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## DELIVERABLE (D-N°3.1)

### Detailed plan for the COMET WP3 Initial Research Activity – list of research projects and goals, participants and timing

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## Executive Summary

According to the description of work (DoW) for the COMET project, the first deliverable in WP3 is a detailed plan for the COMET WP3 Initial Research Activity (IRA) in the form of a list of research projects and goals, participants and timing. The work should be performed during the first five months of the project. According to the DoW, the WP3 IRA should be performed focusing on improved parameterisation of key processes controlling the transfer of radionuclides, with a specific focus on dynamic modelling approaches. It is also guided by Challenge one of the Strategic Research Agenda of radioecology: "To Predict Human and Wildlife Exposure in a Robust Way by Quantifying Key Processes that Influence Radionuclide Transfers and Exposure" with its four associated research lines.

Due to the EC requirement that the start date should be 1 June instead of the proposed 1 September, the initial exchange of ideas between the partners was done by email prior to the kick-off meeting 27-29 August 2013. During the kick-off meeting, the suggested research projects were reduced in number and grouped into six topics. These topics were subsequently discussed with our external Steering Committee at a meeting in October 2013. The topics and their objectives, in short, are the following:

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

The topics are described in detail in chapter 2 and the associated resources allocated are given in chapter 3. The IRA will start in month 7 and last for 24 months.

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# 1. Background

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

Challenge one of the Strategic Research Agenda of radioecology is *"To Predict Human and Wildlife Exposure in a Robust Way by Quantifying Key Processes that Influence Radionuclide Transfers and Exposure"*. It is this challenge that the WP3 IRA will largely address. Linked to challenge one, there are 4 research lines (RL):

*RL1: Identify and mathematically represent key processes that play an important role in the environmental transfers of radionuclides influencing the exposures of humans and wildlife;*

*RL2: Acquire the data necessary for parameterisation of the key processes controlling the transfer of radionuclides;*

*RL3: Develop transfer and exposure models that incorporate physical, chemical and biological interactions, and enable predictions to be made spatially and temporally; and*

*RL4: Represent radionuclide transfer and exposure at a landscape or global environmental level with an indication of the associated uncertainty.*

These research lines are important to address a range of challenging radioecological situations considering existing (e.g., uranium mining and milling sites, NORM sites), planned (e.g., newly built power plants, (geological) waste disposals) and emergency/post-emergency exposure situations.

In the project description of COMET, one stated aim of WP3 is to *"Undertake joint research activities to improve and validate radioecological models [...] for a better protection of humans and the environment in existing, planned and emergency exposure situations."*

Under WP3.1 it is stated that *"An initial research activity will be performed focusing on improved parameterisation of key processes controlling the transfer of radionuclides, with a specific focus on dynamic modelling approaches"*. During the first 5 months of COMET, the task has been to produce this document, i.e. D3.1: *Detailed plan for the COMET WP3 Initial Research Activity – list of research projects and goals, participants and timing.*

The guiding documents used to produce this deliverable are:

- The project description of COMET;
- The Radioecology SRA; and
- COMET D2.1: Towards a first phase Radioecology Alliance RTD implementation plan as input for the preparation of the Competitive Call organised in collaboration with OPERRA.

We have not repeated text from the above documents which justify radioecological research in general and prioritize the research lines, as they can be consulted if necessary for clarification.

The number of person months allocated to WP3.1 in the project description is limited with partners having only 1 to a maximum of 7 person months. To achieve maximal outputs and foster collaboration, most partners have decided to include in-kind contributions to be able to perform more research in the IRA. The inclusion of work performed by PhD students in several partner organisations also increases the total resources allocated to WP3.1. This is clearly shown in the table summarising the resources for each suggested topic in the last chapter. Some activities are linked to other ongoing research in partner organisations, without duplicating the effort; they will provide added value to COMET in the form of additional resources and links to other communities, in particular NERIS.

This document has been produced through input and suggestions on possible research topics by the COMET partners, followed by discussions and finally a decision to form six topical groups to address various research needs. These topics were subsequently discussed with our external Steering Committee at a meeting in October 2013 and are presented in chapter 2.



## 2. Suggested topics

### 2.1 Marine modelling

#### 2.1.1 Background/justification

The Fukushima nuclear accident in 2011 constitutes the most important accidental release of artificial radionuclides to the marine environment that has occurred. Contamination of every marine component (water, sediment and biota) has been observed. Measurements carried out by the operator TEPCO and the Japanese authorities show the evolution of the contamination in these media over the past 2.5 years. 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. In the aftermath of an accidental situation where radioisotopes in the different marine compartments have not equilibrated, time-dependent radioecological models of transfer are required. Such situations offer the opportunity to validate and improve models that are, or have the potential to be, included in decision support systems (DSS) for emergency situations. Two main radioecological modelling topics are to be taken into account in this task: transfers and exchanges between water and sediments (dispersion modelling, including hydrodynamic and sediment modelling); and modelling of transfers to biota.

Dispersion models have been used to predict radionuclide dispersion from the Fukushima accident in the marine environment as part of on-going programmes with expert teams of modellers dedicated to their improvement and inter-comparison (e.g. IAEA-MODARIA, Science Council of Japan model comparison, UNSCEAR assessment projects). COMET partners are participating in these teams and will thus ensure complementarity and mutual benefit of performed research between these teams and in the COMET IRA. Such dispersion models are relevant to, and are developed for, a wide range of applications outside radioecology and maximal use will be made of it.

#### 2.1.2 Objectives

The COMET Marine group proposes to focus on radioecological transfer modelling and plans to use existing models as a basis for the work with a view to improving some of them to achieve more sophisticated models, e.g. trophic transfer modelling, and by combining transfer modelling with sediment modelling. This will improve the estimates of concentrations in biota, the external and internal exposure assessment of biota, and the internal dose assessment to humans due to consumption of seafood. This work addresses the Research Lines 1, 2 and 3 of Challenge 1 of the Radioecology SRA.

#### 2.1.3 Methodology

The proposed project for the next 2 years includes the 3 different topics listed below with a short description on the methodology envisaged.

Task 1: Transfer to biota – biokinetic modelling (IRSN, SCK•CEN, CIEMAT, UMB, NRPA, SU)

Implementation and use of classical radioecological models based on dynamic transfer equations to evaluate concentrations in marine organisms (fishes, molluscs, crustaceans). Improvement of radioecological parameters (concentration factors and single or multicomponent biological half-lives) for  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{90}\text{Sr}$  (the main radionuclides contributing to dose in the medium to long term, with available data in Fukushima marine species) including sensitivity and uncertainty analyses. This task includes the construction of a database with parameter values (using existing reviews, recent project results and results derived from lab experiments) and comparison with observed half-lives from the

Fukushima monitoring dataset. This work will be extended to other radionuclides that could be important in post-accidental situations due to their potential dose contribution or their potential environmental accumulation, e.g. Pu, Am, Ag).

#### Task 2: Transfer to and from sediments (SCK•CEN, NRP, CIEMAT)

The uncertainties in sediment  $K_d$  values are well known to be orders of magnitude. Thus, including a more accurate representation of sediment processes in dynamic transfer modelling is justified. This work will follow ongoing studies adapting an existing dynamic model (D-DAT, see references 1 to 3). The tool is being presently adapted to include depletion of radionuclides adsorbed onto suspended particulates (particle scavenging), molecular diffusion, pore-water mixing and bioturbation (modelled effectively as a diffusive process) represented by a set of coupled differential equations which are related to the biological uptake/turnover processes. In the course of the IRA, the model will be completed and tested with the available data to see if it is capable of reproducing radionuclide activity concentrations in sediments, giving a more realistic calculation of concentrations in biota and improving the dose assessments for humans and biota based on improved  $K_d$  values.

#### Task 3: Ecosystem modelling – trophic transfer (SU, IRSN)

This task will comprise process-oriented modelling for mid- and long-term predictions taking account of ecological and environmental processes. It will include modelling of trophic transfers to pelagic fish, including food-web transfers and establishing if there is real potential for biomagnification in the Fukushima foodchain. This task will investigate the relative importance of transfer through the food chain compared with direct transfer from the water and sediments.

##### 2.1.4 Timeline

- Establish parameter database, months 7-14.
- Biokinetic transfer modelling, and comparisons with available Fukushima time series, months 14-20
- D-DAT model development, months 7-20.
- D-DAT model application, months 20-26.
- Trophic transfer modelling, months 7-31.

##### 2.1.5 Milestones and deliverables

Tasks	Milestones and Deliverables
Task 1: Transfer to biota – biokinetic modelling	M1: Parameters database established M14 D1: Journal article submitted M20
Task 2: Transfer to and from sediments	M2: Completed development of advanced version of D-DAT model including sediment dynamics M20 D2: Journal article submitted M26
Task 3: Ecosystem modelling – trophic transfer	D3: Journal article submitted M28

## References

Vives i Batlle, J. et al., *Dynamic Model for the Assessment of Radiological Exposure to Marine Biota*. Journal of Environmental Radioactivity, 2008. **99**: p. 1711-1730.

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Vives i Batlle, J., Wilson, R.C., and McDonald P., *Allometric Methodology for the Calculation of Biokinetic Parameters for Marine Biota*. Sci Total Environ, 2007. **388(1-3)**: p. 256-269.

*Radionuclide Transfer in Marine Coastal Ecosystems, a Modelling Study Using Metabolic Processes and Site Data*, Journal of Environmental Radioactivity, 2013. In press.

## **2.2 Forest modelling**

### **2.2.1 Background/Justification**

Following severe nuclear accidents such as the Chernobyl, Kyshtym and Fukushima accidents large forested areas have been contaminated by fallout. These ecosystems are essential for wildlife, and contribute also to the human food chain through harvesting. In contrast to cultivated areas, rather few countermeasures (e.g. Cs binder in salt licks or boli) are feasible for forested ecosystems. As the ecological half-lives tend to be rather long in forested ecosystems, suitable methods for the assessment of the long term impact and consequences are a high priority.

In forest models, it is essential to capture the key processes regulating the entry (source term) and dispersion, circulation/transformation, storage/accumulation and transport/ fixation/exit of radionuclides in the abiotic and biotic parts of the ecosystem. Uncertainties in traditional models come from lumping together poorly known transfer processes into single effective transfer rates or transfer coefficients ( $K_d$ , washout coefficients,  $TF_{agg}$  etc.), ignoring variations in the source term and assuming that equilibrium conditions are relevant. Thus, key factors contributing to uncertainties comes from

1. Poor input data: Time series are highly needed to identify time-dependent changes in ecological half-lives associated with forested areas, and for validation of updated forest models.
2. Poor dynamic process descriptions (e.g.,  $K_d$ , TF). For soil-water interactions it is important to distinguish between irreversible (mineral lattice) and reversible processes and assess the mobility of components in the system available for interactions over time. For soil – plant transfer (TF), foliar interception, root uptake/exudation, translocation and transpiration all play important roles in the time-dependent internal transport, accumulation and dose distribution in plants. As these processes are plant species dependent, the system is rather complex and uncertainties in model parameters (e.g., the Penman – Monteith equation) must be taken into account. For plant-animal transfers, available time series data are more limited. The collation of literature would reveal if information can be utilized with respect to transfer to specific wild animals (e.g. wild boar).
3. Model uncertainties: Mathematical uncertainties caused by using complex, non-linear model solution methods should not be ignored (even if good model development and testing can counteract this), nor should uncertainties in the choice of what processes are relevant, and how processes are described, which is a modeller-dependent factor. It is quite possible that the greatest uncertainty results from this "modeller uncertainty". This can be addressed by using formal methods to develop comprehensive conceptual models as a basis for mathematical model development.

The work will focus on the entry of radionuclides into forest ecosystems. In particular, it will address the influence of the source term (particles, colloids, ionic species) associated with accidental (fallout, dry/wet deposition) or routine releases (groundwater/soil water contamination) on ecosystem

transfers. Furthermore, information on dynamic processes is essential: time-dependent soil – water interactions (dynamic  $K_d$ 's) will be examined for different soil types under different environmental conditions, and time-dependent soil – plant transfers will be assessed for biota selected according to radiosensitivity (e.g. pine), or for their importance for wildlife intake (e.g. grasses, lichens) and for human dietary intakes (e.g. mushrooms, berries). Finally, more information on time-dependent soil-plant-animals transfer would be highly beneficial for modelling purposes.

### 2.2.2 Objectives

For the COMET Forest group, the overall objective is to reduce the uncertainties in assessments of short- and long-term impacts of radioactive contamination in forested areas. The focus will be on key processes and variables/factors contributing most to the overall uncertainties. Due to the dynamic nature of the processes, most emphasis will be put on time-dependent soil-water interactions (dynamic  $K_d$ ) and soil - plant transfers (TF, TC, CR), where biota will be selected according to radiosensitivity, importance for wildlife intakes and for human dietary intakes. The plant-animal pathway will only be considered for specific animals (e.g. wild boar). Results will be implemented in existing models as a basis for model development, for parameterization of key processes controlling the transfer of radionuclides and to improve estimated external and internal exposures of biota. The work is in accordance with Research Lines 1 and 3 of the SRA for Radioecology.

### 2.2.3 Methodology

Within the person months allocated, there is no possibility to cover all aspects of the research needed within the Forest modelling IRA. Therefore, the focus will be on the following tasks:

Task 1: Input data improvement: Collating data/time series on short- and long-term dynamics of radionuclide migration in forest ecosystems (NUBIP, GIG, STUK, UMB)

Task 2: Dynamic process information: identifying key dynamic processes/factors/variables influencing forested ecosystem transfers (Chernobyl Center, NRPA, STUK, CEH)

Task 3: Model improvement: Further development and testing of forest models (SCK•CEN, NRPA, CEH, IRSN, BfS)

To fulfil these tasks, data sets from Task 1 and dynamic process information from Task 2 will be fed into the model development Task 3. Task 1 will focus on time series/data sets from several countries affected by the Chernobyl accident (Ukraine, Finland and Norway), and data/results will be compiled and utilized for forest model development and testing purposes. Also forest NORM data will be included where appropriate. Evaluation of the source term and the influence on ecosystem transfers will be made. In Task 2, mobility data ( $K_d$ , sequential extraction data) from selected sites (e.g., Chernobyl) will be reviewed, and additional dynamic experimental work will be performed when needed. Similarly, soil-plant transfers for selected plant species at selected sites will be reviewed, and additional experimental data will be produced if highly needed. Information on selected plant – animal pathways (e.g., wild boar) will be evaluated. In Task 3, results from Task 1 and 2 will be utilized to improve data input, to modify default values or replace constants with functions of time and to further model development. Time series from Task 1 can also be utilized to test the improved models/model compartments. The input from Task 1 and 2 as well as the updated models should reduce the overall uncertainties in assessments.

### 2.2.4 Timeline

- Workshop (Oslo or video meeting) to identify available time series, data gaps, experimental work needed, model selection and distribution of tasks, month 9.

- Sensitivity analysis to identify processes/variables/factors contributing to overall uncertainties, months 9 - 12.
- Water – soil interaction/mobility data base, identifying data gaps, initiating experimental work needed, months 10-20.
- Soil-to-plant transfer data base and plant-to-animal transfer data base, identifying data gaps, initiating experimental work needed, months 10-20.
- Deriving improved input data and variable/parameter/factor values, describing/parameterizing the dynamics, months 12-29.
- Improving model/model compartments by implementing improved input data and process-oriented information, months 12-28.

### 2.2.5 Milestones and deliverables

Tasks	Milestones and deliverables
Task 1: Input data improvement: Collating data/time series on short- and long-term dynamics of radionuclide migration in forest ecosystems (NUBIP, GIG, STUK, UMB) <ul style="list-style-type: none"> <li>• Perform sensitivity analysis to identify processes/variables/ parameters/factors contributing most to the uncertainties.</li> <li>• Evaluate how different source terms would affect the transfers.</li> </ul>	M1: Sensitivity analysis: Factors contributing to uncertainties in forest model prediction, month 13.
Task 2: Dynamic process information: identifying key dynamic processes/factors/variables influencing forested ecosystem transfer (Chernobyl Center, NRPA, STUK, CEH) <ul style="list-style-type: none"> <li>• Data gaps and soil-water/soil-plant experiments</li> </ul>	M2: Selection of improved input data/variables/factors and process information to be included into models, month 20.
Task 3: Model improvement: Further development and testing of forest models (SCK•CEN, NRPA, CEH, IRSN, BFS)	M3: Improve model/model compartment by implementing improved input data and process oriented information, month 25.
Tasks 1-3	D1: Testing of forest models documenting reduced overall uncertainties in prediction, month 30.  D2: Two papers submitted to peer-reviewed journals, month 30

## 2.3 *Human food chain modelling*

### 2.3.1 Background/Justification

The new ICRP recommendations state that model predictions should be able to assess the first year dose to humans from all exposure pathways. One important pathway is through the human food chain. Decision Support Systems such as RODOS and ARGOS have modules to calculate food contamination and consequent doses to humans based on the ECOSYS model developed by Müller and Pröhl (1993). The Terrestrial Food Chain and Dose Module (FDMT) is used in both ARGOS and

RODOS to predict doses from ingestion. One of the deficiencies in FDMT as currently implemented, is the fact that most default parameters are based on German values that may not be appropriate for other regions in Europe. These relate to e.g. seasonal crop development, transfer values, agricultural practice and dietary habits. Effects of using Nordic dietary habit data for predicting ingestion doses were investigated by Hansen et al. (2010) and showed the importance of using regional parameter values when possible.

Other models also exist for dose calculations from food contamination like the French SYMBIOSE: a simulation platform for performing radiological risk assessment (<http://www.irsn.fr/EN/Research/Scientific-tools/Computer-codes/SYMBIOSE/Pages/The-SYMBIOSE-platform-3838.aspx>). The default parameter values here are largely based on French conditions, but could be adapted to other regions. Default radionuclide transfer factors used in SYMBIOSE is taken from IAEA (2010). SYMBIOSE has been used e.g. to simulate terrestrial food chain transfers in the 80 km zone around the Fukushima Dai-ichi NPP (Simon-Cornu et al. 2011).

Region-specific parameters are lacking for many countries leading to rather uncertain predictions of doses from the human food chain. Generally, the Mediterranean area has been understudied so far and the derivation of region-specific values for e.g. Spain would greatly improve the prediction capability. Other regions, such as the Nordic countries, have climatic conditions, and agricultural and grazing practices that differ significantly from the central European ones. Again, a parameterisation of region-specific values would be necessary to provide sound predictions on food contamination over time and consequent doses to humans.

The data base for element or nuclide-specific model parameter values varies significantly with most empirical data being available for 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.

Specific soil properties could also significantly impact on the transfer of radionuclides, in particular for radiocaesium. In general, highly organic peaty soils with low levels of caesium-fixing clay minerals (e.g. illite), low potassium levels and high concentration of ammonium tend to have higher transfer of caesium to plants than many other types of soil. Unusually high transfer of radioactive caesium from soil to plants, with slow decrease in contamination over time, is typical for European wet peat ecosystems and need to be addressed specifically.

In recent years, the use of probabilistic modelling has 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 dose assessment model, the use of Bayesian networks could aid the separation of uncertainty and variability in model parameters.

### **2.3.2 Objectives**

The research undertaken in this topic aims at improving parameter values for human food chain modelling in line with RL2: *Acquire the data necessary for parameterisation of the key processes controlling the transfer of radionuclides*. The implementation of region-specific values in food and dose models would reduce the uncertainty in predicted contamination levels and doses from food ingestion. The application of Bayesian methods to these parameters has the possibility of reducing the uncertainties even further.

In line with RL1 and 3, studies on long-term dynamics of soil-to-plant transfer for specific soil types and for long-lived radionuclides will be performed with the objective of increasing our mechanistic understanding of the key processes and improving our capability for realistic predictions at a temporal scale of many years.

### 2.3.3 Methodology

The project is divided into four tasks:

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

Task 2: Bayesian methods to derive more robust values where data are lacking or have a large variability (IRSN and NRPA).

Task 3: Long-term dynamics of radiocaesium mobility and plant availability for peat soils with unusually high transfers (NUBIP, NERC, IRSN and NRPA).

Task 4: Long-term dynamic soil-to-plant transfers for Tc-99, Pu and Am (NUBIP, NERC and IRSN).

In the first task we will compare SYMBIOSE and FDMT and run the models for selected countries with default parameter values. A sensitivity analysis will be performed to prioritise which data to focus on. Parameter values in SYMBIOSE and FDMT will be compared, as well as with most recent data in-house or in the literature. The focus will be on the main dose contributing radionuclides Cs-134/137, Sr-90 and I-131. The aim is to derive parameter values that are appropriate for Nordic and Mediterranean ecosystems. These include both radioecological parameters and production/management/habit parameters. The models will then be run with the improved values and the results compared with the results with default parameters to investigate the effect of region-specific parameters on model output. Validation of the models will be part of WP3.4 at a later stage.

Linked to this, the second task will explore how Bayesian statistics can improve the parameterisation of models. Bayesian statistics will be used to estimate probability distributions of transfer parameters and it will also address how we can pool site-specific and generic data.

In the third task we will increase knowledge on long-term dynamics of radiocaesium mobility and plant availability in peat systems. In Ukraine, determination of the dynamics of  $K_d$  and of plant contamination after injection of the water-soluble form of  $^{137}\text{Cs}$  in peat-bog soils are investigated at an experimental site. These data will be compared with existing Norwegian field and experimental data for specific soil types with very high radiocaesium transfers.

The fourth task will be a continuation of earlier experiments in the Chernobyl exclusion zone (CEZ) to reveal changes of transfer with time due to the radionuclide “ageing” to improve the dynamic modelling of soil-to-plant transfers for the long-lived radioisotopes of Pu and Am, and for Tc-99. The work will include planting wheat at the experimental CEZ site, measurements of radionuclide activity and determination of the model parameters. The relevance of these derived parameters for other regions will be evaluated.

The results from the experimental work in tasks 3 and 4 can be used to improve input parameters in FDMT and SYMBIOSE for these radionuclides and special conditions.

### 2.3.4 Timeline

- Meeting in November-December (in Oslo or by video conferencing) to agree on details and initiate work, month 7.
- Sensitivity analysis, months 7-17.

- Reasons, dynamics and mechanisms of abnormally high transfer of <sup>137</sup>Cs, months 7-28.
- Dynamic soil-to-plant transfers for Tc-99, Pu and Am, months 7-29.
- Derive improved parameter values, months 18-29.
- Bayesian modelling, months 11-28.

### 2.3.5 Milestones and deliverables

Tasks	Milestones and deliverables
Task 1: Derive human food chain parameter values that are appropriate for Nordic and Mediterranean ecosystems	M1: Sensitivity analysis of parameters in FDMT and SYMBIOSE finished, month 17
Task 2: Bayesian methods to derive more robust values where data are lacking or have a large variability (IRSN and NRPA).	M2: Evaluation of the Bayesian approach month 29.
Task 3: Long-term dynamics of radiocaesium mobility and plant availability for peat soils with unusually high transfers.	D1: Analysis of long-term dynamics of radiocaesium mobility and plant availability in peat systems. Journal paper submitted, month 28.
Task 4: Long-term dynamic soil-to-plant transfers for Tc-99, Pu and Am	D2: Journal paper submitted, month 29.
Tasks 1-4	D3: Sets of improved parameter values for Nordic and Mediterranean ecosystems for Cs-134/137, Sr-90, I-131, Tc-99, Pu and Am-241 month 29.  M3: Summary of the results, month 30 (as input to the D3.2 "COMET IRA...results and impact")

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## **2.4 NORM - Parameterization of key processes ruling environmental behaviour of naturally occurring radionuclides**

### **2.4.1 Background**

The radiation risk to man due to the presence of naturally occurring radionuclides in (1) ores and minerals, (2) wastes and by-products of NORM-related activities such as mining, P-industry, oil and gas industry, etc., and (3) environments impacted by releases of these industries, has long been a matter of interest for scientists. However, the overwhelming majority of authors rarely elaborate upon the measurement of natural radionuclide activity concentrations in particular compartments within the environment. Data on  $K_d$  and media-to-biota transfer factors (TF) for natural radionuclides are scarce<sup>1</sup> and their statistical significance is quite weak. Available data are recommended to be used for screening purposes and applied in the absence of site-specific information, resulting in large uncertainties. Difficulties arise not only from a lack of data but also from inadequacy of traditional modelling concepts when applied to NORM. The wastes produced in NORM industries, besides their radioactivity, are often a complex mixture of different chemical compounds/minerals and elements, including heavy metals and other toxic compounds. Usually the natural decay chains (uranium or thorium) are not in secular equilibrium and isolated sub-chains may be observed. The mutual ratio of particular natural radionuclides (and also other contaminants) present in waste depends on the raw materials processed, technological process applied, local geological and hydro-geological conditions and the time they were created, especially when sequential decay of natural radionuclides is considered. All these facts imply that the behaviour of naturally occurring radionuclides at their final deposition site or in environments impacted by waste streams is expected to vary. A promising strategy to develop a sufficient understanding of environmental transfers and exposure processes is to identify and then explicitly model key processes that influence the behaviour of the natural radionuclides, in line with the logic of the four Research Lines of Challenge 1 in the radioecology SRA.

### **2.4.2 Objectives**

Within the proposed NORM IRA, the objective is to contribute to Challenge 1 of the SRA, RL 1-3, 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. The resulting knowledge is required to explain the behaviour of natural radionuclides in environmental compartments including dynamic processes and to create a capability to assess resulting exposures of both non-human and human populations with substantially reduced uncertainty.

### **2.4.3 Methodology**

Quintessential to achieving the objectives are archival data, field investigations at the Polish Observatory Site and dedicated lab experiments. The proposed project for the next 2 years includes 5 different tasks. They are listed below with a short description of the methodology envisaged:

1. Identification and parameterization of chemical and physical processes influencing natural radionuclide accumulation, mobility and bioavailability after release into the environment;
2. Selection and physical and chemical characterization of testing sites and samples;

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<sup>1</sup> Although some comprehensively compiled examples exist such as those by Vandenhove et al. (2009a,b), UNSCEAR (2008), IAEA TRS 472 (2010). The most advanced compilation of radionuclides transfer factors is probably ICRP 114.

3. Linking radionuclide accumulation, mobility and bio-availability with environmental characteristics;
4. Assessing and modelling transfers to non-human biota;
5. Model validation and improvement.

#### Task 1 – Identification and parameterization of processes (GIG, SCK•CEN, CIEMAT)

Natural radionuclides present in mine water after its release into the environment are subject to different chemical and/or physical processes influencing their final fate. The processes of concern are e.g. precipitation, sedimentation, adsorption, absorption, ion exchange, desorption, leaching, erosion, sequential decay etc. Based on physical and chemical rules and real environmental conditions, these processes may be parameterized and mathematically represented and then used for construction of a model of radionuclide behaviour. The literature reports and archival data gathered during routine mine water discharge monitoring and long-term observation of NORM-contaminated sites will be analysed in the light of current scientific understanding of these fundamental processes.

#### Task 2 – Site/sample selection and their physical and chemical characterization (GIG, BfS, SCK•CEN, UMB)

The site selection and characterization will mainly focus on the Polish Observatory Site. Specific locations will be selected and fully characterized including a whole range of physico-chemical and geochemical properties. Selected sites will provide relevant conditions for: verification of models of particular processes identified in the framework of Task 1; investigation of relationships between environmental characteristics and radionuclide mobility and bioavailability planned in Task 3 and Task 4.

Taking into account the unique properties of the Polish Observatory Site, this task will especially focus on: accumulation of radionuclides in sediments and behaviour of progeny created due to sequential decay that are initially not present in mine water in significant amounts, but later on appear in aquatic/terrestrial ecosystems (i.e. Pb-210/Po-210 and/or Th-228).

Sequential or selective extraction techniques will be applied to assess the phases that the radionuclides and pollutants considered are associated with.

#### Task 3 – Influence of environmental characteristics on radionuclide mobility (GIG, SCK•CEN)

In this task, the mobility will be linked with environmental characteristics to better parameterize assessment models. The in situ  $K_d$  will be determined for selected radionuclides and heavy metals and the relation will be assessed between soil or environmental properties and  $K_d$  values. The radionuclide/contaminant speciation will be predicted using geochemical speciation models. Through these geo-chemical speciation models, the influence of co-contaminants on availability may be assessed. Field studies may need to be complemented by laboratory studies using substrates prepared in the frame of Task 2. Parameterized  $K_d$  values will be developed using advanced statistical tools.

#### Task 4 – Assessing and modelling transfers to fauna and flora (GIG, UMB, SU, SCK•CEN)

Flora and fauna prevailing at the sites will be sampled and monitored for radionuclides, pollutants and a number of nutrients (e.g. cations). Herbaceous plants (e.g. grasses, *Impatiens glandulifera*) will be sampled and monitored.

We will evaluate to what extent concentrations of naturally occurring radionuclides in wildlife and transfer factors observed can be predicted based on the environmental parameters monitored. A parametric transfer factor model for the biota of concern will be established if the results allow. Field observations and experiments may need to be complemented with laboratory experiments to better control influential parameters (like climate).

The transfer factor (TF) from substrate to plant tissues can be assessed by direct measurement of pollutant concentrations in substrate and relevant plant tissues at the chosen sites. To complete the proposed tasks, at least two vegetation seasons are necessary, spring 2014 – autumn 2015.

#### Task 5 – Model validation and improvement (GIG, BFS, CIEMAT, SCK•CEN)

Results obtained from the experimental studies will be used to check how theoretical predictions of existing models and empirical data match each other. The model-data inter-comparison may suggest modifications to improve the models. There are many models that focus on migration, transport and distribution of pollutants in the environment like, such as HYDRUS and CROM. These models will be used to test the accuracy of the water transport of natural radionuclides in mine water.

#### 2.4.4 Timeline

- Identification and parameterization of key processes influencing natural radionuclide behaviour, months 7- 14.
- Site selection and sample characterization, months 9-20.
- Influence of environmental characteristics on radionuclide mobility, months 9-24.
- Assessing and modelling transfers to fauna and flora, months 12-30.
- Model validation and improvement, months 15-31.

#### 2.4.5 Milestones and deliverables

Tasks	Milestones and deliverables
Task 1: Identification and parameterization of processes	MS1: Process identification finalized, month 14,
Task 2: Site/sample selection and its physical and chemical characterization	MS2: Site selection and characterization of samples finalized, month 20
Task 3: Influence of environmental characteristics on radionuclide mobility	MS3: $K_d$ values and availability analysis finalized, month 24
Task 4: Assessing and modelling transfers to fauna and flora	MS4: Transfer data obtained, month 30 D1: Data set of characterized samples, $K_d$ and TF values, month 31 (publication in scientific journal submitted)
Task 5: Model validation and improvement	D2: Parameterized $K_d$ and transfer models and improvement of existing models, month 31, report

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## **2.5 Particle Behaviour**

### **2.5.1 Background**

Physico-chemical forms of radionuclides determine their behaviour in the environment, transfers in the food chains and doses to humans. Besides the most mobile water-soluble forms, the radionuclides can be also present in the so-called hot particles (HP), ranging from submicron size to millimetre fuel fragments. In the last case, the radionuclide mobility and bioavailability depends on the rate of HP weathering and dissolution. Therefore, measures of HP dissolution in various media are necessary for characterization of the radionuclide source term, and reliable modelling of this process can be considered as a first and one of the most important steps for improving the models describing the radionuclides behaviour/transfers in the environment and for reducing the uncertainties of dose assessments.

A major fraction of the radionuclides released during historical nuclear events has been associated with particles. Near-field contamination of the Chernobyl nuclear power plant (ChNPP) is mainly caused by radionuclides associated with the irradiated nuclear fuel particle matrix (Kuriny et al., 1993; Salbu et al., 1994; Kashparov et al., 1996, 2003).  $^{90}\text{Sr}$ ,  $^{238-241}\text{Pu}$  and  $^{241}\text{Am}$  were essentially released during the accident in the fuel particle (FP) matrix. After the Chernobyl accident due to the gradual dissolution of FP in soil the  $^{90}\text{Sr}$  mobile fractions and activities in plants increased with time; the rate of increase depended on particle characteristics, the acidity of the media and the presence of oxygen (Kashparov et al., 1999, 2000, 2004). Also, it depended on the initial degree of transformation of the FP matrix (i.e. on the type of FP, structure and oxidation state of the U matrix). For example, in the bottom sediments of the ChNPP cooling pond (neutral media and absence of oxygen) FP have not yet dissolved. Decommissioning of the cooling pond implies a beginning of lowering the water table in the second half of 2013. It will result in drainage of the highly radioactive sediments and FP will begin to dissolve. The leached radionuclides thus will become involved in the dynamical migration processes. It gives us an opportunity to follow the evolution of FP, which is very important for the accurate prognosis of long-term radionuclide behaviour in the 30-km Chernobyl zone.

### **2.5.2 Objectives**

The overall aim of the project is to improve our ability to describe the processes of Hot Particles transformation and radionuclide leaching in various media, which influences on bioavailability and thus transfer over time. It links to Research Line 1 of challenge 1 in the Radioecology SRA.

### 2.5.3 Methodology

The project is divided into 4 tasks.

#### Task 1: Chernobyl Fuel Particles behaviour in soil (NUBIP, UMB)

Validation of the model and specification of the parameters of FP dissolution in soils for description of the long-term dynamics of the plant contamination with  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in natural conditions at the agricultural areas near the CEZ.

#### Task 2: Chernobyl Fuel Particles behaviour in the radioactive waste trench in the CEZ (NUBIP, IRSN, UMB)

Validation of the model and specification of the parameters of FP dissolution in the radioactive waste trench in the CEZ for description of the long-term dynamics of radionuclide leaching from FP and becoming involved in biogeochemical migration (root uptake by plants, migration to the aquifer).

#### Task 3: Chernobyl Fuel Particles behaviour in the drained bottom sediments (NUBIP, UMB)

Validation of the model and specification of the parameters of FP dissolution in the drained bottom sediments in the CEZ for description of the long-term dynamics of radionuclide leaching from FP and prognosis of radiological situation at the drained parts of the cooling pond and in the water reservoirs remaining after decreasing of the water table in 2013-2015.

#### Task 4: Hot Particles containing NORM (NUBIP, UMB)

Studies of properties of the NORM-containing particles from the tailing dumps of former uranium enterprises.

In the tasks 1-3 we will sample HP-containing materials (soil at the agricultural areas near the CEZ, radioactive waste from the trench in Red Forest (CPS) and drained bottom sediments at the experimental plot in the CEZ and at the drained area of the cooling pond, respectively). In these samples, RN activities ( $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ) will be measured and  $^{90}\text{Sr}$  exchangeable fractions will be determined. Based on exchangeable fractions the total activities of dissolved FP will be determined and non-dissolved FP fractions will be derived. Also, we will determine agrochemical characteristics in soils (exchangeable Ca, pH) and the pH of soil solutions in wastes and sediments. Non-dissolved FP in the samples will be characterized by means of sequential extractions. Certain amounts of FP will be extracted from the samples and their matrix will be studied by means of electron microscopy and other methods. Besides, RN specific activities will be measured in the plant samples collected at the agricultural areas (grain, task 1) in order to characterize the TF dynamics. Obtained data on the non-dissolved FP fractions will be used for validation of the model of FP dissolution and for specification of its parameters. Thus, the key information for the long-term description of dynamics of the RN bioavailable and mobile fractions will be obtained.

In the fourth task, we will examine presence of HP in the NORM-containing materials sampled from the tailing dumps of former uranium enterprise by means of track autoradiography. Certain amounts of HP will be extracted from the samples and their matrix will be studied by means of electron microscopy and other methods.

### 2.5.4 Timeline

- Sampling the FP-containing materials (soil, wastes, sediments) and plants, months 5-15.

- Sampling the NORM-containing material, track autoradiography and extraction of HP, month 11.
- Characterization and extraction of FP from soil, waste and sediments, month 11.
- Measurements of radionuclide activities in samples, determination of agrochemical parameters of soils, pH of materials and exchangeable fractions of <sup>90</sup>Sr, months 5-17.
- Characterization of HP from the NORM-containing material, month 23.
- Study of HP matrix (all extracted HP), months 22-23.
- Determination of FP dissolution rates and dynamics of plant contamination with <sup>90</sup>Sr and <sup>137</sup>Cs, validation of the model of FP dissolution, months 17-23.
- Analysis and generalization of the obtained results, preparation of inputs to D3.2 and journal articles, months 24-32.
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### 2.5.5 Milestones and deliverables

Tasks	Milestones and deliverables
Task 1: Chernobyl Fuel Particle behaviour in soil	M1: Set of parameters for FP dissolution in soils, characterization of FP matrix, month 17. D1: Journal article submitted, month 24.
Task 2: Chernobyl Fuel Particle behaviour in the radioactive waste trench in the CEZ	M2: Set of parameters for FP dissolution in the radioactive waste trench, characterization of FP matrix, month 24. D2: Journal article submitted, month 32.
Task 3: Chernobyl Fuel Particle behaviour in the drained bottom sediments	M3: Set of parameters for FP dissolution in the drained bottom sediments, characterization of FP matrix, month 24. D3: Journal article submitted, month 30.
Task 4: Hot Particles containing NORM	M4: Characterization of HP and their matrix, month 24. D4: Journal article submitted month 32.
Tasks 1-4	D5: Summary of results as input to D3.2, month 32.

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## **2.6 ICRP reference sites - developing alternative transfer models for wildlife**

### **2.6.1 Background/justification**

A large number of organisms and radionuclides have to be considered within assessments of the potential impact of radioactivity on the environment. It should not be surprising that there is a lack of empirical data to derive transfer parameters for many organism-radionuclide combinations; this is especially the case for the Reference Animals and Plants (RAPs) which form the basis of the ICRP framework (ICRP, 2008) that is likely to be used by many states in the future. To compensate for the lack of data, in ICRP Publication 114 (Strand et al. 2009), it is suggested that a series of sites be selected and sampled to determine transfer parameters for the various RAPs. NERC have a paper in press (Strand et al. 2009) on the topic of terrestrial RAPs having been sampled and analysed from a forest in northwest England). NRPA, together with UMB, are conducting similar work at a site in Norway.

Whilst such studies provide a considerable amount of data for these poorly studied organisms and radionuclides (e.g. see Barnett et al. 2013) 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 application of an alternative approach, in part developed under STAR (Beresford et al. 2013), using a Residual Maximum Likelihood (REML) mixed-model regression. This would provide, for each element, the relative concentration (or transfer parameter) values for the RAPs taking into account inter-site variation to enable predictions to be made for organisms for which no data are available from those for which data are available.

Another criticism of the existing data for the transfer of radionuclides to wildlife is their considerable geographical bias. This is true even within Europe with virtually no, if any, data in the international database used by ICRP and IAEA to derive their recommendations (Copplestone et al. in-press), originating from Mediterranean ecosystems. Similarly, virtually none of the existing terrestrial wildlife data originate from Japan. There is also considerable, and increasing, use of stable element data to parameterise models of radionuclide transfer to wildlife with the assumption that stable element data will give reasonable estimates of the long-term equilibrium transfer of radionuclides. However, recent evaluations of the comparative transfer parameter values of stable and radioisotopes for wildlife have demonstrated differences (Barnett et al. in-press; Wood et al. in-

press; Beresford et al. 2013) although there are potential biases in the existing data which may contribute to this.

### 2.6.2 Objectives

The proposed IRA will contribute to COMETs aim to reduce model uncertainty through fostering the sharing of infrastructure, on-going studies, and datasets, together with targeting additional activities to address the issues highlighted above. The results will contribute to the Radioecology SRA's research lines 1 to 3 by providing the data to derive a taxonomically based model of radionuclide transfer independent of site specific factors.

### 2.6.3 Methodology

In brief we will:

Task 1: Establish terrestrial sites in Mediterranean Spain (NERC, CIEMAT), the Chernobyl Exclusion zone (Chornobyl Center, NERC) and Japan (Fukushima University).

Task 2: Sample species representative of terrestrial RAPs and soils from the selected sites.

Task 3: Analyse samples to determine stable element and gamma-emitting radionuclide concentrations (all sites), and  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ , Pu-isotope and  $^{90}\text{Sr}$  activity concentrations in samples from the Chernobyl site (NERC, Chornobyl Center, FU). Note gamma-emitting radionuclide and stable element analyses of samples from Chernobyl and Spain will be analysed under funding secured by NERC separately. Analyses of samples from Chornobyl for Pu-isotopes and  $^{90}\text{Sr}$  are additional, although partially supported by in-kind contributions as is the analyses of samples from Japan.

Task 4: Prepare a combined database (including data from existing UK and Norwegian sites) and data collected from the studies above (CIEMAT, Chornobyl Center, FU, NERC, NRPA, UMB).

Task 5: Prepare and submit refereed paper(s) and publish databases on-line (CIEMAT, Chornobyl Center, NERC, NRPA, UMB).

We envisage that under Task 4 of WP3 (Integration, validation and implementation of RTD results) we will use our integrated dataset to derive REML adjusted (i.e. site independent) transfer values. Analyses of the data will also enable us to determine if stable element data give reliable estimates of radionuclide transfer (the data obtained will allow Cs, Sr and I to be evaluated; additional data held by NRPA and UMB will be used in this evaluation). The results should achieve a high impact and take-up in this rapidly developing area of radioecology. As FU are increasing their staff complement, the work in Japan will start later than at other sites. The Japanese study will serve as a validation dataset of the model derived from the other sites.

### 2.6.4 Timeline

- Select Chernobyl field site, months 6-7
- Select Spanish field site, months 8-9
- Select Japanese site (month 10-11)
- Conduct field studies Chernobyl and Spain, months 10-16
- Sample analyses Chernobyl and Spain, months 16-22
- Data analyses, months 21-28
- Preparation and publication of on-line data set(s), months 21-30



- Preparation and submission of referred papers, months 24-30

The activity as described is achievable within the two year time framework envisaged for WP3's IRAs.

### 2.6.5 Milestones and deliverables

Task	Milestones and deliverables
Task 1	M1: Select Chernobyl site (M7) M2: Select Spanish site (M9) M3: Select Japanese site (M11)
Task 2	M4: To have completed sampling in Chernobyl and Spain (M16) M5: To have defined sampling and analyses schedule for Japanese site (M20)
Task 3	M5: To have analysed all samples from Chernobyl and Spain (M22) D1: Description of and preliminary results from Japanese site (M30)
Task 4	M6: Combine and analyse all datasets (M28)
Task 5	D1: Published dataset (with doi number) for Spanish site M30 <sup>1</sup> D2: Published dataset (with doi number) for Ukrainian site M30 <sup>1</sup> D3: Submit paper(s) to refereed journal describing the study sites and discussing results M30

<sup>1</sup>As far as reasonably possible publication of the datasets will be timed to coincide with that of the accompanying referred paper.

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### 3. Resources To Be Committed

The table shows the number of person months (PM) charged to the EC and as in-kind, including the work performed by PhD students, for each topic and partner.

Partner	IRA Topic	Marine modelling	Forest modelling	Human food chain	NORM	Particle Behaviour	ICRP ref. sites	Total
SCK	PM paid by EU (at 75%)	0.25	0.25		1			1.5
	PM in kind	0.5	0.5		0.5			1.5
	PhD		1		6			7
STUK	PM paid by EU (at 75%)		0.2	1.8				2
	PM in kind							0
NRPA	PM paid by EU (at 75%)	1.5	0	4			1.5	7.0
	PM in kind	9	2	1			3	15
IRSN	PM paid by EU (at 75%)	3	0.5	1.25		0.25		5
	PM in kind	4	0.5	2		1		7.5
	PhD	8		10				18
NERC-CEH	PM paid by EU (at 75%)		1.75	2			0.75	4.5
	PM in kind		1.5	1.5			4	7
CIEMAT	PM paid by EU (at 75%)	0.07		0.46	0.40		0.07	1
	PM in kind	0.27		1.18	1.27		0.50	3.22
SU	PM paid by EU (at 75%)	2			1			3
	PM in kind	1			1			2
BfS	PM paid by EU (at 75%)		0.5		0.5			1
	PM in kind	13	1.5		1.5			16

<b>UMB</b>	<b>Pm paid by EU (at 75%)</b>	0.5	1		2.5	2.5	0.5	7
	<b>PM in kind</b>	1	3		5	3	1	13
<b>GIG</b>	<b>PM paid by EU (at 75%)</b>		1		3			4
	<b>PM in kind</b>		0.2		0.6			0.8
	<b>PhD</b>				two students			
<b>Chornobyl center</b>	<b>PM paid by EU (at 75%)</b>		3.5				1.5	5
	<b>PM in kind</b>		0.5				1.3	1.8
<b>NUBIP</b>	<b>PM paid by EU (at 75%)</b>		0.75	2.25		3		6
	<b>PM in kind</b>		0.25	0.5		0.75		1.5
<b>FU</b>	<b>PM paid by EU (at 75%)</b>	1					3	4
	<b>PM in kind</b>						2	2
<b>Sum per topic total</b>		45.09	20.4	27.94	24.27	10.5	19.12	<b>147.32</b>
SUM EC funded per topic		8,32	9.45	11.76	8.40	5.75	7.32	51.00
SUM in-kind per topic		28,77	8.45	4.68	9.87	4.75	7.8	64.32