locations. The relevant information is provided in this section, and further details can be found in D3.6

KS Shinsei Maru Research cruise (KS-14-20). October 2014. We participated in the 2014 Shinsei Maru "KS-14-20" cruise organized by Japan off Fukushima in October 2014. The cruise was carried out from 17 October 2014 (Aomori) to 26 October 26 2014 (Yokosuka). Samples of water were analysed for ^{37}Cs, ^{134}Cs, ^{90}Sr, ^{129}I, ^{236}U and Pu-isotopes. Biota samples were analysed for ^{134}Cs and ^{137}Cs.

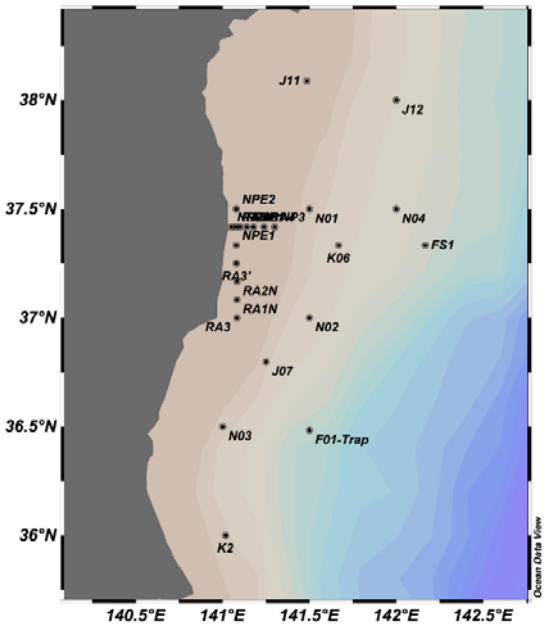

Figure 1.2.1 Stations sampled for the analyses of radionuclides during the KS14-20 cruise in 2014.

KS Shinsei Maru Research cruise (KS-15-13). October 2015. We participated in the 2015 Shinsei Maru "KS-15-13" cruise organized by Japan off Fukushima in October 2015. We collected 124 samples at 19 different stations including 121 seawater samples and 3 sediment cores, in this case including groundwaters. In addition to the artificial radionuclides, the natural quartet radium isotopes was analysed to quantify the fluxes of chemical elements associated with the offshore transport via submarine groundwater discharge (SGD).

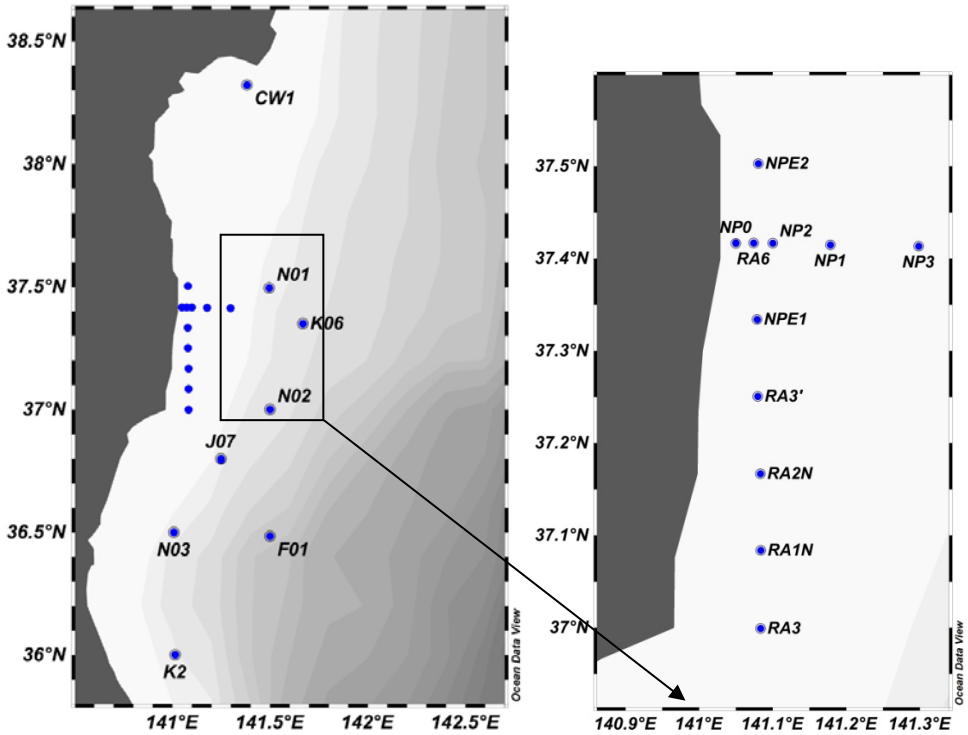

Figure 1.2.2. Stations sampled for the analyses of radionuclides during the KS-15-13 cruise in 2015.

Impacts on marine biota: model development

Further development in the marine biota dynamic transfer model D-DAT was carried out, principally reorganising the user interface, adding additional radionuclides (^{90}Sr, ^{129}I, ^{131}I, ^{134}Cs, ^{137}Cs, ^{236}U) and organisms (zooplankton) as well as updating the biokinetic database with the necessary new information. An Excel tool (MARISSA) for batch dose rate calculations from measured data (biota, water, sediment) was developed as part of the D-DAT family of codes. This can be used instead of the ERICA tool when radionuclide concentrations in biota are available from monitoring, the advantage of MARISSA is that it can process a large amount of data in batch mode. MARISSA was then used to analyse a from the FRAME cruises. Dosimetry data as well as the transfer parameters were updated to ERICA tool v. 2 standards.

1.2.3 Results

Data on concentrations of the suite of isotopes analysed in the samples collected off Japan during FRAME is provided in D3.6.

There are four major sources of FDNPP-derived radionuclides to the environment. The largest and earliest source occurred via the initial venting and explosive releases of gases and volatile radionuclides to the atmosphere, which led to fallout on both land and to the ocean. Atmospheric fallout peaked around March 15 (Chino et al., 2011; Morino et al., 2011; Huh et al., 2012; Stohl et al., 2012), and transport models indicate that >80% of the fallout fell on the ocean surface, with the highest deposition in coastal waters near the FDNPP. However, there no data on atmospheric fallout over the ocean is available to measure this estimate. Soon after initial releases to the atmosphere discharge there was the likely smaller direct discharge of contaminated material to the ocean during emergency cooling efforts at FDNPP, that resulted in run-off over land, enhanced flow of contaminated groundwater, and stagnant water leakage from the basement of the reactor buildings into the ocean. This secondary releases peaked around April 6, 2011, when the highest FDNPP-derived radiocaesium activities were observed in the ocean close in proximity to FDNPP (e.g., Buesseler et al., 2011). Ongoing radionuclide releases to the ocean from land through rivers, runoff, and groundwater flow must also be considered. These sources are influenced by an array of complex processes, but they are significantly smaller than the initial atmospheric fallout and subsequent direct discharges.

Estimates of the total atmospheric fallout of ^{137}Cs are provided in Table 1.2.1. They vary greatly because of the uncertainty in the transport and deposition parameters in the atmospheric models as well as the lack of observations required to conduct inverse calculations. The estimates of direct discharges of radiocaesium to the ocean average 3.5 ± 0.7 PBq. Estimates tend to converge to between 15-20 PBq for the combined FDNPP inputs of ^{137}Cs from atmospheric fallout and direct discharge to the North Pacific. This represents an additional input of about 25% more ^{137}Cs than existed in the North Pacific prior to the FDNPP accident (69 PBq) from nuclear weapons testing (Aoyama et al., 2015). Adding in an equal amount of ^{134}Cs would double the radiocaesium inventory derived from the FDNPP in 2011.

Radiocaesium inputs to Japanese coastal waters also occur via riverine sources and surface water runoff (Chartin et al., 2013; Nagao et al., 2013; Evrard et al., 2015). In a review of Cs transfer from land to the ocean, Evrard et al. (2015) summarized modeling simulations indicating that up to 10-12 TBq of particle associated ^{137}Cs was transferred from land to the ocean after the initial release, specifically during the first year, which would represent less than 1-2% of the total inventory deposited on land. Models suggest that up to 155 TBq of ^{137}Cs will continue to be exported to the ocean over the next century (Pratama et al., 2015). It is estimated that by late 2011, inputs of radiocaesium were ~4-5 orders of magnitude lower than those immediately at the time of the accident (Tsunume et al., 2011; Estournel et al., 2012, Bailly du Bois et al., 2012, Rypina et al., 2013; Charette et al., 2013, Kanda et al.,

2013). However, the Cs activities in the coastal ocean off Japan have been remained consistent over the past few years, exceeding background values, which suggests on-going non-steady state inputs.

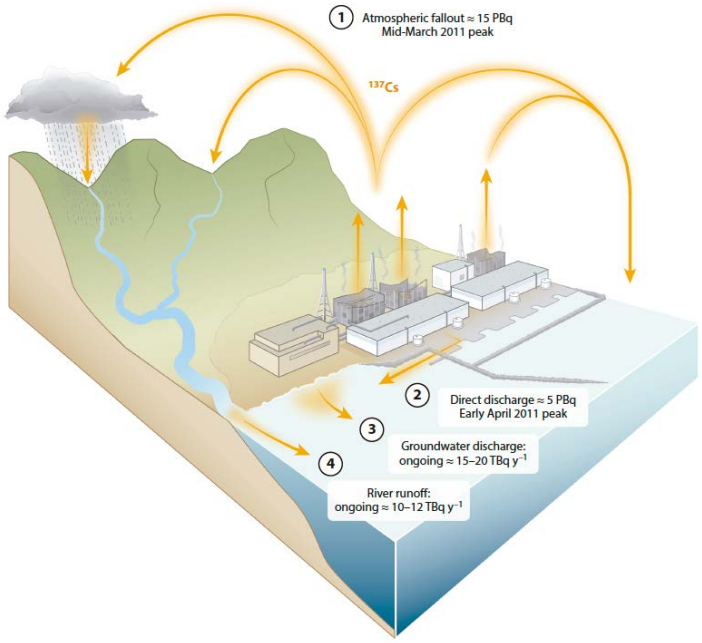

Figure 1.2.3. Schematic of Fukushima Daiichi–derived sources of ^{137}Cs to the environment. Atmospheric fallout (1) and direct discharge (2) are shown as total petabecquerels (PBq) released in the first month after the accident (median values from Table 12). Groundwater fluxes (3) and river runoff (4) are approximate ranges for the first year after the accident. Notice that (3) and (4) are expressed in terabecquerels (TBq). From Buesseler et al. (2017)

Table 1.2.1.: Source estimates for ^{137}Cs from the Fukushima Daiichi nuclear power plants. From Buesseler et al. (2017)

Reference	Total atmospheric fallout (PBq)	Atmospheric fallout on ocean (PBq)	Direct discharge to ocean (PBq)	Total in North Pacific (PBq)
Chino et al. 2011	13			
Katata et al. 2012	11			
Mathieu et al. 2012	20.6			
Stohl et al. 2012	36 (23–50)			
Terada et al. 2012	8.8			
Kobayashi et al. 2013	13	7.6	3.5	
Saunier et al. 2013	15.5			
Winiarek et al. 2014	19.3			
Katata et al. 2015	14.5			
Kawamura et al. 2011		5	4	
Bailly du Bois et al. 2012		11.5	27 ± 15	
Estournel et al. 2012		5.8 ± 0.1	4.3 ± 0.2	
Tsumune et al. 2012, 2013			3.5 ± 0.7	
Charette et al. 2013			13.5 ± 2.5	
Miyazawa et al. 2013			5.6 ± 0.2	
Rypina et al. 2013			16.2 ± 1.6	
Aoyama et al. 2016	15.2–20.4	11.7–14.8		15.2–18.3
Inomata et al. 2016				15.3 ± 2.6
Tsubono et al. 2016		10.5 ± 0.9		16.1 ± 1.4

Submarine groundwater discharge represents a long term source of radiocaesium to the ocean, in particular from the contaminated site at FDNPP, in addition to river inputs. Kanda (2013) calculated an average release rate of 93 GBq d^{-1} for the summer of 2011 and 8 GBq d^{-1} in the summer of 2012 (1 GBq = 10^{-6} PBq) and Tsumune et al. (2013) and Aoyama et al. (2015) calculated a discharge rate of about 30 GBq day^{-1} in 2013 and 10 GBq day^{-1} in 2014). These rates e are small relative to those for the first month after the initial releases, when ^{137}Cs inputs were 5 to 6 orders of magnitude higher (15-20 PBq/30 days= 0.5 – 0.7 x 10^6 GBq d^{-1}).

As part of FRAME, and using naturally occurring radium isotopes we estimated relatively constant Cs fluxes of about 10^{10} Bq/d between May 2013 and October 2014. Significant increase in ^{137}Cs activities were observed in seawater during the fall 2015 after typhoons events, leading to a larger flux of about 10^{11} Bq/d. Sanial et al (submitted) measured the highest concentrations of ^{137}Cs (of up to $23 \cdot 10^3$ Bq·m^{-3}) in groundwater samples collected between 2013 and 2016 beneath sand beaches up to 100 km away from the FDNPP. Consequently, the beach groundwater must be a source of ^{137}Cs to the ocean, that could be of about 0.6 TBq yr^{-1}, a magnitude similar to the ongoing releases of ^{137}Cs from the FDNPP site, as well as the input of Fukushima derived dissolved ^{137}Cs in rivers.

Levels of ^{90}Sr from the FDNPP in atmospheric fallout were, at most, four orders of magnitude lower than ^{137}Cs measured on land due to its low volatility (Povinec et al., 2012; Steinhauser et al., 2014; Tanaka et al., 2014). Estimates of ^{90}Sr in the ocean ranged from 0.04 to 1.0 PBq (Casacuberta et al., 2013; Povinec et al., 2012), spills of liquid radioactive waste at the FDNPP site have been reported since the accident, with corresponding anomalies in the ^{90}Sr activities in seawater. Castrillejo et al. (2016) reported ^{90}Sr concentrations in seawater samples collected in September 2013 ranging from 0.6 to 8.9 Bq·m^{-3}. The initial ^{137}Cs/ ^{90}Sr ratio of 39 ± 1 in seawater win June 2011, significantly higher than the global atmospheric fallout ratio of 1.6, decreased with time, averaging 3.5 in 2013 in waters within 100 km of the FDNPP (Castrillejo et al., 2016). This results from either continuing accidental spills and/or the higher mobility of Sr, as Cs is more strongly adsorbed onto soil particles, while Sr remains largely dissolved and thus relatively enriched in any ongoing releases via groundwater. Castrillejo et al. (2016) estimated that FDNPP leaked ^{90}Sr at a rate of 2.3 – 8.5 GBq d^{-1} into the North Pacific Ocean in September 2013, a figure that is 2-3 orders of magnitude larger than the amount of ^{90}Sr transported by rivers from land to ocean.

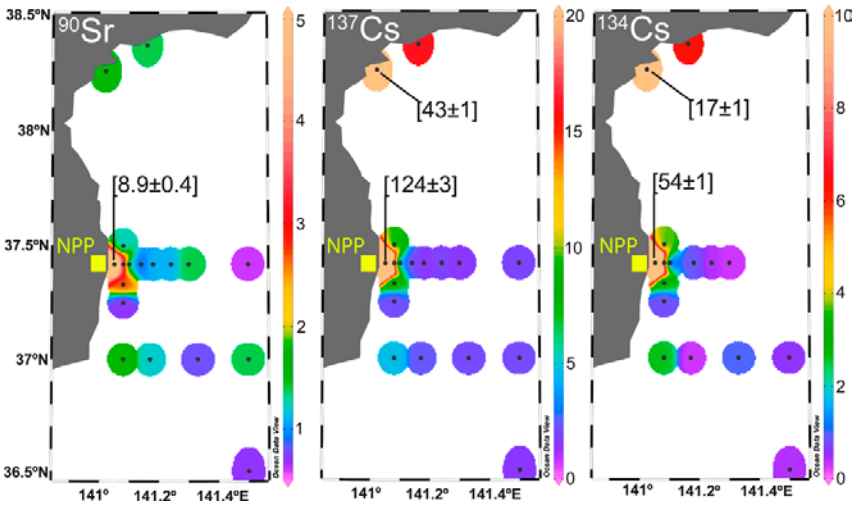

Figure 1.2.4. Surface concentrations of ^{90}Sr, ^{137}Cs, and ^{134}Cs in September 2013 (in Bq·m^{-3}). Pre-Fukushima concentrations from literature (decay corrected to sampling) are 0.9-1.1 Bq·m^{-3} for ^{90}Sr (IAEA, 2005; Povinec et al., 2012) and 1-2 Bq·m^{-3} for ^{137}Cs (Aoyama and Hirose, 2004). From Castrillejo et al. (2016)

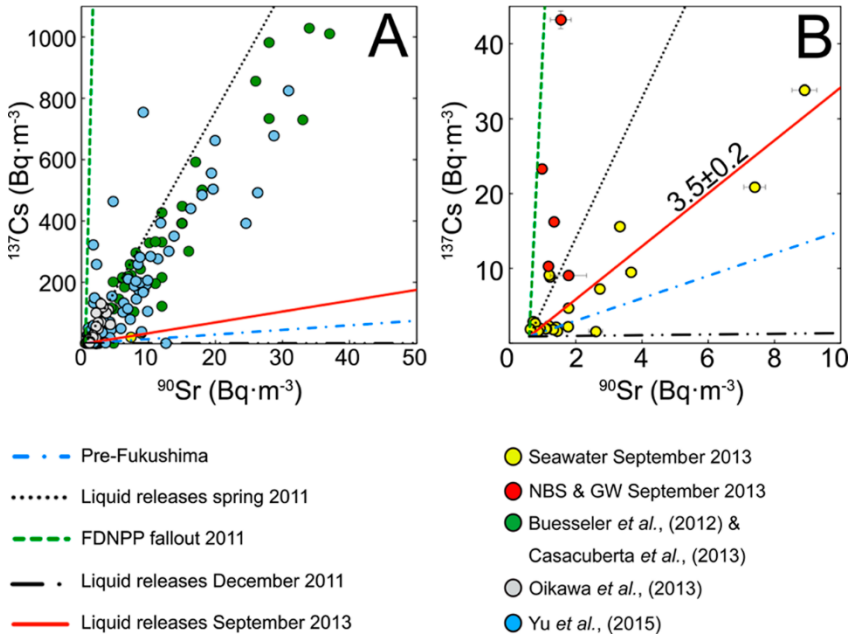

Figure 1.2.5 (A) ^{137}Cs/^{90}Sr activity ratios in seawater (SW), northern beach samples (NBS), and groundwater (GW) collected in September 2013 together with data published elsewhere (Buesseler et al., 2012; Casacuberta et al., 2013; Oikawa et al., 2013; Yu et al., 2015). (B) Magnified image of SW, NBS, and GW samples collected in September 2013. ^{137}Cs/^{90}Sr activity ratios of each end-member are included: pre Fukushima levels from global fallout due to nuclear weapon tests (~1.5; UNSCEAR, 2000), FDNPP liquid releases in the spring of 2011 (i.e., ratio = 39 ± 1; Casacuberta et al., 2013), FDPP fallout in 2011 (i.e., ratio ~1000),21 and FDNPP liquid releases in December 2011 (ratio = 0.016; Povinec et al., 2013; TEPCO, 2015). Also indicated are the continuing liquid releases from the FDNPP causing the highest seawater concentrations measured in samples collected within 6 km off FDNPP in September 2013 (ratio = 3.5 ± 0.2). From Castrillejo et al. (2016)

Zheng et al. (2013) had shown that FDNPP derived ^{137}Cs/^{239,240}Pu ratios are 1 million or greater, from measurement of leaf litter and soils and suing isotopic ratios of Pu, and it is estimated that only a relatively small amount of Pu, on the order of 1.0 -2.4 × 10^9 Bq ^{239,240}Pu, was released into the environment from the damaged FDNPP reactors (Zheng et al., 2012). We have not detected any evidence of Pu contamination from the FDNPP (Casacuberta et al., submitted). Also, Casacuberta et al. (submitted) showed that no additional ^{236}U due to releases from FDNPP could be observed in waters off Japan (Figure 1.2.6).

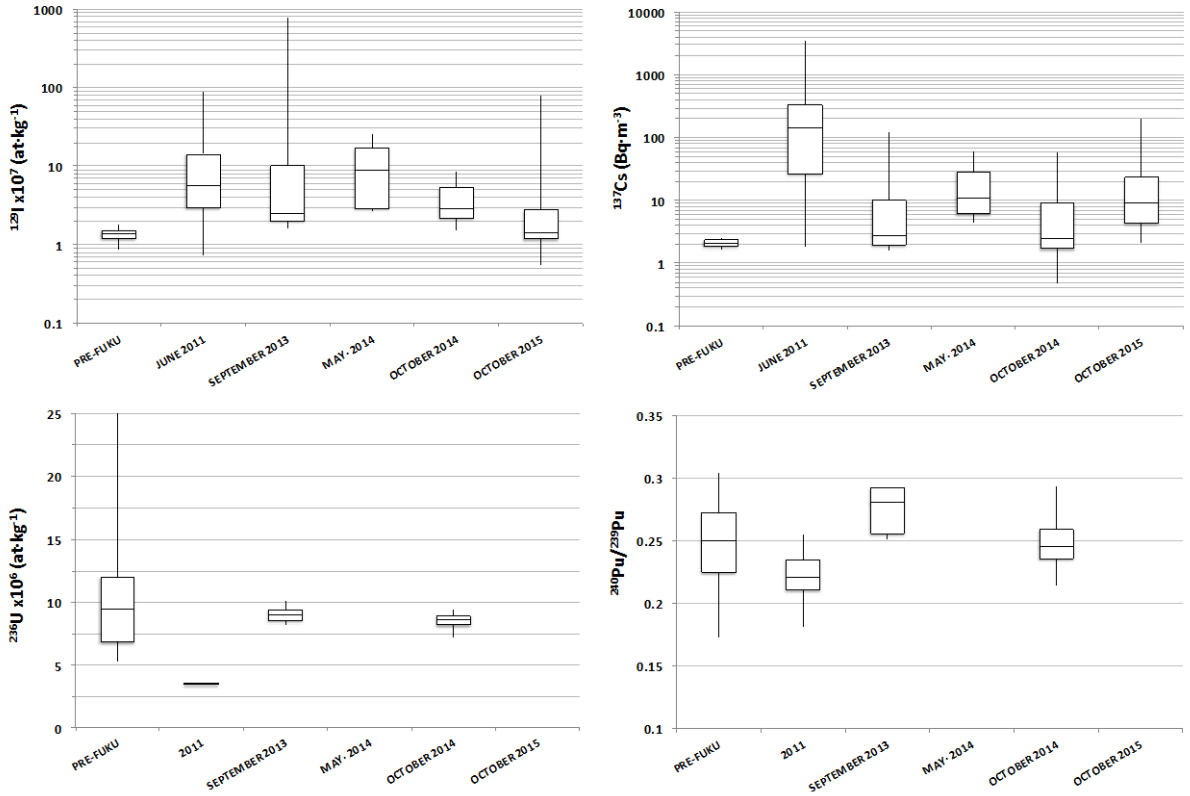

Figure 1.2.6. Evolution of the concentrations of ^{129}I (a), ^{137}Cs (b), ^{236}U (c) and the ^{240}Pu/^{239}Pu atom ratios in shallow waters (upper 200 m) off Japan. From Casacuberta et al. (submitted)

Approximately 1 kg of the long-lived ^{129}I ($t_{1/2}$ = 15.7 million years) was also released from FDNPP, mainly through direct discharges into the ocean (Guilderson et al., 2013). Based on seawater samples collected in 2013, 2014 and 2015, Casacuberta et al. (submitted) showed that concentrations of ^{129}I were systematically above the pre-Fukushima levels at stations located close to the FDNPP, with a maximum value of 790·10^{7} at.kg^{-1}, that exceeds all previously reported ^{129}I concentrations in the Pacific Ocean (Figure 6). The total amount of ^{129}I released after 2011 was calculated from the ^{129}I/^{137}Cs ratio of the ongoing ^{137}Cs releases and estimated to be about 100 g, which adds to the 1 kg released after the accident in 2011.

In the North Pacific Ocean, the ^{137}Cs activity in surface waters before March 2011 was 1–2 Bq m^{-3}, due to atmospheric nuclear weapons testing (Aoyama et al., 2008; 2011). In March 2011, ^{137}Cs levels increased rapidly to almost 70·10^{6} Bq m^{-3} by early April near the FDNPP and decreased by more than 3 orders of magnitude within about a month and to ≈10,000 Bq m^{-3} through early 2012 (Buesseler et al., 2011) and to ≈1,000 Bq m^{-3} from 2013-2015.

The dispersion of ^{137}Cs is mainly controlled by physical ocean processes, although a small fraction of the fallout was transported with atmospheric winds more rapidly across the North Pacific and the rest of the globe. The major flow of the contaminated plume moved eastward under the influence of the southward flowing Oyashio Current and the northward and eastward flowing Kuroshio Current. The eastward progression of the FDNPP plume was driven by the North Pacific Current. The movement of the leading edge of the plume indicates an average speed of propagation of 7 km day^{-1} (8 cm s^{-1}) until March 2012, and 3 km day^{-1} (3.5 cm s^{-1}) from March 2012 through August 2014 (Aoyama et al., 2015). These results are consistent with a drifter-based estimate of the horizontal spread of the FDNPP plume (Rypina et al., 2014).

Throughout much of the Pacific Ocean, the FDNPP plume was rapidly mixed throughout the upper 100-150 m of the water column and has only more slowly begun to penetrate to intermediate water depths (Buesseler et al., 2012; Smith et al., 2015; Yoshida et al., 2015). However, subsurface transport of FDNPP Cs occurred in various Pacific mode waters that are formed through wintertime cooling and buoyancy loss (Aoyama et al., 2015; 2016a). Ocean circulation models all suggest a broadening and diluting FDNPP radiocaesium plume arriving in the eastern North Pacific in 2013-2014.

1.2.4 Discussion

Radionuclides associated with sinking particles and in seafloor sediments

A small fraction of the FDNPP derived radiocaesium is associated with particulate organic matter and clay particles that may accumulate on the seafloor. Time-series sediment traps at 500 and 4800 m depth off Japan showed FDNPP Cs (evidenced by ^{134}Cs) in late March and beginning of April 2011 (Honda et al., 2013), due to atmospheric fallout Cs. At time-series sediment traps placed later in 2011 some 100 km east of FDNPP the signal was also detected with total ^{137}Cs fluxes as large as 100 to 3000 mBq m^{-2} d^{-1}. Subsequent samples over a 3 year period indicated that typhoons were likely responsible for the resuspension of shelf sediments that were subsequently transported with the southward flowing currents to the F1 trap site. When compared to the inventory of ^{137}Cs in coastal sediments however, this laterally supported flux accounted for an annual loss term of 1% or less of the shelf sediment inventory (Buesseler et al., 2015)

Radioactive contamination in bottom sediments from sites off the east coast of Japan are dominated by ^{134}Cs and ^{137}Cs (Kusakabe et al., 2013; Otosaka & Kobayashi, 2013; Thornton et al., 2013; Ambe et al., 2014; Black & Buesseler, 2014; Otosaka & Kato, 2014; Sohtome et al., 2014; Nagaoka et al., 2015). Measurements of Pu and Am activities and isotopic ratios in sediments reflect atmospheric weapons testing rather than a smaller FDNPP source (Zheng et al., 2012; Bu et al., 2013; Wu et al., 2014; Oikawa et al., 2015). Inventories of ^{90}Sr in bottom sediments are likely 3 orders of magnitude lower than the ^{90}Sr inventories measured in the water column due to the low affinity of ^{90}Sr to particles (Periañez et al., 2013). Sedimentary ^{137}Cs inventories range from <50 Bq m^{-2} (Ito et al., 2007) to >100,000 Bq m^{-2} in sediments closest to the FDNPP, showing a decrease with increasing distance from the FDNPP and increasing water depth Potential remobilization and transport and/or redistribution due to bioturbation are processes that can occur, and thus sediments may act as both a repository and a source in the long term. In total, seafloor sediments contain less than 1% (130± 60 TBq) of the ^{137}Cs activity initially released from FDNPP (Kusakabe et al., 2013; Black & Buesseler, 2014), but represent now more than 5-10 times the current total inventory of Cs in the overlying waters (36° to 38°N, water depths to 200m; Buesseler, 2014).

Radioecological investigations of the uptake of radionuclides by Fukushima marine biota

We reviewed the available data in the literature and official Japanese websites to obtain the radiological state end evolution of the radioactivity levels in biota. Radionuclide concentrations in fish off Fukushima are highly variable but remain significant, although have decreased drastically during the five years after the FDNPP accident.

An exponential decrease of activity concentrations with time for all the main categories of marine biota (pelagic, benthic and molluscs) is observed. Issues regarding scattering of the data, lack of information or the first year and the difficulty of relating water and biota data were addressed. Modelling work with D-DAT (Vives i Batlle, 2016) suggests that concentrations in biota decrease much more slowly (than expected given the biological half-lived, likely due to hold-up processes by bottom sediments, confirming the role of the sedimentary deposits as a main source-term. Fugitive emissions can also affect the variability in the data.

Reverse-modelling allowed to calculate integrated releases of 103, 30 and 3 PBq for ^{131}I, ^{137}Cs and ^{90}Sr, respectively The UNSCEAR report puts these estimates in the ranges 70 – 120 and 8 – 14 PBq for ^{131}I and ^{137}Cs (no data for ^{90}Sr); hence our model gives a reasonable prediction (UNSCEAR, 2014). Our calculations indicate that the ^{137}Cs in sediments near the FDNPS have an associated clearance half-time of 118 days, whilst biota activity concentrations decrease with time consistent with ecological half-lives ranging from 70 – 90 days (macroalgae and zooplankton) to 200 – 220 days (crustacean and pelagic fish). This variability is likely due to differences in trophic level and occupancy pattern in the water column (Iwata et al., 2013, Vives i Batlle, 2016).

A dual component D-DAT model with further calibration, including a 20% reduction of the transition time between 'fast' and 'slow' elimination in the biokinetic model, allowed to substantially improve the quality of the predictions when compared to field data, although biota data scattering does not allow to verify the short-term variations predicted by the model. This kind of verification can only be obtained from a controlled single pulsed release study (Vives i Batlle et al., 2008), and such data was not available for this project.

Radiological impact to marine biota

Available data for 2012 – 2014 show that the most impacted fish species nearest to the FDNPS exceeded ERICA benchmark levels, with a median dose rate of μ45 Gy h^{-1} (Johansen et al., 2015). In 2013, our calculations showed that the highest internal dose rates to greenling were 17 - 44 μGy h^{-1} for ^{134}Cs and 32 – 82 μGy h^{-1} for ^{137}Cs, although most dose rates were clearly below the maximum and a median of 1.38 μGy h^{-1} with 95% of results under 50 μGy h^{-1} was obtained. The external dose rates were estimated to be significantly lower at 0.12 μGy h^{-1} for ^{134}Cs and 0.05 μGy h^{-1} for ^{137}Cs (Vives i Batlle, 2015). High values are the exception rather than the rule and do not signal risks to fish at the level of population (Copplestone et al., 2008).

For the period 2012 -2015, dose rates decrease exponentially, with attenuation half-times in the order of 10^3 days, although the trend for lower doses may be influenced by a decrease in sampling frequency. The highest dose rates are for benthic fish, exposed to contaminated sediments; pelagic fish received substantially lower exposures. The dose rates in recent times are well below the ERICA no-effects dose screening level of 10 μGy h^{-1} (Beresford et al., 2007, Brown et al., 2008, Andersson et al., 2009).

Biota dose rate calculations on the samples collected during the October 2014 Shinsei Maru cruise show that the highest internal dose rates are for ctenophore > benthic fish (rockfish > greenling and flounder > pelagic fish (mackerel) > plankton, and dose rates to other than benthic fish are predominantly dominated by internal exposure, then external exposure to sediment and then external exposure to water. The average internal dose rates to benthic fish landed in the Fukushima prefecture after 2012 have decreased by one order of magnitude between 2012 and 2016 down to about 2 nGy h^{-1}.

The total dose rates to benthic fish are dominated by external exposure to sediment, showing that sediments are now the main source of radiation exposure to the benthic biota. The highest total doses are of the order of 0.15 μGy h^{-1} for rockfish, but again all doses are below the ERICA no effects benchmark. Hence, the biological species considered are not at risk from this radiation. It still remains necessary to investigate the high spatial variability of the data and whether there are potentially higher doses linked to residual emissions in some local hot-spots.

Radiological impact to humans

The committed effective radiation dose to adult human consumers of contaminated biota was also calculated, using as a source the activities in fish obtained in the FRAME October 2014 cruise. The results show that the hypothetical ingestion of fish at the levels measured would pose no radiological significance, given that the doses are a fraction of the average annual dose limit to a member of the public of 1 mSv. By category, the highest doses correspond to eating 56.6 kg y^{-1} of benthic fish alone,

giving 30% of the 1 mSv annual dose to members of the public, but in practice the actual doses will be much lower, due to the conservative assumptions made in the estimates.

1.2.5 Further work

A recent upsurge of available data shall allow further modelling investigations from which a realistic representation of the radiological state end evolution of the environment can be derived. Further work should then focus on the following aspects:

Use the data from the Japanese NRA, JAEA, MAFF and TEPCO, to apply the model to different radionuclides (e.g. Sr, I, actinides), seeking an improved version of the model parameter database that can also reproduce the biota transfer for these isotopes. This should include a sensitivity analysis and calibration of the multi-component biological half-lives in D-DAT to optimise model predictions for radionuclides in biota in order to reconcile short (accident) and longer (post-accident) terms.

- Improved extrapolation approaches to cover gaps in data shall soon allow a more accurate reproduction of the ecological half-lives observed in the field with the D-DAT model, explained as the dynamic exchange of radionuclides between seawater, sediments and the biota (particularly for benthic species within sediment-associated food chains).
- A further development not included in the present project but recommended for the future is the migration of D-DAT to a FORTRAN platform and associated development of user-friendly interface, leading to a formal public release of the model.
- There is the possibility of including into D-DAT aspects of foodweb modelling, such as sediment and food ingestion.
- Further development of MARISSA include adding ^{210}Po and ^{239,240}Pu and improve the averaging algorithm that assigns nearby water concentration to discriminate vertical profiling of radionuclides in the water column and to couple the tool with a system for contour plotting of the results.

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Forest Modelling

2.1 Background

In forest radioecology, the impact of recent nuclear accidents (Chernobyl and Fukushima) gave evidence of the ability of forests to induce significant external and internal exposures to humans 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 high 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 possible, and the existence of a nutrient cycle that may carry radionuclides leads to persistent radioactive cycling and pollution. Whilst modelling work to understand this has been 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 fundamental need for improving and guiding forest model development and the validation of models through a variety of scenarios to assess the models' accuracy and transferability.

In the COMET WP3, IRA on forest modelling, the main objective was to generate guidance on how to reduce the uncertainties in the modelling of short- and long-term impacts of radioactive contamination in forested areas through the production of guidance for modellers on topics such as model sensitivity analysis and parameterisation of key processes controlling the transfer of radionuclides, as well as advice on model parameter requirements to facilitate the interaction between forest modellers and experimentalists, whilst working on the ongoing development of some forest transfer models as part of the ongoing activity of the research groups involved in the project.

The focus was therefore on producing guidance documentation and dealing with key modelling processes and variables/factors contributing most to the overall uncertainties in forest modelling. Due to the dynamic nature of these processes, emphasis was placed on time-dependent soil-water-atmosphere interactions (as expressed, for example, in the evapotranspiration as an engine for water fluxes in trees, interception of airborne radionuclides, parametric Kd approaches to simplify the soil hydrology, etc.) and soil - plant transfers (TF, TC, CR), with forest vegetation selected according to its importance for wildlife and human intakes. Starting from existing models as a basis for the guidance, the group discussed process identification and mathematical representation, as well as a parameterisation of key processes controlling the transfer of radionuclides.

The work performed was in full agreement 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.

2.2 Methods and materials

The objective of forest radioecology modelling is to mathematically simulate the distribution, cycling and sinks of radionuclides in forest environments. In developing a forest model for radionuclides, it is important to capture the essential fluxes governing entry, circulation, storage and exit of substances to the trees – in other words, the biogeochemical cycling. A schematic, based on the IAEA Biomass report (IAEA 2003), is given in Figure 2.1 below. There is a dual purpose to forest modelling:

- Research modelling: understanding the role of trees as 'biological pumps' cycling radionuclides along with the water. This involves implementing the governing equations of processes

controlling the movement of water (e.g. evapotranspiration, groundwater flow, sap flow) and energy (e.g. solar irradiation, changes in temperature). Then, it is necessary to link radionuclide transport to these fluxes.

- Assessment modelling: Calculating the pathways of radionuclides intercepted by the forest, providing a basis for the assessment of dose to man and the wildlife. The calculation of doses to humans through external exposure and ingestion of forest foodstuffs also relies on the prediction of radionuclides transfer dynamics within forest compartments (Rantavaara, Calmon et al. 2001), though the models used are less detailed from a mechanistic point of view.

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 an example of research, process-based model. It 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 becomes available. In the meantime the model has been used in process sensitivity analysis studies, and on determining data requirements for models of this type. As such, it provided input for the "parameter wish-list" document and the forest modelling guide, in order to illustrate the process selection and associated parameter requirements. There was also the development by IRSN of 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 process. This too was used as an example for the aforesaid guidance documentation.

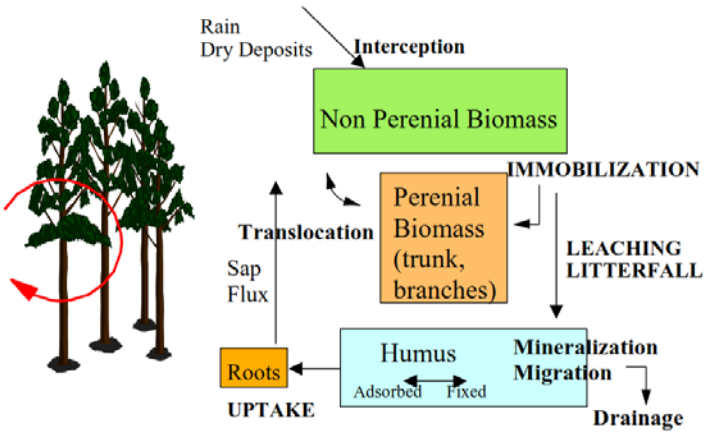

Figure 2.2.1.: Processes regulating the cycling of radionuclides by forest vegetation

The above modelling activities brought to evidence that there are three main areas to consider:

- The transfer of radionuclides from the unsaturated zone of the soil. This is important when the source of radioactivity is in the soil, e.g. ground disposal of radioactive waste. Another situation of interest is long-term (i.e. years to decades) prediction of contamination in forest systems contaminated by atmospheric deposits (for which memory of the initial input is progressively lost).
- The interception by the trees of radionuclides from the atmosphere (important for accidents).
- The deposition of radionuclides transported by the litter fall to the forest floor, leading to exposure to forest products and to man.

In principle, a forest model needs to consider the root zone, which in many cases extends below the upper organic layer. How much water is available in the soil is a key factor for determine the water uptake by the tree, as well as to explain the mobility of elements from the decomposing litter to the soil. However, the relative importance of modelling interception or soil hydrological processes depends on model applicability - if the model aims to study the deposition and interception in the initial phase of an accident, the soil hydrology and the Kd are of secondary importance. However if the model aims at understanding the longer-term cycling of radionuclides, then the soil transport becomes progressively a more important process.

The parameter requirements of a forest radioecology model vary greatly depending on the degree of complexity of the model, which in turn is dictated by the model's purpose. Therefore, there is no "one size fits all" parameter list. Research models require a higher number of parameters, carrying information closely linked to the governing equations used to describe the processes. This can include physical, biological (i.e. ecophysiological) and chemical parameters which need to be derived from observations and field measurements in contaminated environments, alongside laboratory experiments, because it is virtually impossible to trace down every single parameter in the equations to basic physical or chemical constants.

On the other hand, assessment-type models tend to be simpler, with many being systems of linear, first order differential equations in which the exchange between compartments is mathematically represented by first-order kinetics. This means that there is a 'rate equation' with a constant transfer rate which is generally an empirical or semi-empirical parameter. A 'box model' can be constructed, which is very simple and has a reasonably small number of these rate constants. Note however that this is an approximation; one may have transfer rates that can be parametrised as a function of environmental co-variables/co-factors (like time series of meteorological data or biomass fall, etc.) and the transfer between compartments does not necessary follow a first order kinetics representation in every case.

A good example of a typical 'box model' of the type that could be used in assessments is the model of ^{36}Cl cycling in a coniferous stand (Van den Hoof and Thiry 2012), developed at SCK•CEN (Belgium). This is a model with rates of transfer k_{ij} between compartments C_i and C_j. Although there are many types of such simple box models, it often (but not always) shares the fundamental characteristics for a model of this complexity, such as (a) it is dynamic, with the fluxes of biomasses, energy or nutriments following first-order kinetics, (b) the behaviour of the radionuclide is similar to the behaviour of the stable element (c) the transfer rates are time-independent, (d) the fluxes between compartments are controlled by the element content in the donor compartment and (e) as an initial condition, sometimes all the model compartments except the initial source term are generally assumed to be empty (although ordinary differential equation-based models can be started/run from a pre-existing contamination in the forest system).

Another example of a typical assessment model is given by TREE4 model - Transfer of Radionuclides and External Exposure in FORests (Calmon, Thiry et al. 2009, Calmon, Gonze et al. 2014). In the course of the 4th European Commission framework programme (1995-1999), the French Institute for Radiological Protection and Nuclear Safety (IRSN) and the Finnish Radiation and Nuclear Safety Authority (STUK) designed and developed a forest module for the RODOS system (Ehrhardt and Weis 2000). The development of the model relied on post-Chernobyl observations made in western European forests. The objective was to develop a rather simple approach to help decision-making by rapidly estimating the consequences of accidental atmospheric fallout, with a special emphasis on the short-term phase (i.e. first few months). This dynamic model accounts for the physical and biological processes that control the fate of radiocaesium during the short-term phase: dry deposition onto vegetation and forest floor, interception of wet deposit by vegetation and vegetation depuration

through litterfall, throughfall and stem flow. The approach has been tested in the frame of IAEA research programmes BIOMASS, 1997-2001 and EMRAS, 2003-2007, dedicated to forest models inter-comparison and parameters review (IAEA 2002, Shaw, Venter et al. 2005, Calmon, Thiry et al. 2009, IAEA 2010). Preliminary results obtained with the TREE4 approach on radiocaesium transfer and ambient dose rate in Fukushima forest biotopes have been recently reported (Calmon, Gonze et al. 2014, Gonze, Calmon et al. 2014, Gonze, Renaud et al. 2014, Calmon, Gonze et al. 2015).

The evolution of radionuclide inventories in each forest pool (expressed in Bq m^{-2}) is calculated along with mass fluxes between them (Bq m^{-2} s^{-1}). The approach relies on the resolution of mass balance equations with mathematical parameterisations specific to each process considered in the conceptual model. Parameter values are tabulated for three different types of forests: deciduous broadleaf, evergreen and mixed forests, the latter being characterised by the percentage of deciduous and evergreen.

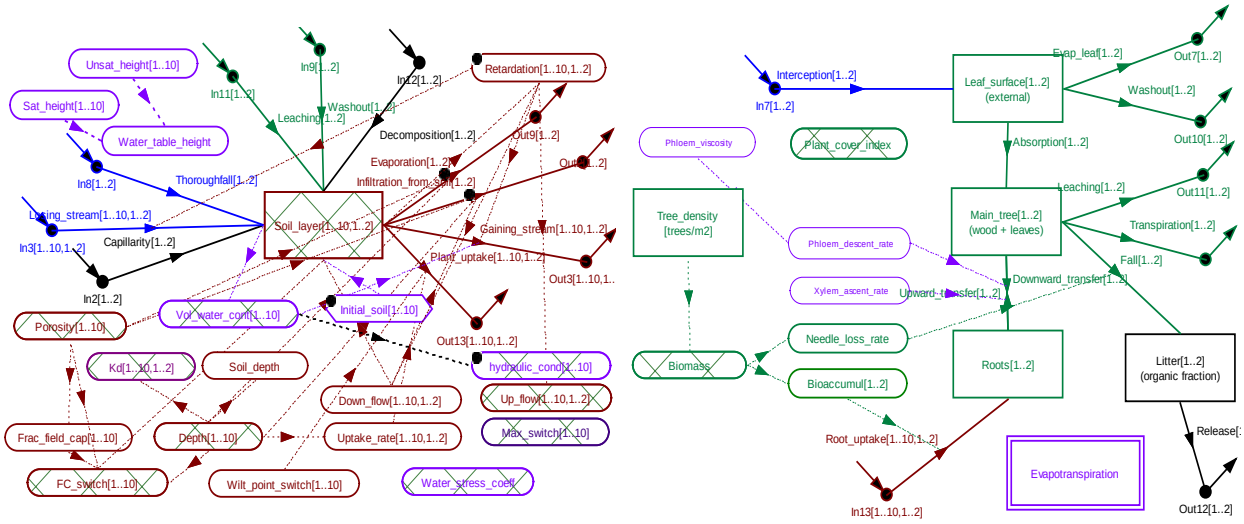

Figure 2.2.2.: Schematic of ECOFOR model as set-up in ModelMaker 4, including hydrological (left) and vegetation (right) main sub-models. The meaning of the symbols is as follows: Compartments (rectangle), variables (rounded rectangle), sub-model (double rectangle), flow (arrow) and influence (dotted arrow)

2.3 Results

This IRA on Forest modelling 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) and the production of the Handbook titled 'Guidance on Forest models for contaminated sites'. The Handbook is a Deliverable of the IRA Forest.

Models fit for purpose – outcome of the discussions

The workshop on "Models fit for purpose" gave good interaction between modellers and experimentalists and the summary of the discussions are given below:

Purpose and relevance of forest models

There was a stimulating debate on the general relevance of radioecological models for forests, which raised the following points. Forests are considered to be a challenge for the radiation protection of humans only in rare cases. Forests tend to stabilise the radiological situation, thus preventing the

contamination of surface water. After a nuclear accident or at NORM sites, forests could be useful to reduce the spread of contamination. At sites, where radioactive waste is deposited close to the surface, trees will most likely not be accepted, since deep tree roots might damage barriers and facilitate the transport of radionuclides.

Desired output

The discussion pointed out that regulators usually prefer tiered approaches to human and environmental risk assessments, taking into account the purpose of the assessment and the specific situation to be evaluated. High-quality assessment models should provide results (e.g. contamination levels and doses to humans and non-human biota) that are as realistic as reasonably achievable. The desired endpoints are different parts of trees and edible forest products, such as wild mushrooms, wild berries and game. Time-dependent transfer factors may pose a problem, especially in the case of radiostrontium. From a regulator's point of view, a forest model should also provide the basis for calculating doses to non-human biota. In a way, this part of the discussion focused on the assessment application, but neglected somewhat the research aspect of modelling, probably because there was a large proportion of end users, but not so many modellers/researchers.

Model complexity and data requirement

Although the process-based model ECOFOR has been designed to be "sufficiently complex to be realistic but sufficiently simple to be practical", some participants of the workshop remarked that this model is too complex and requires data for too many model parameters. ECOFOR, whilst more complex and process-based than a simple box model with constant transfer rates, is not as complicated as some forest models that include plant biology and chemical processes at the molecular level (Deckmyn, Verbeeck et al. 2008, Deckmyn, Campioli et al. 2011). Such forestry models can have hundreds of parameters and be very complex because they include advanced ecological processes (e.g. the physico-chemical and biological behaviour of nutrients, micronutrients with applications to woodland management). AECOFOR, on the other hand, represents a research model of the type sufficiently complex to be realistic and sufficiently simple to be practical for future use in environmental assessments.

There is just no easy solution in the form of a model requiring only a handful of parameters to represent such a complex environment as a forest. For this reason, ECOFOR is considered to be a research model rather than an assessment model, but in reality it tries to bridge the gap between the two, aiming to be a platform for model abstraction in the search towards a simple radioecological assessment model for forests. It certainly provided a sound basis for identifying in this IRA the most relevant processes (process sensitivity). Another key point of the discussion was that the development of assessment models should be guided by the question, what endpoints are important for assessing the radiological risk of humans and the environment. One participant suggested to make a bridge to ecology and ecotoxicology, since these scientific disciplines are expected to face similar challenges when investigating processes or assessing risks.

Sensitivity and uncertainty

Several participants emphasised the importance of sensitivity analyses and the quantification of the overall predictive uncertainty. In the case of water-mediated transport processes the element-specific and specie-specific partition coefficient K_d is expected to be the most important model parameter. For sensitivity analyses, but also when evaluating the applicability of a forest model to other sites, it is essential to differentiate between input parameters (which usually are site-specific) and model parameters (which should ideally be generally applicable). Attention should also be paid to the source term and the physico-chemical forms of emitted radionuclides. After the Chernobyl accident, for example, large activities (3-4 tons of spent U fuel) were released as particles ranging from submicrons to fragments from the reactor, whereas the source term of the Kyshtym accident (nitric acid waste solution) was quite different. Thus, the behaviour of radionuclides such as Cs-and Sr-isotopes

associated the Chernobyl explosion (imbedded in particles) were quite different from what was observed in the Mayak PA surroundings.

Model-specific criticism

The forest model ECOFOR developed at SCK-CEN focusses on water-mediated transport processes. Participants remarked that water fluxes are important for radiocaesium, but not so much for radiostrontium, and that radionuclide speciation is essential to implement in the model. The model concept is therefore expected to limit the application to other radionuclides. Concerning the French model TREE4, the question was raised if the good agreement of model predictions and measurements demonstrates the quality of the model or is only the result of "calibrating" the model output to measured data. According to the model developer (present at the meeting), good agreement has been observed for three data sets. The most important parameters, interception fraction and weathering rate, are only moderately variable. These two main parameters for short-term predictions cannot directly be measured and therefore have been determined by "calibrating" the model to measured data obtained at Japanese forest sites. The model predictions agree well with data obtained at a forest site in Southern Germany after the Chernobyl accident. The experience after the Fukushima-Daiichi nuclear power plant accident demonstrated that predicting the spatio-temporal deposition pattern remains a challenge.

Wish-list for modelers and experimentalists

Based on the above discussions and the feedback from the modellers, a wish list of modelling requirements to experimentalists for three types of forest models was generated.

Especially for assessment models, a difficulty is often to manage uncertainty/variability in the parameter values. One major point is that for each parameter, there is a set of environmental characteristics that determine its numerical value. Therefore, the transfer parameter values are conditioned by physico-chemical, biological and ecological characteristics of the forest ecosystem considered, and thus are intrinsically site-specific. The most influencing environmental factors are known to be the local meteorological conditions (e.g., rainfall and snowfall time series, mainly) and the vegetation characteristics (e.g., stand density and age, tree species composition, tree component biomasses or area indexes and biomass dynamics at both intra-annual and inter-annual scales).

Therefore, part of this variability/uncertainty can be better resolved by introducing parameterisations that explicitly account for environmental conditions/co-factors (ex: throughfall rate driven by the effective rainfall rate) and a sub-model dedicated to the description of the forest structure and functioning, through for example allometric relationships and population density dynamic models. These kind of refinements were shown to significantly increase the realism of such assessment model predictions and its capability to capture the spatio-temporal variability of radionuclide dynamics in Fukushima contaminated forests (Gonze, Calmon et al. 2016).

Based on the above, the wish list of modelling requirements in the case of flux-based models, like TREE4 (Calmon, Thiry et al. 2009, Calmon, Gonze et al. 2014), is very similar to that described above for the ^{36}Cl box-model. Some further parameters, including the speciation concept, are required.

ECOFOR provides a good example of a process-based box model of the type that could be used for research when hydrological processes are important (Vives i Batlle, Vandenhove et al. 2014). This is because the model was designed with radioactive waste disposal in mind. The model structure is indicated in Figure 3. This model assumes a simplified representation of the hydrology with water infiltration, Darcy flow and the Lucas-Washburn equation which describes capillary flow, rather than adopting the general but more complex representation by the Richards' equation, which is difficult to solve numerically. The movement of water through the soil layers is then modelled using a 'tipping bucket' approach in which the soil column is represented by ten computational layers and water flows from to and from a layer are controlled by a an algorithm based on the above hydrological equations

and each layer's actual volumetric water content, total porosity and field capacity, with excess water exiting to groundwater recharge.

The hydrological model is coupled to a plant sub-model where water uptake via roots is driven by evapotranspiration as calculated by the Monteith model (Monteith and Unsworth 2007) and fluids circulate through the plant upwards (xylem upflow governed by the Poiseuille equation) and downwards (phloem downflow along an osmotic pressure gradient) allowing translocation of the radionuclides with these flows. Water interception by the canopy, washout, absorption and leaching are considered as transfer factors and litterfall plus litterfall decomposition are modelled by an empirically-derived linear transfer rate. Element transport is linked to water via retardation processes in soil (with link to the K_d of the element, assumed to be dependent on soil moisture) whilst empirically-derived selectivity coefficients link element fluxes to the water fluxes in plants in an approach similar to the BioRUR model (Casadesus, Sauras-Yera et al. 2008).

Of all the components of the model, it is the hydrology that is the most difficult to simplify. In the ideal case one would have to solve Richard's Equation which depends on very few parameters, but this is very complex to solve mathematically. By adopting simpler strategies, the model is more practical and requires less parameters and computational effort, but a price is paid in terms of reducing the applicability range of the model because of the use of some approximations (such as the aforesaid Darcy flow and Lucas-Washburn capillarity equations, for example) (Vives Batlle 2016).

Handbook on guidance for forest models

The concluding achievement of the Forest Working group has been the production of the Handbook titled 'Guidance on Forest models for contaminated sites'. The Handbook is a Deliverable of the IRA Forest and constitutes a final synthesis of the expert thinking and consultation performed during the COMET IRA on forest modelling. At the COMET Final event in Bruges on April 25th-27th 2017, the Handbook was presented with an oral presentation and a poster. The Forest modelling group is finalising a publication intended to be submitted for peer review, which describes the most important aspects presented related to forest models in the Handbook. This publication addresses the fact that a wide community (regulators, young researchers in the field of environmental modelling, stakeholders) are needing advice and expert contribution when developing and handling models for contaminated forest sites. Based on this, the IRA Forest working group decided that it would be helpful to exploit the possibilities given within COMET and the Forest IRA by providing such support by means of a written guidance.

The Handbook has been prepared in a way that the reader can develop a forest model or to apply an already existing one. Modelers and experimentalists have worked close together to create a document in which the reader can find both, general and specific information and useful suggestions, on how to choose model complexity, how to identify model requirements depending on the model's purpose and how to deal practically with the challenges that inevitably modelling of radionuclides in such an ecosystem poses.

The Handbook has 90 pages document divided into 12 chapters: each chapter deals with specific aspects related to forest model development and application. After introducing historical background information on radioactive contamination in forests, the first part of the handbook (chapter 3-7) discusses the possible objectives of forest models, gives a description of pathways of radioactive pollution and provides a guidance on developing a conceptual model by means of FEPs list and interaction matrix. The second part of the Handbook (chapter 8-9) introduces the distinction between empirical and process-based models and gives an overview of the processes related to radionuclide transfer in forests, which need to be accounted for and their possible mathematical representation. Chapter 9 is a summary of the major available models and two of these are described in a detailed way. They are examples on how processes are implemented with an empirical and with a process-based approach. The last part of the Handbook (chapter 10-12) focuses on the specific challenges of

forest models. These are mainly related to the large heterogeneity of forest ecosystems. Model validation and calibration, the distinction between variability and uncertainty, spatial scaling issues, limitation of process knowledge in forests are analysed. Often limitations of experiments and field sampling make it difficult to obtain the required data for testing a model. This is discussed in chapter 12, where a general guidance to experimentalists is provided, highlighting the need for high quality, well-documented data.

2.4 Discussion

Different types of forest models implement processes in a different way and require different types and numbers of parameters. The problem is compounded by the fact that in developing a model for forests, it is necessary to couple processes in three domains: soil, the vegetation and the atmosphere. The hydrological problem in particular is not amenable to easy mathematical representation. This complex situation requires approximations and simplifications. Under these conditions, it is not possible to give a 'one size fits all' wish list of modelling parameters for the modelling of radionuclides in forests.

However, some generalisations can be made, and the IRA group approached the challenge by describing parameter requirements with three examples ranging from simple box models for assessments to more advanced research models, from which the basic parameter requirements according to model type, their importance and the sources of information have been deduced and presented.

With respect to sources of information, in current monitoring programmes, it is often the case that data on radionuclide concentrations in tree parts and surrounding soils and their associated biomasses are determined, but the principal 'accompanying parameters' describing the environment (soil hydrological characterisation, soil K_ds, evapotranspiration controlling parameters) are often missing. A lot of what is referred to as model uncertainty is caused by not measuring these 'hidden' variables and thus being unable to explain differences that would be quantified by integrating this ancillary information in the model. This can be addressed by complementing regular sampling (to measure radionuclides) along with ongoing monitoring of these parameters, using monitoring stations with data logging capabilities linked to basic instruments (e.g. pluviometers, photosynthetically active radiation (PAR) sensors, sap flow meters, feed from a local meteorological station, soil moisture sensors and piezometers). This is the approach used in a recent study (Gielen, Vives i Batlle et al. 2016).

When attempting a predictive modelling approach for forests, it may well be the case that all the required site-specific parameters are not available. The following process can then be attempted. In the first instance one should try to utilise bibliographic reference parameters, whereupon by literature search one may hopefully be able to find a range for the parameters (minimum, maximum and median). Starting with the median value, the modeller can vary each parameter within the permitted interval and, through this calibration process, assert the optimum value that fits best the model output to the available reference data. If no reference data are available, it is at least possible to calculate an uncertainty band for the model predictions.

If the model output does not fit observation unless the parameters are outside their permitted interval, then there is a need to change the structure of the model. Usually, the problem is that the actual model representation is too complex. The only solution then is to simplify by reducing the model's scope and complexity/level of detail (model abstraction), because simple models require less data and the results are easier to interpret since the structure of the model is better understood. Model simplifications should always be down to a level that still maintains sufficient accuracy for addressing the modelling objectives.

2.5 Impact and further work

The most important future challenge in European forest modelling post-COMET 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 (i.e. following a heuristic approach). 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 process 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, exposing them 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, ^{210}Po, ^{226}Ra, radioiodine, plutonium and uranium isotopes. Possibilities to compare data (measurements) and model calculations exist for Cs (and sometimes Sr and Pu), but not so much 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 can this 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 differently will radionuclides from underground sources 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 foodchain 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.

The Handbook 'Guidance on Forest models for contaminated sites' will contribute to the organisation of work of any potential forest modeller, because it provides in a readily usable manner a comprehensive amount of information for using forest models or developing new ones. The Handbook should be used as a tool to check aspects related to forest models and to look for solutions when issues arise during modelling activity. Furthermore, it may help the reader to conduct a successful sampling campaign to collect necessary data for model validation and calibration. The Handbook will be used as lecturing material at the Master course in radioecology at the NMBU and it will be distributed as a hard-copy.

The Handbook considers numerous aspects, which are often overlooked when developing, choosing or assessing the quality of a model. In particular, the link between modellers and experimentalists and the mutual benefits of a close interaction are highlighted. The advantages and disadvantages of empirical process-based approaches are stressed and the lack of knowledge of some processes e.g. mycorrhizal symbiosis and its effects on radionuclide uptake by plants. Suggestions are provided on how to address separately the quantification of the uncertainty and the variability.

In the future, models that are more realistic will be necessary given the increased amount of realism recommended in the EURATOM safety standards. To achieve this, it is on one hand necessary to improve understanding of the processes involved in radionuclide transfer in forests and hence to focus more on a process-based approach. On the other hand, it is fundamental to be able to quantify the uncertainty related to the model.

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Human Food Chain Modelling

3.1 Background

Risk assessment, in case of nuclear emergency, is confronted with uncertainties on the transfer of radioactive substances in terrestrial ecosystems and to human population through the food chain, which could affect the relevancy of decision-support. The extent of the repercussions of Chernobyl and Fukushima accidents highlighted the difficulty of managing the consequences of such disasters, specifically to accommodate the different sources of uncertainty within decision-making processes.

The overall objective of the Human food chain working group was to improve human food chain modelling in agricultural systems:

- through regional customization of relevant 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.

More details describing the key issues of the work are given below. The summary was based on the "Detailed plan for the COMET WP3 Initial Research Activity" (D3.1).

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 (see section 3.2 for more about the models). 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, with slow decrease in contamination over time, is typical for European wet peat ecosystems and need to be specifically addressed. Regarding element- or nuclide-specific model parameters, most empirical data concern isotopes of Cs, I, and Sr. It is thus 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 the use of probabilistic modelling generated a substantial interest in 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 and dose assessment model, the use of Bayesian networks could aid the separation of uncertainty and variability in model parameters.

The key issues addressed above are in line with the objectives of COMET WP 3 (as discussed in D3.1). 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 (*"To Predict Human and Wildlife Exposure in a Robust way by Quantifying Key Processes that Influence Radionuclide Transfers and Exposure"*) – research line (RL) 2, and to some extent RL1 and RL3 (Hinton et al., 2013). Most parts of the work proposed by our working group is also included as intended activities for the "Human Food Chain Roadmap" of the European Radioecology Alliance.

3.2 Methods and materials

More about the dynamic models – FDMT and SYMBIOSE

The **F**ood and **D**ose **M**odule **T**errestrial is used to predict transfer of radioactive substances in food chains – following radioactive fallout (concentrations in various products and doses to the population are calculated). The model is based on the ECOSYS dynamic model developed in the early 1990s (Müller and Pröhl, 1993). A large number of adjustable parameters are included in this module, where some are dependent on site and situation, whereas others have a more general validity. In the broadest sense, parameters in FDMT can be categorised (based on Raskob et al 2000; Pröhl and Müller, 2005) as being either: Element-independent – i.e. food products, plant growing and harvesting times, animal feeding characteristics, human consumption habits. Element dependent – i.e. translocation factors, soil leaching/fixation, soil-plant transfer factors, transfer factors to animal products, processing of feed-/foodstuffs. Nuclide-dependent – i.e. physical half-lives, various dose factors. For a detailed description of FDMT parameters including default values used, we refer to Müller et al. (2004). FDMT as implemented in ARGOS/JRODOS does not include features enabling probabilistic simulations by the user.

SYMBIOSE models the fate and transport of radioactive substances in environmental systems to assess their risks to humans, accounting for uncertainty and variability (Gonze et al., 2011; Simon-Cornu et al., 2015). This platform can be used in a wide range of situations for assessing risks induced by radioactive releases from nuclear facilities under normal operation, accidental or decommissioning conditions. Environmental models in SYMBIOSE address atmospheric, terrestrial, freshwater and marine systems as well as the major transfer processes at their interfaces. The modelled exposure pathways are external radiation (in the plume and outside of the plume) and internal contamination (inhalation, percutaneous transfer for tritium, accidental ingestion of sea sand and sea water, ingestion of foodstuffs, including drinking water, leafy vegetables, potatoes, cow milk, beef, hen eggs, river fish, sea fish, etc.). Since IRSN is both the developer and the user of SYMBIOSE, uncertainty and sensitivity analyses of the model can be performed (as opposed to FDMT).

Research approach

As described in D3.1, Four tasks were specified for the work within our IRA. These are summarised in Table 3.2.1. Partners included in each task are also shown. Some modifications have been done to the initial descriptions mainly in connection with the updated description of work for COMET (where FRAME and RATE were included), these are mentioned were relevant in the following.

Table 3.2.1.: Work overview (see main text for details)

Task	Short description	Participants
1	Derive human food chain parameter values appropriate for Nordic and Mediterranean ecosystems	STUK, NRPA, CIEMAT, IRSN
2	Use Bayesian methods to derive more robust values where data are lacking or have a large variability	IRSN, NRPA
3	Study long-term dynamics of radiocaesium mobility and plant availability for peat soils with unusually high transfers (laboratory experiment)	NUBIP
4	Investigate long-term dynamic soil-to-plant transfers for Tc-99, Pu, and Am (field experiments)	NUBIP, NERC, IRSN

Regional customization (task 1)

In this part, we focussed on Spain as representative of the Mediterranean area, whereas southern Finland and three climatic zones of Norway represented Nordic areas (as shown in Figure 3.2.1). The zones entitled Z1, Z2 and Z3 were derived using the start of growing season, defined here as the

approximate time the average temperature in a specified area exceeds 5°C (Thørring et al., 2016; Nielsen et al., 2009).

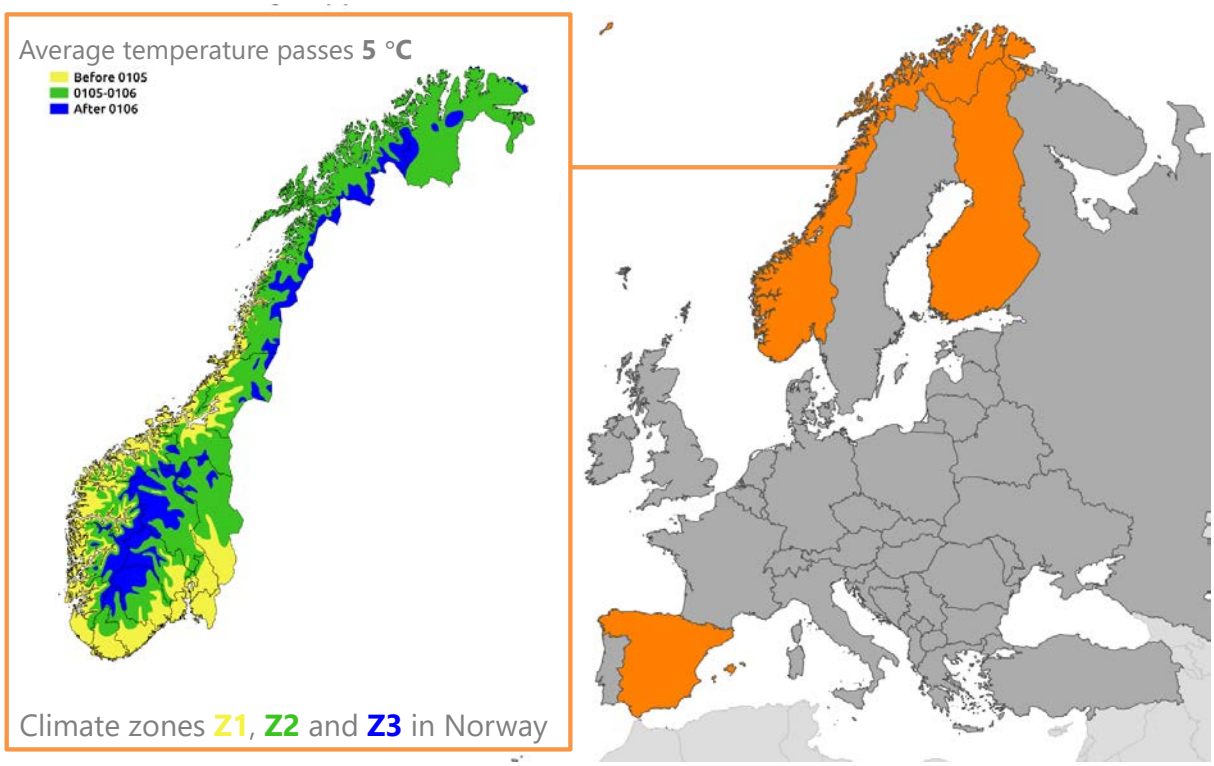

Figure 3.2.1.: Countries considered in the study. Norway were divided in three climatic zones (maps by Mari Komperød and Tanya H. Hevrøy, NRPA)

FDMT: Regional customization of FDMT parameter values has previously been done for seven eastern European countries (Raskob et al. 2000; Slavik et al. 2001), and relevant data for the Nordic countries (Hansen et al. 2010; Andersson et al. 2011) and Ireland (RPII, 2007) are also available. As described in several publications (e.g. Müller and Pröhl, 1993; Müller et al., 1993; Hansen et al., 2010), the model outcome from ECOSYS (the basis of FDMT) is highly sensitive to variations in a number of input parameters. Since FDMT in ARGOS/RODOS does not include features enabling sensitivity analysis by the user, we decided to use findings from the studies above in our prioritization of regional adaptation of parameters. Still, the uncertainty of model results is – as dealt with in Müller et al. (1993) – case specific, and does not provide general answers regarding the model uncertainties. However, recommendations regarding adaptation of FDMT parameters for specific regions or sites have also been developed within the JRODOS or ARGOS communities. These recommendations are largely based on expert judgment from the developers of ECOSYS or other experienced users of FDMT (Pröhl and Müller, 2005; Raskob et al 2000; Raskob, 2014). Generally, parameters are rated as being of high, moderate or low priority in relation to regional updating.

Numerous approximations, interpolations, extrapolations and "educated guesses" were required to derive parameter values applicable for use within FDMT. For details regarding such assumptions, we refer to Thørring et. al (2016).

SYMBIOSE was regionally adapted using updated parameters from Finland, Norway (Z1) and Spain from the FDMT work (described above). However, differences in modelling choices influences the direct applicability of FDMT parameters within SYMBIOSE, as further described in Thørring et al. 2016.

Case studies: To study the effect of the parameter value updates on foodstuffs activity concentrations and intake doses for different age groups, two "simple" scenarios were specified: a dry deposition scenario and a wet deposition scenario with a specified amount of rainfall (10 mm). For both scenarios,

the deposition date was set to 1 August[1]. Four radionuclides were included: Cs-134, Cs-137, Sr-90 and I-131, with an assumed 1000 Bq m^{-2} deposition of each nuclide. Both models – FDMT and SYMBIOSE – were run for a 5-year period. Initial runs, using default parameter values, were performed in early 2014 as a modelling test (example shown in Figure 3.2.2). Unfortunately, CIEMAT did not participate, and only results from the "wet" scenario was available from STUK (the other JRODOS user).

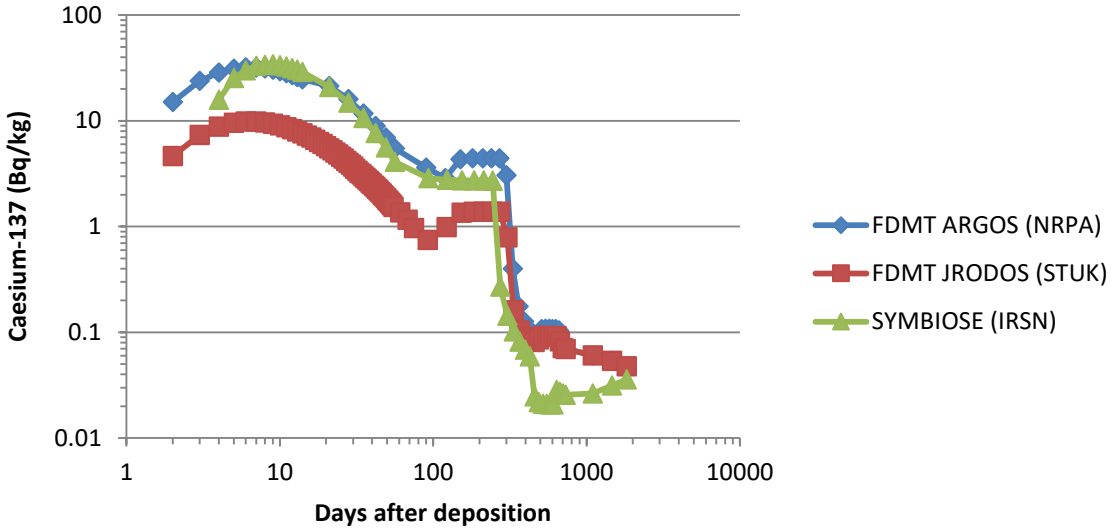

Figure 3.2.2.: Cs-137 in cow milk (from initial runs, wet scenario). The deviation between STUK and NRPA is attributable to amount of rainfall used – 10 mm and 3 mm, respectively (see main text below for explanation)

Final runs, using defaults and updated parameter values, were performed in late 2016. Note that for Norway only Z1 was considered in the model runs. For these model runs, STUK and CIEMAT used JRODOS July 2014 Update 3 version, whereas NRPA used ARGOS 9.4 with FDMT/AgriCP version 3.0. Unfortunately, it was not possible within ARGOS to change the default 3 mm amount of rainfall to 10 mm for the wet-scenario. Consequently, default 3 mm was used for the Norwegian model runs, which resulted in higher interception of fallout radionuclides (and about three times higher contamination values for the first few months of the modelling period as compared to Finland and Spain).

SYMBIOSE V2.2.3 was run using five different sets of "regional" data – default values of SYMBIOSE (assumed to be representative of France), default values of FDMT (assumed to be representative of Central Europe), and the values proposed for Finland, Norway and Spain within COMET.

Validation of models for task 1 was excluded from COMET (cf. the updated description of work).

Advanced statistical methods (task 2)

The study performed in task 2 was in four parts: (a) Development of a methodology to take into account uncertainties within environmental and food risk assessment models using Hierarchical Bayesian Models (HBM); (b) Application of this methodology to weathering loss, i.e. the reduction of radionuclides concentrations in plant foliage due to wind, rain, biological dilution due to growth etc.; (c) Propagation of parametric uncertainty to predict fate of radiocaesium in leafy vegetables growing in Fukushima prefecture; (d) Propagation of parametric uncertainty for the hypothetical deposition scenario of task 1.

[1] Other dates were also considered for Norway

(a) HBM methodology

A methodology was proposed to account for parametric uncertainty. This relies on:

- *Bayesian inference*, a branch of statistics well suited to express parametric uncertainty, as the evidence about the true state of the world is expressed in terms of degrees of belief (a distribution). The posterior distribution (or updated probability estimate) is the combined result of the observed data and *a priori* information (prior).
- *Hierarchical modelling*, i.e. statistical modelling written in multiple levels (hierarchical form). Hierarchical modelling is used when information is available on several different levels of observational units. This hierarchical form of analysis is appropriate to separate uncertainty (our degree of belief formulating our lack of knowledge) and variability (differences between plants, radionuclides…).
- *Meta-analysis*, i.e. statistical analysis that combines the results of multiple scientific studies. HBM (Hierarchichal Bayesian modelling) is one of the convenient approaches to perform meta-analyses.

This methodology was applied to three radioecological parameters of the foliar pathway: dry interception (Sy et al. 2015), wet interception (Gonze & Sy, 2016) and – in the frame of this COMET IRA – weathering loss parameter, see part (b).

(b) HBM meta-analysis about weathering loss

Weathering loss is an important parameter in accidental situations following foliar interception of radionuclides. For the present work, the weathering half-life database compiled by NRPA (Andersson et al., 2011), was expanded by IRSN to include more publications. This updated set of weathering half-life values, covering the period from the 1960s to 2014, was divided into three sub-sets: iodine field studies (I), and other radionuclides – laboratory (L) or field studies (F) – as shown in Table 3.2.2.

Table 3.2.2.: Main characteristics of the weathering half-life database (n=317). Number of data for each category is shown in parentheses

Database	Elements	Physical forms	Plant types
Iodine/field (I)	I (115)	Gaseous (34) Particle (25) Unknown (56)	Herbaceous vegetation (9) Pasture grass (106)
Other elements/field (F)	Ce (2), Cs (81), Rb (8), Ru (2), Sr (34), Tc (8), Zr (1)	Particle (136)	Herbaceous vegetation (16) Leafy vegetables (3) Non-leafy vegetables (6) Pasture grass (111)
Other elements/lab (L)	Ce (1), Cs (17), Co (10), Mn (10), Ru (10), Sr (18)	Particle (66)	Leafy vegetables (25) Non-leafy vegetables (37) Pasture grass (4)

For each dataset, hierarchical Bayesian models were constructed, sharing common features:

- Logarithms of the weathering loss values are assumed to be i.i.d realizations of a Gaussian distribution with unknown variance and mean parameter.
- Each model captures simultaneously between plant and between radionuclides variability
- Each model accounts specifically for the biological dilution effect (λ_i^{BD})

The models providing best fit were selected upon the application of Deviance Information Criterion (DIC) (Spiegelhalter et al., 2002). The hierarchical framework incorporates a level of complexity, which

enables studying the specific variations across elements, their physical forms and the different considered plant species.

(c) Case-study: leafy vegetables in Fukushima prefecture

The impact of uncertainty and variability about radioecological parameters characterized by HBM (and others characterized by Simon-Cornu et al., 2015) was assessed by SYMBIOSE, with stochastic simulations and sensitivity analyses applied on case studies. The first case study aimed to predict fate of radiocaesium in leafy vegetables after the accident of Fukushima nuclear power plant, within 80 km around this nuclear power plant, where the spatial dispersion of Cs-134 and Cs-137 deposition and the dry deposit proportions were assumed to be known. Different numerical simulations using the Monte Carlo sampling method (with samples size set to 1000) were performed in the SYMBIOSE platform, in which ordinary differential equations are solved, using the 4th order Runge–Kutta method. Deposits were modelled from March 15th, 2011, to April 1st, 2011 and transfers were modelled from March 15th, 2011, to March 15th, 2012. The sensitivity analyses were performed by computing Spearman correlation coefficients (SCC) and Morris indices for different dates and different meshes. For more details about the methods used, we refer to Sy et al. (2016).

(d) Case study: the task 1 dry scenario

Probabilistic simulations were also performed using SYMBIOSE for a hypothetical deposition scenario that was basically the same as the "dry" scenario described in task 1 (considering the first year after fallout, only). The uncertainty distributions assigned to the dry interception fractions were derived from Sy *et al.* (2015). Moreover, uncertainty distributions assigned to root uptake and transfer of radionuclides from feed to animal products were taken from Simon-Cornu *et al.* (2015). Uncertainty in the outputs (concentration values and the effective doses) was characterized by Monte Carlo simulation, and quantified using the uncertainty ratio (UR) – i.e. the ratio between the 95th percentile and the median of the distribution of concentration values and of the distribution of effective doses. Spearman correlation coefficients were calculated to assess sensitivity of model outputs to changes in the input parameters. For more details about the methods used, we refer to Sy (2016).

Long-term dynamics of soil-to-plant transfer (tasks 3 and 4)

The work performed in these tasks where field experiments that could be relevant in relation to modelling. As stated in D3.1, the intention was to use the data to "improve input parameters in FDMT and SYMBIOSE", but for the revised list of milestones and deliverables this activity was removed, and IRA-Human Food Chain D3 was confined to task 1 output (i.e. Thørring et al., 2016).

In task 3, the focus was on Cs in peat soil. A two-year laboratory experiment using peat-bog soils of Rokytne 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 – as shown in Figure 3.2.3. Caesium-137 soil to plant transfer factors of natural grasses reach 0.1–0.2 m^2/kg. The present study compared root uptake of 'freshly added' and 'old' Chernobyl radiocaesium using spiny rush (*Juncus acutus* L.) as model plant.

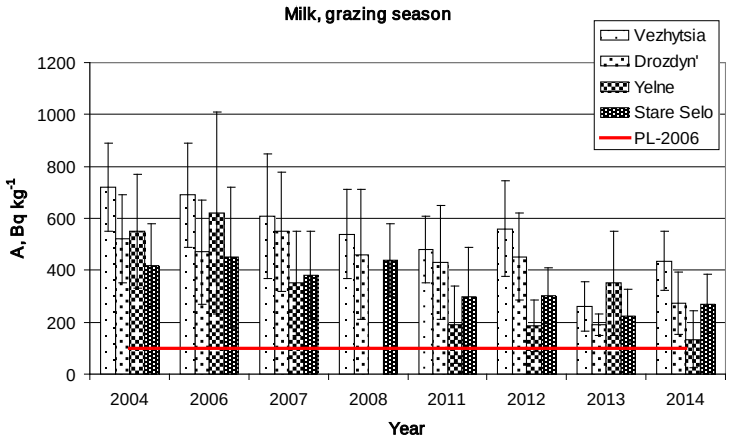

Figure 3.2.3.: Dynamics of Cs-137 milk contamination for different settlements (arithmetic mean, standard deviation, n>20

For task 3, comparison with other relevant areas (e.g. Norway) was not performed. This was, again, due to modifications in the updated work description. Thus, IRSN, NERC and NRPA did not – as initially planned – take part in this activity.

In task 4, the focus was on less studied radionuclides (as compared to Cs-137, Cs-134, Sr-90, I-131). The studies were generally continued experiments in the Chernobyl exclusion zone (CEZ). The aim 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 – Podzoluvisol, Greysem, Phaozem, Chernozem. Results from the Trans-uranic elements will not be given in the present report.

For Tc-99, plant uptake and vertical migration / soil retention was studied after two consecutive applications of the radionuclide to uncontaminated soils (using lysimeters) in 2005 and 2006. The work included in COMET covered the period 2005–2014.

Detailed description of sampling procedures, extraction methods and measurements for the experiments where considered out of the scope of the present report. For more details, we therefore refer to Kashparov et al. (2015) for task 3 and Levchuk et al. (2015) for task 4.

3.3 Results

A short summary of results from this IRA was included in D3.2. More details from all work tasks are given in the following.

Regional customization (task 1)

Based on the recommendations regarding regional updating (Pröhl and Müller, 2005), and to a lesser extent on sensitivity analyses (e.g. Müller et al., 1993), important parameters in relation to regional adaptation were identified (Table 3.3.1). Except for "Uptake from soil" (which is element-specific), they all belong to the FDMT "element-independent" category. Note that all nuclide-specific parameters are considered low priority in relation to regional updating (i.e. they have general validity).

Table 3.3.1.: Summary of important parameters in relation to regional adaptation

Category	Parameter
Contamination of plants due to direct deposition	• Relevant growth periods • Leaf area indices (LAI) • Yields • Period of preparing winter feed
Animal parameters	• Animal specific feeding rations

Human habits	• Age-dependent consumption rates • Seasonality of consumption rates
Uptake from soil	• Transfer factors • Migration rates

Due to limited amount of time for this task, we could not consider all important categories, and we, therefore, decided not to include updates in radionuclide soil-plant transfer factors in COMET – even though this parameter is highly important, as has e.g. been demonstrated by the long-term trends of Cs-137 in Norway. Regarding the updated parameters, short comments/descriptions and a couple of examples are provided below. For full details, we refer to Thørring et al. (2016).

Leaf area index (LAI) characterises the plants´ stage of development, and is defined as the area of plant leaves (m^2) present on a unit area of ground (m^2) (Müller and Pröhl, 1993). The LAI is plant dependent, and varies strongly through the year in relation to plant growth stage. We performed regional customization for LAI, growth periods, harvest, and yields for regionally relevant cereals, vegetables and to some extent fruit and berries. Grass for harvest were also included.

Regarding feedstuffs and animal feeding regimes, practice varies between countries. We modified pasture periods in accordance with the growing season in respective countries / climate zones. Type and amount of concentrates were also considered for each country. Table 3.3.2 shows grazing periods for cows in various regions (default values for FDMT and SYMBIOSE are also included). Note that the grazing period for Spain is from early March to end of November, whereas the grazing period for Norway Z3 is from late June to End of August. This is obviously crucial information when considering possible impact of radioactive fallout.

Table 3.3.2.: Grazing period for cows in different regions

	Spain	Finland	Norway			FDMT	SYMBIOSE
			Z1	Z2	Z3		
Start	Mar 1	Jun 1	May 15	Jun 1	Jun 20	May 11	Apr 1
End	Nov 30	Sep 15	Sep 14	Sep 14	Aug 31	Nov 9	Oct 14

Finally, we have included a table comparing diets for adults from Finland, Norway, and Spain (with FDMT defaults). How comparable these country-specific data are, is difficult to say, since this will depend on the statistical information available for each country, and on the assumptions made to get from e.g. “bread” in dietary surveys, to the very detailed requirements of FDMT for cereals – as evident from Table 3.3.3.

Table 3.3.3.: Average food consumption (g/day) for adults

Category	Food product	Finland	Norway	Spain	FDMT default
Cereals	Spring wheat, whole grain	*	72	134	2.6
	Spring wheat, flour	55	96	***	15
	Winter wheat, whole grain	*	**	21	23
	Winter wheat, flour	4.8	**	6.8	130
	Rye, whole grain	*	6.5	***	8.7
	Rye, flour	42	6.5	*	35
	Oats	8.2	13	***	5.6
Vegetables	Potatoes	73	67	76	160
	Leafy vegetables	19	28	135	94
	Root vegetables	34	53	29	33
	Fruit vegetables	72	56	94	47
Fruit and berries	Fruit	145	140	179	120
	Berries	22	14	6.4	14
	Milk	272	317	352	230

Milk products	Condensed milk	**	**	1.2	18
	Cream	17	22	1.8	16
	Butter	16	5.5	1.1	18
	Cheese (rennet)	39	34	***	26
	Cheese (acid)	*	4.4	22	17
Meat	Beef (cow)	24	15	117	27
	Beef (cattle)	*	24	***	55
	Veal	*	1.0	***	2.2
	Pork	29	53	19	108
	Lamb	1.4	11	***	3.9
	Chicken	33	41	41	17
Other	Eggs	20	26	8.5	43
	Beer	268	85	37	610

*Not known / not considered in the statistics, **insignificant / assumed to be zero, ***not given separately. See Thørring et al. (2016) for all assumptions made for each country.

FDMT model runs – notes and selected results

The initial plan for this report was to compare results using updated parameter values for Finland, Norway and Spain, but since the default runs gave different results for all countries, especially for the "dry" case (as shown in Figure 3.3.1), we decided to present selected results from the different countries, separately. Obviously, JRODOS and ARGOS use different approaches, and specifying the "simple" scenarios did not prove to be so simple after all – not even between users of the same version of JRODOS.

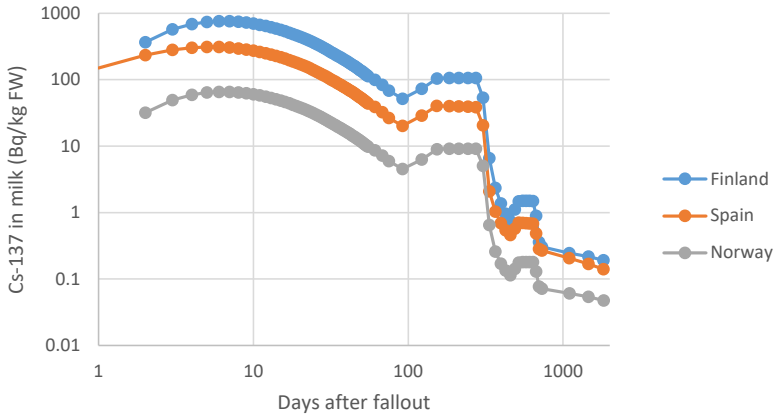

Figure 3.3.1: Dry deposition case (Cs-137, 1000 Bq/m^2) predicted levels in milk using FDMT defaults

"Bugs" were encountered in both JRODOS and ARGOS, making some of the model outputs from our runs somewhat dubious. However, details regarding the modelling is regarded outside the scope of this summary report. Anyway, the main purpose/output of the work in task 1 was the actual parameter updates (as given in Thørring et al., 2016).

Below, some results from the model runs by STUK, NRPA and CIEMAT are presented separately:

Finland: Calculations were performed for cow's milk, leafy vegetables, spring and winter wheat, beef (cow), lamb meat and pork as well as for different feedstuffs. An order of magnitude – and sometimes bigger – differences corresponding to certain points (or intervals) of time could be observed between some activity concentrations and/or nuclide specific doses estimated using Finnish and JRODOS default parameter values. The greatest total disparity was found in the wet-scenario one-year doses that originated from eating pork – depending on the nuclide and age (adult, one-year old child) the difference was up to a few orders of magnitude (the Finnish doses – though still being small – were bigger). This is likely due to two facts: First, winter barley used to feed pigs in Central Europe was

already harvested before the deposition while in Finland the harvest of spring barley, which is a basic feedstuff item for pigs in Finland, took place a couple of weeks after the release. Second and more important, in dry conditions the intake of radionuclides by inhalation by pigs seems to be the dominant contributor to the contamination of pork during the first few months while in wet conditions this pathway causes much smaller activity concentrations in pork (because of the much smaller nuclide concentrations in the ground-level air). In the wet scenario, this in turn greatly emphasized the effect of Finnish pigs eating contaminated feedstuff earlier than the pigs in Central Europe (FDMT defaults used).

All the other differences found could also, at least to some extent, be traced back to the differences in animal feeding habits, growing and harvesting seasons of foodstuff cereals and plant-based feedstuff items and in consumption figures. However, as mentioned above there were some difficulties encountered during the JRODOS calculations.

Generally, both in Central European and Finnish conditions nuclide specific doses received in the wet scenario were much smaller than those received in the dry scenario. The doses (one year doses or lifetime doses), that were in all cases bigger in Central Europe, were those from winter wheat and leafy vegetables, while in Finland the doses from spring wheat were always bigger. Milk (Figure 3.3.2) was the major dose contributor in all calculations.

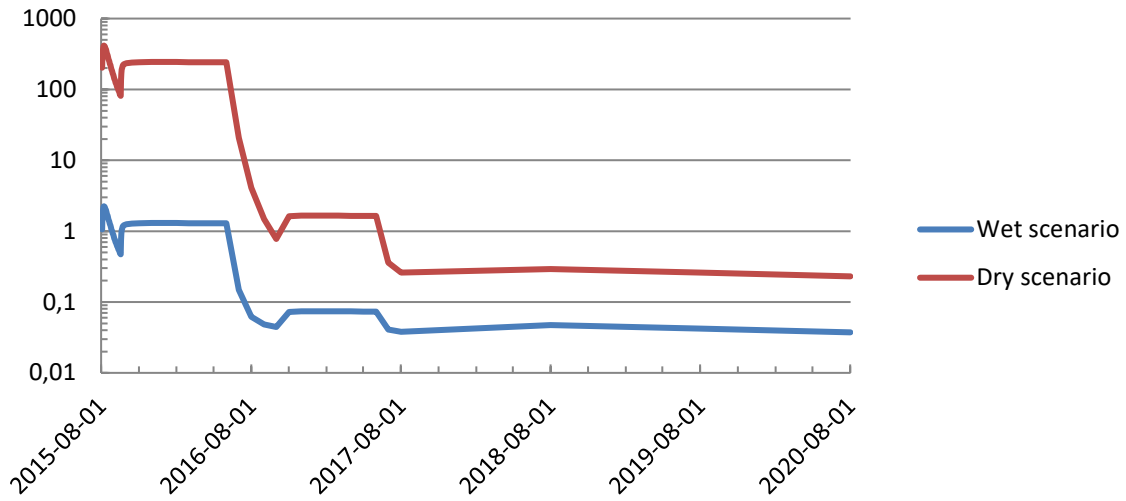

Figure 3.3.2.: Cs-137 concentration (Bq/kg) in cow milk (dry and wet scenarios) calculated by FDMT model (JRODOS) with Finnish parameters. One-year doses for small children from Cs-137 in milk were 0.26 mSv in the dry scenario and 1.4 µSv in the wet scenario. For adults the corresponding doses were 3-4 per cent higher.

Norway: For the first period following the 1 August deposition, predicted activity concentrations in foodstuffs were generally higher for the dry-scenario (as shown in Figure 3.3.3). Comparison of runs using default and Norwegian updates showed that e.g. levels of Cs-137 in winter (or spring) wheat were 2–4 times higher using the updated parameters (not shown). In contrast, levels in lamb meat and cow milk were slightly lower compared to default (0.7) – probably reflecting the lower grass intake for lamb and cows in Norway (and the use of supplementary feed for cows – spring barley and oats). The same trend was observed for I-131 and Sr-90 (in milk).

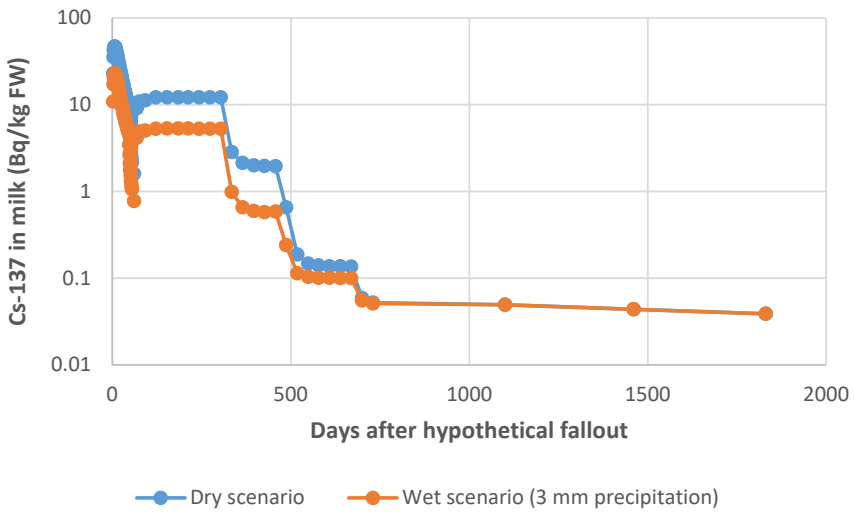

Figure 3.3.3.: Cs-137 concentration (Bq/kg) in cow milk (dry and wet scenarios) calculated by FDMT in ARGOS with Norwegian (Z1) parameters for the whole 5-year prediction period

For milk and lamb meat, additional deposition dates were considered in our model runs. Radioactive fallout during the summer pasture period (May–September) had largest impact on the Cs-137 levels – reaching almost 50 Bq/l for cow milk and more than 400 Bq/kg for lamb meat (using Norwegian update, Figure 3.3.4). The lamb slaughter period in Norway is September–October, so the actual deposition and slaughter date is crucial for the contamination level in the meat produced. Deposition date is also important for levels of radionuclides in winter feed for cows (i.e. hay – or actually grass silage). Highest levels in hay (and consequently in milk) were found using fallout 1 June, decreasing in the following order: 1 July > 1 August > 1 September > 1 May.

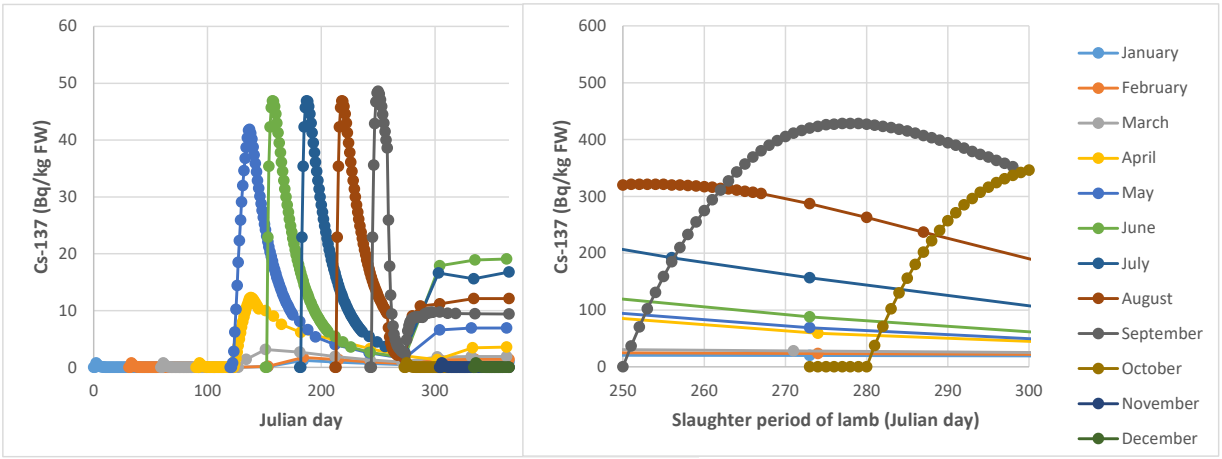

Figure 3.3.4.: Cs-137 levels in milk and lamb meat using different deposition dates (Dry scenario)

For the late part of the 5-year period considered in our runs, soil-plant transfer is the main factor controlling levels in plant and animal products. Since neither transfer factors (TF) nor soil migration rates were changed for our model update (see Section 3.2), no significant differences in results were observed between runs using default or updated parameter values for this period.

Using cow milk as an example, the accumulated effective ingestion dose from Cs-134, Cs-137, I-131 and Sr-90 in milk were approximately 2 times higher using updated parameters for all age groups except 1 year olds, where the corresponding ratio was around 0.5. The reason for this is probably that default milk intake for this age group is rather high (560 g/day, assuming no milk substitutes).

Spain: Calculations were carried out for Cow milk, Beef (cow), Leafy vegetables, Winter wheat, Flour wheat, Pork, and Grass intensive, Hay intensive and Grass extensive. The results obtained show clearly that the highest values occur in dry scenarios, both Default and Mediterranean, rather than in wet scenarios. The magnitude and temporal development of the activity concentrations of these foodstuffs are clearly season dependant. During the selected deposition date, 1st August, the winter cereals in the Mediterranean areas are already harvested, so the activity concentrations of Winter wheat and Flour wheat, in the following years, come from the root absorption of the radionuclides deposited on the bare soil and are several orders of magnitude lower (see Figure 3.3.5).

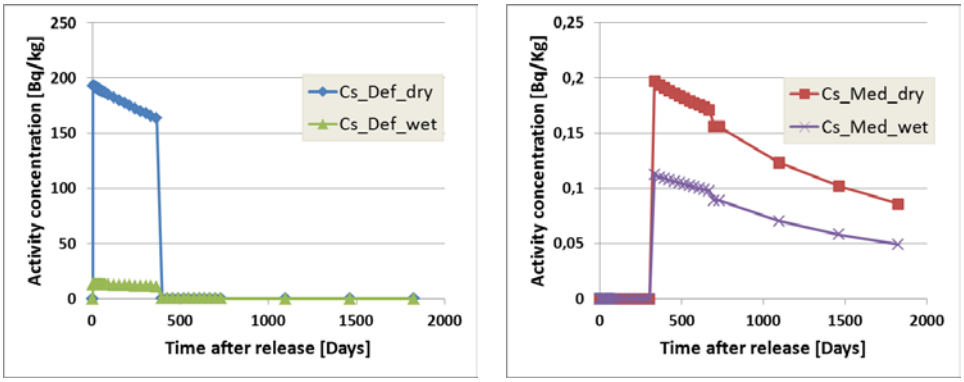

Figure 3.3.5.: Activity concentration of caesium isotopes with time in Winter Wheat for the scenarios "Default" and "Mediterranean"

Seasonality will also affect the activity concentrations of the animal products following the time schedule of the animal diet (grazing periods and feed stuffs ingestion). Figure 3.3.6 compares the time development of the activity concentrations of caesium isotopes in Cow milk and Beef cow.

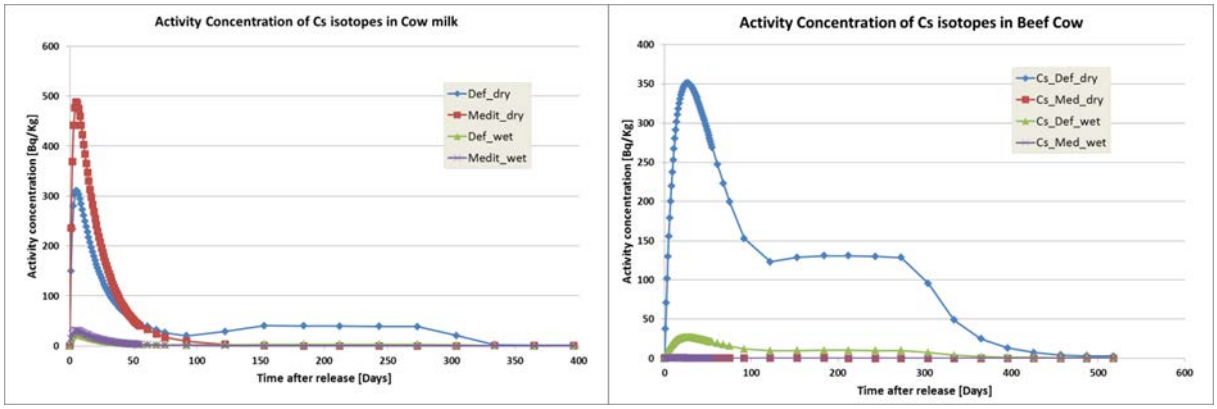

Figure 3.3.6.: Activity concentration of caesium isotopes with time in Cow milk and Beef cow for the scenarios "Default" and "Mediterranean"

The ranking of activity concentrations from the highest to the lowest values, among the different scenarios show the following trend:

- Dry default > Dry Mediterranean >Wet default > Wet Mediterranean, for the Leafy vegetables
- Dry Mediterranean > Dry default > Wet Mediterranean >Wet default, for the intensive and extensive Grass, as well as Pork and Cow milk.
- Dry default > Wet default > Dry Mediterranean > Wet Mediterranean, for the Hay intensive products, Beef cow and Winter wheat products.

Regarding the ingestion doses (Figure 3.3.7), the values of the doses from Mediterranean scenarios are lower than the respective values from Default scenarios, for the three groups of isotopes studied (caesium, iodine and strontium isotopes). Only the values for Cow milk are in the same range of magnitude. In the case of Leafy vegetables, Pork and Winter wheat the doses are between one and two orders of magnitude lower (except for iodine, which is irrelevant in the Mediterranean scenarios). In the case of Beef cow and Wheat flour, the values are reduced by 3-4 orders of magnitude.

Regarding the contribution of the different foodstuffs in each age group, the Default scenarios show, for the age group of 1 year, that Cow milk is the product that contributes most to the doses, for the three radionuclides considered. For the age group above 10 years the doses come mostly from the ingestion of Leafy vegetables, followed by Cow milk and Wheat flour. In the Mediterranean scenarios, Cow milk ingestion, in every age group, is the product that contributes most to the doses, followed by Leafy vegetables. There is also a small contribution of Cs and I from Pork and Beef ingestion.

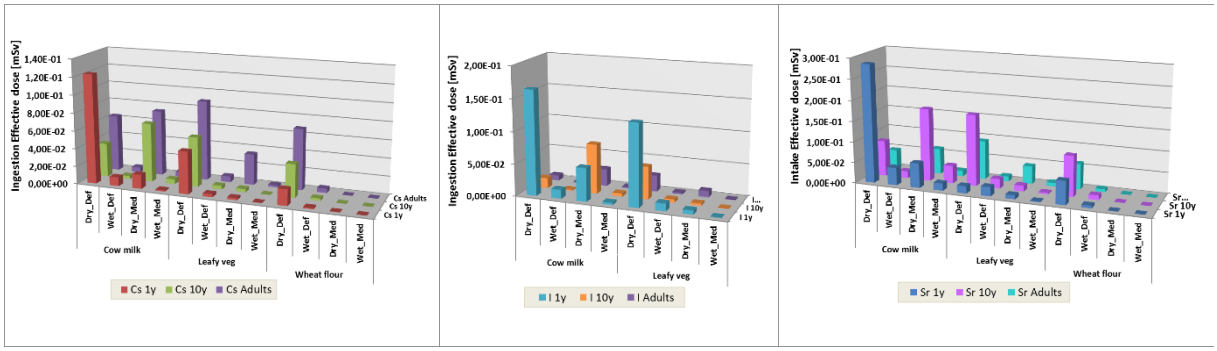

Figure 3.3.7.: Contribution to the effective dose by ingestion (whole 5-year period) from the most relevant foodstuffs in relation to isotopes group and age group. From left to right, the graphs show caesium, iodine and strontium isotopes, respectively.

SYMBIOSE model runs – notes and main results

Iodine-131 is the radionuclide for which the highest differences were observed from one scenario to another, and this is particularly true for the very young children (1 year old). This dose is mostly explained by consumption of leafy vegetables and cow milk.

Leafy vegetables: Considering that the default SYMBIOSE model does not explicitly account for LAI for leafy vegetables, the predicted levels of Cs-134, Cs-137, I-131 and Sr-90 in leafy vegetables are identical whatever the country. One month after dry deposition, they are ca. 25 Bq/kg for Cs-134, Cs-137, and Sr-90, and 1.3 Bq/kg for I-131. For the wet scenario, predictions are lower: 5 Bq/kg for Cs-134, Cs-137, and Sr-90, and <0.1 Bq/kg for I-131.

In the SYMBIOSE default database, an adult eats 87g/day of fresh leafy vegetables (73% of them locally produced) and 15 g/day of canned leafy vegetables (40% of them locally produced), these canned leafy vegetables being stored 3 months before consumption. Thus, in the 3 first months, the local consumption is only 87x0.73=63.5 g/day, which is 1.5 times less than the assumption in FDMT, in which an adult eats 94 g/day of fresh leafy vegetables (100% locally produced). These SYMBIOSE and FDMT consumption data are intermediate between the Finnish and Spanish data (resp. 19 g/day and ca 135 g/day).

Thus, differences from one country to another in predicted doses linked to leafy vegetables ingestion are proportional to these food diets, ranging for an adult from about 1 µSv (for 1 kBq/m² dry deposited of anyone of the simulated radionuclides) in Finland to 10 µSv (for 1 kBq/m² dry deposited of anyone of the simulated radionuclides) in Spain. For the wet scenario, they are 5 to 10 times lower.

Cow milk: In August (date of the simulated accident), cows are assumed to be outside, grazing, in all countries. Thus, in the first month, differences between countries in predicted activities in cow milk are linked to the quantity of grass assumed to be consumed daily (with low consumption of grass in Finland and Norway, where cows are supplemented with other feedstuffs). Dynamics in the following months depend on the winter feed diet and the harvest dates of these feedstuffs. In the French (SYMBIOSE) scenario, cows are assumed to eat maize silage harvested late September. Under this assumption, the decrease of the milk activity stops as soon as the cows stop grazing.

The French scenario is then more pessimistic than others for the prediction of activity in milk, but the default food diet of SYMBIOSE is less pessimistic (as total consumption of cow milk includes consumption of fresh milk and canned milk, with longer storage and lower local production). For the one-year-old child, the default assumption is that they drink no fresh milk, only canned milk.

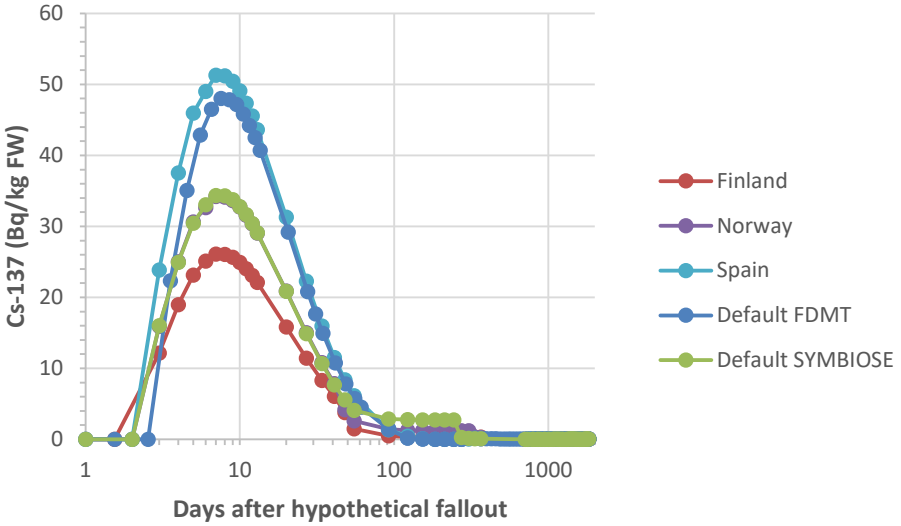

Figure 3.3.8.: Predicted concentrations of Cs-137 in milk for the wet deposition

Accounting for all differences, doses linked to Cs-137 in cow milk in the wet scenario range for an adult from about 1 µSv (for 1 kBq/m² wet deposited) in France to 5.3 µSv (for 1 kBq/m² wet deposited) in Spain (Figure 3.3.8). In the dry scenario, they are 6 to 10 times higher.

Advanced statistical methods (task 2)

(a-b) HBM methodology, applied to weathering loss

The Bayesian models developed demonstrated the influence of plant species and/or type of radionuclide on weathering loss, depending on type of data set (i.e. iodine field studies, other radionuclides – laboratory or field studies, see Table 2). In addition, this approach enables taking into account, in an explicit way, the effect of biological dilution (when relevant). To illustrate this, Figure 3.3.9 shows the prior and posterior distribution for one of the parameters expressing this biological dilution.

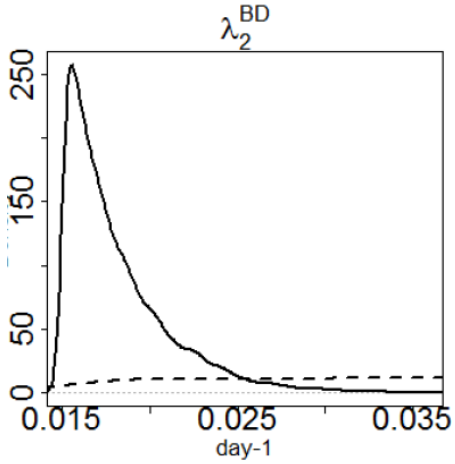

Figure 3.3.9.: Prior (--) and posterior (-) distribution of the parameter expressing biological dilution of radionuclides other than iodine in (field) pasture grass

For more info e.g. regarding summary statistics of the simulated posterior distributions of the parameters and latent variables of the different Bayesian models we refer to Sy (2016).

(c) Case-study: leafy vegetables in Fukushima prefecture

Sy et al. (2016) provided a three-dimensional (temporal variability, spatial variability, and parametric uncertainty) analysis of predicted levels of Cs-137 in leafy vegetables within 80 km around the Fukushima Dai-ichi nuclear power plant. The conclusions of the sensitivity analysis was that in the earlier phase (from April to September 2011) the interception fractions (dry/wet) and the weathering loss parameter were the most influential input parameters on uncertainty, with regards to both sensitivity analysis methods (the SCC values and Morris indices). The SCC value calculated for the weathering loss parameter is negative and reflects the fact that the higher the weathering loss value the lower the concentration of Cs-137 in leafy vegetables. In addition, the impact of this parameter grows increasingly over time.

(d) Case-study: the task 1 dry scenario

The results obtained from the Monte Carlo simulations cover Cs-137, Cs-134, Sr-90 and I-131 in nine food items – i.e. fresh leafy vegetables, canned leafy vegetables, flour, beef, pork, sheep meat, fresh cow milk, canned cow milk and goat milk. Only selected results are provided in the following.

Figure 3.3.10 illustrates the dynamics of Cs-137 in fresh cow milk and leafy vegetables. For milk, the uncertainty regarding the nuclide decreases rapidly the first weeks after the hypothetical fallout, but increases again the following months. These fluctuations, explained by seasonal variations of feeding conditions (i.e. indoors with contaminated or not contaminated forage, outdoors with less and less contaminated grass.), are more immediate for milk and smoother for meat (not shown).

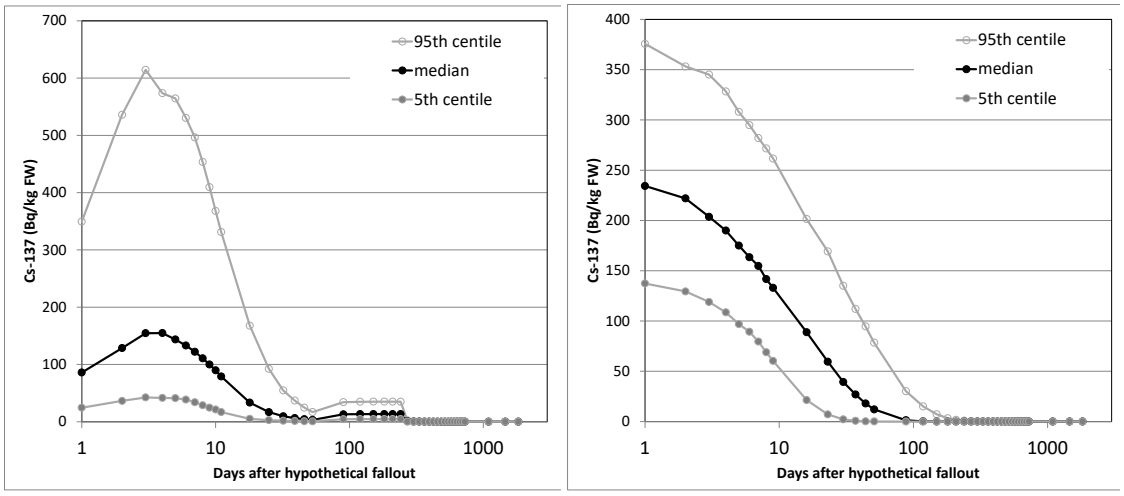

Figure 3.3.10.: Time development of Cs-137 (Bq/kg FW) in cow milk (left) and leafy vegetables. The bandwidth illustrates the dynamics of the uncertainty (90% CI)

The dynamics of the effective dose of Cs-137 received by an adult and a young child are illustrated in Figure 3.3.11. During the first month following the deposition, the effective doses of caesium isotopes received by an adult are found to be higher than the doses received by a young child, whereas the effective doses of ^{131}I and ^{90}Sr (not shown) are higher for young child.

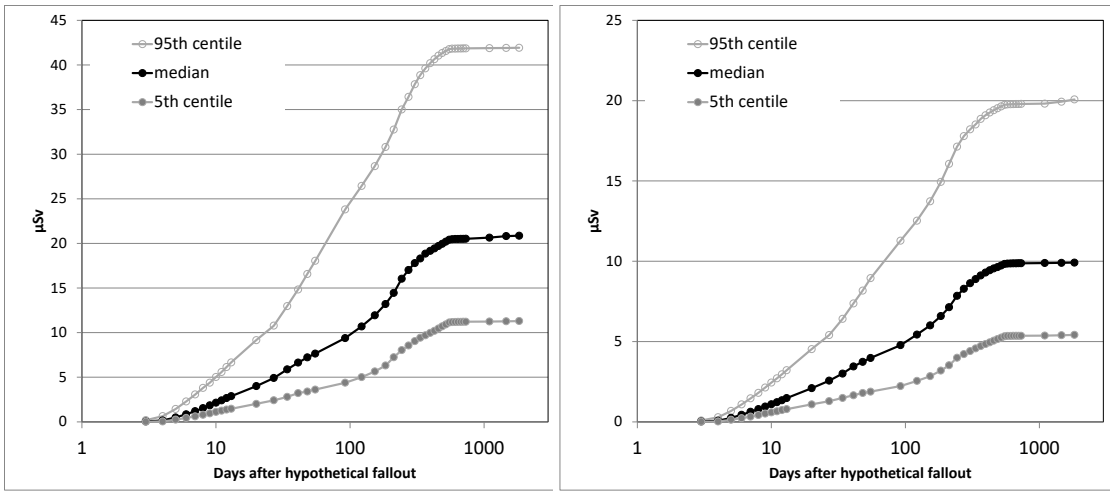

Figure 3.3.11.: Cumulative effective doses (µSv) of ^{137}Cs received by an adult and a young child. The bandwidth illustrates the dynamics of the uncertainty (90% CI)

Table 3.3.4 display the Spearman correlation coefficients (SCC) calculated between the effective doses and different uncertain input parameters, respectively. Only the most influential parameters with respect to SCC values for Cs-134/137 are presented.

Table 3.3.4.: Spearman correlation coefficients calculated between the effective ingested dose of Cs-137 received by an adult. Only the parameters with highest impact on the distribution of effective doses are displayed.

Input parameter	1 month	3 months	1 year
Foliar Biomass – Grass	-0.54	-0.49	-0.38
Weathering loss – Grass	-0.31	-0.30	-0.19
Weathering loss - Leafy Vegetables	-0.31	-0.38	-0.29
Dry Interception - Leafy Vegetables	0.31	0.31	0.26
ETF - Cow Milk	0.46	0.33	0.42

For the effective doses, the foliar biomass of grass and leafy vegetables, the dry interception, the weathering loss parameter and the equilibrium transfer factor to animal products are the most influential parameters. These observations confirm the influence of the foliar pathway on the effective doses as previously suggested by SCC values calculated for the concentrations of caesium, iodine and strontium in food items (not shown here) and by the contribution of each food item on the predicted effective doses. For more results and detailed discussion on results, we refer to Sy (2016).

Note that the results presented in this section is somewhat different from those given in Sy (2016). Recently, i.e. posterior to delivery of COMET milestones and deliverables and the defence of M. Sy's PhD (Sy, 2016), it was discovered that the parameter describing dry foliar interception of all radionuclides by maize silage was erroneous (overestimated) in SYMBIOSE[2]. This error has been corrected for the results shown here.

Long-term dynamics (tasks 3 and 4)

Main findings from the Cs in peat soil experiments: As shown in Figure 3.3.12, the ratio of TF_{fresh} to $TF_{Chernobyl}$ – initially around 3 – reached equilibrium after 1–2 years. The effective half time of the fast component (T_{eff1}) was thus estimated at 0.2–0.3 years. The decrease was due to reduced levels of bioavailable forms of radiocaesium with time.

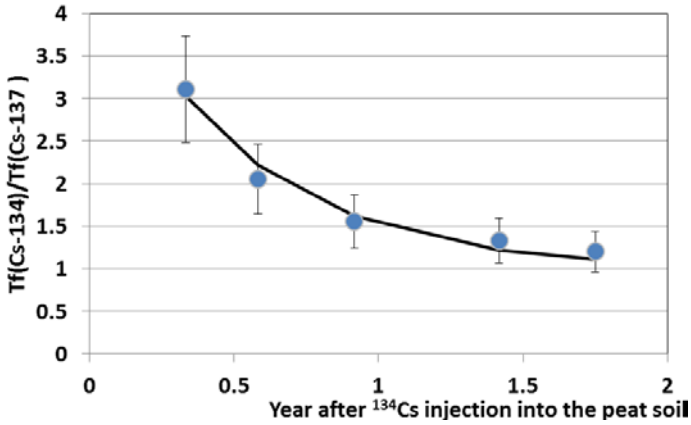

Figure 3.3.12.: Ratio of 'freshly added' and 'old' Chernobyl radiocaesium uptake by Spiny rush (*Juncus acutus* L.) as a function of time

[2] See: https://gforge.irsn.fr/gf/project/symbiose/tracker/?action=TrackerItemEdit&tracker_item_id=29999&start=50)

A paper has been published in Ukrainian (Maloshtan et al., 2015). More details can also be found in Kashparov et al. (2015).

Main findings from the Tc-99 field experiments: One year after the first administration, 20–50 % of the Tc-99 remained in the top soil (0–30 cm). This was mostly due to vertical migration with soil water, but the effect of plant uptake was significant (even though variable). After 6–7 years, only 11–18 % of the Tc-99 added (in two stages) remained in the upper 20 cm of the soils, corresponding to an effective half removal time of 2–3 years. Water-soluble fractions of Tc-99 decreased from 20–30% in 2006 to 4–9 % in 2014 (Figure 3.3.13).

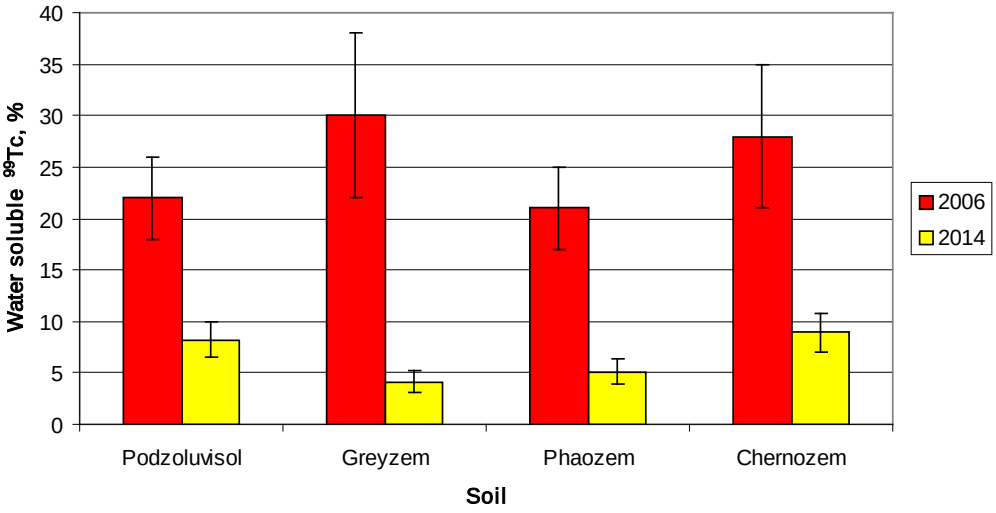

Figure 3.3.13.: Time dependence of water soluble fractions of ^{99}Tc during the experiments

Plant uptake did not differ between soil types during the first two years after Tc-99 administration. In subsequent years (with no Tc-99 administration), plant accumulation was higher for Podzoluvisol and Chernozem compared to Greyzem and Phaozem. Six years after the last administration of Tc-99, concentration ratios for wheat grain ranged six fold from Podzoluvisol to Phaozem (Figure 17). Lettuce leaves and radish roots showed highest uptake, with CR in the range of 140–190 and 100–150, respectively, for freshly contaminated soils. In contrast, concentration ratios for potato tubers were 0.4–2.3, and summer wheat grain 0.8±0.3 over the first 1–2 years. In 2012–2014, the CR values of wheat grain were one order of magnitude lower than for the fresh contamination (Figure 3.3.14).

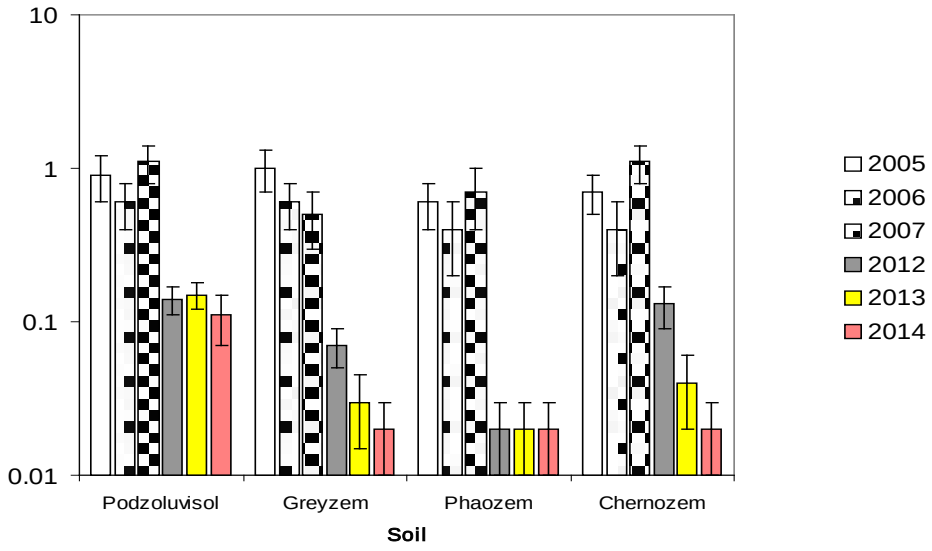

Figure 3.3.14.: Time dependences of ^{99}Tc concentration ratio for wheat grain

More details about the work can be found in Levchuk et al. (2015). Furthermore, a paper has been submitted concerning this work.

3.4 Discussion

The possibility of implementing local foodstuffs in DSS such as JRODOS and ARGOS is fundamentally important in relation to regional customization. This issue is particularly important for regions (of Europe) where important types of agricultural produce are not included in FDMT – such as in Mediterranean areas. For instance, CIEMAT initially considered the inclusion of rice within the present work, but this was not possible due to continuous failure of the system. Similar problems also apply to animal feedstuffs. In e.g. Finland and Norway, grass silage is an important feedstuff for cows, beef cattle and lambs. For the regional customization, STUK tried to include grass silage as a feedstuff in JRODOS, but again, the system failed. Consequently, Finland and Norway had to make a rough conversion to hay-equivalents in the present work since this feedstuff is included in FDMT. Such difficulties of implementing "new" food- and feedstuffs need to be sorted out in the near future.

As described in the previous section, there were some "modelling challenges" in relation to the FDMT task 1 case studies, and consequently we could not perform direct comparison of output from different countries. These findings, however, should be further discussed e.g. in the JRODOS/ARGOS communities (we are after all talking about the two standard European Decision Support Systems).

For the SYMBIOSE runs comparison between countries was possible, and we could also compare task 1 output with results from task 2 (using the dry scenario). The overall finding was, that the uncertainty linked to between-country variability in the "element independent" input data/parameters used in task 1 (see Table 3.3) leads to uncertainty in the results in the same order of magnitude as the uncertainty in the element-dependent radioecological parameters used in task 2 (i.e. interception, weathering, transfer to animal products). The regional customization and the advanced statistics work done in this IRA, is thus two tracks of similar importance to reduce uncertainty in the results.

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. This remaining issue would necessarily have to be considered outside COMET (see also next section).

Based on the updated list of milestones and deliverable (as discussed earlier), the objectives of this work group have been generally met. All Milestones and deliverable reports have been finalised in accordance with the updated plan.

Since the focus of this IRA has been the terrestrial environment, there is an obvious link to the Forest IRA. For instance, many domestic and semi-domesticated animals in Norway graze in forested areas during summer. Beyond that, most of the work in the other WG are relevant for the human food chain (as demonstrated by Figure 3.4.1).

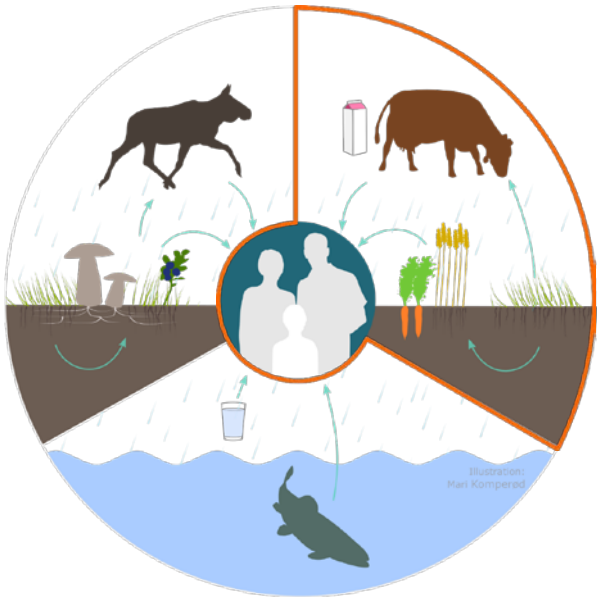

Figure 3.4.1.: Overview of Human food chain transfer (Figure by Mari Komperød, NRPA)

3.5 Impact and further work

Particularly the work on regional customization and Bayesian statistics are highly relevant in relation to emergency preparedness (and other radiation protection platforms). In parallel to the work performed in COMET, the topic of regional customization is also considered within the on-going EURATOM-project "Harmonising Modelling Strategies of European Decision Support Systems for Nuclear Emergencies" (HARMONE) (funded by the Second call of OPERRA). HARMONE is using a slightly different approach, specifying a number of radioecological regions matching biogeographical regions of the European Environmental Agency (OPERRA Deliverable D5.37). Note that, COMET parameter updates from Thørring et al. (2016) have been used within HARMONE. Moreover, the output from our work and HARMONE constitute the basis for further work within the European Radioecology Alliance, as described in task 2 of the Human food chain Roadmap. This work is expected to improve predictions on a regional basis and lead to an enhanced ability to plan and predict the effect of remediation.

Regional updates in soil-plant transfer parameter values should be properly addressed in the future. This also applies to the 'special case' of peat (or organic) soils used as pastures in task 3, 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 Alliance Human Food Chain Roadmap (and the initial work description of D3.1). Other possibilities for R & D 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 Decision Support Systems through probability modelling is also addressed in the Human Food Chain Roadmap (task 4) – including the use of Bayesian statistics.

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NORM modelling

4.1 Background

The radiation risk to humans due to the presence of naturally occurring radionuclides in non-nuclear industries has long been a matter of interest for scientists. In contrast, the radioecology studies associated with NORM have been until now quite limited and the majority of authors rarely elaborate upon the measurement of natural radionuclide activity concentrations in particular compartments within the environment. It is recommended that available data be used for screening purposes and applied in the absence of site-specific information. Such conservative approaches suit regulations, if the presence of natural radionuclides is well below regulatory criteria, i.e. clearance levels or derived dose limits. On the other hand, the application of such conservative approaches often leads to results exceeding these criteria and overregulation in NORM situations. To organise an authority control system that will be both adequate and effective is difficult considering that NORM residues are different in terms of appearance, as they tend to have more in common with industrial waste than radioactive waste, and a great deal of uncertainty associated with exposure assessments should be expected. This uncertainty arises not only from the lack of reference data but also from the inadequacy of traditional modelling concepts when applied to NORM. For these reasons regulatory authorities, industry operators and other stakeholders require more realistic – therefore less uncertain - assessment procedures to be used in dose assessments as well as humans and wildlife assessments in these specific situations.

The main problems with the application of existing approaches, in the case of NORM, occur because of the large number of different NORM sources that can be found. While in 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., and these mostly have worldwide or regional impact, in the case of NORM the number of different sources of natural radionuclides potentially impacting the environment is much more varied. There are hundreds of different NORM generating plants scattered around the world, such as uranium production sites and civil activity like oil and gas production, mining, and road and tunnel construction. Releases to the environment are often observed and the residues generated in each industrial NORM activity has its own peculiarity depending on either their physical or chemical form and the environmental compartment affected. Moreover, regardless of the origin, NORM always contains a suite of natural radionuclides which are subjected to sequential decay resulting in the creation of different isotopes as well as elements varying in chemical and physical properties. Additionally, when carrying out impact or risk assessment one must remember that natural radionuclides are ubiquitous in the environment and only exposure exceeding the natural background is subject to authority control. Additional complexity is caused by the fact that natural radionuclides enclosed in NORM never occur alone, but other contaminants will usually be present, giving a multiple stressor scenario.

NORM legacy sites are important to mention, these sites are where NORM was disposed into landfills without due regard to environmental protection and without the concern about natural radioactivity or environmental issues which exists today. As time went by secondary biological succession, which is not present at all on a fresh NORM landfill, is often observed on legacy sites. This changes the exposure scenario significantly and the risk to wildlife must be considered not only in the surroundings affected by natural radionuclides migration from NORM site but also in the borders of the site.

All these facts imply that the behaviour of naturally occurring radionuclides released from NORM generating industry at their final deposition site or in environments impacted by waste streams can be expected to vary. A promising strategy to develop sufficient understanding of environmental transfers and exposure processes is to identify and then explicitly model key processes that influence the

behaviour of natural radionuclides, in line with the logic of the four Research Lines of Challenge 1 in the radioecology SRA.

Within the proposed NORM IRA, at the very beginning the objective was to contribute to these research lines, by acquiring data necessary for the parameterization of the key processes controlling the behaviour of naturally occurring radionuclides in the environment and by improving existing models or developing parametric models linking observed accumulation, mobility (K_d), and transfer (TF and fluxes) with environmental parameters and processes. Therefore, five different research tasks have been defined. They are listed below:

6. Identification and parameterization of chemical and physical processes influencing natural radionuclide accumulation, mobility and bioavailability after release into the environment;
7. Selection, and physical and chemical characterization of testing sites and samples;
8. Linking radionuclide accumulation, mobility and bio-availability with environmental characteristics;
9. Assessing and modelling transfers to non-human biota;
10. Model validation and improvement.

However, the number of existing models application for NORM situations assessment and/or evaluation is very limited. Existing application are focused on specific natural radionuclides behaviour in environmental compartments (mainly uranium, rarely radium) and there is not direct link with technological processes generating them. Such approach limits significantly their usefulness for NORM generating industry assessment and evaluation. In such situation, significant part of efforts has been devoted for identification of generic technological processes and scenarios, when NORM causes non-negligible risk to environment in order to create well defined circumstances and border conditions for further either implantation (and improvement) of already existing models or development new ones adjusted to specific NORM features. Gathered data were not sufficient for validation of possible application existing models describing radionuclides behaviour in the environment in the way similar to other IRAs and this task remains for further research. That is why, IRA NORM slightly differ in the scope of research performed and final results obtained from other IRA turned into reality in frame of this WP.

4.2 Methods and materials

The following were important to achieving the objective: archival data, field investigations and dedicated lab experiments. Based on available data and experience of NORM WG members basic terms reflecting possible exposure scenarios have been defined and combined with the sequences of technological processes applied in NORM generating industry. This made possible Identification of critical processes common for each kind of NORM, and development of a structured procedure applicable for an environmental impact assessment independently from NORM industry. Finally, a generic description of possible exposure scenarios has been identified creating a warp sufficient for development of specific models focused on natural radionuclides in relation to the source (technological process).

Field investigations were completed on NORM observatory sites. In the frame of STAR NoE, it was decided that some areas were to be used as long-term observatory sites. One of them is in Poland, at areas affected by NORM released from mining industry.

Observatory site characterization: Upper Silesia Coal Basin (USCB) in southern Poland is an area where, as well as many other pollutants resulting from long term activity of heavy industry, enhanced concentrations of natural radioactivity are observed. It is mainly radium isotopes and their decay products which are observed near to coal mines due to radium rich formation water release. Despite the recent significant limitation of coal mining activity, more than 400 MBq per day of both radium

isotopes (^{226}Ra and ^{228}Ra) is still discharged into inland water with mine effluents [Chałupnik et al., 2017]. The range in concentrations of total dissolved solids (TDS) may vary in discharged waters from 2 to 3 g/l up to 120 g/L. The most frequently found ions in these brines, include Cl^- and Na^+. Apart from Cl^- and Na^+, abundant concentrations of Ca^{2+}, Mg^{2+} and SO_4^{2-} can be found in brines together with significant concentrations of Ba, Sr, Pb, Fe, Mn, Ca, Mg, Na, K [Bzowski and Michalik, 2015]. As coal mining activity in this region has been ongoing for longer than 150 years, this result in many "hot spots" contaminated by radium where the observed level of radium isotopes in bottom sediments as well as soils around watercourses reaches level of kBq/kg and air kerma rate 1 metre above ground level is often 100 times greater in average than natural background in this area (up 6 µGy/h) [Michalik, 2008; Michalik et al., 2002].

One of the most interesting objects for radioecological observation are the former natural lakes that had been applied for pisciculture and then adapted for mining industry use. These lakes were used for mine water retention and clarification before release into rivers. It resulted in a layer of bottom sediments containing a high content of radium isotopes and subsequently their decay product. After the mine water discharge was stopped and no remedial activity was performed some of them represent natural lakes with contaminated bottoms, to some extent.

Initial Research Activities (IRA) for NORM, have been developed and research began at the NORM OS in Poland focussing on:

- the contamination of terrestrial environments affected by the discharge of mine water:
- the characterisation of the current state of Rontok lake which is contaminated due to the discharge of radium rich water from coal mines considering:
    - comparison of radionuclide (^{226}Ra and ^{210}Pb) distribution in bottom sediments (1999-2016),
    - assessing the Kd for ^{226}Ra
- ^{226}Ra and ^{210}Pb Transfer Factors to selected biota,

the application of FEPs and IM analysis for the identification of the main processes determining radionuclides' behaviour in a contaminated fresh water lake ecosystem with respect to a conceptual model for risk assessment.

4.3 Results

Identification of generic processes and scenarios when NORM create risk to environment

In NORM generating industries the technology applied determines the properties of residues and the way it is released, therefore identification of key processes influencing the environmental behaviour of radionuclides must start just at a plant. Analysed examples showed that waste streams present in the majority of NORM generating industries are usually kept within the borders of the plant and create additional exposure mainly to workers directly involved in the technological processes. The *sine qua non* condition when NORM can seriously influence the environment is the existence of either massive amounts of waste deposited outside of a plant or effluents in gaseous or liquid form. Therefore, assessment methods that are practical and transferable between different NORM sites can be classified into three generic exposure scenarios accounting for possible interactions and environmental burden, namely:

- the existence of enormous amounts of waste deposited directly into the environment
- release of contaminated water,
- release of contaminated air/dust.

When applying these conditions, the expansive list of already identified NORM creating industries can be easily classified into just a few categories, where even the simultaneous occurrence of generic exposure scenarios is considered. For each category, the characteristic exposure scenario can be defined as can common criteria for environmental impact assessment [Michalik et al., 2017]. These three basic exposure scenarios must be analysed in terms of secondary processes determined by the interaction of releases with the receiving environment, however it can be done in a uniform manner with no regard to industrial process. To define appropriate parameters for safety assessment studies, an approach based on features, events and processes (FEPs) is widely used, however, the behaviour of a radionuclide in the environment is determined mainly by its chemical properties and the properties of the receiving compartment of the environment. After release, the effects of dilution or accumulation of natural radionuclides may occur. Fumes and liquid effluents are usually diluted and the derived activity concentration of natural radionuclides in the receiving compartment of environment is lower than in the released NORM. This is sometimes used as a final solution for NORM and is applied in industries where other treatments of residues is still not justified from an economical and/or technical point of view, e.g. the oil and gas industry, especially in off-shore platforms. The opposite situation is created in the case of solid residues when disposal can lead to radionuclide accumulation in a landfill site. Landfills are often regarded as the final solution for NORM waste, therefore the proper construction of a landfill site can significantly limit the burden placed on the surrounding environment. Currently, the majority of industries that generate NORM/TENORM finish their activity with a focus on the safe disposal of their residues at this stage.

With no regard to the type of NORM and the form of the released material, subsequent FEPs taking place after primary dilution (or accumulation) should be considered in prospective assessments in order to provide evidence that the environment is protected in a proper way [Michalik and Mora, 2017]. Keeping in mind that NORM usually contains a suite of natural radionuclides which are different elements and subject to sequence decay, the complexity of a possible exposure situation may increase significantly. Moreover, interaction with biota and human activity not related to previous industrial activity creates many additional FEPs that influence radionuclide migration, which are frequently element specific and determined by existing external circumstances, in an abiotic environment. However, given that the environmental effect at the abiotic environment level is usually assessed based on the observed final activity concentration of a radionuclide, all FEPs after release should be considered from a dilution–accumulation balance point of view (Fig. 4.3.1)

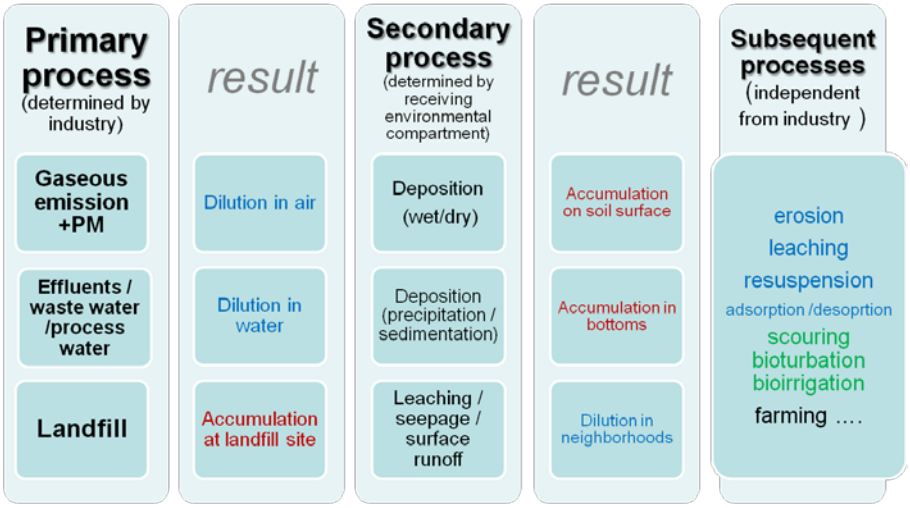

Figure 4.3.1.: Possible sequence of processes ruling radionuclide behaviour after release into the environment (subsequent processes are named FEPs in the text).

The altered distribution of radionuclide concentration in environmental media influences direct exposure scenarios and may be used to measure environmental effects, especially when clearance levels set in relevant regulation have not been exceeded. However, migration into living organisms as a primary source of internal exposure is usually important, when taking into consideration the fact that natural radionuclides are frequently alpha emitters. A living organism acquires matter and energy from the external environment and converts it into different parts of their bodies. That is why identification of the processes responsible for the transfer of radionuclides from biotope to biocenosis is crucial for proper exposure assessment. From the first point of view ingestion, inhalation and interception are the generic processes leading to the accumulation of radionuclides in biota. However, when looking into processes occurring in an ecosystem (no matter what its scale is) it is easy to notice that actual migration from an abiotic environment into biota is just taking place at the level of the primary production controlled by photosynthesis. This fact is important not only for an organism (plant) that is subjected to this process but also for all other ones that are at a higher level in the trophic chain. In contrast to the accidental direct ingestion of abiotic matter, ingestion of organic matter with already incorporated radionuclides is the main secondary process leading to radionuclide accumulation in biota, including humans. When the radionuclide activity concentration of organisms occupying a trophic level are known, their further migration is quite easy to evaluate, based on limited parameters, as the dietary pattern, which is usually easily recognized at each level of an ecosystem.

For the description of soil-to-plant transfers in assessment models, a transfer or concentration factor (TF) is often used. However, the absorption of elements into a plant is not a simple process and a quantity should be expressed by a multivariable function based on mechanistic knowledge instead of a simple factor. In general, the process responsible for the migration from soil to plant consists of two sub-processes that depend on different conditions/parameters and are ruled by different phenomena. The first one is migration from a solid phase to a liquid phase in soils and the second one, is the absorption of the liquid phase of soils to plant roots. What is happening inside a plant after this, is another matter completely which is ruled by the plant's metabolism and the radionuclides' distribution among different plant tissues is not as important when the radiation dose is calculated (considering that there is no additional concentration of the concerned radionuclides in the edible parts). When a simple transfer process is divided into many sub-processes, an additional advantage is created that allows the presence of radioactive elements as a source of synergy as well as antagonism (e.g. ion competition effects) to be included in the effects analysis. However, this significantly multiplies the number of FEPs needed to be identified (Fig. 4.3.2).

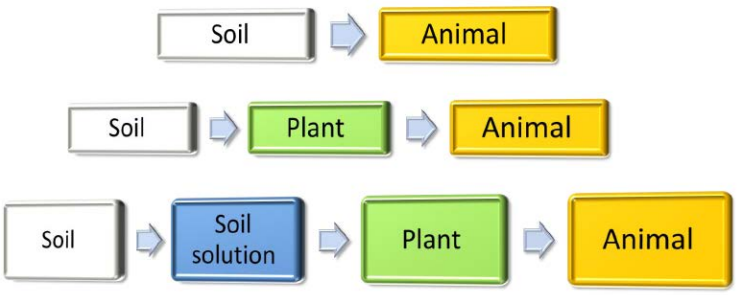

Figure 4.3.2.: Roughly multiplied sequences of processes ruling radionuclide transfer from soil to biota

Within the proposed NORM IRA, the objective was to contribute to Challenge 1 of the SRA by obtaining data concerning the behaviour of natural radionuclides released from the NORM generating industry. The unified approach, described above, has been applied and the data necessary for the parameterization of the key processes controlling the behaviour of naturally occurring radionuclides in the environment was acquired. Field experiments focused on the areas affected by the release of

radium rich water into terrestrial and aquatic environments. This, in turn, reflects the aforementioned generic scenario 2.

Terrestrial environment

Contamination of the terrestrial environment caused by mining activity in Poland is observed along watercourses which were receiving mine water. Soil on the banks was contaminated due to either a local flood or the relocation of bottom sediments in order to keep a watercourse patent. For the preliminary test an area along the "Rów Chwałowicki" stream in Świerklany county was chosen (fig. 4.3), where enhanced dose rates were previously measured in the range 0.2 - 5 µSv/h.

Figure 4.3.3.: Dose rate measurement and soil sampling points

After the COMET experts' field visit in 2015 it was decided not to use this area for longer observation because the contaminated area is too small (the length is less than 500 m and contamination is observed only very close to the stream bed). However, the observed contamination of soil was so high that 4 soil bulk samples (about 15 kg each) were collected to prepare substrates for further laboratory tests. Samples were collected at points where the dose rate varied in more or less the whole range of observed at this area values (Fig. 4.3.3). The fifth sample of soil was collected later near to Rontok lake, where the activity concentration of radium was expected to be greatest.

Soil samples were characterised in terms of chemical, mineralogical composition and radionuclide concentrations; then they were prepared as a substrate for further planned experiments (leaching and TF). The maximum ^{226}Ra activity concentration measured reached 13.2 kBq/kg (dry mass).

In spite of field experiment no longer being planned in the frame of the IRA NORM WG, FEPs analysis was carried out to identify main process responsible for soil contamination. When considering the source of observed contamination, two processes were suspected to be responsible for the contamination, i.e. either sorption in soil flooded by mine water or bottom sediments graded on the watercourses' banks. Despite the small number of samples available, radium correlations were checked alongside other parameters that were measured to identify the source of contamination. Pearson's correlation coefficient was calculated according to the formula:

$$r_{XY} = \frac{cov(X,Y)}{\sigma_X \sigma_Y}$$

EQUATION 4.1

High correlation factors between radium and barium, barite and strontium content were noticed. This proves that much of radium present in the soil resulted from the process of the co-precipitation of radium with barium (or strontium) as radium-barium-strontium sulphate. As bottom sediments can be enriched with radium isotopes due to this process, it means that the soil contamination observed was caused by bottom sediments graded on the stream banks. The lack of radium correlation with amorphic substance and the content of organic matter (loss on ignition) additionally proves that sorption on soil (organic matter), in this case, did not play a significant role in radium accumulation.

Aquatic ecosystem

Contamination of the aquatic environment is mainly observed in former settling ponds which were used for mine water retention before release into rivers. The lake chosen for investigation as an observatory site was well characterised in 1999 just after the discharge of mine water into it was stopped. Archived data can be compared with current data and, helping with FEPs, the identification of the main processes determining radionuclides' behaviour in contaminated a fresh water lake ecosystem is possible.

Since 1977 the pond has been used for the sedimentation of suspended solids from mine waters. Approximately 70 million cubic metres of saline water has been discharged into the pond during the last 22 years. The content of the suspended solids varied from 0.3 up to 2.4 g/dm^3. The area of the pond is quite large – about 36 ha (Photo 4.3.1).

Radium-bearing waters were pumped into reservoir – along with significant amounts of barium and traces of sulphate ions [Chałupnik et al., 2001]. For example, in 1998 about 5600 m^3/day of such water was released into the pond. The co-precipitation of radium and barium took place spontaneously and as a result insoluble deposits of $BaSO_4$ + $RaSO_4$ settled at the bottom of the pond. Such sediments were characterised by highly enhanced levels of radium isotope concentrations. 28 samples of bottom sediment were collected in a regular grid covering all the available surface of the lake. Table 4.3.1 summarises the results of the measurements, performed by gamma spectrometry, of radium activity in bottom sediments and in the surrounding area. In the report concerning the measurement of gamma dose rates, values were quoted of up to 42 µGy/h near the point of inflow of waters into the settling pond (phot. 4.1). With the increase of the distance from the discharge point a decrease in the dose rate was observed – down to 2 µGy/h at 15 metres from the end of the pipeline. In the remaining part of the area, lower values of gamma dose rates were measured that did not exceed the level of 1.2 µGy/h.

Photo 4.3.1: Former mine water discharge point into Rontok lake

Table 4.3.1.: Radionuclide activity concentration in the Rontok lake area – 1999, 28 sampling points

radionuclides (typical range for soil)	Activity concentration [Bq/kg]		
	Bottom sediments (28 samples)	Soil from banks	Soil around the banks
^{226}Ra (7–50)	67–49 151	30–3508	29–42
^{228}Ra (10–50)	62–6388	40–2482	33–47
^{224}Ra (10–50)	65–8990	38–1791	30–44
^{137}Cs	< 3–1014	< 3–79	77–238
^{40}K (100–700)	155–848	414–664	377–560

In the framework of COMET, sediment samples were collected in the same grid, if possible. Samples were collected using a core sampler to assess the vertical distribution of radionuclides. The comparison of the distribution of radium isotopes in 1999 and 2015/16 in the top layer of sediments is presented in Figure 4.3.4.

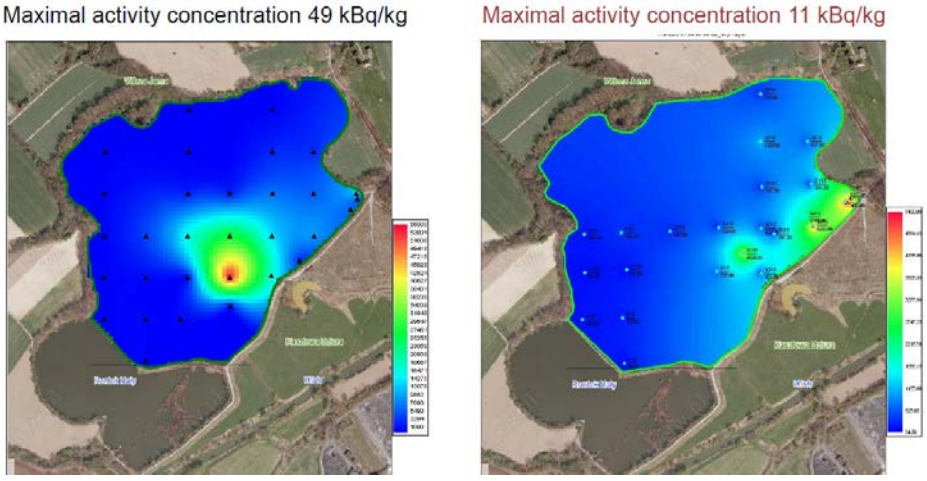

Figure 4.3.4.: Comparison of radium distribution in bottom sediments, left -1999, right 2015/16

In the majority of the sampling points, the current activity concentration of ^{226}Ra is significantly lower than in 1999. Rontok is a flow-through lake fed by small local watercourses and the most probable scenario leading to the observed situation can be explained based on different processes of turbation, including bioturbation as many species of aquatic biota are currently present there. This hypothesis is also supported by the fact that the highest radium activity concentration is now observed in places where the thickness of sediments layer measured in 1999 was greatest.

However, slightly different information is provided by current measurements of the activity concentration of ^{210}Pb. The distribution of this radionuclide corresponds to the distribution of ^{226}Ra.

Moreover, the average ratio of ^{210}Pb to ^{226}Ra in the top layer of sediments is about 0.72, quite uniform, with a standard deviation of 0.15. This reflects the expected value of ^{210}Pb activity concentration created due to the radioactive decay of radium, if the assumption is made that at the beginning only radium isotopes were present in sediments. In the layers of sediments below, the ^{210}Pb/^{226}Ra ratio is slowly decreasing reaching a value close to 1, when radium and lead activity concentrations are close to natural levels and the observed excess of radium is negligible. Therefore, no significant migration of lead is observed, be it spatial or along the sediment profile. This is contrary to previous conclusion that turbation processes are responsible for significant deficiency of radium isotopes in sediments. The only explanation is that radium deficiency has resulted from a sudden increase in water flow through the pond caused by, for example, heavy rainfall which re-suspended the top layer of tiny sediments and moved them away.

The low mobility of ^{210}Pb was also confirmed by measurements of activity concentration of this radionuclide in water. At three points in Rontok lake, water was sampled from the top layer and close to the bottom. Before being measured, water was filtered to remove the suspended matter and then chemically prepared to measure the activity concentration of the natural radionuclide. Liquid scintillation spectrometry and alpha spectrometry were applied to measure ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po. At the same points bottom sediments were sampled and activity concentration of ^{226}Ra, ^{228}Ra, ^{228}Th and ^{210}Pb was measured by gamma spectrometry.

Results showed that there is no significant difference in radioactivity content in particular layers of the water and no correlation is observed between radionuclides in water and sediments below. The depth of the Rontok ranges from 0.4 m to 2.0 m. This allowed it to be classifies it as a polymictic lake, where thermocline is not present and water is mixed. Moreover, as Rontok is also a flow-through lake, there is no visible spatial correlation between radioactivity content at the bottom and in the water. Both facts explain results obtained.

Among the radionuclides measured in water the activity concentration of ^{210}Pb is only slightly enhanced when compared with the average activity concentration observed in fresh water. Unfortunately, there are no results of direct polonium measurement in sediments. However, taking into consideration that sediments are at least several years old the expected activity concentration of polonium is equal to ^{210}Pb activity concentration. After sequential decay ^{226}Ra….^{210}Pb ->^{210}Po nuclides of polonium are created as free ions and their expected mobility is high. However, different concentrations of lead and polonium in water must be explained, considering additional deposition from air.

To verify experimental results about the activity concentration of radium in water, a laboratory experiment was carried out which focused on K_d assessment. Three samples of sediment (collected in Rontok lake) were chosen to be used in this test.

K_d values were calculated using the equation [IAEA, 2010] presented below:

$$K_d = C_s/C_l$$

EQUATION 4.2

where: K_d – partition coefficient [L/kg]
C_s – activity concentration in the solid phase [Bq/kg]
C_l – activity concentration in the liquid phase [Bq/L]

Uncertainty was calculated using:

$$\Delta K_d = \Delta C_s/C_s + \Delta C_l/C_l$$

EQUATION 4.3

the roughly calculated relationship between K_d value and radium activity concentration in the solid phase is presented in figure 4.3.5.

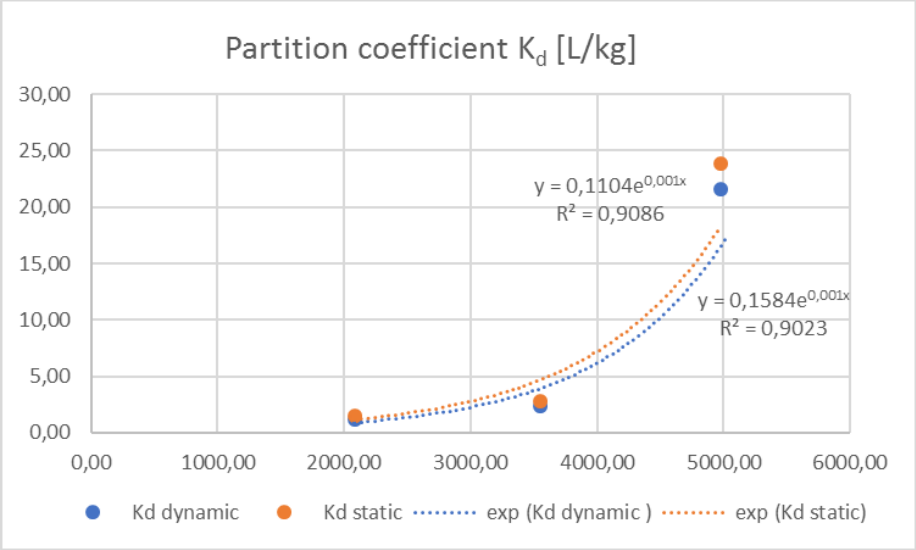

Figure 4.3.5.: Partition coefficient for ^{226}Ra

The final thing to be measured was the transfer of radionuclides from the abiotic environment to biota. As defined before, the main process of transfer to biota is only occurring at the level of primary production. In the case of Rontok lake, the most abundant plant observed is common reed (*Phragmites australis*). In fact, in Europe it is an invasive species however it is now a dominating species surrounding fresh water lakes. Four samples of green parts from above the water were collected and one rhizome was taken at the same points as the sediment samples. This allowed the transfer factor from sediments to common reed tissue to be calculated. The results for ^{226}Ra presented in Fig. 4.3.6 were calculated for the dry mass of both sediments and plant.

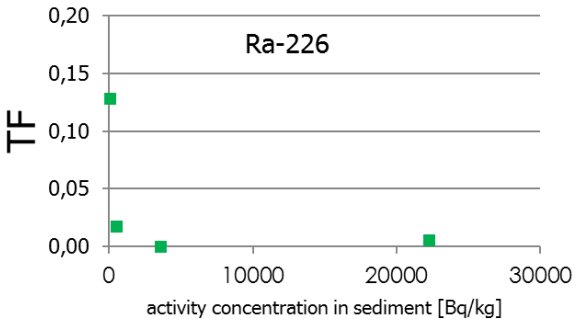

Figure 4.3.6.: TF for ^{226}Ra

There is no positive correlation of TF values with radionuclide activity concentration in sediments. The very similar level of radium activity concentration in green parts, which does not vary in relation to radium activity concentration, suggests that the intake of radium is limited by the plants' metabolisms. On the other hand, bigger TF values in the case of plants growing on sediments with a low content of radium (close to the natural level observed in soil) suggest that radium originating from mine water occurs in a different form less bio-available than the radium present in sediments due to natural processes. This is also supported by the radium's low mobility and the K_d coefficient calculated earlier.

In the case of lead, the situation is very similar, however there is no reason to have different ^{210}Pb speciation in sediments. ^{210}Pb created as a decay product of radium and ^{210}Pb deposited from the air should have very similar chemical forms. The lack of relationship observed between the lead concentration in sediments and plants is, probably, caused by the fact that the overwhelming majority of lead in plants is provided by the interception process.

4.4 Discussion

Despite the high content of radium isotopes in mine water, direct radiation risk, in the majority of analysed cases, is negligible. In comparison with the radium content in the common soil, which is reported to be 25 Bq/kg, the activity concentration commonly observed in mine waters is very similar. The highest activity concentration observed in mine water from Polish coal mines, i.e. about 400 Bq/dm^{3,} is still below the clearance level set for solids in the recent European BSS. That is why the subsequent processes finally leading to radionuclides accumulation are most important. Besides possible accumulation by aquatic biota (if present), the most efficient processes leading to radionuclide concentration are precipitation and sorption. Experimental data, obtained during IRA, as well as other data collected during many pieces of research carried out in GIG prove that the overwhelming amount of radium observed in environments, affected by mining activity, occurs in the form of sulphates [Bzowski and Michalik, 2015]. This means that the precipitation process is mainly responsible for radium accumulation.

However, considering that the solubility product of pure radium sulphate is equal to 4.2×10^{-11} other conditions must be met to get sediments with a high content of radium under common conditions that occur in the environment. In practice, the concentration of ions Ra^{2+} and SO_4^{2-} is always lower than its solubility product in the range of temperatures occurring in environments that receive water released from a mine. One Bq/dm^3 of ^{226}Ra expressed as molar concentration equals 1.2×10^{-13} mol/dm^3, therefore the highest radium molar concentration observed in mine water is about 4.66×10^{-11} mol/dm^3 (390 Bq/dm^3). Despite such a high content of radium, the amount of sulphates necessary to start radium sulphate precipitation reflects concentration about 1 mol/dm^3, this equals 100 g/dm^3. Such a high content of sulphates does not occur in the environment often. Assuming that the possible concentration of sulphates in water is 3 g/dm^3, which is often observed, the necessary radium activity concentration needed to start radium sulphate precipitation is about 12 000 Bq/dm^3, which is also not

so common. Therefore, precipitation radium dissolved in mine water as radium sulphate because of natural processes is almost impossible (Fig.4.4.1).

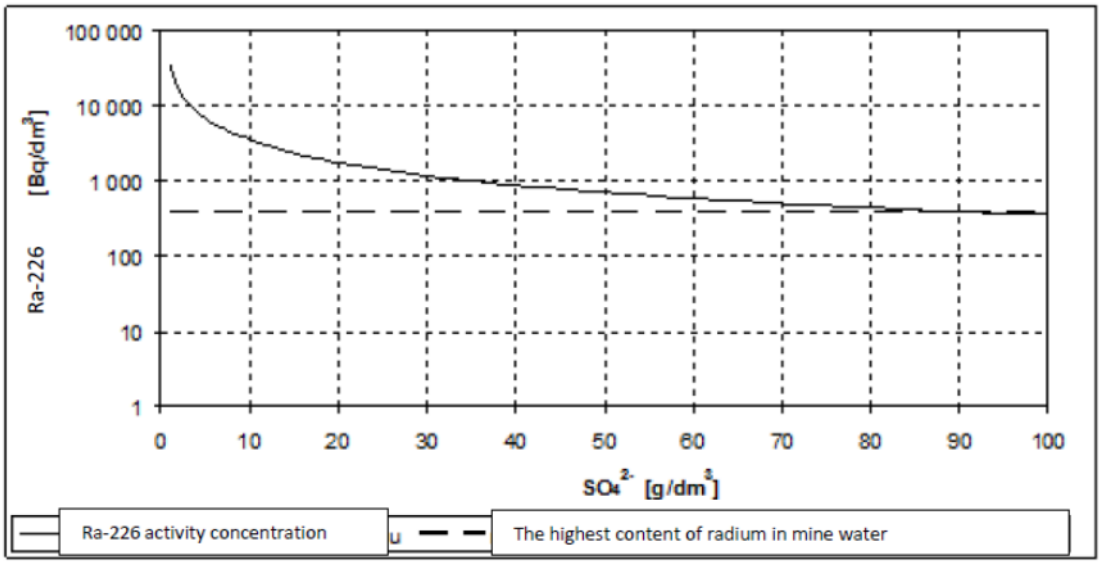

Figure 4.4.1.: Concentration of radium and sulphates necessary to start radium sulphate precipitation

On the other hand, barium sulphate which has a solubility product equal to 1.0×10^{-10} will start precipitation when barium ion concentration is about 0.5 mg/dm^3. Considering that barium has high chemical affinity with radium the process of co-precipitation, based on Fajans–Paneth–Hahn law, often occurs and forms a barium-radium sulphate:

$$Ba^{2+} + Ra^{2+} + 2SO_4^{2-} \rightarrow BaRa\,(SO_4)_2$$

This phenomenon is well known and commonly applied in radiochemistry for radium extraction from solutions. From an environmental point of view the occurrence of barium in water, which is quite common and often reaches concentrations at the levels of grams per litre, is crucial. In formation waters especially, the main conditions influencing radium behaviour after mine water discharge are associated with fossil fuel barium presence.

As results show, the observed contamination of the terrestrial as well as the aquatic environment is caused by the presence of barium-radium sulphate. This fact influence subsequent processes can be observed in the environment. Due to the very low solubility of this sulphate, radium mobility and bio-availability is very limited. That is, why migration of this radionuclide in the environment is mainly determined by processes of erosion or mechanical turbation and transfer to biota is very limited. Results obtained for Rontok lake are in line with conclusions from other pieces of research, focused on the properties of radium rich sediments from the mining industry [Michalik et al., 2009; Bzowski and Michalik, 2015; Michalik 2008; Leopold et al., 2007].

However, based on radium-barium sulphate properties only the behaviour of radium can be explained. Long-lived decay products as ^{210}Pb and ^{210}Pb (to some extend) are not bonded to sulphate crystal lattice and should be more mobile. In the case of lead experimental data from Rontok lake, these expectations were not confirmed and additionally highlighted the problem about how to distinguish the lead originating from radium in sediments from lead that resulted from the permanent deposition of this radionuclide which originates from the decay of short-lived, fugitive radon gas that was released from the soil. The slightly enhanced activity concentration of polonium in the Rontok lake water confirms

the expectations concerning this nuclide's mobility and its final transfer to biota should be analysed. This is important due to the high radiotoxicity of ^{210}Po.

The current situation proves that ^{228}Ra and its decay products do not contribute to the exposure significantly. Considering that ^{228}Ra's half-life is about six years and the fact that in mine water and then, subsequently, in emerging sediments there is no ^{232}Th at all (or merely a few Bq/kg) in the long term this radionuclide will decay to a negligible level. In the case investigated where the activity concentration was observed after 16 years (about 3 ^{228}Ra half-lives), the maximal activity concentration of this radionuclide is about ten times lower, which corresponds to the radioactive decay law. However, from short term perspective exposure to ^{228}Ra and its progeny is not negligible.

The remaining question which is unanswerable, based on the Rontok lake case, is what would happen to radium when barium is not present? Available data proves that sorption in clay minerals does not play an important role [Bzowski and Michalik, 2015]. However, sorption on organic matter in soil (or sediments) cannot be excluded. To check this possibility, a laboratory experiment has been carried out. The sorption of radium isotopes from coal mining wastewaters in the soil was investigated. The mining wastewater, which is being discharged to the natural environment, currently containing high activity concentration of radium isotopes ^{226}Ra and ^{228}Ra as well as other cations and anions, was used for this purpose. The sorption of radium isotopes was observed in organic garden soil. The determined sorption yield is about 43%. It is quite a high value and, considering that the radium in this case is assumed to be not so strongly bonded to the matrix as in the case of barium sulphate, this process can significantly affect further radium migration when contaminated soil is exposed to the environmental conditions. However, in the case of the mining industry, the direct contact of water with soil is very limited due to the applied system of mine water discharge. This again proves that analysing the potential environmental effects on the analysis of the particular technology applied in NORM generating industries and way of effluents discharge must be considered first.

4.5 Impact and further work

Defining basic terms reflecting possible exposure scenarios and combine them with the sequences of technological processes applied in NORM generating industry IRA-NORM activities let one identify of critical processes for each kind of NORM and then develop of a structured procedure applicable for an environmental impact assessment independently from NORM generating industry. This laid the grounds of a strategy to better understand the key processes that result in the release and that influence the environmental behaviour of naturally occurring radionuclides [Michalik et al., 2017; Michalik and Mora Canadas, 2017].

Unfortunately, terrestrial observatory site available in Polish observatory site did not provide appropriate conditions to verify the assumed approach. To cover this gap, an activity in the new observatory site in Belgium, which by far better reflects landfill exposure scenario, has been launched.

On the other hand, examples of aquatic environment and related archive and new collected data create a good opportunity to characterise NORM situations caused by discharge of radium rich effluents into inland water. From the many processes identified applying FEPs for a generic lake ecosystem, in light of the peculiarities of the contamination caused by the mining industry, only a few were identified as important from an environmental point of view. Apart from the process of radium and barium co-precipitation which is responsible for the primary accumulation of radium, other processes occurring in abiotic matter that should be considered when an environmental impact assessment is carried out towards an aquatic ecosystem, are:

- resuspension (bottom sediments to suspended sediments) diagenesis, scouring, bioturbation, bio-irrigation,
- runoff, wind and water erosion (dried sediments, bank sediments to suspended sediments),

- deposition of ^{210}Pb created in the ground layer of the atmosphere due to the decay of radon exhalation from the ground

The sequential decay of radium is the source of exposure changes. The data obtained suggests that, considering the very low radium and lead mobility and bio-availability, further migration into the trophic chain is most important in the case of ^{210}Po. However, the interception of ^{210}Pb (and probably ^{210}Po) created in the ground layer of the atmosphere due to the decay of radon which is released from the ground is in fact not related in any way to the analysed situation. Yet, it is source of the problems that will be encountered when the proper assessment of the TF of these radionuclides must be performed.

The efficiency of the radium accumulation process can be assessed based on the chemical characteristic of the environmental compartment affected and co-precipitation rules. K_d and TF for radium can be parameterised by the evaluation of the mobile part of radium based on, for example, the Tessier test [Leopold et al., 2007].

Priorities for further R&D

In view of NORM roadmap, it is shown that all IRA already done or going on cover only a tip of an iceberg of needs and expectations. In detail, concerning identified generic exposure scenarios, the most important gaps have pointed out as:

- insufficient knowledge on key processes in NORM landfills causing that too cautious/conservative approaches are commonly used for risk assessments in these cases and,
- understanding and modelling performance of transport and immobilisation processes of NORM radionuclides in surface and ground waters, especially in the unsaturated zone are still far from being satisfactory, and thus should be improved.

The main future challenges identified are related to the facts that:

- diversity in the characteristics of contaminated NORM sites is crucial. In spite of rudiments of a uniform frame of evaluation expected by regulatory authorities were prepared, every NORM case shows differences in radionuclides transfer due to different chemical, physical and physical-chemical properties; therefore there is a need to look into different parameters influencing transfer of NORM;
- ecosystem transfer of NORM is still not clearly understood and studies on bioavailability, migration from biotope to biocenosis and possible effects of NORM on wildlife considered together with other stressors (multiple stressor approach) are lacking.

The lack of site and industry (technological process) specific data indicating the need to continue research at multiple NORM sites for more abundant and accurate situation-specific data. With this, the predictions on the potential health risk for humans and wildlife originating from NORM sites can be made much more precise, and in addition, the uncertainty of such calculations can be significantly reduced. Moreover, besides strict research activities, the acceleration of the transfer and application of newly generated knowledge on the environmental behaviour of NORM radionuclides - generated in the laboratory or in the observatory sites - into management and re-use options of NORM residues is strongly expected. Last, a bridge between experimentalists and modellers must be made simultaneously by the both parties to improve both sides of the same thing.

In specific case on coal mining industry further investigation should be focused on specific conditions leading to the possible mobilisation of radium bonded to sulphates, sorption of radium (and decay products) in soil, and subsequent remobilisation and migration of polonium into biota.

Relevance for emergency preparedness community or other RP platforms

Release of radium with either effluents or any kind of radium rich leakage and all subsequent processes - sorption on soil and further remobilisation under different environmental conditions - remains crucial for many NORM cases. However, radium rich leakages are significant also from long term perspective in case nuclear spent fuel repositories. Radium in this case is important radionuclide when consider that fissile products included in spent nuclear fuel will have decayed in few hundred years.

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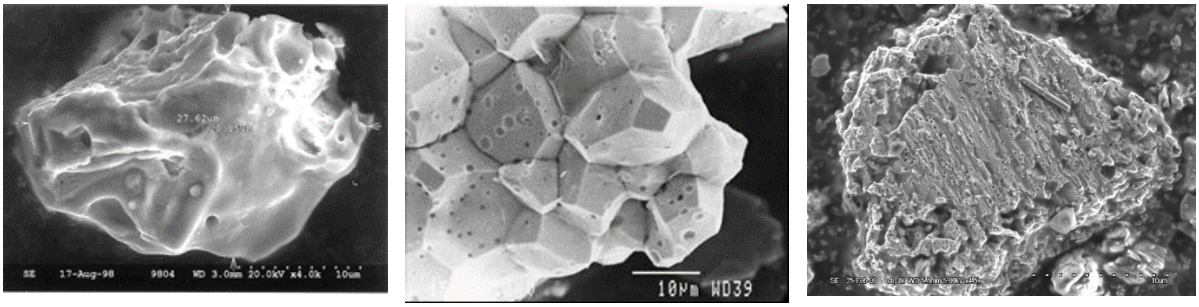
Particle behaviour

5.1 Initial Research Activity

5.1.1 Background

Fuel particles (FP) within the radioactive fallout were previously considered to be a specific "peculiarity" of the Chernobyl Nuclear Power Plant (ChNPP) accident (Sandalls et al., 1993), although radioactive particles have been released from all severe nuclear events (IAEA, 2011). About 3 tons of spent U fuel was released to the environment during the Chernobyl accident. Contamination of the near Chernobyl exclusion zone (ChEZ) was mainly composed of radionuclides associated with particles of irradiated nuclear fuel, ranging from submicrons to fragments (Kuriny et al., 1993; Salbu et al., 1994, 1999, 2000 and 2001; Kashparov et al., 2003, Kashparov 2003). Certain radionuclides such as ^{95}Zr, ^{95}Nb, ^{99}Mo, ^{141,144}Ce, ^{154,155}Eu, ^{237,239}Np, $^{238\text{-}242}$Pu, ^{241,243}Am, ^{242,244}Cm were released from the accidental unit only as fuel particles. More than 90 % of ^{89,90}Sr, ^{103,106}Ru and ^{140}Ba activity also were released as fuel particles. All these radionuclides were deposited on the ground as fuel particles varying with respect to physical and chemical properties (size, composition, crystalline structures, oxidation state) and were initially characterized by very low mobility in the environment. During the initial explosion (high temperature, high pressure, no oxygen), U particles containing construction materials (Zr, C) released to the West appeared very inert, while U particles released to the North, East and South during the subsequent fire (high temperature, normal pressure, presence of air) were oxidized and more subjected to weathering. After deposition, weathering of particles occurred and associated radionuclides were mobilized with time. Fuel particles (separate grains, crystallites of uranium oxide) with a median diameter of about 4-6 μm and their aggregates can be divided into 3 groups (Fig.5.1.1.1) according to their composition, U oxidation states and subsequent dissolution rates under natural conditions (Kashparov et al., 1996, 1999, 2004).



a b c

Figure 5.1.1.1.: SEM images of fuel particles: (a) ZrU_yO_x fuel particles; (b) UO_2 fuel particles; (c) UO_{2+x} fuel particles (Zhurba et al., 2009)

From the radiological point of view (influence of external irradiation to humans and animals, inhalation of internal irradiation, and per oral intake of radionuclides into organisms), the physical and chemical characteristics of fuel particle associated radionuclides are mostly expressed in terms of:

- the presence of radioactive contamination of the territory, including the physico-chemical form;
- the inhalation intake of radionuclides during the passage of radioactive clouds and the resuspension by wind, radionuclide uptake, metabolism in organisms and the associated doses;
- the migration of radionuclides in soil that determine the dynamics of the external irradiation dose rate and the contamination of surface and ground waters, as well as the changes in the biological availability of radionuclides. This in time determines the levels of contamination in plants and agricultural products and the intake of radionuclides into human organisms;

- the per oral intake of radionuclides in agricultural animals and wild-life, metabolism and associated doses
- per oral intake in man due to radioactive contamination of agricultural and forest products.

After deposition, weathering of particles and radionuclide remobilization depends on particle characteristics and environmental conditions. Analysis of the physical - chemical properties affecting fuel particle dissolution has shown that the rate of dissolution for particles of the same group can be correlated, to the largest extent, with the oxidation degree of UO_2 fuel and the acidity (pH) of the water in the environment as seen in Fig 5.1.1.2 (Kashparov et al., 1999, 2004). Particle weathering can be followed by the observed increase of ^{90}Sr exchangeable fraction (%) in soils and by the subsequent uptake in vegetation (Oughton et al., 1993, Kashparov et al., 1999, 2013). Autoradiography and radiochemical analyses of samples have shown that about 90-99 % of the ^{90}Sr activity in the bottom sediments of the ChNPP cooling pond (pH about 7) is still (2015) associated with fuel particles. During the post-accident period, less than half of the total activity of radionuclides has been leached from the FP matrix into soil from numerous radioactive waste storage sites within ChEZ (Kashparov et al., 2012).

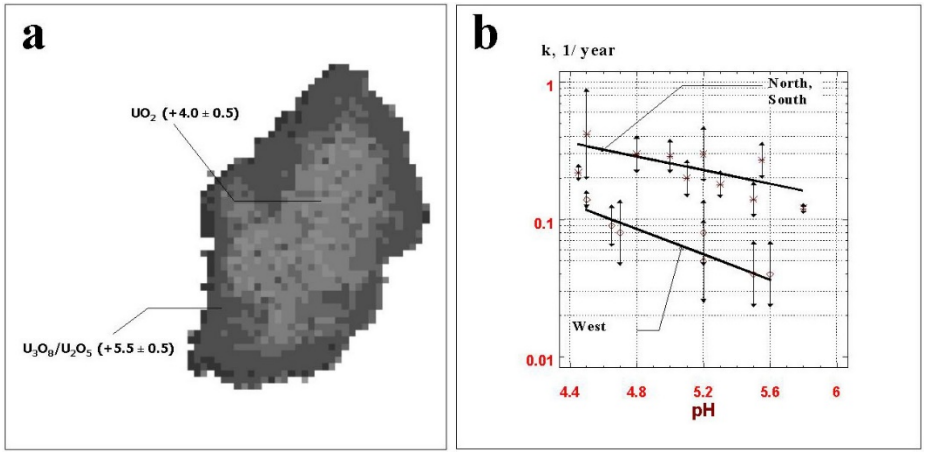


Figure 5.1.1.2.: Particles released from the Chernobyl reactor and associated weathering rates. (a) Oxidised fuel particle (UO_2 cores with oxidised U_3O_8 and U_2O_5 layers) released during the reactor fire obtained from 2 D micro-XANES (Salbu., 2001, Salbu et al., 1999, 2000 and 2001), (b) Weathering rate constants as functions of pH for fuel particles released during the explosion (lower) and during the fire (upper) (Kashparov et al., 1999)

The parameters of the dynamic models of the dissolution of fuel particles in soils and sediments and at drained areas describe the mobility and potential bioavailability of radionuclides. Refinement of these parameters by systematization of existing data and by obtaining novel data on the long-term environmental behaviour of Chernobyl fuel radioactive particles (solubility in the soil and transfer of radionuclides to plants) is essential for assessing environmental impact as well as radiation protection of humans and the environment.

Additional work has been performed related to NORM particles. The Pridneprovsky Chemical Plant (PChP) was one of the first Soviet enterprise for processing uranium ores, built in 1947 in the suburb of Dniprodzerjisk in Ukraine. Up to 65 % of all uranium ore of the Soviet Union was processed at the PChP. During the remediation of the PChP, the impact and potential risk associated with release of high alpha-activity NORM particles should be taken into account.

Four tasks were specified for the work within the IRA on particle behaviour:

- Task 1: Chernobyl Fuel Particle behaviour in soil;
- Task 2: Chernobyl Fuel Particle behaviour in the radioactive waste trench in the CEZ;

- Task 3: Chernobyl Fuel Particle behaviour in the drained bottom sediments.

These tasks included the characterization of FP matrix and a set of parameters for estimating the dissolution of fuel particles in soils (NUBiP of Ukraine, NMBU), in the radioactive waste trench (NUBiP of Ukraine, NMBU, IRSN), in the drained bottom sediments (NUBiP of Ukraine, NMBU).

- Task 4: Radioactive particles containing NORM. This task includes characterization of fuel particle properties (NUBiP of Ukraine, NMBU).

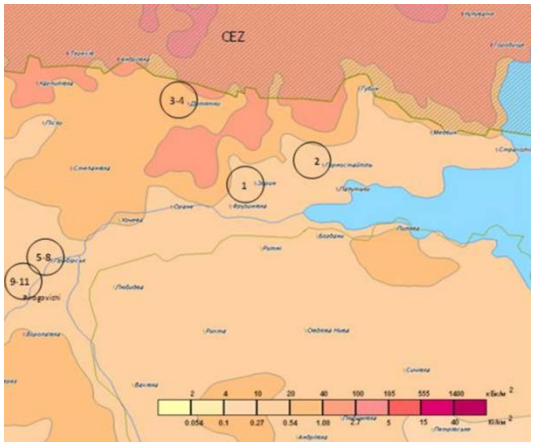
5.1.2 Methods and materials

Soil and grain sampling has been performed at agricultural areas near the ChEZ in Aug 2013 and Aug 2014 (Fig. 5.1.3). Additional sampling took place May 2016.

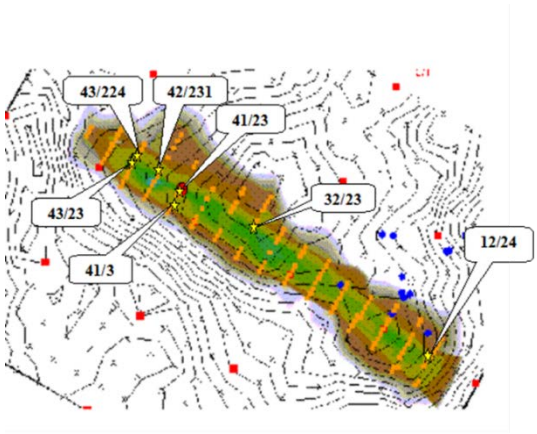
Soil sampling in the trench of the temporary storages of radioactive waste was carried out in 2014-2015 at the depths up to 3 m (Fig. 5.1.3). Taking into account the specific heterogeneity and spatial distribution of the radionuclides in the trench, the samples were collected in 5 points along the trench axis near the sampling points used in 2000-2001 (Kashparov et al., 2012).

On the drained part of the ChNPP cooling pond, 6 research sites were chosen (Fig. 5.1.3). In these point samples of the dried sediments from different soil layers at the depths of 0-25 cm and samples of the first vegetation were collected. In addition, an experimental field plot for the simulation of drainage of the sediment and their exposure in natural conditions has been created.

Samples of NORM sludge at various depths in three tailing sites (Zahidne, Centralnyy Yar and Dniprovske) at the Pridneprovsky Chemical Plant (Dneprodzerginsk, Ukraine) were collected during the “dry” auger drilling of the boreholes (Fig. 5.1.2.1).



a)



b)

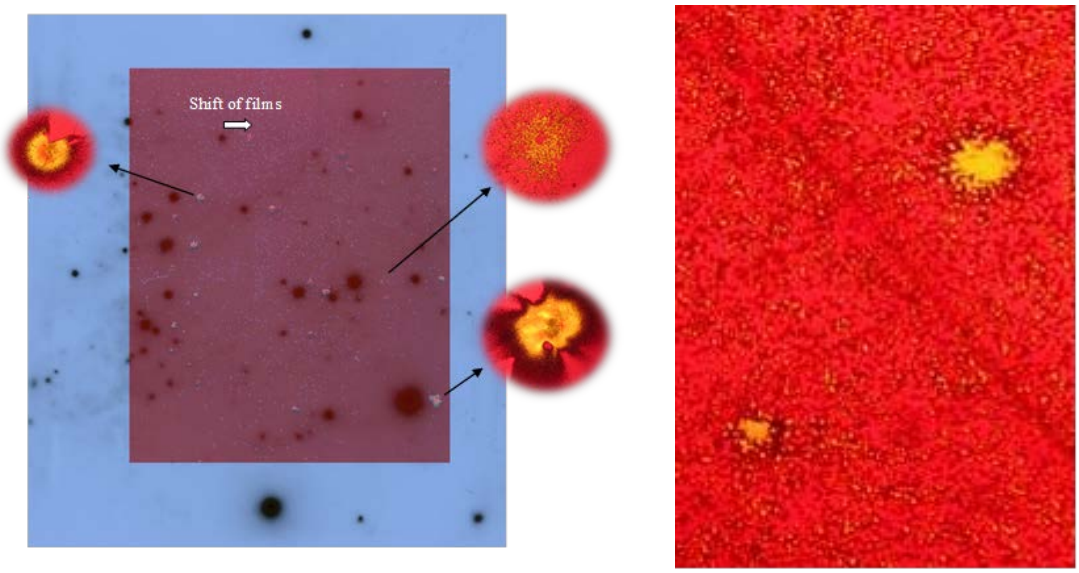
c)

d)

Figure 5.1.2.1.: Sampling units: a) – task 1; b) –task 2; c) – task 3 and d) – task 4

For estimating the fraction (%) of radionuclide (^{90}Sr, ^{154}Eu and ^{241}Am) associated with fuel particles of various types in composite soil samples, we applied sequential extractions using ^{85}Sr (in water-soluble form) as a tracer for monitoring the extraction yield. This method was developed for analyses of radioactive contaminated topsoil, nuclear fuel, radioactive waste and bottom sediments (Kashparov et al., 1999, 2003). In the composite sample of soils, the ^{90}Sr, ^{137}Cs, ^{154}Eu and ^{241}Am activity concentrations were measured by means of gamma and beta spectrometry. In the first stage, extractions by 2M NH_4Ac (exchangeable forms of the radionuclides) and 6M HNO_3 heated to 98°C during 2 hrs according to the standard radiochemical method (total content of the radionuclides in soil, including those in the UO_{2+x} and UO_2 particles) were carried out. For estimation of the fraction of chemically extra-stable particles (U-Zr-O) activity concentration of ^{90}Sr, ^{154}Eu and ^{241}Am in residue after extractions of the composite soil sample by 6 M HNO_3 were measured by means gamma and beta spectrometry. X-ray microanalysis of such particles after extractions of the composite soil sample was undertaken by NMBU.

Autoradiography and α-track radiographic studies were carried out for visualizing high α-active and β-active radioactive particles in samples of the bottom sediments of cooling pond of the ChNPP and in NORM of the tailings of Pridneprovsky Chemical Plant (Dneprodzerginsk, Ukraine), and estimation of their dispersed composition (Fig. 5.1.2.2).



a) b)

Figure 5.1.2.2.: X-ray film and film LR 115 of α-track radiography after joint exposure on the sample of bottom sediments of cooling pond ChNPP (a) and α-track radiography of sludge NORM samples from tailing "Dniprovske" (b)

5.1.3 Results

Task 1 and Task 2. In 2013-2015, the efficiency of the models related to the dissolution (weathering) of Chernobyl fuel particles elaborated 15-17 years ago and the reliability of the associated predictions for the dynamics of plants contamination with ^{90}Sr were verified. The theoretical/modeling estimations of 1997-2001 of exchangeable fraction (%) of ^{90}Sr activity in soils with different pH for the Southern trace of fallout (Fig. 5.1.3.1) coincided with the experimental results obtained in 2013-2015 (Kashparov et al., 1999, 2004, 2013, Otreshko et al., 2014).

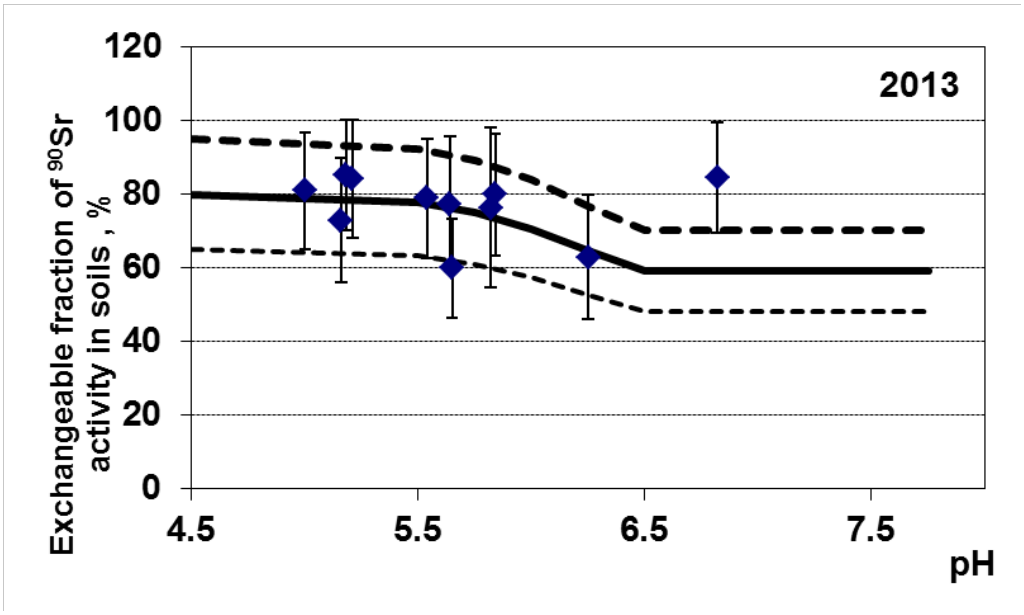


Figure 5.1.3.1.: Exchangeable fraction (%) of ^{90}Sr activity in soils for Southern trace of fallout (UO_{2+x} chemically low stable particles) and theoretical dependence (solid line) in 2013 (mean±SD)

The process of fuel particle dissolution in soils can be well described by a first-order kinetics equation without considering FP dispersal composition:

$$dA(t)/dt=-(k+\lambda)\cdot A(t),\ \ \Delta FP(t)=A(t)/(A_0\cdot exp(-\lambda\cdot t))=exp(-k\cdot t)$$

EQUATION 5.1

where: ΔFP - part of the radionuclide activity in FP; $A(t)$ and A_0 – activity of UO_2 and UO_{2+x} particles at time t after the fallout and the initial activity (Bq), respectively; k and λ - transformation and decay coefficients ($year^{-1}$); t – the time particles have been dissolving in soil after the accident (years). Values of k as a function of soil acidity pH have been derived from ΔFP as follows (Kashparov et al, 1999, 2004, 2012):

For the Western trace:

- **$k=0.6\cdot10^{(-0.15\cdot pH)}$ at pH<7.0**
- **k=0.05 at 7.5> pH>7.0**

For the Southern and Northern traces:

- **$k=40\cdot10^{(-0.45\cdot pH)}$ at pH<6.5**
- **k=0.05 at 7.5> pH>6.5**

The period of FP half-dissolution varies from one to 14 years with increasing soil acidity (pH from 7 to 4).

In 2015, the reliability of the predictions of the dynamics of radionuclides leaching, has been verified in an experimental site situated in the so-called Red Forest within the Chernobyl exclusion zone. The main results included the 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/modeling estimations of 2000-2001 of the exchangeable fraction of radionuclide activity in soils (Fig. 5.1.3.3) coincided with the experimental results obtained in 2015.

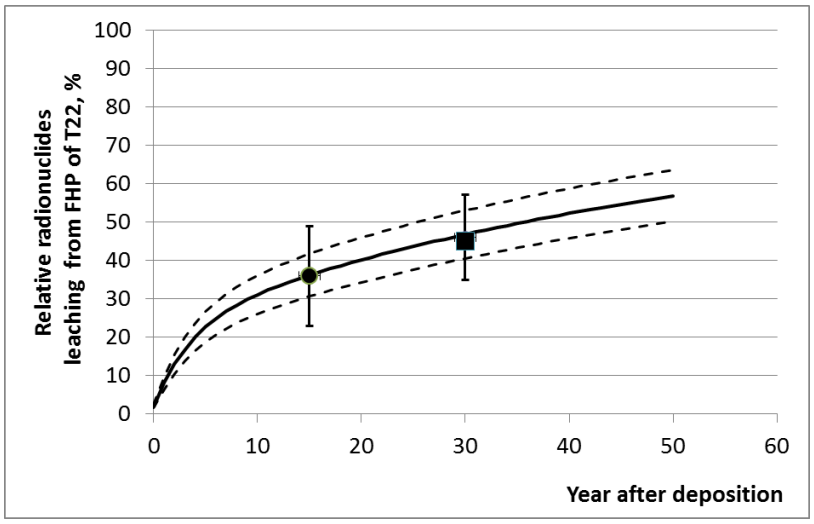


Figure 5.1.3.3.: Relative (%) fraction of ^{154}Eu and ^{241}Am activity of the leached from fuel particles of the waste trench No.22 "Red Forest" in 2001-2002 and 2015 (solid line - theoretical dependence, mean±SD). (Kashparov et al., 2012)

The dissolution process related to fuel particles of various properties (U-Zr-O, UO_2 and UO_{2+x} particles) can be described as a sum of the first-order kinetic equations:

$$\Delta FP(t) = \sum_{i=1}^{4} \Delta FP_i(0) \cdot \exp(-k_i \cdot t)$$

EQUATION 5.2

$$\sum_{i=1}^{4} \Delta FP_i(0) = 1$$

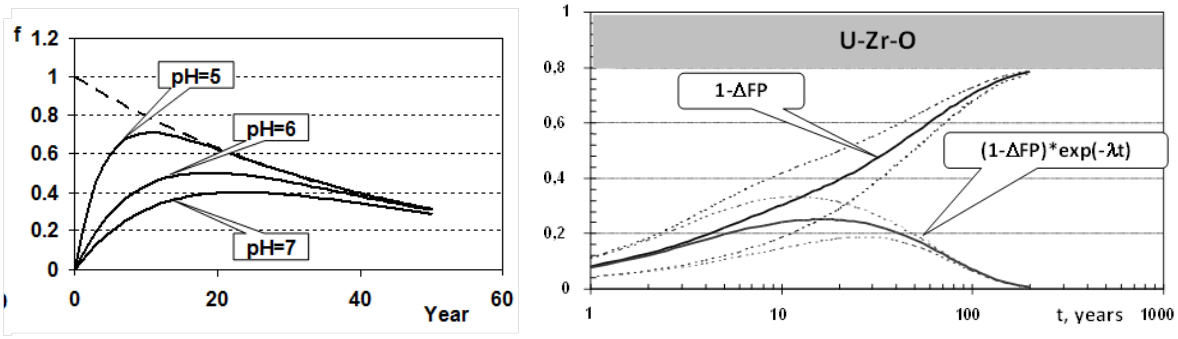
EQUATION 5.3

where: $\Delta FP_i(0)$ is the initial fraction (part of the activity) of the i-type particles (genesis: i=1 for **U-Zr-O** particles, i=2 for **UO_2** particles, i=3 for **UO_{2+x}** particles and i=4 for condensed particles of **^{90}Sr** outside FP); k_i – transformation constant of the particles of i-type (year^{-1}); t – time after the beginning of the dissolution process (years).

Results showed that in TSRW Red Forest (2.5 kilometers South-West of the ChNPP) ^{90}Sr and ^{154,155}Eu, $^{238-241}$Pu and ^{241}Am released during the initial radioactive contamination of the trench (at t=0) were present in four different physico-chemical forms:

1. **U-Zr-O** particles**: $\Delta FP_1(0)=0,20\pm0,10$** of ^{90}Sr total activity and **$\Delta FP_1(0)=0,22\pm0,10$** of ^{154,155}Eu, $^{238-241}$Pu and ^{241}Am total activity (***$k_1 \approx 0$***)
2. **UO_2** particles: **$\Delta FP_2(0)=0,57\pm0,15$** of ^{90}Sr and ^{154,155}Eu, $^{238-241}$Pu and ^{241}Am total activity (***$k_2=0,018\pm0,001$ year^{-1} for pH=5***)
3. **UO_{2+x}** particles**: $\Delta FP_3(0)=0,21\pm0,15$** of ^{90}Sr, ^{154,155}Eu, $^{238-241}$Pu and ^{241}Am total activity (***$k_3=0,28\pm0,05$ year^{-1} for pH=5***)
4. Condensed particles of **^{90}Sr** outside FP**: $\Delta FP_4(0)=0,02\pm0,01$** of ^{90}Sr total activity (***($k_4=\infty$).*** **$\Delta FP_4(0)=0$** for ^{154,155}Eu, $^{238-241}$Pu and ^{241}Am.

The results of the COMET project showed that the part of ^{90}Sr activity in the mobile form (the potential biologically available form) has reached its maximum values for the post-accident period due to fuel particles dissolution in topsoil and in radioactive waste of the trench materials (Fig. 5.1.3.4).



a) b)

Figure 5.1.3.4.: Relative (%) dynamics of ^{90}Sr contents in the mobile forms in topsoil (a) and in radioactive waste of the trench materials (b)

The dynamics of the contamination of grains and wood with ^{90}Sr is mainly determined by the kinetics of fuel particle dissolution and by changes in the mobile radiostrontium concentration in the root-layer. A monotonic increase of the concentration ratio (CR) of ^{90}Sr for the grain (Fig. 5.1.3.5) was observed in the survey area due to dissolution of the fuel particles and this coincided well with model predictions (Kashparov et al., 2012, 2013, Otreshko et al., 2014 and 2015).

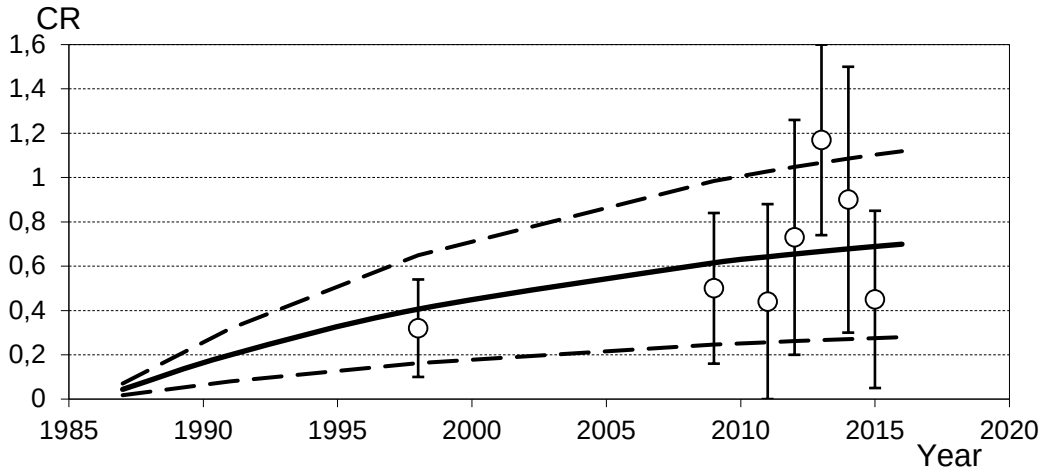


Figure 5.1.3.5: Dynamics of the ^{90}Sr average CR in grain (date in 1997-2011 from Kashparov et al., 2012) and theoretical estimates (solid line) for pH=7 (Fig.1) and average CR=1.0±0.5 in 2013 (Otresko et al., 2014)

Task 3. The investigation of the dissolution of fuel particles in the drained bottom sediments started in 2013 in the framework of the COMET project, before the beginning of a decrease in the water level of cooling pond of the ChNPP in 2015. Today, the water level has decreased by 4 m and the drained very contaminated bottom sediments were exposed to air (Fig. 5.1.3.6). Assessments of physico-chemical forms of ^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239,240}Pu, ^{241}Am and the composition as well as the spatial distribution of fuel particles in bottom sediments of cooling pond were carried out. According to the obtained results, more than 98 % of ^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239,240}Pu, ^{241}Am were in non-exchangeable forms in the slimy bottom sediments (Fig.5.1.3.7). About 70-80 % of total ^{90}Sr, ^{241}Am and plutonium isotopes activity may still be located within the matrix of the fuel particles (Protsak and Odintsov, 2014).

a) b)

Figure 5.1.3.6.: Water level of the ChNPP cooling pond in July (a) and in September (b) 2015

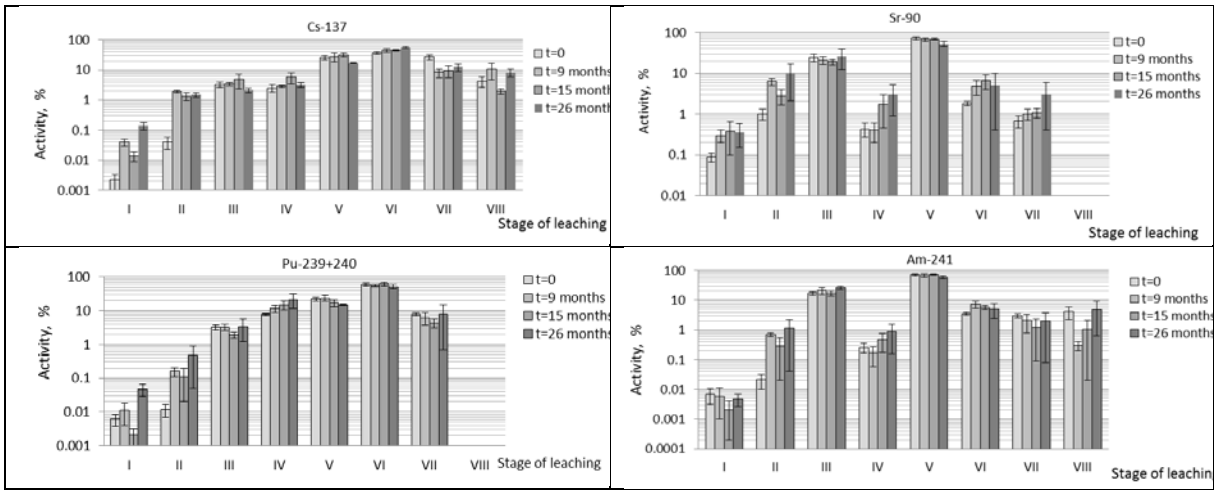


Figure 5.1.3.7 The dynamics of ^{137}Cs, ^{90}Sr, $^{239+240}Pu$ and ^{241}Am activity among fractions (%) leached from the bottom sediments of CP ChNPP after their draining (t=0) and exposure during 9 months (t=9), 14 months (t=14) and 26 months (t=26) in the natural conditions on the experimental site (stage of leaching: I - distilled water; II - 1 M CH_3COONH_4 (NH_4Ac); III - 1 M HCl; IV - 0.2 M $(NH_4)_2C_2O_4$ + 0.1 M $H_2C_2O_4$; V - 8 M HNO_3; VI - Residue after stage 5 was washed at 550 °C during 6 hours, treated by the acid mix of 8 M HNO_3 + 10 M HCl for 2 hours at 95° C; VII - Residue after stage 6 was leached by the acid mix: 8 M HNO_3 + 4 M HF for 2 hours at 95° C; VIII - insoluble rests).

Task 4. The obtained results allow the estimation of the main physical-chemical parameters and elemental composition of core material of PChP tailings. The highest differences between the element concentrations in the tailing dumps were found for such elements as Al, S, K, Ca, Mn, Fe, As, Sr. Specific activities of the uranium-thorium series radionuclides decreased in the PChP tailings sequence: Central Yar > Dneprovskoe > Zapadnoe. The highest activity concentration of the uranium-thorium series radionuclides was found in the Centralnyy Yar tailing of PChP (for ^{226}Ra up to 100 Bq/g). Within the framework of the COMET project, a distribution of α-emitting radionuclides in sludge of PChP tailings was investigated by autoradiography, using track detectors (Fig. 4X.). During this study, single spots with a significantly high activity concentration were revealed, as well as a inhomogeneous background distribution of the α-activity. These spots indicated the presence of radioactive particles. The size of the investigated particles was from 2 µm to 20 µm. The activity concentration of α-emitting radionuclides in these particles was of the same order of magnitude as the activity concentration of α-emitting radionuclides in the Chernobyl fuel particles.

5.1.4 Discussion

Task 1. The dissolution rate of fuel particles in waste trenches is an important source term parameter of long-term dynamic models simulating root uptake in plants and trees and potential migration of radionuclides, including plutonium, to groundwater. Such processes need to be implemented in associated with radiation protection assessment of the human and environment after Chernobyl accident (Kashparov et al., 2012, 2013, Levchuk et al., 2012, Otreshko et al., 2014 and 2015).

Review and detail analysis of available data on the long-term environmental behaviour of radioactive particles (soil, plants, bottom sediments etc.) derived from research carried out in Ukraine following the Chernobyl accident (Kashparov et al., 2004, 2012,2013) have been accomplished. Radioactive fuel particles specific for the Chernobyl accident can be divided into 3 groups according to their dissolution rates under natural conditions (Kashparov ey al., 2004, 2012), as described above.

In 2013, efficiency of the models related to the Chernobyl fuel particles dissolution elaborated 15 years ago, and the reliability of their predictions for the dynamics of plants contamination with ^{90}Sr, were experimentally verified within the COMET project. The theoretical/modeling estimations of 1997-2000 of the exchangeable fraction (%) of ^{90}Sr activity in soils with different pH for the Southern trace of fallout (UO_{2+x} chemically low stability particles) (Kashparov et al., 2004) coincided with the experimental results in 2013 (exchangeable fractions of ^{85}Sr in soils was 80±15 %).

The fraction (%) of ^{90}Sr activity in biologically available forms (e.g., uptake in grain) has reached its maximum values for the post-accidental period due to fuel particles dissolution. Values of concentration ratios (CR) and aggregated transfer factors of ^{90}Sr from soil to rye and oat grain are inversely proportional to exchangeable calcium content in soil. Then, the transfer factors and dependences are in good accordance with those that had been obtained in our previous works and with generalized data of the IAEA for sandy soil.
In all samples collected in 2013, the activity concentration of ^{90}Sr (24 – 59 Bq/kg) exceeded the permissible level of ^{90}Sr for food grains (20 Bq/kg) due to fuel particle dissolution. Monotonic increase of CR of ^{90}Sr was observed in the grain in the survey area due to dissolution of the fuel particles and this coincides well with model presictions. The dynamics of the contamination of vegetation grain and wood with ^{90}Sr is mainly determined by the kinetics of fuel particle dissolution and by changes in the mobile radiostrontium content in the root-layer. Depending on the fuel particle dissolution rate, root contamination of plants by ^{90}Sr is an important factor in radiation protection of the human and environment after Chernobyl accident (Otreshko et al., 2014). Thus, the presence of fuel particles represent a delay in the ecosystem transfer, and the apparent Kd and subsequent uptake in vegetation (CR) changes over time. The apparent Kd decreases and CR increases due to the particle weathering rates and the subsequent remobilization of particle associated radionuclides.

Task 2. In the years 2015, the robustness of the models of Chernobyl fuel particles dissolution, elaborated 15 years ago, have been verified with respect to the dynamics of ^{90}Sr, ^{154}Eu and ^{241}Am activity remobilized from fuel particles in the radioactive waste trench. The theoretical/modeling estimations of 2000-2001 of the exchangeable fraction (%) of the radionuclides activity in soils coincided with the experimental results obtained in 2015. The results showed that the activity of the mobile form of ^{90}Sr in the trench has presently reached the maximum value, and the activity concentration of radionuclides in groundwater and surface water should not change significantly.

Task 3. The low dissolution rate of radionuclide leaching from sediments of the ChPNN cooling pond is controlled by a prolonged slightly alkaline pH in the medium. In turn, this is caused by the presence of a large number of remnant shells of zebra mussels in bottom sediments of the ChPNN cooling pond (Fig.5.1.4.1). It can be stated that a sharp increase of the mobilization of radionuclides should not be expected from the newly exposed sediments (Protsak and Odintsov, 2014). Thus, a significant increase of radionuclide mobility in the sediments, which was associated with the beginning of the destruction of the fuel particles after drying and 26 month exposure in the natural conditions, was not observed, contradicting the existing forecasts (Bulgakov et al., 2009).

A significant part of the radionuclide activity was not extracted from the bottom sediments collected from the CP ChNPP, even under the "extra strong" extraction conditions (stage of leaching V-VII, Fig. 10X). This fact indicates that part of radionuclides are kept in a chemically very stable form as U-Zr-O particles, and for this reason these radionuclides will not be transferred into the environment for many years.

Thus, it can be assumed so far that 70 to 90 % of the radionuclide activity in the bottom sediments of CP ChNPP are still associated with fuel particles (half-lives more than 50 yrs). The obtained results

indicate the spatial heterogeneity of forms of radionuclides in sediments of the ChNPP cooling pond. In this regard, more detailed studies should be performed in the future.

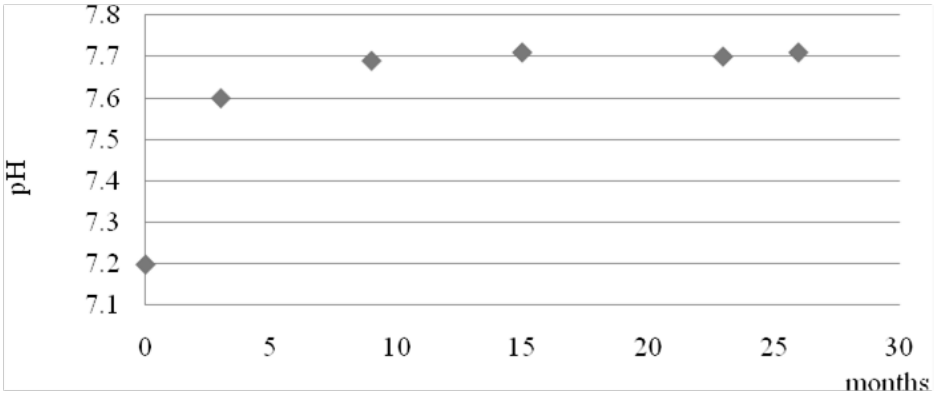


Figure 5.1.4.1.: Dynamics of pH in medium of modified bottom sediment at the experimental site

Task 4. By autoradiography using track detectors the homogeneity of the distribution of α-emitting radionuclides in sludge of PChP tailings has been investigated. It has been found that beside a homogeneous background distribution of the α-activity, single spots with significant higher activity concentration exist in form of α-active particles. Each gram of NORM contained hundreds of radioactive particles. The concentration of radionuclides in the sludge from different tailings was much higher than background level. Thus, handling TENORM may cause problems in dry processes, where NORM scales and dust become air borne. The main hazards associated with α-active particles are inhalation and ingestion routes of entry. The study of the characteristics of the α-active particles in tailing dumps should therefore be continued.

The key objectives of the WG: parameters of the dynamic models of the FP dissolution in soils and in the radioactive waste trench have been checked and clarified for the long-term prognosis of the mobility and potential bioavailability of radionuclides.

The parameters of the FP dissolution in the drained bottom sediments have not been revealed due to a very slow dissolution rate that cannot be estimated during the COMET project period.

The presence of radioactive particles is an important parameter of the source term, while the particle properties influencing weathering/dissolution and remobilization of particle associated radionuclides are essential for ecosystem transfer. The implementation of particle codes and relevant particle characterization input are essential for emergency preparedness models, and also for estimating long-term dynamic models such as root contamination of plants for IRA working groups “Human food chain modelling” and “Forest modelling”.

5.1.5 Impact and further work

Based on the results from the COMET project, a long-term prediction of the activity concentration of ^{90}Sr in agricultural (grain) and forest (firewood) products can be obtained for the territories affected by the Chernobyl accident and other accidents with a release of irradiated nuclear fuel particles. Future needs relate to predicting the long term behaviour of more exotic radionuclides also associated with the fuel particles.

Since the dissolution of the fuel particles in the drained bottom sediments of cooling pond of ChNPP is very slow to be estimated for today, the studies of this process should be continued under real conditions.

The study of the characteristics of the α-active radioactive particles (respirable size < 10 μm) containing NORM from Pridneprovsky Chemical Plant (Dneprodzerginsk, Ukraine) tailings dumps and assessment of their radiological hazard should be continued. Similar studies of radioactive particles are also needed at other NORM sites.

The greatest attention should be paid to the study of the effect of radioactive particles on organisms, including the manifestation of inflammatory processes and the development of microdosimetry, as pointed out in the COMET Position paper (Salbu, et al., *in press*).

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5.2 RATE - RAdioactive particle Transformation processes

5.2.1 Background

A major fraction of refractory radionuclides such as uranium (U) and plutonium (Pu) released to the environment from the nuclear weapon and fuel cycles is present as particles ranging from sub-microns to fragments. Such particles can carry a substantial amount of radioactivity (e.g., fission and activation products, transuranics) and associated metals, and can act as point sources. Furthermore, U particles (progenies, metals) are present at NORM sites.

Research indicates that particle characteristics such as composition, atom and element ratios depend on the emitting source, while particle size, structure and oxidation states are closely linked to the release scenarios. Following deposition, ecosystem transfer of particle associated radionuclides are delayed compared to mobile radionuclide species; i.e., ecosystem transfer would be delayed until particle weathering and remobilisation of associated radionuclides occur. The apparent soil-water distribution coefficient (Kd) will therefore change over time, and the thermodynamic constant concept should be replaced with rate functions. At the same time the particles and their associated radionuclides can be retained uptake by selective biota

Thus, the present project has focused on particle characteristics, weathering rates, remobilization and prediction of ecosystem transfer of radionuclides associated with U and/or Pu containing particles originating from selected key sources (nuclear weapons tests, safety tests, conventional detonation of nuclear or DU weapons, nuclear reactor accident, NORM) as well as in linking particle characteristics to retention in biological systems

Utilizing advanced techniques, leaching experiments in which well characterized particles are exposed to *abiotic* and *biotic* degradation agents have been performed. The parameterization should reduce uncertainties in model predictions on ecosystem transfer and environmental impact associated with

particle contaminated areas, linking particle properties to sources, and linking particle properties to weathering rates under different environmental conditions.

Objectives

Overall objectives of the present project have been to fill knowledge gaps related to transformation processes influencing weathering of radioactive particles released to different ecosystems and the subsequent release of associated radionuclides, to reduce the uncertainties in environmental impact assessments of particle contaminated sites. To achieve the objectives, focus was put on

- Linking particle characteristics to specific source and release scenarios by utilizing particles provided from different particle contaminated sites by the partners and by utilizing advanced techniques for characterization of these particles.
- Analysis of transformation processes affecting particle composition such as size, structure, and oxidation states due to ***abiotic conditions*** (e.g., pH, conductivity, complexing agents, physical-chemical properties of the particles, mechanical disruption) and due to ***biotic conditions*** (e.g., microorganisms/fungi affecting redox conditions, degradation after uptake in selected organisms, chelate formation), by performing well defined leaching experiments under controlled conditions, and *In situ* measurements of particle breakdown products. The work is based on developed protocols.
- Linking particle characteristics to ecosystem processes (weathering rates, time-dependent Kd's, selected uptake experiments (CR/BCF)).

The structure of this report will follow the three main tasks carried out along the project that are the following:

Task 1: *To produce a particle database from archived nano- and micrometre sized radioactive particles released from different sources, varying with respect to composition, particle size, crystalline structure and oxidation states.*

Task 2: *To develop and define protocols for leaching of radioactive particles, to use and apply them afterwards to develop information on the solubilisation, remobilization and ecosystem transfer potential of particle associated radionuclides*.

Task 3: *To study and analyze the phenomenon of retention or uptake of particle associated radionuclides in various biota from contaminated areas.*

5.2.2 Methods and materials

Particle characterization

The particles included in the generated database and available for the abiotic and biotic transformation processes on the frame of the present project have been localized, identified, isolated and characterized using a set of non-conventional techniques in radioecology that will be summarized in the following paragraphs.

In particular, the characterization of the particles has been carried out mostly by using microanalytical techniques based in the use of different types of beams (electrons, ions and neutrons). Information about elemental composition has been obtained by analyzing X-ray emissions generated by X-ray beams (μ-XRF), proton beams (μ-PIXE) and electron beams (SEM-EDX). In the great majority the SEM-EDX technique was used, combined with one of the other two techniques. Electron microscopes available for all the partners to perform SEM-EDX, tandem accelerators available at ANSTO and the University of Seville, and synchrotron facilities approached as external users to perform μ-XRF were used.

μ-XRF can be performed at laboratory scale using typically as excitation sources X-ray tubes or radioisotope sources together with focusing and collimating optics to produce the beam, or alternatively, X-ray beams enough intense to allow very sensitive microanalysis can be obtained by using much more brilliant synchrotron radiation (electromagnetic radiation from infrared to X-rays) generated by the acceleration of light elements in synchrotron facilities. (Salbu et al., 2000).

The use of very intense X-ray beams has additionally the advantage that in addition to μ-XRF other complementary techniques giving structural, morphological and oxidation states information can be applied (Salbu et al., 2001). In this sense, μ-XRF can be combined with:

a) μ-X ray absorbed near edge structure technique μ-XANES) to obtain information about oxidation states of elements present in the analysed particle.

b) μ-X ray diffraction (μ-XRD) to obtain information on structural data in crystalline materials, and

c) μ-X ray computed tomography (μ-XRCT) for the visualization of local structures. This last technique is based in the fact that the intensity of the X-ray beam passing through the sample (the intensity of the transmitted X-ray) depends on density, thickness and elemental composition.

In addition to the microanalytical techniques based in the use of excitation beams, another set of techniques based in atom counting and covered by the general term of mass spectrometry (MS) techniques have been applied. On the frame of the RATE project two main mass-spectrometric techniques (AMS and ICP-SF-MS) have been used for ultrasensitive determination of ^{239}Pu and ^{240}Pu in selected particles. The AMS facilities available at the University of Seville and ANSTO and the high-performance ICP-MS systems available at NMBU and DTU have been used. Both techniques (AMS and ICP-SF-MS) are characterized for being destructive techniques: radiochemical separation of the Pu existing in the particles after their complete dissolution, and conditioning of the Pu isolated fraction to the requirements of the applied MS technique (in dissolution for ICP, as a solid cathode in AMS) is needed.

Complementing to the mentioned mass-spectrometric techniques (ICP-SF-MS, and AMS) conventional nuclear techniques (NT), based in detecting emitting radiations, have been used for isotopic Pu characterization in the hot particles (X-Ray spectrometry and Alpha-particle spectrometry). The drawbacks of these nuclear techniques (NT) are the relatively high limits of detection in comparison with the mass-spectrometric techniques (MS), but on the other hand one of them (X-ray spectrometry) is a non-destructive technique that can be applied before the performance of the characterization and transformation studies.

Abiotic and biotic protocols

In order to decrease the overall uncertainty in assessing the health and ecological impacts of radionuclide releases in particle contaminated areas, RATE has developed and defined protocols for leaching of radioactive particles, to use and apply them afterwards to develop information on the solubilisation, remobilization and ecosystem transfer potential of particle associated radionuclides. The intention was to identify key parameters affecting properties of anthropogenic and NORM containing particles (either when changing water qualities, temperature and utilizing abiotic leaching agents or when they are exposed to biological systems such as microorganisms influencing redox systems or rumen liquids from grazing animals) as a way to determine transformation kinetics (weathering rates, time dependent Kds, etc).

In order to generate values directly comparable and reproducible between laboratories, special efforts have been devoted to define joint, scientifically agreed protocols for abiotic and biotic transformation studies between the different RATE partners. One of the main outputs of the project has been these protocols which will be detailed in the next pages.

With basis in the protocols defined by consensus, leaching experiments in which radioactive test particles have been exposed a) to abiotic reagents such as rain water, sea water, irrigation water and synthetic solutions mimicking body fluid such as simulated stomach fluid, and b) to biological systems such as rumen liquids from grazing animals, have been performed associated to this project by the different partners. In addition, and in parallel to the transformation experiments applied on single radioactive particles (RP), similar experiments (with also a well-defined and specific protocol) has been applied on aliquots from bulk samples from which the particles were isolated.

The defined abiotic protocol has been constructed with basis in several key points that will allow reaching to unique and original information. In this sense we can indicate, that:

a) The radioactive particle (RP) submitted to the leaching experiment is characterized prior and, if it is possible, after the transformation experiments by non-destructive techniques, by using as a minimum gamma spectrometry. Additional techniques such as electron microscopy, labtop nanotomography and SR based nano or micro- XRF/XANES/XRD have been used prior to transformation experiments for RP characterization.

b) The contact time RP-solution will be dependent on the type of solution used, at the same time that the temperature at which the particle leaching is conducted must be recorded. When using water solutions, room temperature should be applied, ranging between 20-23 ºC; while with HCl 0.16 M (gastric fluid simulant) or with rumen gastric fluids the temperature should be adjusted to the "human body temperature", 37ºC ±1; therefore, a thermostatic bath must be employed.

c) In the abiotic leaching experiments, information about three different fractions in each obtained leachate is gained: low molecular mass, colloidal and particulate fractions.

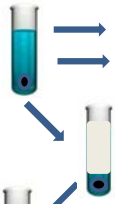
d) The Pu isotopic content in the leachate fractions obtained is determined by Accelerator mass spectrometry (AMS), while in the case of NORM particles, the amount of Th or U leached in the experiments is determined by ICP-MS. By applying these techniques, it is possible to reach limits of detection that cannot be approached by the application of more conventional radiometric techniques such as gamma-ray or alpha-particle spectrometry.

The detailed proposed abiotic Particle leaching experiment is illustrated in the following diagrams:

Flux diagram of abiotic protocol with Radioactive Particle (RP)

1. Place RP at the bottom of a 50 mL Nalgene polypropylene Oak Ridge centrifuge tube.
2. ɣ spectrometric measurement prior the experiment.
3. Add 20 mL solution (i.e water, acidic solution) (T_0: experimentation starts).

T1 First Contact time (if solution is water 1h, acid solution 2h).



Transfer 10 ml of the upper volume to a centrifuge tube (Aliquot 1- T_1).

Transfer 8 ml of the upper volume to a scintillation vial (Aliquot 2-T_1).

Remaining in the tube is 2 ml and the RP.

Addition of 18 ml fresh solution.

T_n n Contact time (if solution is water then n=2 is 1day, n=3 is1 week, n=4 is 1 month)
(if acid solution then n=2 is 1 day, n=3 is 1 week).

Transfer 10 ml of the upper volume to a centrifuge tube (Aliquot 1- Tn).

Transfer 8 ml of the upper volume to a scintillation vial (Aliquot 2-Tn).

After each contact time, it remains 2 ml and the RP
and 18 ml of fresh solution is added.

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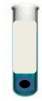


After the nth contact times, at the end the RP and a final 2 ml solution is remaining in the tube.

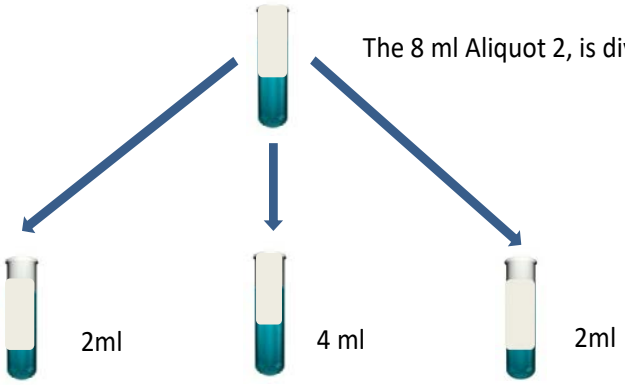
1) Transfer RP and the 2 ml residual solution to a 20 ml filter funnel with a 0.45 µm filter and filter directly into a vial for analysis.
2) Rinse the filter funnel with 2x 2 ml deionized water.
3) Measure the RP which is now residing on the 0.45 µm filter by gamma spectrometry.

Flux diagram of abiotic protocol with RP (Continued)

TREATMENT PROTOCOL FOR ALIQUOT 1

1. The 10 ml aliquot is centrifuged at minimum 3000 g for 30 minutes.
   The centrifugation allow separation of the dissolved and colloidal phase.
2. Transfer 5 ml of the upper supernatant volume into a vial and keep it for analysis.
3. The supernatant of Aliqout 1 represents a low molecular fraction (LMM).

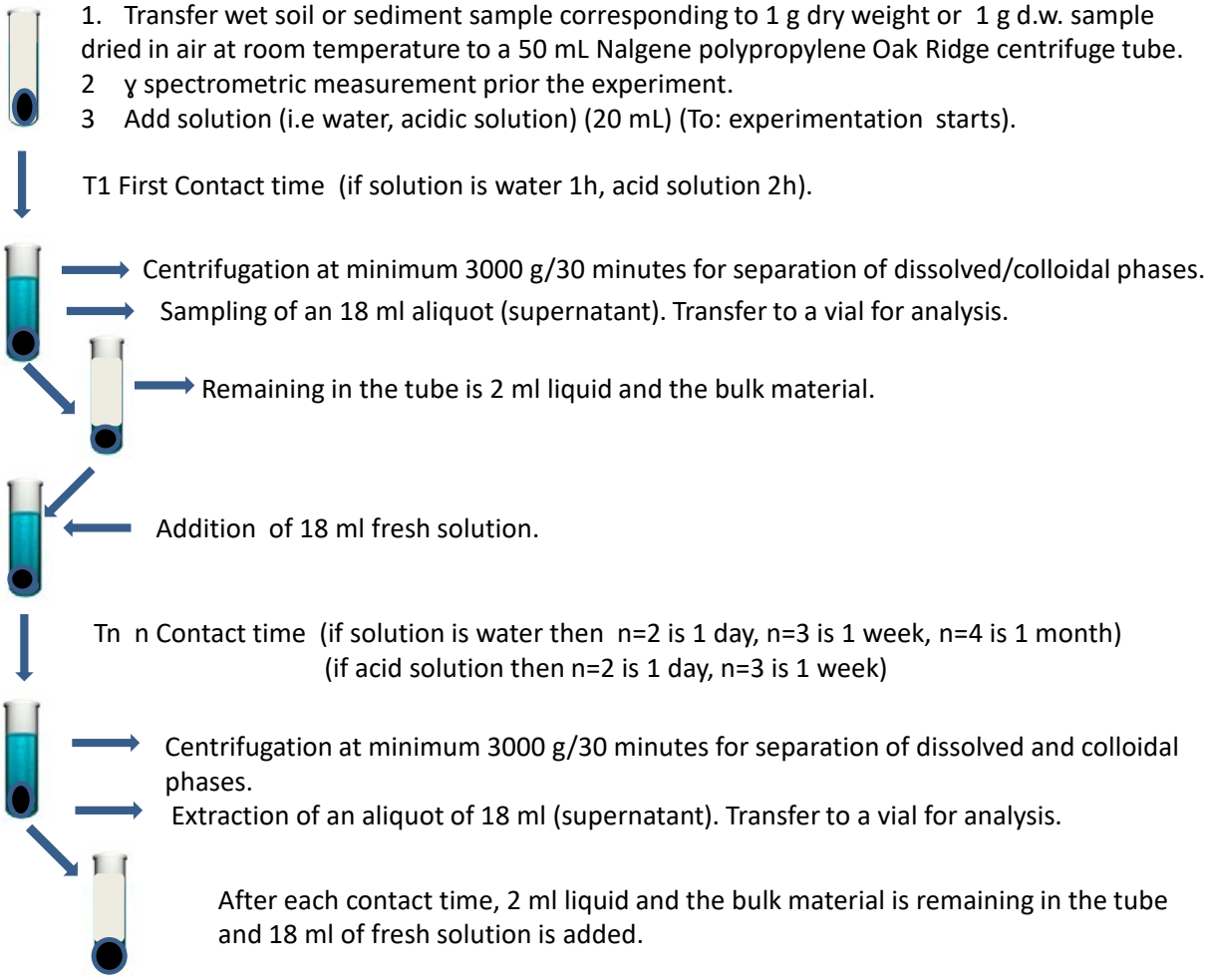
TREATMENT PROTOCOL FOR ALIQUOT 2



Fraction 1	**Fraction for 0.45 µm syringe filter filtration: Use 1 ml for conditioning of filter and discard the filtrate, use 1 ml for sampling of filtrate and keep it for analysis.**
Fraction 2	**Fraction for membrane ultracentrifugation. Use the first 2 ml for conditioning and discard the filtrate, keep the filtrate from the subsequent 2 ml for analysis.**
Fraction 3	**This 2 ml fraction is kept for total analysis.**

A more simplified abiotic protocol has been generated for application to the bulk samples. Obviously, all the solutions used should be prepared from 18 MΩ water and p.a. reagents. The protocol is described in the following illustration:

Flux diagram of bulk leaching protocol



Finally, and along the project, we have applied the following biotic protocol, using liquid rumen from cow as a leachate, to a variety of soil contaminated samples

1. Transfer wet and fresh soil or sediment sample corresponding to 1 g dry weight or a 1 g d.w. sample dried in air at room temperature to a 50 mL centrifuge tube with polypropylene screw closure.
2. Add 50 mL of the cow rumen solution. This rumen should be collected the same day that the leaching experiment is going to be performed and should be transported from the collection place to the lab maintaining its original temperature. In the laboratory, the temperature and the pH should be measured and recorded.
3. The amount of cow rumen added to the soil sample will fill completely the centrifuge tube, being that needed to avoid the presence of air inside the tube in the next steps.
4. The contact time between the sample and the rumen is restricted to 24 hours. The temperature at which the sample is leached must be on the other hand maintained at 39 ºC, and during the leaching time, gentle rolling agitation (ca 50-70 rpm) should be used similarly to the RP abiotic leaching. In this way, the movements and temperature of the cow animal are simulated. With this end the centrifuge tube and the rolling agitator should be introduced in an incubator system.
5. After 1h of leaching, the experiment should be interrupted in order to open the centrifuge tubes thereby avoiding a build-up of gas pressure.
6. After 24 hours contact time, centrifugation is performed at 10000 rpm. The first 20 ml of supernatant are taken for analysis while the remaining liquid is kept for additional analyses.

It is necessary to remark that all the rumen gastric juice leaching experiments have been performed on soil aliquots. The turbidity of the rumen juice prevents for the moment the application to small sized radioactive isolated particles due to the difficulties to follow and control their location and integrity during the leaching experiments.

Screening heterogeneities in biota

Along the RATE project the phenomenon of retention or uptake of particle associated radionuclides in various biota from contaminated areas have been studied, centering the efforts in terrestrial areas affected by Pu/U releases to the environment in particulate form with origin in accidental or provoked conventional detonations of nuclear weapons: Maralinga (Australia), Palomares (Spain) and Thule (Greenland). Additionally, the retention capacities of radioactive particle by mussels have been analysed and evaluated through the performance of laboratory experiments.

For the performance of these studies the set of microanalytical techniques previously described and used in the characterization of the micrometer size particles have been used in combination with more conventional radiometric (gamma and alpha spectrometry) and non-radiometric (autoradiography) techniques.

5.2.3 Results

Radioactive particle Database

In the frame of the RATE project, the research has been focused on radioactive particles characteristics, weathering rates, remobilization and prediction of ecosystem transfer of radionuclides associated with U and/or Pu containing particles originating from selected key sources (nuclear weapon tests, safety tests, nuclear reactor accidents, NORM....) (Salbu, 2001), and with this end, a deliverable associated to this research has been the creation of a database with the information about identified and isolated

hot-particles that has been available for performing the transformation studies. The detailed description of this database was covered by the COMET deliverable 3.4

The database is formed by selected isolated hot particles with quite different origin which has been made available by the different partners of the RATE project. This database is not exhaustive, being important to indicate that the partners have in their archives (or has access to) contaminated soil samples from different origin from where they can identify and isolate additional particles for additional transformation studies, if they are needed. But, in spite of this fact, we can indicate that this database is a unique selection of particles of well characterized anthropogenic and naturally occurring nanometre-millimetre size particles representing different historical sources and releases scenarios, which allowed obtaining very rich and original information for prediction of ecosystem transfer of particle-associated radionuclides

The particles included in this database and available for the abiotic and biotic transformation processes on the frame of the present project have been localized, identified, isolated and characterized using a set of non-conventional techniques in radioecology previously described. This database is formed schematically by radioactive particles with the following origin:

Table 5.2.3.1.: Origin and number of radioactive particles included in the database

Origin of the Particles	Number of particles
Nuclear Weapon Tests	**12**
Conventional detonations	**21**
Accidents in Reactors and Nuclear Reprocessing Plants	**8**
Reactor Accidents: Chernobyl	**3**
NORM Sites	**10**
TENORM facilities	**8**

Detailed description of the database can be found in Deliverable 3.4.

Abiotic and Biotic Transformation Processes

Along the project, leaching experiments have been applied to particles and soil aliquots from quite different origin, although they can be classified mostly in three different important groups:

a) Pu/U particles and contaminated soils corresponding to locations affected by accidental weapon tests (Palomares, Spain and Thule, Greenland) or controlled/safety detonation tests (Taranaki, Australia).

b) Pu/U particles and contaminated soils collected in areas where atmospheric weapon tests were performed (Semipalatinsk, Kazhajistan ; Trinity, United States ; Maralinga and Montebello, Australia).

c) Th enriched Particles and soils from a High Background natural Radioactivityy Area (HBNR) located in Scandinavia (Fen, Norway)

The experiments were centred in the indicated areas in order either to cover an important gap at European level, or at the same time to complement the studies performed in parallel by the IRA radioactive particle group of COMET in areas such as the affected ones by the Chernobyl accident.

In the following Table is compiled all the information about the isolated radioactive particles and corresponding soil aliquots that have been analysed on the frame of the RATE project. Hundreds of solutions to be measured by ICP-MS and/or AMS have been obtained. In this report only some few clear and representative examples should be shown in the following pages.

From the table below it can be deduced that the previously defined protocols have been applied to isolated radioactive particles but also to soil aliquots, and that different leaching agents have been applied: rainwater, HCl 0.16 M (mimicking the human gastric fluids), rumen cow, GI tract simulation fluid, etc.

Site	Particle	Soil aliqouts	Rainwater extraction (centrifugation only)	Rainwater extraction (fractionation)	Rumen liquid (cow)	0.16 M HCl	GI tract simulation (enzymes)
Taranaki	**1**			**1**		**1**	
Taranaki		**1**	**1**			**1**	
Palomares Z2	**1**					**1**	
Palomares Z2		**3**				**3**	
Palomares Z6	**1**					**1**	
Palomares Z6		**5**				**5**	
Thule terrestrial	**2**	**4**		**2**		**4**	
SPTK-4 (Semipalatinsk)	**3**			**1**	**1**	**2**	
Australian glassy particles	**7**			**7**			**4**
Australian TS		**7**	**7**		**7**	**3**	
Fen	**3**			**3**			
Fen		**6**	**3**		**3**	**3**	
Chernobyl	**1**				**1**		

Only as an example, the results obtained by analyzing the total fraction in the leaching with 0.16 M HCl of two contaminated soil aliquots with origin in Palomares and Thule, respectively, are shown in Figure 5.2.3.1.

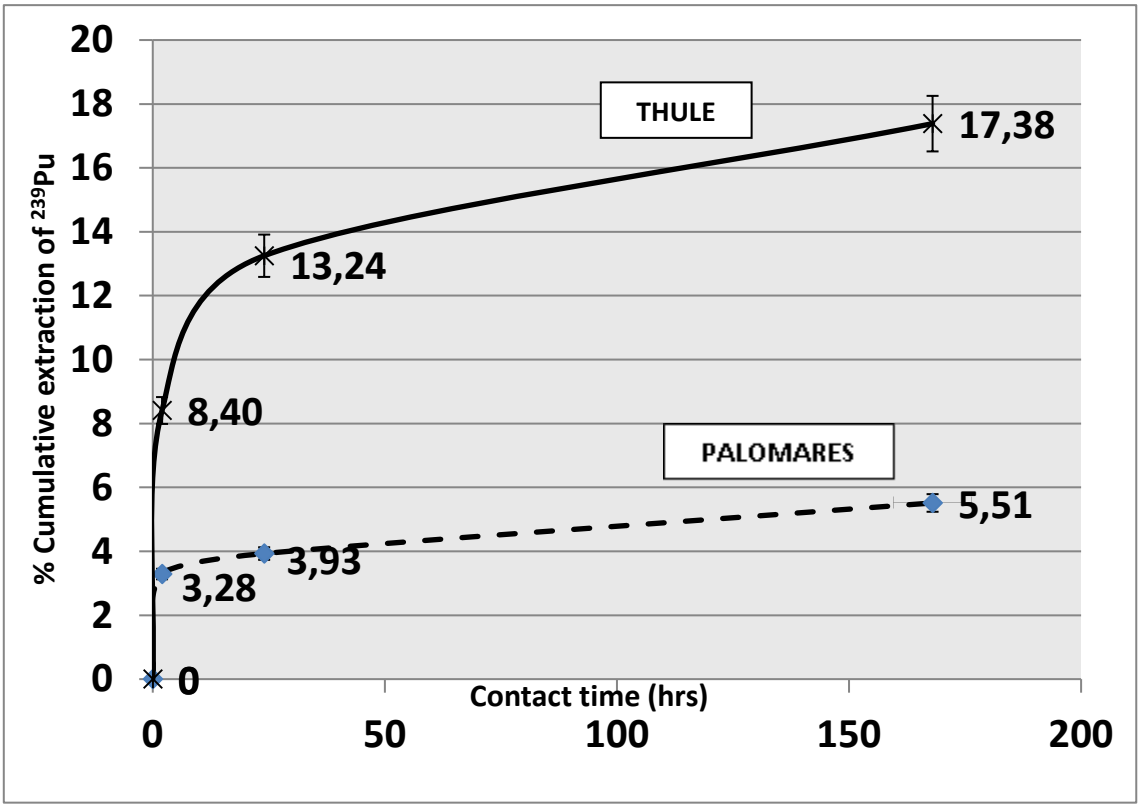
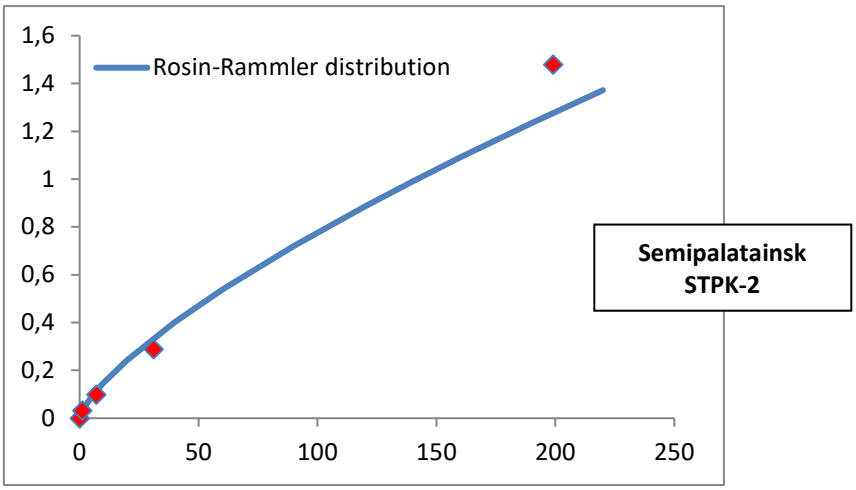


Figure 5.2.3.1.: % of cumulative extraction of Pu vs contact time after the treatment of Pu contaminated soil aliquots from Palomares and Thule with 0.16 M HCl

Results obtained in other leaching experiments with 0.16 M HCl solutions to radioactive particles from Semipalatinsk and Alamogordo are shown in Figure 2. The three radioactive particles submitted to the experiment were previously characterized forming part of the radioactive particle database created associated to the project: in the case of Semipalatinsk correspond to milimetre particles isolated from contaminated soils at the area called Ground zero where atmospheric nuclear weapon tests were performed, while the Alamogordo particle, correspond with the weapon test product commonly known as Trinitite.



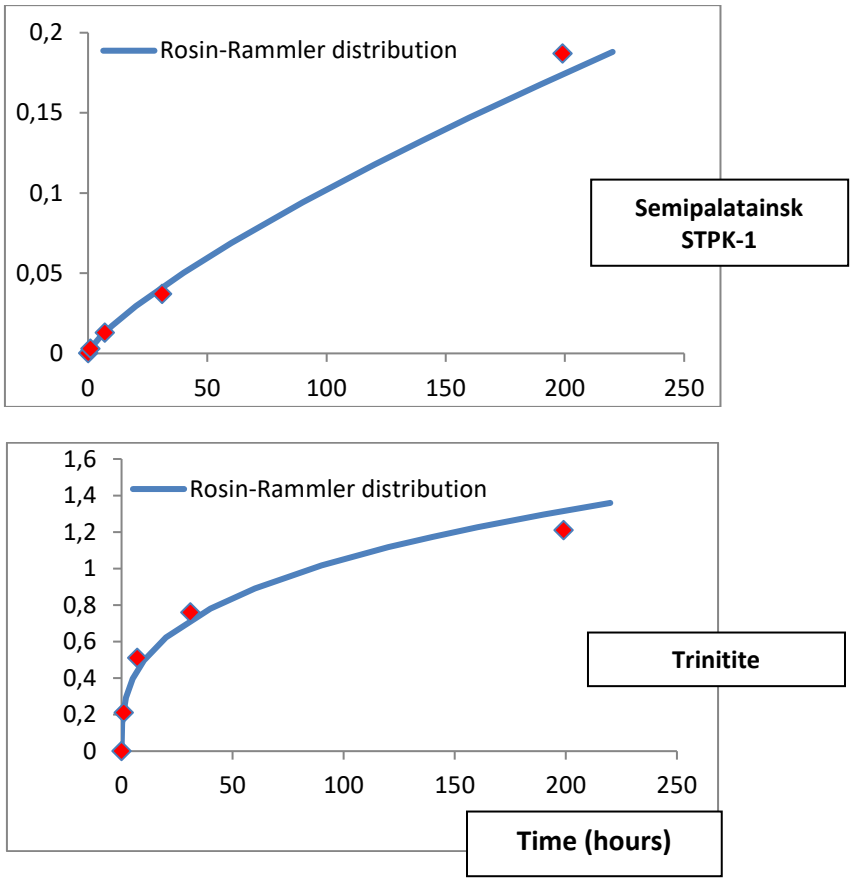


Figure 5.2.3.2.: % of cumulative Pu extracted from Semipalatinsk and Trinity radioactive particles with 0.16 M extractant solutions

Similar experiments have been performed with isolated particles from contaminated soils collected in Thule (Greenland) and Palomares (Spain), but in this case using rainwater. The cumulative solubilized Pu from these particles after one week of leaching did not reach in any of the experiments, 1% of the Pu associated to the particles, confirming the reactive behaviour of the contamination released in the two accidents (Jimenez-Ramos et al.2008), and that since the initial deposition of the contamination in the sixties, no big changes have been produced inducing increments in transformation processes.

The results obtained in the leaching of Palomares and Thule contaminated soil samples ratify the conclusions obtained from the analysis of the isolated radioactive particles. Negligible amounts of Pu were put in dissolution in these leaching experiments, being more related the small percentage dissolved with dissolution of material that with the conversion of the Pu to ionic form. In addition, in these soil bulk samples competitive effects trying to capture the dissolved Pu should not be neglected. The typical cumulative desorption curves observed in the leaching over time of isolated particles (as the shown ones in the Figure 2) will follow the same structure but with lower percentages of dissolution for their associated soils due competitive re-adsorption effects.

On the other hand, contaminated soil samples collected in Palomares and Thule have been also submitted to in vivo biotic experiments with rumen cow at 39ºC, indicating the results that after 24h or leaching the amount of Pu found associated to the leachates were quite small in proportion to the present in the bulk soil. This fact confirms the refractory behaviour of the Palomares contamination even 50 years after the accident and after the consequent dispersion of contamination, in opposition to the observed in the leaching of soils affected by weapon fallout where the contamination is in more

bioavailable form, and high percentages of dissolution are obtained in biotic experiments with rumen cow.

Screening heterogeneities and evaluating particle uptake in biota

Through a joint collaboration between the University of Seville, CIEMAT and NMBU groups it has been evaluated and analysed the degree of contamination and possible uptake of radioactive particles by one of the main living species which can be found in the semi-dessertic environment affected by the Palomares accident: snails

A sampling campaign was performed in the spring 2015 where a good set of representative snails were collected in the affected area. After collection, all the snails were cleaned individually with deionised water in order to remove possible external contamination, being afterwards fed during some days in the laboratory with inert material in order to collect their faeces and provoke the removal of all the material present in its digestive system at the moment of collection. The snails after the cleaning of their digestive system were sacrificed in order to separate the body mass from the shell.

The whole-body tissue of each separated individual was introduced in a small vial and the whole-body ^{241}Am content in each snail was determined by high-resolution gamma-ray spectrometry. The ^{241}Am activities for the different snails analysed, contained in each body, are compiled in Table 5.2.3.1.

Table 5.2.3.1.: ^{241}Am activity content in snails collected in Palomares.

Identification Code	Whole-body ^{241}Am, mBq	Identification Code	Whole-body ^{241}Am, mBq
Snail 2	**6 ±1**	**Snail 13**	**18 ±1**
Snail 3	**7 ±1**	**Snail 14**	**132 ± 3**
Snail 4	**15 ±1**	**Snail 15**	**80 ± 2**
Snail 5	**14 ±2**	**Snail 16**	**2 ± 1**
Snail 6	**46 ± 3**	**Snail 17**	**68 ± 2**
Snail 7	**174 ±3**	**Snail 18**	**63 ± 2**
Snail 8	**89 ± 3**	**Snail 19**	**48 ± 2**
Snail 10	**10 ± 1**	**Snail 20**	**87 ± 3**
Snail 11	**2 ± 1**	**Snail 21**	**19 ± 2**
Snail 12	**2 ± 1**	**Snail 22**	**32 ± 2**

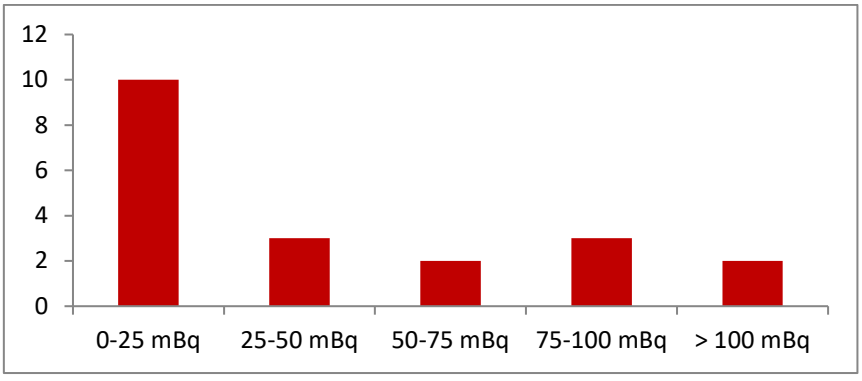


Figure 5.2.3.3.: Histogram showing the distribution of the snails analysed as a function of its ^{241}Am whole-body activity

The faeces collected before sacrifying the snails have been analysed for the determination of its ^{241}Am content by gamma-ray spectrometry. In these faeces the total collected mass and the associated determined ^{241}Am activity were respectively 1.20 grams and 4 ± 1 Bq.

An additional experiment with some of the snails collected in Palomares was performed after the determination of their ^{241}Am level of contamination. The snails 14, 15 and 18 were subjected to a dissection by a specialist in order to search for the internal distribution of the ^{241}Am in the different parts of their bodies.

Each snail body was divided in four different parts.

a) **ADI:** Initial part of the digestive apparatus (mouth, esophagus and stomach).
b) **ADF:** Final part of the digestive apparatus (intestines, anus).
c) **HP:** Hepatopancreas; digestive gland, that in gastropods provides the functions which in mammals are provided separately by the liver and the pancreas.
d) **R** Rest of the animal, formed mostly by muscles

The results obtained in the dissection experiments are compiled in the following table:

Table 5.2.3.2.: ^{241}Am internal distribution in snails collected in Palomares

	^{241}Am, mBq	**% of whole-body activity**
Snail 14		
ADI	**14 ± 1**	**10**
ADF	**68 ± 4**	**47**
HP	**41 ± 2**	**28**
R	**21 ± 4**	**15**
Snail 15		
ADI	**8 ± 1**	**14**
ADF	**8 ± 1**	**14**
HP	**36 ± 3**	**64**
R	**4 ± 1**	**7**
Snail 18		
ADI	**12 ± 1**	**17**
ADF	**18 ± 2**	**27**
HP	**36 ± 4**	**52**
R	**3 ± 1**	**5**

The snails have in their feeding costumes the ingestion of soil to extract particularly some essential constituents for the formation and growing of their shells. It is then also interesting to analyze and to determine the levels of radionuclide contamination that can be found in the shells of some of the snails collected in Palomares, having in consideration that in addition to the possible incorporation of ^{241}Am through the internal route in the snail followed by the material generating the shells, some small radioactive particles can be adhered to their surfaces. A total of eight selected shells from the snails described in Table 5.2.3.3, were measured independently by high-resolution with gamma-ray spectrometry to determine their ^{241}Am levels. The obtained results are the following

Table 5.2.3.3: ^{241}Am levels determined in shells from snails collected in Palomares

Snail	^{241}Am, mBq, in the shell
Snail 2	**4 ± 2**
Snail 4	**20 ± 2**
Snail 5	**28 ± 2**
Snail 6	**24 ± 1**
Snail 7	**400 ± 21**
Snail 8	**28 ± 2**
Snail 13	**2 ± 1**
Snail 18	**25 ± 3**

In addition to the studies shown concerning snails, the actinide transfer from the Palomares soil contaminated area to higher levels of the trophic chain has started to be analysed, taking rabbits as the representative mammal living in the area. In one of the visits performed to the Palomares area along this project it was observed that some of the small hills located in the more contaminated remaining areas were perforated by several dens, while accumulated rabbit faeces were found in some specific points.

Some of these faeces were collected, submitted to a detailed visual screening to remove possible traces of vegetation and soil particles, and finally measured (dividing the bulk sample in different aliquots) by gamma-ray spectrometry with an ultralow-background system equipped with a well detector. In all the aliquots analysed a clear signal of the gamma-peak of ^{241}Am was observed. Faeces samples with total masses ranging in the interval 3 to 5 grams were collected in different zones of the affected area, being obtained the following results

Table 5.2.3.4.: ^{241}Am activity concentrations, in Bq/g, found in faeces rabbit samples collected in different contaminated areas from Palomares.

Sample Code	^{241}Am, Bq/g
EC-02-01 (zone 2)	**0.93 ± 0.04**
EC-02-02 (zone 2, agricultural .land)	**0.08 ± 0.01**
EC-02B-01 (zone 2B)	**0.08 ± 0.01**
EC-03-01 (zone 3)	**1.93 ± 0.04**
EC-06-01 (zone 6)	**0.08 ± 0.01**

On the other hand, studies analyzing the degree of contamination and possible uptake of radioactive particles by living species in the area affected by the well-known Thule accident (Dahlgaard et al., 2004, Eriksson et al., 2008) have being also performed in the RATE project, by the DTU partnership. In particular, this partner is concentrating the efforts in the evaluation of the levels of actinides existing in faeces generated by Muskox specimens and in searching for the presence of heterogeneities and hot-particles in them. The muskox is an Artic mammal and is the last member of a line of ovibovines that first evolved in temperate regions of Asia and adapted to a cold tundra environment late in its evolutionary history.

The levels of ^{241}Am have been determined by gamma-ray spectrometry in several decens of muskox faeces samples, being compiled all the obtained results in the Figure 5.2.3.4 (levels of ^{210}Pb determined in the same set of samples are also shown for comparison purposes).

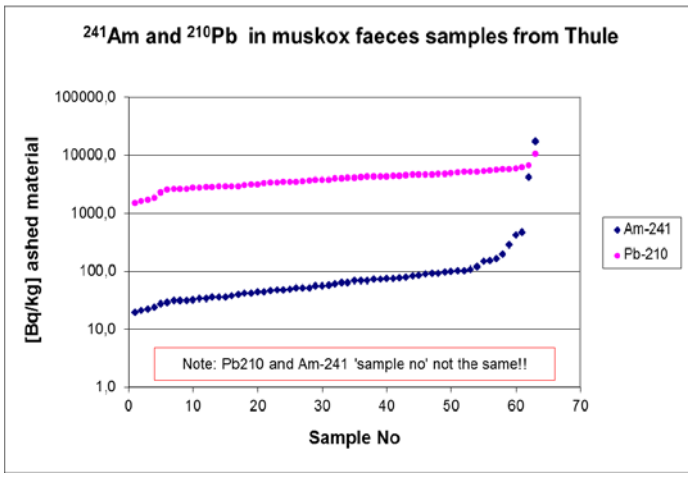


Figure 5.2.3.4.: ^{241}Am and ^{210}Pb activity concentrations (Bq/kg) in muskox faeces samples from Thule

All the analysed muskox faces samples included in this study show levels of ^{241}Am over the limit of detection, but in most cases the levels are quite moderate, under 100 Bq/kg. In a quite simple trophic chain as the existing one in the artic environment, there is some transfer of actinide to the Muskox specimens. In a limited number of the aliquots analysed, the levels of ^{241}Am found are clearly higher than the average. In a couple of them the ^{241}Am activity concentrations are clearly higher than 1000 Bq/Kg, being potential candidates to contain a fraction of its contamination in particulate form.

On the other hand, and in spite of the obvious continuous oral intake, body parts (liver, kidney, bones, lungs, spleen and meat) from a single muskoxen showed no detectable ^{241}Am with detection limits below 2 Bq/kg . Addditional studies about internal Pu are still under development.

Finally, and despite the high potential for accumulation and the potential of radioactive exposure, very few studies looking for the retention of radioactive particles in filter-feeders have been performed until now. As far as we know, and worldwide, only these type of studies have been and are being done by one RATE partner, NMBU, The main results and conclusions obtained until now in this particular subject are described in the following paragraphs. The work developed until now has been mostly laboratory work, although some results about the presence of radioactive heterogeneities in mussels collected on the deck of one submarine dumped in Artic waters has also been performed.

The Russian nuclear submarine K-27 was launched in April 1962. Powered by two liquid metal (Pb-Bi) cooled reactors, remained operational until 1968 when suffers a reactor event damaging approximately 20% of the portside reactor's fuel. After attempts to repair the submarine, a decision was made to decommission it and the submarine was ultimately scuttled in September 1981 in the shallow waters of Stepovogo Fjord, Nova Zelmya, at an estimated depth of 30 m. More than three decades later, the K-27 has become a topic of discussion as part of plans for remediation of the Arctic Seas, and some monitoring surveillance programs are periodically performed in the surroundings to control the absence of radioactive leakages.

In one monitoring program carried out in 2012, some mussels as well as some sediment material were collected on the deck and in the vicinity of the dumped submarine (Gwynn et al., 2016). The soft tissues of the mussels and the sediment material were recently screened looking for possible radioactive

heterogeneities by obtaining digital autoradiograms. In both set of autoradiograms (mussels and sediments), some signals indicating the presence of few radioactive particles were observed. The origin and composition of these particles are not known, although it can be indicated that the analysis by ultra-low background gamma-ray spectrometry indicates that all common fission and activation gamma-emitters were below the limit of detection in both materials. Further analyses are needed, although a first conclusion confirming the possible retention of radioactive particles by filter-feeders can be highlighted.

With these obtained results in mind, a detailed laboratory experiment was planned and executed in order to demonstrate that radioactive particles of variable size can be retained in a selected filter-feeder specie (blue mussels) under laboratory conditions, and that retained radioactive particles may induce adverse effects if the contact dose is sufficiently high. In these laboratory experiments, spent fuel particles released to the environment associated to accidents in the nuclear reprocessing plant of Dounreay and containing elevated amounts of ^{137}Cs and ^{90}Sr, were introduced into mussels in suspension with plankton food or through implantation in the extrapallatial cavity (Jaesckhe et al., 2015).

For the introduction of radioactive Dounreay particles to the mussel via pipette with 0.5 ml of plankton food, the mussel valves were opened, in air, using a small tool, and the particle, with the plankton food, placed inside the mussel. Once inside the mussel it was not known whether the particle would move, how quickly or to where. In this way, associate with food a total of 16 particles were introduced. From them, a total number of 10 particles were expelled (taken precautions to avoid losses or simple ejections due to gravity) while 6 particles were retained for 70h.

Of the 6 particles retained, a total of 5 were found “loose” within the mussel, i.e found in the siphon and gills, with the notable exception of one particle that was ingested and found in the stomach. It was not determined if the 5 particles found “loose” within the mussel have been caught by gills and bound within mucus, but it appears likely if the experiment has continued such particles would be collected by the mussel and ejected without a short timeframe.

On the other hand it was not found for the 10 expelled radioactive particles any correlation with size. The rejection of these radioactive particles could probably be more related with the absence of organic content in them and with their high density values.

Due to the high rate of loss of particles introduced via fed from the mussel, often within 24 h from introduction, a second series of experiments were performed where the particle was implanted into the mussel tissues Introduction in the extrapallial cavity is far less likely to occur than introduction via food but not impossible, as evidenced by the presence of pearls. Pearl formation could occur if a filtered particle were to become embedded into the mantle or trapped in the extrapallial cavity. In such events the inclusion of a radioactive particle into a pearl or into the shell would make the incorporation permanent with all the consequent implications in dose assessments.

We can then conclude indicating that the mussels actively collected the introduced particles and ejected them from the body as mussels are capable of selecting filtered particles, rejecting and expelling undesirable or unpalatable particles from the body (Jaesckhe et al., 2015).

The rate of retention of radioactive particles that may be encountered in blue mussels when filtering seawater is low. Even within relatively short time of few days, an introduced particle may rapidly be lost or ejected from the body. Present observations suggest that mussels are capable of actively detecting and removing such particles, although further research is needed in order to obtain a proper frequency distribution pattern of particle uptake.

The implantation of particles generates localized doses, situation totally different that the attributable when homogeneous distribution of radionuclides occur. Consequently, current methods used for risk assessments are inadequate for radioactive particles exposures. Knowledge is lacking about the ecological implications of radioactive particles released.

5.2.4 Discussion

Linking particle characteristics to specific source and release scenarios

Task: *Produce a particle database from archived nano- and micrometre sized radioactive particles released from different sources, varying with respect to composition, particle size, crystalline structure and oxidation states.*

A major fraction of refractory radionuclides such as Uranium (U) and Plutonium (Pu) released to the environment from the nuclear weapon and fuel cycles is present as particles ranging from sub microns to fragments. Such particles can carry a substantial amount of radioactivity and associated metals, and can act as point sources.

The radioactive particles that historically can be found or have been released to the environment can be classified in the following categories (IAEA, 2011).

1. NORM-particles related to front end of nuclear weapon and fuel cycles (e.g. U-mining)
2. NORM-particles related to non-nuclear industry
3. NORM-particles related to U and Th rich minerals.
4. Fuel particles, or particles of fissile material formed by mechanical disruption of the original bulk of radioactive material, ranging in size from submicrons to fragments (U/Pu particles)
5. Condensed particles or molecular aggregates and particles upon condensation of volatile radionuclides
6. Discrete radioactive particles or clusters formed within the fuel during normal operations or during the release (Ru-particles, activation products).
7. Colloids or pseudocolloids either released directly or formed in the environment (1 nm-0.45 µm)

In the frame of the RATE project, the research has been focused on radioactive-particles characteristics, weathering rates, remobilization and prediction of ecosystem transfer of radionuclides associated with U and/or Pu containing particles originating from selected key sources (nuclear weapon tests, safety tests, nuclear reactor accidents, NORM....)(Salbu, 2001), and with this end, a deliverable associated to this research has been the creation of a database with the information about identified and isolated hot-particles that are or has been available for performing the transformation studies: biotic and abiotic experiments. The detailed description of this database was covered by the COMET deliverable 3.4

The database compiled is formed by particles available by the different RATE partners to perform transformation studies. These particles can be classified in the following groups attending to its origin and/or way of generation.

a) Radioactive particles originating from Nuclear Weapon Tests

b) Radioactive particles from accidental or provoked conventional detonation tests

c) Radioactive particles originating from Reactor Accidents

d) Radioactive particles from accidents in Nuclear Reprocessing plants

e) Radioactive particles collected in NORM sites

f) Radioactive particles generated in TENORM industrial activities

A general description of the particles forming part of each of the mentioned groups is performed in the following paragraphs. Detailed information of the particles forming part of the database can be found in Deliverable 3.4.

Radioactive particles originating from Nuclear Weapon Tests

Radioactive particles with origin in the nuclear weapon test sites of Semipalatinsk (Kazakhstan), New Mexico (USA) and Maralinga, Emu and Montebello (Australia) form part of the RATE database.

A total of 3 isolated particles from Semipalatinsk form part of the database: Two of them were collected in the area called "ground-zero" where high levels of contamination remains due to local fallout associated to the atmospheric tests, while the third one was separated from a bulk soil collected around the crater called "Telkem´2" in an area identified as containing high concentration of transuranic nuclides.

The particles isolated from Ground-Zero are representative of the found ones in this area. They are glassy, and vitrified due to the high temperature release scenario and in general contain rather low gamma-activities (Lind, 2006). In addition to transuranic, some fission gamma- emitters of relatively long half-life as ^{137}Cs, ^{60}Co and ^{152}Eu can be detected and quantified. The radioactive particle from Telkem´2 that form part of the database is also representative of the collected and analysed in the area. The Telkem´2 particles appeared to be large grains of soils where U and Pu are distributed at concentrations lower enough to prevent detection of these elements with microanalytical techniques such as SEM-EDX and PIXE (Conway et al., 2007).

A couple of small fragments are also available, and form part of the database, with origin in the 20kT Trinity nuclear weapon test (ground surface shot) carried out in July 1945 near Alamogordo (New Mexico, USA). These fragments (called colloquially as Trinitites) are formed by the melting of products from the weapon with soil.

In addition, a total of 8 particles collected at different test sites in Australia form part in the database. From 1952 to 1957, the British nuclear weapons testing program conducted 12 detonation tests, called "Major trials" in Australia. These tests caused radioactive contamination with nuclear weapons debris, including Pu into the surrounding environment. Most of the contamination was present in particulate form with areas presenting very high number of particles per surface area (Child at al., 2013). The eight isolated particles (are relatively large with dimensions in the range 200-800 □m, and their activities in ^{241}Am are in the range 0.5 – 5 Bq.

Radioactive particles from accidental or provoked conventional detonation tests

In this section of the database are grouped isolated Pu/U particles with two main different origins but generated/formed in the same way. This group is formed by particles dispersed by accidental explosion/burning of nuclear weapons in aircraft accidents, as well as by Pu particles generated on conventional explosive/burning (i.e. non-nuclear) high-explosive dispersal events performed at test sites. Particles from Palomares, Thule and Taranaki/Maralinga form this group.

Uranium and plutonium containing particles dispersed in the terrestrial environment of Palomares, Spain, following the accident in 1966 of a B52 bomber are available, provided by CIEMAT. The Palomares particles were isolated mainly from soils collected in the main different zones (the so-called zones 2 and 3) contaminated each one by the conventional explosion of one of the weapons affected.

Following a similar B52 accident in 1968 at Thule (Greenland), and the dispersion in particulate form on the sea and on the land of the material forming part of the nuclear weapons involved, a lot of expeditions and studies have been carried out in the area, mostly by one of the partners (DTU). This

partner has previously isolated and characterized hot particles from the affected area (Eriksson, 2002) and has made available for the transformation studies performed in RATE, selected hot particles mainly from terrestrial origin.

The number of particles isolated from the Palomares and Thule scenarios and included in the database is elevated due to the special interest in the project to perform comparative evaluations of transformation biotic and abiotic experiments, because both cases are characterized by similar source materials and similar formation mechanisms, but the released particles have been residing for 50 years in very different environmental compartments. In both cases thermonuclear bombs crashed after fire in the plane. In both cases the bombs detonated conventionally with a subsequent explosive fire, dispersing micrometre particles where enriched U and weapon grade Pu coexist, but in a non-homogeneous mixture (surface inhomogeneities in both Palomares and Thule particles have been observed (Lind et al., 2007)). In both cases the particles are characterized by low $^{239}Pu/^{235}U$ (0.62-0.78) and $^{240}Pu/^{239}Pu$ (< 0.07) atom ratios, and in both cases the particles have similar morphology (are particles with high porosity and very fragile). The particle matrices in both cases (Palomares and Thule) are U and Pu oxide mixtures (the original metallic U and Pu in the bombs was oxidized in the explosive fires) and their persistence during 5 decades is an indication of its in general inert behaviour under the existing conditions. The evaluation of this inert behaviour and the confirmation of possible increments in their solubility over time is one of the objectives to be analysed on the frame of RATE project

The set of Palomares and Thule particles are complemented with two Pu particles isolated from soils collected at Taranaki site, Maralinga (Australia) where 12 non-nuclear explosion tests ("Minor trials") were performed at the beginning of the 1960´s. These tests dispersed more than 22 kg of Pu, resulting in four radioactively contaminated deposition plumes radiating from the test site (Johansen et al., 2014) in a similar release mechanism to Palomares and Thule release scenarios.

Radioactive particles originating from reactor accidents

It is a well-known fact that dispersion in the environment of radioactive particles occurred as a result of the Chernobyl accident (Kaspharov et al., 2003). In fact, and for several years, the presence of radioactive particles was referred to as a peculiarity of the Chernobyl accident, without to have in consideration the formation of particles in previous source and released scenarios (weapon tests, releases of nuclear reprocessing plants…).

The Chernobyl Nuclear Power Plant (ChNPP) accident took place on April 26, 1986. As a consequence of the initial explosion, mechanical destruction of the UO_2 fuel occurred under high pressure and high temperatures, and deposition of fuel particles took place to the west of the reactor. During the subsequent fire, volatile fission products and U fuel particles were released under moderate temperatures and oxidising conditions, and deposition of particles occurred to the north, northeast and south of the plant (Kashparov et al., 1999). As a result, about 3-4 tons of U fuel with variable burn-up were released into the atmosphere, either as fuel particles, as particles containing construction material or as condensation particles (Salbu et al., 1994; Salbu 2001; Kashparov et al., 2003). Radionuclides such as ^{95}Zr, ^{95}Nb, ^{99}Mo, $^{141,144}Ce$, $^{154,155}Eu$, $^{237,239}Np$, $^{238\text{-}242}Pu$, $^{241,243}Am$, $^{242,244}Cm$ were released from the accidental unit only in the form of fuel particle matrix, while more than 90 % of $^{89,90}Sr$, $^{103,106}Ru$ and ^{140}Ba were also released as fuel particles.

In the database several Chernobyl particles provided by NMBU have been included.

Radioactive particles from NORM/TENORM sites

The presence of radioactive particles in the environment has been generally associated to their releases from nuclear sources including nuclear weapon tests, accidents with nuclear weapons, reactor accidents and accidents from reprocessing plants, as it has been shown in this report. Radioactive particles existing in the environment from NORM (Naturally Occurring Radioactive Materials) sources, with few exceptions, have been much less in focus, although the failure to recognize the presence of

these particles in an ecosystem can have a number of serious consequences with independence of the nuclear origin or not of these particles. The possible sources of NORM particles are huge around the world, being present either in unperturbed areas enriched in radionuclides of Uranium and/or Thorium series or in contaminated areas by the releases of wastes generated in conventional industries that treat NORM raw materials (these wastes are commonly known as TENORM: Technologically Enhanced Naturally Occurring Materials).

On the frame of the RATE project, were available for the transformation studies a good number of particles from different NORM/TENORM sites. In particular:

a) The database is enriched by a couple of particles isolated from an alum shale formation in Norway. The alum shale contains enriched levels of Uranium with their daughters in secular equilibrium, being in the alum shale formation the U levels quite heterogeneous due to the presence of U inclusions in some particles or fragments (Skipperud et al., 2016). The two alum shale particles have been provided by NMBU, isolated from the ore material extracted in the process of construction of a tunnel (RV4) at Gran, Hadelend, Norway.

b) Ten particles have been isolated, and were available for the transformation studies, from soils collected in an undisturbed naturally Th-rich site. This site is located in south of Norway, called Fen/Sove, forming part of a complex of alkaline rocks and carbonates enriched in rare earths, niobium and thorium (Popic et al., 2014). As soil radionuclide concentrations are determined by those in the parents materials and by physical and chemical phenomena, the Fen/Sove soils were shown to have significantly elevated ^{232}Th and moderate ^{238}U concentrations.

c) A couple of particles have been also provided by NMBU from U mining areas of Central Asia (Lind et al., 2011, Lind et al., 2013). Following the cold war, extensive U mining and production took place at selected sites in Central Asia as a vital part of the nuclear weapon program in the former Soviet Union. In these areas all types of Uranium recovery were undertaken: open-cast extractions, underground mining, in-situ leaching with sulphuric acid solutions, etc. were used. The two particles were particularly isolated from soils collected at the former U site at Kadji Sai, Kyrgyzstan after their identification and characterization using micronalytical techniques, searching for information on the solid state speciation of uranium and other metals which can improve the basis for assessing environmental impact of TENORM activities.

In addition, several U particles were isolated along the production process of a phosphoric acid plant located in Huelva, South West of Spain and provided to the database by USEV. In this paradigmatic NORM industrial activity are treated phosphate rock minerals enriched in uranium with their daughters in secular equilibrium. The particles included in the database were isolated from waste material accumulated/generated in the digestion and filtration units of the plants (Bolivar et al., 2009), and should be reflecting inclusions of U in the original raw mineral that due to its refractory behaviour are not dissolved in the production processes (García-Tenorio et al., 2015). These U particles have a well defined crystallization structure with sizes variable but always at the micrometer level.

Abiotic and Biotic Transformation Processes

After their release to the environment, the radioactive particles represent point sources of potential radiological significance to both human and non-human biota. These radioactive particles represent a significant external and internal radiation risk, being important to highlight that there is nowadays a high degree of uncertainty in modelling their re-suspension, transport, deposition as well as in the knowledge about their potential leaching (with all the associated consequences in determining the dynamics of radionuclides between different environmental compartments).

As it has been indicated previously, weathering rates of particles will depend on the particle composition, structural changes occurring during the event, and on local chemical conditions occurring after deposition, given that dynamics of radionuclide transfer of particle-associated radionuclides are

often irregular in nature compared with areas where there are no radioactive particles present. It is possible then to conclude that for particle contaminated areas, the overall uncertainty in assessing the health and ecological impacts of radionuclide releases may be unacceptable to provide accurate and reliable dose assessments high.

With the clear objective to decrease the overall uncertainty in assessing the health and ecological impacts of radionuclide releases in particle contaminated areas *the first objective in this frame inside the RATE project has been to develop and define protocols for leaching of radioactive particles, to use and apply them afterwards to develop information on the solubilisation, remobilization and ecosystem transfer potential of particle associated radionuclides*.

The intention is to identify key parameters affecting properties of anthropogenic and NORM containing particles (either when changing water qualities, temperature and utilizing abiotic leaching agents or when they are exposed to biological systems such as microorganisms influencing redox systems or rumen liquids from grazing animals) as a way to determine transformation kinetics (weathering rates, time dependent Kds….).

Some previous leaching studies (mostly abiotic) have been done in the past, showing that the degree of solubilisation is very much dependent on the source of material and nature of environmental breakdown scenarios analysed. Only as an example we show in Figure 5.2.4.1 the leaching behaviour of several particles treated with HCl 0.16 M mimicking the pH of gastrointestinal fluids. The percentage of solubilisation goes from a few to nearly a hundred per cent, being evident that if no knowledge exists about the solubilisation behaviour of the analysed particle, the uncertainty in the evaluation of their radiological and environmental impact is extremely high.

In spite of the mentioned facts, it is evident that only fragmented and no directly comparable efforts have been done in the past in relation with the leaching studies being applied. In most cases “local” laboratory protocols have been used that are difficult to replicate, or in some cases avoid the possibility of results intercomparisons.

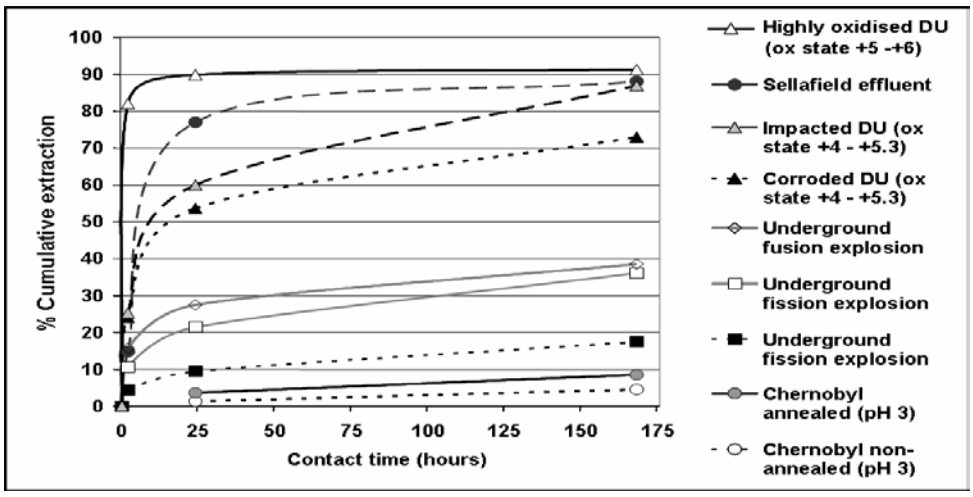


Figure 5.2.4.1.: Leaching experiments with 0.16 M HCl solution performed by CERAD/NMBU (Lind, 2006)

In order to avoid and overcome similar problems, and with the end to generate values directly comparable and reproducible between laboratories, special efforts have been devoted to define joint, scientifically agreed protocols for abiotic and biotic transformation studies between the different RATE partners. One of the main outputs of the project are these protocols which have been detailed in the methods and materials section.

With basis in the protocols defined by consensus, leaching experiments in which radioactive test particles have been exposed a) to abiotic reagents such as rain water, sea water, irrigation water and

synthetic solutions mimicking body fluid such as simulated stomach fluid, and b) to biological systems such as rumen liquids from grazing animals, have been performed associated to this project by the different partners. In addition, and in parallel to the transformation experiments applied on single radioactive particles (RP), similar experiments (with also a well-defined and specific protocol) has been applied on aliquots from bulk samples from which the particles were isolated.

Along the project, leaching experiments have been applied to particles and soil aliquots from quite different origin, although it can be classified mostly in three different important groups:

- Pu/U particles and contaminated soils corresponding to locations affected by accidental weapon accidents (Palomares, Spain and Thule, Greenland) or nuclear weapon tests without fission yield (Taranaki, Australia).
- Pu/U particles and contaminated soils collected in areas where atmospheric weapon tests were performed (Semipalatinsk, Kazhajistan ; Trinity, United States ; Maralinga and Montebello, Australia).
- Th enriched Particles and soils from a High Background natural Radioactivity Area (HBNR) located in Scandinavia (Fen, Norway)

In the results section, the total dissolved fraction from leaching with 0.16 M HCl of two soils with origin in Palomares and Thule, respectively, were shown in Figure 1.

From the results it was possible to deduce that only a quite small proportion of the Pu existing originally in the Palomares sample is dissolved, indicating that under these abiotic conditions the particle has a refractory behaviour. This behaviour is on the other hand in qualitative terms in agreement with the known fact that in the remaining contaminated area by the Palomares accident the transfer and remobilization of the actinide contamination to vegetation, plants and water is quite limited as is reflected in the historical database of monitoring analysis carried out in the area by CIEMAT. In this sense, the average value of the plutonium solubility in water in five soil samples taken in 1986 was 0.01%, while the solubility increase in one to two orders of magnitude in identical water solubility studies performed with soils taken in 1999 and 2000, but still continuing in very limited values.

The obtained results are also in qualitative agreement with the results obtained by the University of Seville by performing bioavailability laboratory experiments based on the use of single extractants on contaminated Palomares soil. In particular, and in order to make an empirical and preliminary assessment of the Palomares Pu availability by plant uptake, and from among the great variety of single chemical extractants which can be found in the literature which serve for this purpose, 1 M $MgCl_2$ was selected as the most suitable extractant. The extraction method was applied to a set of 6 Palomares soil aliquots with a different degree of Pu contamination, where in all the cases the available fraction lower than 7% of the total Pu associated to the bulk material. The contamination levels in the analysed soil aliquots cover a range as ample as 4 to 200 Bq/kg.

It is interesting also to remark the different behaviour of the soils to the same leaching agent and under the same experimental conditions of the Palomares and Thule soil aliquots, with a major weathering observed for the Thule sample (Figure 5.2.4.2). But in both accidents, similar B52 aircrafts carrying thermonuclear bombs were involved and similar weapons dropped from the planes with the subsequent conventional detonation and explosive fire. And in both cases the disseminated contamination is mostly in particle form, with particle composition coexisting of both Uranium and weapon grade Plutonium. In fact, determinations performed in particles from both sites by ICP-MS, and/or AMS show low $^{239}Pu/^{235}U$ (0.62-0.78) and low $^{240}Pu/^{239}Pu$ ratios.

In addition, the characterization of the Thule and Palomares particles has shown that they have quite similar morphology (scanning electron microscopy), they present similar oxidation states (very similar XANES profiles) and in both cases they present a clear peculiarity: the mixture of enriched uranium and the weapon grade is non homogeneous (inclusions).

The different leaching behaviour observed could be associated then mostly to physical characteristics: in particular to the fragility observed in the radioactive particles isolated from both places, that can be more significant in Thule due to the influence of the more variable and hard ecosystem conditions.

The leaching values determined with radioactive particles from Semipalatinsk and Alamogordo and also shown in the section devoted to results (Figure 5.2.2) cannot be considered surprising given the physical structure of the analysed particles. They correspond to fused material with a glassy structure where, as it was reflected for similar particles in the characterization database, the actinide elements are present as small inclusions embedded in the glassy structure. The data obtained gives indication that the extracting agent most probably do not interact with the mentioned inclusions as the glassy surface acts as a protective cover preventing the solubilisation of the imbedded plutonium. On the other hand, special mention is deserved by the results obtained in the same experiment for the $^{241}Am/^{239+240}Pu$ activity ratios. As it can be observed in the Figure 5.2.4.2, corresponding to one of the Semiplatinsk particles, this activity ratio is not constant over time, with higher relative solubilization values at the beginning of the experiment for ^{241}Am in relation to the plutonium. These results are indicating that in spite of the low solubilization found for both elements, a proportion of ^{241}Am is in more available form than $^{239+240}Pu$.

The decrease over time of the mentioned ratio is likely to reach the activity ratio value determined in the bulk particle after their destructive analysis. The $^{238}Pu/^{239+240}Pu$ activity ratio determined from the cumulative amounts extracted of these elements along the leaching experiment is also in good agreement with the determined one in the bulk particle.

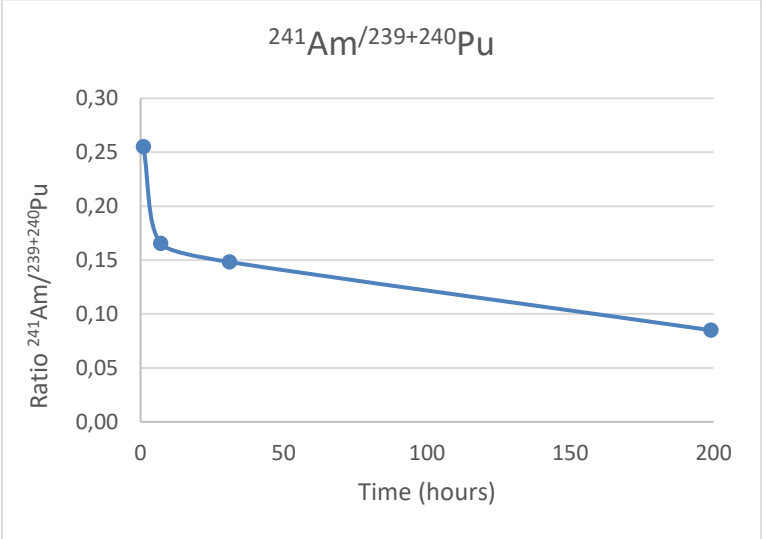


Figure 5.2.4.2.: $^{241}Am/^{239+240}Pu$ activity ratio in a leaching experiment with 0.16M HCl applied to a hot-particle from Semipalatinsk

The results obtained particularly in the analysis of these weapon test particles show clearly how essential and necessary is the performance of these type of experiments to avoid clear bias and overestimations in ecosystem transfer of actinides in these areas if conventional and non-specific transfer factor parameters are adopted. In particle contaminated areas, their leaching and weathering rate behaviour is very much dependent of their origin, composition and chemical and physical characteristics.

The need of specific data for a proper modelling of actinide ecosystem transfer in particulate contaminated areas is high given the strong dependence of the particle behaviour on its origin and of physic chemical parameters. This also suggest the necessity to develop independent experiments in

small defined and different areas inside broader contamination regions affected historically by several nuclear events. This is the case for example of the Semipalatinsk region where hundreds of different nuclear tests with different objectives were performed. The great majority of conducted tests disseminated particles along the region, but for example the particles generated in the Ground Zero region by atmospheric weapon tests are not comparable with the particles formed in the excavation experiments generating the Telkem craters. These different origins and the different physico-chemical compositions of the relevant particles can be reflected in total different lixiviation behaviour, that should be taken in consideration for a proper radioecological evaluation.

We will finish indicating that biotic leaching experiments using cow rumen have been also applied to different aliquots of a soil sample collected in the High Background Natural Radioactivity Area of Fen (Norway), enriched in Th. Concordant results from the five different aliquots treated have been obtained, being extracted about 200 ng of Th with rumen extractant from 1 gram of treated aliquot. That means that the Th weathering rate from the soils with the used gastric juice is extremely low, a fact that has been also observed when other aliquots from the same soil were treated with rainwater by applying the defined abiotic protocol.

In conclusion, more than 50 abiotic and abiotic experiments, applying the defined protocols, have been performed along the RATE project. Quite detailed and rich information can be obtained from the hundreds of samples generated in the experiment that are now undergoing analysis to obtain the maximum information for publication. But as a whole, the more interesting and general conclusions that have been inferred from the performed experiments are the following:

a) It has been confirmed that the characteristics and magnitude of the transformation processes are clearly site dependent, with the transfer parameters generated and their time evolution varying several orders of magnitude depending of the origin of the radioactive particle analysed. The weathering rates observed are clearly source related, although in general for the studies performed with particles resulting from close-in fallout around weapon tests and/or in conventional detonation of nuclear weapons, the lixiviation observed are clearly smaller that the observed historically in ecosystems affected by weapon tests fallout depositions in ionic form. This fact indicates a refractory tendency of the Pu/U released particles in these type of events.

b) In broad areas affected historically by a great number of different nuclear events, the transformation processes affecting particles across the site can be extremely different attending to the local characteristics of the event which has generated the particle under analysis. In contaminated places such as Semipalatinsk the experimental behaviour of particles collected for example from the ground zero area where a lot of atmospheric nuclear tests were carried out, are totally different that the behaviour observed for particles collected in the Telkem craters formed in excavation tests. In some areas affected by deposition of radioactive particles of different origin quite local specific studies should be done in order to perform proper radioecological evaluations.

c) In areas affected by similar sources of U/Pu particles, the results obtained in the transformation processes can be also dependent upon the affected ecosystem characteristics. Some of the results obtained for example with Thule and Palomares particles, are inducing this comment. In both cases the source and the characteristics of the accident producing the dissemination of the radioactive particles are similar, the morphological and structural properties of the particles are similar but differences exist in the climatic conditions and ecosystem characteristics (Artic vs semidessertic ecosystems). These release scenario dependence, however should be confirmed by additional studies

d) The transformation processes analyses performed with NORM particles and associated soils, reaffirm the need to perform specific studies in each contaminated area. NORM particles, found naturally in High Background Natural Radioactivity Areas or released to the environment forming part of waste releases have a behaviour very much dependent of its chemical form that should be analysed at local level, for each source, if a proper radioecological evaluation is desired. The analysis of

radionuclide transfer from NORM particles to waters or plants by using transfer parameters derived from areas where the natural radionuclides can be in ionic or bioavailable form can induce to enormous bias and not proper description of the real processes occurring in the analysed ecosystem.

Screening heterogeneities and evaluating particle uptake in biota

Although is a well-known fact that a significant fraction of refractory radionuclides released to the environment is present as radioactive particles , there has been historically a clear tendency to perform simplistic conservative dose assessment in the particle contaminated areas because there was a lack of knowledge about the behaviour of these radionuclide-bearing particles (García-Tenorio et al., 2016). In particular, the uptake of radionuclides by wildlife to assess ionizing radiation exposure, has been generally quantified using concentration ratios (CR) taken from databases constructed with basis in studies generally done in areas only affected by radionuclide homogeneous distributions mostly in ionic form (i.e. worldwide weapon fallout), not being recognized,

a) that radioactive particles contains a significant fraction of the bulk sample activity, leading to sample heterogeneities problems because in general are inhomogeneously distributed, and

b) that the ecosystem transfer will be in general delayed until weathering and remobilization of associated particle radionuclides occur.

The CR should be defined then as rate functions, avoiding the simplistic approach of its constancy. In other words, for terrestrial systems, the uptake of radionuclides by wildlife, the soil to animal transfer, is typically quantified by using the concentration ratios (CR) between the organism whole-body activity concentration (fresh mass) and the activity concentration in soil (dry mass). This parameter in particle contaminated areas will suffer from different sources of uncertainty:

a) the whole-body activity concentration can experiment very long variations depending of the possibility of uptake by the living organism, and through inhalation or ingestion, of a significant amount of the radionuclide under analysis associated to a particle. In fact, the radioactive particles represent point sources of potential long-term environmental and ecological significance, existing a risk from inhalation, dermal absorption, skin exposure and ingestion of particles. Some animals as filter-feeders (e.g. molluscs) and soil-dwelling animals (e.g. gastropods) can retain particles in their organism and eventually be ingested by man.

b) the contamination is typically distributed unevenly in the soils and is subject to change over time, being required for a thorough investigation of soil-to-wildlife transfer, a better understanding of how contaminants are distributed in soils, and how such distributions change over time. In addition, in the analysed cases affected by radioactive particles, the contamination is in general in less bioavailable form that in standard reference sites, dampen the effectiveness of radionuclide uptake such that the use of standard reference concentration ratios (CRs) would over-predict animal radionuclide body burden. Soils affected by nuclear weapons testing differs from soils affected by particle deposition in that they present a higher percentage of the radionuclide under analysis in water soluble and readily exchangeable form. And also differ in their rate of change in availability in the soil because the mobility of radionuclides from nuclear weapons testing decrease with time because of fixation by soil components, while in the particle contaminated areas, the availability increase with time due to weathering.

In this frame, and along the RATE project:

a) the phenomenon of retention or uptake of particle associated radionuclides in various biota from contaminated areas have been studied, centering the efforts in terrestrial areas affected by Pu/U releases to the environment in particulate form with origin in accidental or provoked conventional detonations of nuclear weapons: Maralinga (Australia), Palomares (Spain) and Thule (Greenland). Additionally, the retention capacities of radioactive particle by mussels have been analysed and evaluated through the performance of laboratory experiments.

b) an evaluation of the factors affecting the magnitude and uncertainty in the prediction of Pu/U uptake by selected biota in some areas contaminated by Pu/U particles have been performed.

The main conclusions are compiled in the following paragraphs.

Plutonium uptake by wildlife at the Maralinga legacy site, Australia

The experience of ANSTO, a partner of RATE, in the analysis of the uptake by wildlife of Pu in an area affected by the presence of Pu particles with origin in weapon tests performed in the 1960s, has been the ideal starting point of the studies carried out along the project in the mentioned subject. A detailed ANSTO database of CR for different animals in the Maralinga test site (Johansen et al., 2014) has served to emphasize and highlight some key points to be checked and analysed in other affected particle contaminated terrestrial ecosystems as the existing ones in Palomares (Spain) and Thule (Greenland).

The ANSTO database of soil-to animal transfers (as measured by CRs) for mammals, reptiles and arthropods in Taranaki was created 50 years after conventional detonations with basis a well defined sampling and field work strategy (Johansen et al., 2014). The CRs at Taranaki were calculated as the activity concentration in the organism related to that of the composite soil samples (n=5) of the forage-range. The use of multiple soil samples enables the distribution of the CR values over the forage range to be estimated and expressed in terms of geometric means.

For mammals, the CR values were calculated following standard approaches: the whole-organism was calculated from measurements on a range of tissues where Pu had accumulated via circulation system (muscles, bones, liver, kidneys, spleen, heart, lungs and blood). The GI tract was excluded for CR determination. For reptiles in the CR determinations the GI tract was excluded, but the skin was considered. For arthropods, calculations include the entire organism.

In mammals (rabbits) captured at Taranaki, the geometric mean (GM) of CR aligns well with the CR determined from similar species and conditions (such as Nevada Test site), but is lower than the CR-GM of the international mammal data reported in the wildlife transfer database (WTD). These lower values are likely due to the presence of a low-soluble, particulate form of the Pu in Maralinga soils. In fact, it was observed that the CR were not consistent among rabbits captured from various plots, but rather indicated a distinct spatial pattern which appears to relate to the physical form of the Pu contamination. In the plot with the highest Pu concentration in soil, it was found that the activity concentrations within rabbits were high but the CRs were the lowest. At plots with lower activity concentrations in soil the CR values increased approximately linearly by three orders of magnitude. The most likely explanation is the increased density of larger unrespirable low-solubility particles at areas with high levels of Pu contamination, i.e. closer to the test detonation locations. Such particles contribute to the soil concentration in the denominator of the CR, but make a limited contribution to the numerator as they are not readily respired or absorbed. This effect dampens the near-term effectiveness of Pu uptake such that the use of standard reference CR would over-predict animal Pu body burdens where such particles are present (Johansen et al., 2016a).

We can finally indicate that when uptake is compared over time in rabbits at Taranaki, a series of data from the 1970s-2012 indicated that no variation in Pu uptake rate has been observed confirming that the proportion of biologically available forms of Pu have persisted at the site over 30-year time scales.

The commented ANSTO database includes also information about Pu internal distribution in rabbits inhabiting Taranaki weapons test site, Maralinga, Australia (Johansen et al., 2016b). The Pu activity concentrations in the lung tissues were found elevated relative to the body average, indicating the presence of non absorbed Pu mostly incorporated by inhalation of Pu-soil respirable fraction, while high concentrations were also obtained in the Gastrointestinal (GI) relative to the absorbed within the body, fact that is consistent with the low absorption rates for Pu in mammals published by the International Commission on Radiological Protection (ICRP, 2012) which indicate that most ingested Pu is not absorbed across gut mucosal cell, but rather remains in the non-absorbed material becoming

progressively more concentrated as it moves through the intestines. In addition, the rabbits occasionally feed through coprophagic reingestion of soft faeces, which also contributes to concentration build-up as the material is recycled through the digestive system.

We can conclude from this initial section and as a basis for the studies performed at Palomares and Thule that in the Taranaki weapon test site, the uptake of Pu by wildlife appears to be regulated and highly influenced by Pu form as some Pu-containing particles are of not readily respirable, absorbable size. This peculiarity is also observed in the internal distribution of Pu in rabbits with higher Pu levels than expected in lungs and in the gastrointestinal tract due the presence of persistent non absorbed Pu in these organs. Although the Pu bound within larger particles is not easily absorbed by mammals in the near-term, its potential for uptake is shifted (or delayed) from present to future receptors when the particles remain in the environment subject to weathering.

Screening heterogeneities in snails collected at the Pu contaminated terrestrial site of Palomares.

Absolutely all the snails collected show the signal of the accident but in a quite variable level of contamination. Although all the snails were collected from the same contaminated area, (collection surface was not higher than 200 m x 200 m) differences in the levels of ^{241}Am of even two orders of magnitude have been observed. This high variability in the uptake by animals captured in a defined terrestrial area is generally not found when the radionuclide contamination is homogeneously distributed, with the bioavailable fraction in the soils being relatively uniform. The high variability found in our study is associated with the presence of most of the radionuclide contamination in the soils of zone 2 in particulate form, inhomogeneously distributed. The ^{241}Am levels in superficial soils collected randomly in the sub-zone where the snails were collected can differ even in two orders of magnitude.

The variability observed in the whole-body ^{241}Am levels of the snails, and the variability existing for the same radionuclide in the associated soils, make questionable the utility of use and determination of the ^{241}Am concentration ratio (CR) parameter for the area under analysis due to its enormous associated uncertainty. Being the ^{241}Am-CR defined as the ratio between the ^{241}Am activity concentration in the snail (in fresh weight) and the ^{241}Am activity concentration in soil (in dry weight), its value in zone 2 of Palomares will have an associated uncertainty of several orders of magnitude, in contrast with the narrow range of ^{241}Am-CR values compiled in the IAEA database for gastropods inhabiting in areas only affected by homogeneous weapon test fallout contamination (IAEA,2014). The use of CR values defined from experimental areas affected by weapon test fallout on the other hand will over-predict the uptake of ^{241}Am in the majority of the snails analysed from Palomares due to the low bioavailability of the radionuclide contamination but at the same time will not have in consideration that a random number of snails can have much higher ^{241}Am values due to the ingestion of a discrete radioactive particle (Lind et al., 2016)

The ^{241}Am levels found in the analysed Palomares snails on the other hand, can be evaluated as moderates with little evidences existing about the possibility of the presence of some micrometre highly radioactive particles retained in their digestive system. Consequently, if due to their feed habits, some hot-particles are ingested by these animals, it seems that in most cases these particles should flow through their digestive system and then would be excreted in a high proportion.

In order to obtain some evidences supporting this last hypothesis, and to gain additional information about the interaction of these animals with hot-particles disseminated in its living area, two different approaches were followed: In the first one, the faeces collected before sacrifying them have been analysed for the determination of its ^{241}Am content by gamma-ray spectrometry. The faeces results reflect the fact that associated to their feeding habits the snails are ingesting Pu contaminated soils by the Palomares accident, but with the particularity that most of the contamination should flow along their digestive system, being excreted. This conclusion is in agreement with previous results and conclusions obtained by Ciemat in the area (Aragón et al., 2006).

The second approach followed to confirm that most of the ^{241}Am ingested by the snails is finally excreted was the performance of a laboratory experiment, where initially non-^{241}Am contaminated individual snails were, under controlled conditions, fed with a mixture of Palomares soil plus fodder and the proportion of the ingested ^{241}Am found in the faeces determined.

A detailed description of the laboratory experiment is done in the following lines:

a) Two individuals of the specie *Otala Punctata* were collected for the feeding controlled experiment. One of them (snail α) to be fed by a mixture of Palomares contaminated soil and fodder, while the other (snail β) to be fed only with fodder.

b) The two snails were introduced in independent containers with ventilation and periodic humidification with pulverized water. The experiment was done at controlled temperature of 20-21ºC, and with a photoperiod of 12 hours of light per day.

c) The experiment had duration of 14 days. During the first two days of the experiment both snails (α and β) were fed only with fodder. After that, the snail α was fed with a mixture of Palomares plus fodder during 10 days while the snail β continued being fed only with fodder. In the last two days both snails were as at the beginning of the experiment fed only with fodder in order to assure that snail α had excreted all the Palomares soil ingested.

d) After the 14 days experiment, both snails were sacrificed and the separated soft tissues were independently measured by gamma-ray spectrometry looking for ^{241}Am determination. Obviously, the ^{241}Am content in the Palomares soil ingested by snail was determined at the beginning of the experiment.

e) Along the 14 days of the experiment the faeces generated were collected, forming at the end and for each snail a composite sample measured also by gamma-ray spectrometry.

The results of the laboratory experiment were quite conclusive. In the case of snail β, used as control sample, not ^{241}Am was detected in the fodder provided to the snail, in the whole-soft body after being sacrificed and in the faeces collected along the 14 days. In the case of the snail □, the following results were obtained.

^{241}Am activity in the Palomares soil fed by snail □ during the experiment: **213 mBq**

^{241}Am activity in the faeces of snail □, collected along the experiment: **177 mBq**

^{241}Am activity detected in the body of snail □ after being sacrificed: **not detectable**

Having in consideration that the measurements performed in the soil and the snail can be affected by an uncertainty of around 10%, it is clear to observe, a) the coherence of the results obtained from an activity balance point of view, and b) the association of the great majority of the radioactive contamination ingested by the snail to the faeces excreted in total agreement with the conclusions obtained previously by a different approach.

Some of the snails were dissectioned in order to analyze the internal distribution of the actinide contamination. The results obtained in the dissection experiments, which were compiled in the section 1.3 deserves the following comments:

a) The great majority of the ^{241}Am in the analysed snails have been found in their gastrointestinal system (GI), even knowing that the snails were sacrificed after assure that all the remains from material feeded in Palomares ecosystem were excreted. In the three cases analysed, 85-95% of the ^{241}Am is found in the GI, whit a minimal proportion in the tissues. These results indicate that the Plutonium (Americium) incorporated by ingestion by the snails is in a not bioavailable form, being not absorbed and, consequently, no distributed along the whole body.

b) Looking the distribution of the ^{241}Am in the different parts of the GI, not clear/definitive conclusions can be deduced because the number of samples analysed until now is limited, although some tendencies to be confirmed can be inferred. The results show that the proportion of americium in the initial part of the digestive system is clearly lower than the found one in the intestinal system and particularly the hepatopancreas. The proportion of americium in this last glandule is clearly higher that the corresponding one according to its weight if the americium were homogeneously distributed in the animals.

Concerning the experimental determinations performed in shells we can indicate that in absolutely all the shells analysed it has been observed signals of ^{241}Am over the detection limits, fact that cannot be associated to the presence of external soil contamination because as was indicated previously, all the shells were cleaned before their measurement.

However, the levels found are in general quite moderate, being possible to observe some correlation between the levels found in the shell and the previously determined in their associated soft-body. As an outlier, only, in one case, the shell 7, the ^{241}Am levels found were one to two orders of magnitude higher than the average, indicating the possibility of association of a discrete radioactive particle. This possibility was reinforced after crushing the shell in fragments by observing that the great majority of the ^{241}Am was associated exclusively to one small fragment, and was confirmed by doing an autoradiography of the mentioned fragment, as it is shown in Figure 5.2.4.3.

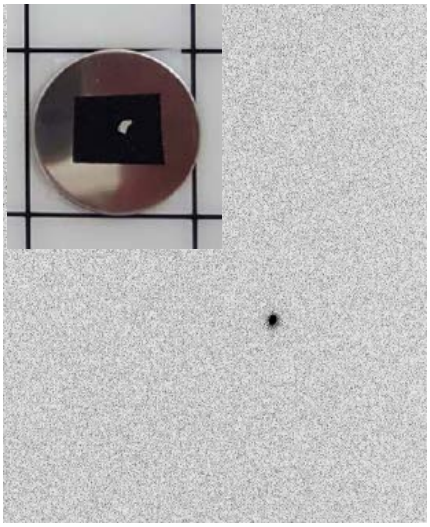


Figure 5.2.4.3.: Shell fragment of snail 7 (upper left), and associated radiography indicating the presence of a radioactive particle in the fragment

The work of identification, location and characterization of radioactive particles in the snail study performed in Palomares was concentrated in three different sample types: whole-body snails, faces and shells. In all the cases, combinations of advanced microscopic techniques not conventionally used in radioecology have been applied. The main results and conclusions obtained in the performance of this work are now presented.

Great efforts were first devoted looking for the identification of radioactive heterogeneities in the whole-body snails that presenting the higher levels of ^{241}Am, without success. All the data conduits to the theory that if some radioactive particles are ingested by the snails, these particles are not retained/incorporated in their bodies and are excreted with the faeces.

Looking for confirmation of the mentioned theory, selected pieces of faeces with relatively high ^{241}Am levels were screened looking for heterogeneities that could be associated to the presence of radioactive particles by applying the nano-computational tomography technique (Nano-CT). The results obtained by the application of Nano-CT (> 400 nm) to one faeces fragment indicates the

presence of a total of 10 high density particles of micrometer size inside, that can be considered as potential candidates to be radioactive Pu/U particles embedded in the faece (Lind et al., 2016).

With the information obtained by Nano-CT in mind, the following step was the application of the micro-XRF technique to the same fragment, looking to obtain multielemental information and particularly the distribution of some potential contaminants by performing 2D mappings. The μ-XRF maps show us how embedded in an organic substratus can be found elements like Ca and Fe characteristics due to their presence in high proportion in the soils of Palomares. And additionally show how some of the high density particles detected previously by Nano-CT correspond with radioactive particles. But interestingly, no all the radioactive particles identified and characterized in the faeces fragment are Pu/U particles with origin in the Palomares accident. Together with Pu/U particles have been identified some U/Th NORM particles that should form part of the Palomares substratus. In these NORM particles, both elements U and Th are present in high proportion (peculiarity that is present only in some NORM minerals, being more usual the presence of only one of the two elements). This result opens a new line of research trying to evaluate the levels of NORM nuclides in the Palomares area, and their contribution in dose assessments studies in the zone.

The screening of radioactive particle heterogeneities in the snail study performed in the contaminated areas of Palomares were finished by analyzing with advanced microanalytrical techniques the small shell fragment of snail 7, where the presence of a Pu/U particle was identified previously by high-resolution gamma-ray spectrometry and by autoradiography.

In the analysis of this shell fragment, the combination of three microanalytical techniques was used:

a) Scanning electron Microscopy (SEM) in the Backscatered Image Mode together with X-ray microanalysis (XRMA),

b) nano computational tomography (nano-CT), and

c) micro X-rays fluorescence (μ-XRF).

This study constitutes a paradigmatic example of the amount and quality of the information gained when several complementary microanalytical techniques are used in combination. The fragment containing the radioactive microparticle was first of all analysed by SEM. An elevated number of attempts were done scanning the fragment to find the microparticle associated with. By means of BEI images, and looking for high Z heterogeneities, several candidate particles forming part of the shell were found, being after that characterized elementary by XRMA. The work carried out was not successful initially in the localization of radioactive particles. The microparticles identified in the shell with high Z are mostly formed either by rare earth elements or are enriched in zircon. All the data indicates that the radioactive particle identified by autoradiography should be embedded in the shell fragment, doing very difficult its localization by SEM-XRMA due to the superficial character of these techniques.

In order to obtain information about heterogeneities inside the shell fragment, first of all the material to be scanned was reduced by dividing it in two parts, keeping for posterior analysis the piece where the high levels of ^{241}Am remains. In fact, this piece was analysed applying the nano-CT and the micro-XRF techniques

Through the application of the nano-computerized tomography (nano-CT) technique it was possible to find several small areas inside the shell with high densities values, being these zones potential candidates to correspond to the radioactive particle searched. With the obtained tomographic information, the micro-XRF technique was constrained in its application to the areas with high density, being in this way quite limited the zones to be scanned and analysed.

The detailed μ-XRF analysis of the marked high density zones, allowed us to locate the radioactive particle searched, that was embedded in the shell fragment. The signal of Pu and U associated to the

presence of these elements in the radioactive particle was generated because with the micro-XRF technique the volume excited by the incoming radiation is much bigger than the generated by SEM. And the situation of the particle covered by intrinsic material in the shell is coherent with the fact that through the application of the SEM-XRMA technique the previous work carried out for the localization of the radioactive particle was not successful. Once the location of the radioactive particle was fixed, SEM-EDX images of the particle embedded was possible to obtain. One of these images is shown in Figure 5.2.4.4.

The peculiar location of the Pu/U radioactive particle embedded in the shell fragment open the discussion about how the particle has finished in the mentioned place. Two theories can be established, with no additional evidences to support in particular one of them as much probable: a) the particle would have been ingested by the snail, and transferred through the route used by the snail to feed its shell with the materials essential for its growing, b) the particle could have been adhered to the surface of the shell and integrated during the growing of the animal by been covered by young formed layers of the snail. But with independence of how the radioactive particle was incorporated to the shell, we should highlight the presence of a good number of microparticle heterogeneities in the fragment analysed with different composition (rare-earths, zircon, iron, Pu/U microparticles). All the data indicates that the incorporation of Pu/U to the shell structures of the snail should not be peculiar, single and improbable event, fact that we are trying to verify by screening a statistically significant number of shells collected in the Palomares contaminated area .

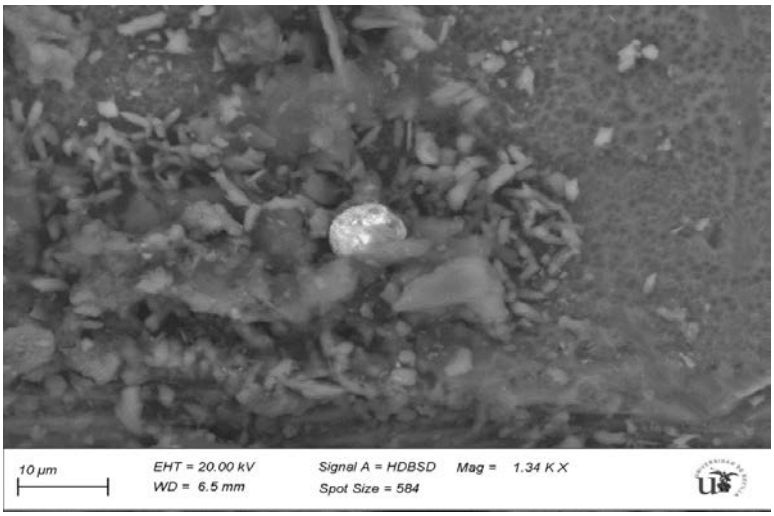


Figure 5.2.4.4.: Identification by SEM of a Radioactive particle embedded in the shell of a living snail collected in the affected contaminated terrestrial area of Palomares (Spain)

5.2.5 Impact and further work

In conclusion, the work performed in RATE associated to terrestrial and aquatic systems affected by radioactive contamination in the form of particles clearly allow highlighting two important points:

a) The usual methods used for risk assessments in radioecology and environmental radiation protection should be adapted for radioactive particles exposures.
b) Some additional knowledge is still needed about the ecological implications of radioactive particles released into the environment, for example about the trophic transfer in the food chain.

Specific on site experimental radioecological studies should be performed in each area affected by particle radioactive contamination, because their behaviour and characteristics is source and scenario release dependent. In this sense, the information gained through the experiments performed in some

analysed contaminated ecosystems has allowed obtaining some specific and time dependent transfer parameters to be linked with models for a more consistent (but no full) radioecological evaluations in the affected areas. But even after the performance of the mentioned experimental studies, the uncertainties associated to parameters defined for prediction for example of transfer to wildlife will be quite high. And the use of the obtained transfer parameters obtained in a specific ecosystem cannot be applied in another one without the performance of experiments demonstrating their similar radioecological behaviour.

As it has been demonstrated in studies performed in Maralinga and Palomares, the Concentration Ratios in rabbits (Maralinga) and snails (Palomares) determined experimentally are in general clearly lower than the defined ones in zones affected by general fallout and their range can cover orders of magnitude. The use of the parameters compiled in ICRP and IAEA databases for wildlife transfer of radionuclides (based mostly in studies performed in areas where the contamination is in soluble and exchangeable form), if applied to ecosystems affected by radioactive particle contamination, generally overpredict the contamination of biota because the contamination associated to particles are in most cases in refractory or insoluble form, being inhibited its transfer. The wildlife transfer factors in the particle contaminated areas will be on the other hand variable, with their values increasing over time due to the increase of the available fraction of the radioactive contaminant (weathering effects).

In addition, once the radioactive contamination is incorporated to the biota, its distribution between different compartments/organs does not follow generally the partitioning models that have traditionally been used for human worker protection purposes. These models are based in laboratory experiments with Pu acute exposures carried out via injection and with short evaluation periods. Specific studies in the areas under evaluation are needed to have in consideration the source and ecosystem dependent characteristics of the radioactive contamination. In areas affected by U/Pu radioactive particle contamination with origin in the conventional destruction of nuclear weapons (Thule, Palomares, Maralinga) it has been demonstrated that a large fraction of the U/Pu incorporated by ingestion by different living biota (rabbits, snails, muskox,…) is not retained, being excreted with the faeces, with the majority of the retained fraction being associated to the gastrointestinal tract due to its low solubility.

The uptake and retention of particles by living organisms in areas affected by this type of contamination is a possibility that on the other hand we have shown in our work that cannot be neglected, with all the dosimetric implications that can be associated. The incorporation of particles through inhalation and/or ingestion by living biota in terrestrial areas and through digestion/filtration in aquatic systems is a type of event with a frequency and with dosimetric implications very difficult to model/predict. Once incorporated to the living biota, the effects from radioactive particles will be concentrated to a smaller area of the body, potentially causing more localized impact. These effects will be very much dependent of the amount and emission type of radioactive substance present in the particle and of the part/organ where the particle is retained.

Additional studies to evaluate localized doses and their effects caused for discrete particles, and to adapt the information gained through the implementation of the project in some well-recognized radioecological models are points that should be fulfilled in the short term future. These aspects are further outlined in the COMET Position paper (Salbu et al., *in press*)

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ICRP reference sites – developing alternative transfer models for wildlife

6.1 Background

Transfer is currently describe in environmental assessment models using the whole-organism concentration ratio (CR_{wo}) (Beresford et al. 2008):

$$CR_{wo} = \frac{Activity\ concentration\ in\ whole\ organism\ (Bq\ kg^{-1}\ fresh\ mass)}{Activity\ concentration\ in\ soil\ (Bq\ kg^{-1}\ dry\ mass) or\ filtered\ water\ (Bq\ L^{-1})}$$

EQUATION 6.1

This parameter has been shown to be highly variable adding considerable uncertainty to assessments (e.g. Beresford et al. 2008).

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. Data availability is especially poor 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 (hereafter referred to as 'Reference 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 by COMET partner NERC-CEH (Barnett et al. 2013, 2014).

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 (Beresford et al. 2013, 2016). This 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 values for organisms taking into account inter-site variation. This enables predictions to be made for organisms for which no data are available from those for which data are available at a given site:

$$Activity\ concentration\ in\ species\ x = \frac{REML\ adjusted\ mean\ for\ species\ x}{REML\ adjusted\ mean\ for\ species\ y} \times activity\ concentration\ in\ species\ y$$

EQUATION 6.2

The initial application of this approach considered Cs and freshwater fish and the resultant model performed better than the traditional concentration ratio approach (Beresford et al. 2013).

Another criticism of the existing data describing the transfer of radionuclides to wildlife is their considerable geographical bias, most data originating from temperate (European and North American) or Arctic ecosystems (Copplestone et al. 2013). There are virtually no, if any, data from Mediterranean ecosystems in the international database (Copplestone et al. 2013) used by both the ICRP and IAEA to derive their recommendations (ICRP 2009 ; IAEA 2014).

When the IRA activities were agreed, NRPA had an on-going sampling and analyses programme at a Reference Site in Norway and NERC-CEH had a programme of work to sample at a further four sites (funded by a UK programme under the TREE project; http://www.ceh.ac.uk/tree). Under this IRA we have brought together these resources and adapted activities such that sampling was conducted in a

Mediterranean ecosystem and additional analyses of samples from a site in the Chernobyl Exclusion Zone (CEZ) were funded under COMET.

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.

6.2 Methods and materials

Sampling has focussed on RAPs for the terrestrial ecosystem (Table 6.2.1). The ICRP defines the RAPs at the taxonomic level of family (Table 6.1) and therefore species representative of these families were targeted. Each Reference Site was selected on the basis that the majority of terrestrial RAPs could be sampled from a relatively small area. In addition to samples of representative RAPs, soils were also collected from each site.

Table 6.2.1.: ICRP RAPs which may occur in the terrestrial ecosystem

RAP	Family	RAP	Family
Wild grass	Poaceae	Deer	Cervidae
Pine Tree	Pinaceae	Rat	Muridae
Earthworm	Lumbricidae	Duck	Anatidae
Bee	Apidae	Frog	Ranidae

The Reference Sites sampled were:

Spain: The main site was a semi-natural grass land (dehesa) which is managed for the hunting of red deer (Cervus elaphus) and wild boar (Sus scrofa). As the site did not include Pine Tree a nearby natural woodland site was also selected. The sites are in the province of Extremadura.

Norway: The site was Tjøtta which is a *c*. 11 km^2 island off the coast of central Norway (marine and freshwater species were also collected from the vicinity but are not considered here) (see Thørring et al. 2016).

United Kingdom: Two sites in north-east England were sampled. Both are managed coniferous plantations: Site K is dominated by Sitka spruce (Picea sitchensis) whilst Site H comprised mixed coniferous species.

CEZ: The Chernobyl Exclusion Zone site was the location of a former village on the edge of the Red Forest (trees were not killed here in 1986).

Species falling into the RAP definitions which were sampled at each site are presented in Table 6.2.2; additional species were collected from some sites but these are not considered here. Species collected during the study reported by Barnett et al. (2014) are also shown in the table as these data are combined with those from the more recent studies in the subsequent analyses presented below; the Barnett et al. study was conducted in a coniferous forest in north-west England (Site G).

Table 6.2.2.: Species representative of ICRP RAPs as sampled at the Reference Sites

RAP	Spain	Tjøtta	Sites H & K*	Site G	CEZ
Wild grass	*Briza minor*	*Deschampsia flexuosa*	*Molinia caerulea, Agrostis gigantea* (K), *Deschampsia flexuosa* (K),	*Molinia caerulea*	*Agrostis gigantea*
Pine Tree	*Pinus pinaster*	*Picea sitchensis, Pinus sylvestris*	*Pinus sylvestris* (H), *Abies alba* (H), *Picea sitchensis* (K)	*Picea sitchensis*	*Pinus sylvestris*
Earthworm	*Lumbricidae spp.*	*Aporrectodea calignosa, Aporrectodea rosea, Lumbricus rubellus, Lumbricus terrestris*	*Lumbricus rubellus* (H), *Dendrodrilus rubidus* (H), *Aporrectodea caliginosa* (K)	*Lumbricidae spp.*	*Lumbricus terrestris*
Bee	*Apis mellifera*	*Apis mellifera*	*Bombus terrestris*, *Bombus pascuorum* (H&K), *Bombus hortorum* (H)	*Bombus spp.*	Bombus spp, Xylocopa spp.
Deer	*Cervus elaphus*	*Alces alces*	*Capreolus capreolus*	*Capreolus capreolus*	Not sampled
Rat	*Apodemus sylvaticus*	*Mus musculus*	*Apodemus sylvaticus*	*Apodemus sylvaticus*	*Apodemus agrarius, Apodemus flavicollis*
Duck	*Anas platyrhynchos*	*Anas crecca Anas platyrhynchos, Aythya fuligula*	Not sampled	Not sampled	Not sampled
Frog	*Pelophylax perezi*	*Rana temporaria*	*Rana temporaria*	Not sampled	*Rana terrestris,*

*Where H or K appears after species name then the species was sampled for that site only.

Samples from the Spanish, Norwegian and UK sites were all analysed by ICP-MS to determine stable element concentrations; radioanalyses were conducted on some of these samples but the results of this are not discussed here. Samples from the CEZ site were analysed to determine activity concentrations of ^{137}Cs, ^{90}Sr, ^{241}Am and Pu-isotopes; samples from this site are currently undergoing ICP-MS analyses via addition funding. For animals, concentrations in the whole-organism (less pelt or feathers and gastrointestinal tract) have been estimated. For Wild Grass and Pine Tree, above ground vegetation and wood (for the Norwegian site data were only available for needles) were analyses respectively (these being representative of the ICRP geometries for these organisms (ICRP 2008). Results were used to estimate CR_{wo} values as defined in Equation 6.1.

For the purposes of this study results for Cs, Pb, Se, Sr and U have been used to establish REML models (it had been intended to also consider Th but insufficient data were available for this element across the different sites). Table 6.2.3 summarises the data available for inclusion in the REML analyses. REML models were fitted to CR_{wo} values using the *IBM SPSS Statistic* package with the fixed factor being the ICRP RAP classification and the random factor being the Reference Site. The analyses were also rerun removing Reference Site (to determine if inclusion of the random factor resulted in the better model).

Table 6.2.3.: Data available for each element for inclusion in the REML analyses

RAP	Spain	Tjøtta	Sits H	Site K	Site G	CEZ
Wild grass						
Cs	Y	Y	Y	Y	Y	Y
Pb	Y	Y	Y	Y	Y	n/a
Se	Y	Y	n/d	Y	n/d	n/a
Sr	Y	Y	Y	Y	Y	Y
U	Y	Y	n/d	n/d	Y	n/a
Pine Tree						
Cs	Y	Y	Y	Y	n/d	Y
Pb	Y	Y	Y	Y	Y	n/a
Se	n/d	n/d	n/d	Y	n/d	n/a
Sr	Y	Y	Y	Y	Y	Y
U	n/d	Y	n/d	Y	n/d	n/a
Earthworm						
Cs	Y	Y	Y	Y	Y	Y
Pb	Y	Y	Y	Y	Y	n/a
Se	Y	Y	Y	Y	n/d	n/a
Sr	Y	Y	Y	Y	Y	Y
U	Y	Y	Y	Y	Y	n/a
Bee						
Cs	Y	Y	Y	Y	Y	Y
Pb	Y	Y	Y	Y	n/d	n/a
Se	Y	Y	Y	Y	n/d	n/a
Sr	Y	Y	Y	Y	Y	Y
U	Y	Y	Y	Y	Y	n/a
Deer						
Cs	Y	Y	Y	Y	Y	n/s
Pb	Y	Y	Y	Y	Y	n/s
Se		Y	Y	Y	Y	n/s

Sr	Y	Y	Y	Y	Y	n/s
U	Y	Y	Y	Y	n/d	n/s
Rat						
Cs	Y	Y	Y	Y	Y	Y
Pb	Y	Y	Y	Y	Y	n/a
Se	Y	Y	Y	Y	Y	n/a
Sr	Y	Y	Y	Y	Y	Y
U	Y	Y	Y	Y	n/d	n/a
Duck						
Cs	Y	Y	n/s	n/s	n/s	n/s
Pb	Y	Y	n/s	n/s	n/s	n/s
Se	Y	Y	n/s	n/s	n/s	n/s
Sr	Y	Y	n/s	n/s	n/s	n/s
U	Y	Y	n/s	n/s	n/s	n/s
Frog						
Cs	Y	Y	Y	Y	n/s	Y
Pb	Y	Y	Y	Y	n/s	n/a
Se	Y	Y	Y	Y	n/s	n/a
Sr	Y	Y	Y	Y	n/s	Y
U	Y	Y	n/d	n/d	n/s	n/a

n/d – not determined by specific laboratory or all replicates below detection limits; n/a – stable element results not available at the time of writing; n/s – not sampled.

6.3 Results

It was possible to fit REML models to all five of the elements considered. For comparing the model with and without the inclusion of Reference Site as the random factor, Akaike's Information Criterion (AIC) was used (the model with the lower value of AIC better describing the data) (Table 6.3.1). For Cs and Sr the AIC indicated a clear effect of site; this was not obvious for the other elements.

Table 6.3.1.: A comparison of AIC values from models fitted with and without the inclusion of Reference Site as a random factor.

Element	AIC – Site as random variable	AIC – No random variable
Cs	967	1638
Pb	660	676
Se	447	449
Sr	832	1596
U	449	449

The output of the REML analyses is a 'REML adjusted mean', this is a relative value indicating how the different RAPs compare to each other rather than being any absolute measure. REML adjusted means for the five elements are presented in Table 6.3.2.

Table 6.3.2.: REML adjusted means for the RAPs and selected elements

Family	Cs	Sr	Pb	Se	U
Anatidae	2.4E-02	7.0E+00	8.8E-03	2.6E-01	1.4E-03
Apidae	7.2E-02	8.5E-02	1.3E+00	2.3E-02	4.5E-04
Cervidae	2.5E-01	1.4E+00	1.1E+00	9.0E-02	2.2E-04
Lumbricidae	3.1E-02	2.5E-01	2.4E+01	1.9E+00	2.3E-02
Muridae	1.6E-01	4.3E-01	1.0E+00	2.0E-01	2.3E-04
Pinaceae	7.9E-02	5.5E-01	5.8E-01	7.6E-03	1.1E-04
Poaceae	1.4E-01	4.5E-01	1.6E+00	6.1E-03	5.3E-04
Ranidae	1.6E-01	1.4E+00	7.3E-01	1.8E-01	3.9E-04

A data set and a journal article based on the work in this IRA have been submitted:

Guillén, J., Izquierdo, M., Young, S., Barnett, C.L., Wells, C., Chaplow, J., Beresford, N.A., Baeza, A., Salas, A., Muñóz-Serrano, A., Corrales-Vázquez, J.M., Muñoz-Muñoz, J.G., Tovar, E. (submitted) Elemental concentrations in representative species of the ICRP's Reference Animals and Plants and associated soils in terrestrial Mediterranean ecosystems in Spain. NERC Environmental Information Data Centre.

Guillén, J., Beresford, N.A., Baeza, A., Izquierdo, M., Wood, M.D., Salas, A., Muñóz-Serrano, A., Corrales-Vázquez, J.M., Muñoz-Muñoz, J.G., (submitted) Transfer parameters for ICRP's Reference Animals and Plants in a terrestrial Mediterranean ecosystem J. Environ. Radioact.

6.4 Discussion

In parallel to this work a separate project (http://tree.ceh.ac.uk/) has begun to establish REML models based upon data within the international Wildlife Transfer Database (WTD) (Copplestone et al. 2013). To date this work has established REML models for terrestrial wildlife species at different taxonomic levels for Cs and at the species level for Sr. The Cs model has been shown to give reasonable predictions; the Sr model has yet to be tested. Comparing these models with those derived using the Reference Site data here does not show good agreement; the example of Cs at the family level is presented in Figure 6.4.1. This lack of agreement is likely to be the result of the different species included in the two models, there is considerable variation between species and the species level model, best describes the WTD data.

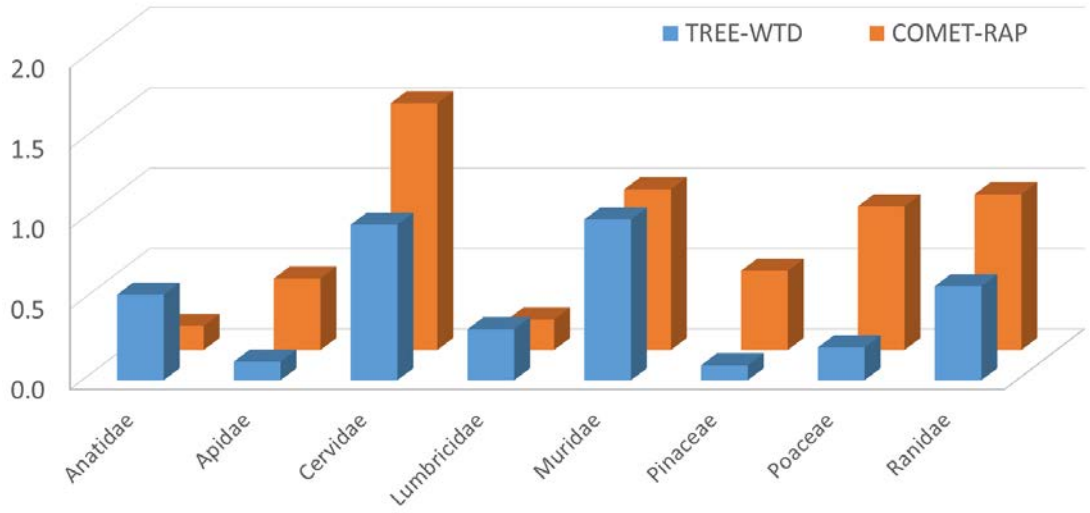

Figure 6.4.1.: A comparison of REML means fitted to data from the WTD and to data for RAP species from the Reference Sites discussed here

The models derived for Se, Pb and U during this work represent the first application of the REML approach to these elements. The lack of site effect may reflect little variation between sites for these elements or perhaps the lower number of data available. Analyses of the larger WTD data for these elements would likely elucidate on this issue.

The application of REML modelling as a method of predicting radionuclide transfer has been suggested for sometime (e.g. Beresford et al. 2004; Willey 2010). However, whilst models had been established for a range of elements and plants the paper of Beresford et al. (2013) was the first to validate a model using predictions against blind test data. The models derived here now need to be tested against data not used to establish them. Though perhaps it would be best first to merge the Reference Site data with the WTD and rerun the models using the larger database.

The application of REML models to marine wildlife has also been considered within the activities of the Marine Roadmap working group with an analyses of marine data for Cs from the WTD having been conducted.

6.5 Impact and further work

The work of this IRA has contributed to the radioecology SRA by enabling a more robust prediction of the exposure of wildlife (there is not a roadmap which is focussed on improving exposure assessments for wildlife).

The investigation of alternative transfer models may ultimately be beneficial to human foodchain modelling; the applicability of REML modelling to food crops is currently being evaluated (http://www.ceh.ac.uk/tree). The work has significantly increased the data available for the ICRP RAPs and will contribute to the development of the ICRPs RAP framework. There is an on-going collaboration with the ICRP to improve transfer parameters for the RAPs and future ICRP reports will consider the use of REML models. Whilst not considered here the studies have yielded, to our knowledge, the first terrestrial wildlife CR_{wo} values for some elements (e.g. I, S, P).

The data generated by the studies described above now need to be incorporated into the international wildlife transfer database (Copplestone et al. 2013) and used in establishing REML models in combination with the existing WTD dataset.

These studies have used ICP-MS analyses to obtain data and CRwo values for stable elements assuming that transfer will be the same as that for radioisotopes under equilibrium conditions. This is an

increasingly common practice as a method by which data for a large range of elements can be obtained relatively inexpensively to derive transfer parameters for radiological models for both wildlife assessment and the human foodchain. However, the assumption that stable element results can be used in this way has not been adequately tested. A small project funded under the ALLIANCE first call will assess the validity of the assumption focussing on Cs and Sr. Note as REML modelling in-effect looks at relative values between organisms within a site the application of stable element data is not questioned (as relative differences should be consistent between stable and radioisotopes even if CRwo values are not).

When considered as a whole the data from the studies used here give us the ability to begin to look at relationships between different elements across biota as a potential approach to predicting radionuclide activity concentrations (see Beresford et al. 2016 for a discussion of ionomics and stoichiometry). Discussions between the data holders on taking this forward after COMET have begun.

6.6 References

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Conclusions

In WP 3 of COMET, significant contributions to improve radioecological modelling capacities have been made, both through model development and through improvement of model parameters. Dynamic modelling approaches have improved our ability to correctly predict ecosystem transfer and uptake of radionuclides, particularly in the early phase after an accident, as well as to assess the potential exposure of and doses to humans and wildlife. Fieldwork and experiments have provided data for model parameters, as well as for model validation. In this respect, access to time series data has been particularly valuable for dynamic modelling, and has allowed us to model radionuclide transfer and uptake in early, intermediate and late phases after radioactive contamination. Furthermore, data from different regional ecosystems, such as the Mediterranean and Nordic ones, has allowed us to customize radioecological models with regional parameters, leading to improved accuracy in model predictions. Hence, the work in COMET WP3 has also been highly relevant for the emergency and post-accident communities.

COMET has contributed significantly towards integration, both within the radioecological community in Europa and internationally, as well as with other scientific disciplines. Institutions from fifteen different countries participated in WP 3, including researchers from countries that have been affected by major nuclear accidents, such as Ukraine and Japan. This provided opportunities for joint experiments and fieldwork, as well as access to unique data for model and parameter improvement. COMET also included partners from different regions in Europe, such as the Mediterranean and Nordic ones, which furthered work towards regionalization of radioecological models to better suit these ecosystems. COMET fostered interdisciplinary collaboration through the two projects chosen in the open call, bringing new perspectives as well as new modelling capabilities into the consortium.

Although the modelling work in COMET WP 3 has made us better equipped to assess the consequences of past radioactive contamination of the environment, as well as to predict the impact of present and future releases, all the topical working groups in WP 3 have identified important future challenges that need continued attention from the radioecological community. The need for continued collaboration between modelers and experimentalist to continue model improvement and to validate models has been highlighted by several groups and will require sustained funding for European radioecological research.