DYNAMICS OF RADIONUCLIDES IN SEMI-NATURAL ENVIRONMENTS

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1. Introduction

This paper reports a review of the implementation and achievements of the SEMINAT Project [1] after the first two years of activities (1996-1997).

SEMINAT (Long-Term Dynamics of Radionuclides in Semi-Natural Environments: Derivation of Parameters and Modelling) is a project funded by the European Commission (Research Contract n°FI4P-CT95-0022) in the frame of the Nuclear Fission Safety Research and Technological Development Programme (1994-1998).

I. Linkov and W.R. Schell (eds.), Contaminated Forests, 17–21.

The main aim of SEMINAT activities with respect to semi-natural systems (meadows and forests) is to deliver working computer models of radionuclide behaviour and fate in semi-natural ecosystems typical of the majority of the European Union countries.

Meadows and forests are typical semi-natural ecosystems. Meadows are used extensively in many countries as pastures for cattle, sheep and goats while forests are important to man since they provide wood, paper, wildberries, mushrooms, game and recreational areas. Foodstuffs and other products from both of these types of ecosystem have exhibited persistently high contamination levels following the Chernobyl accident. It is important a) to understand the reasons for this persistence and b) to estimate radiation doses incurred by human usage of these areas.

During the last years our understanding of radionuclide behaviour in semi-natural ecosystems has been improved significantly, especially for boreal forests and middle European meadow systems which have been extensively investigated. Data sets have been obtained which describe the distribution and the cycling of radionuclides (especially ^{137}Cs and ^{90}Sr) within these systems. However, predictive modelling has largely been restricted to aggregated transfer factors which provide good contamination estimates but only for the sites from which data have been obtained directly. For radiation protection purposes advanced models are essential which are applicable to a broad variety of ecosystems. They are needed for dose estimation, countermeasure implementation and environmental management. They should give reliable estimates of the behaviour of radionuclides in semi-natural systems and of external and internal radiation exposure to man. In order to develop such models it is necessary to understand the basic mechanisms of transfer and migration of radionuclides in meadows and forests. The SEMINAT project will therefore identify critical transfer parameters and quantify their influence on the persistence and migration of strontium and caesium in semi-natural systems. At the same time predictive models will be developed on a modular basis which will provide an integrated predictive capability for both forest and meadow ecosystems. Stand-alone modules will be coupled to provide an ecosystem-level model which will be tested and calibrated against site-specific data collected by groups from different countries.

2. SEMINAT Main Achievement (1996-1997)

SEMINAT project is mainly addressed at the development of dynamic models describing the radionuclide behaviour and fate in semi-natural ecosystems, typical of the majority of the EU countries. To achieve the final aim, the activities were organised in 4 work packages (WP) as follows:

- WP1 - Definition of scenarios
- WP2 - Derivation of fluxes and transfer parameters
- WP3 - Modelling
- WP4 - Results analysis

The main objectives for 1996 and 1997 were:

a) to identify the main parameters driving the radionuclides dynamics in semi-natural ecosystems (WP1);
b) to identify the different scenarios and the experimental protocols for the field and laboratory activities (WP1);
c) to determine representative values for the main parameters, quantifying the spatial variabily at large and small scale (WP2);
d) to develop a working version of the forest model (WP3).

2.1 DEFINITION OF SCENARIOS

In the frame of the WP1, forest and meadow sites were chosen. In 7 of forest sites (Lady wood, Buttersteep wood, Ballistone wood, Tarvisio, Weinsberger, Kobernausser and Novaggio forests) studies on the fluxes of caesium are currently taking place within and between various ecosystem components. In the three forest sites in Germany (Hochstadt, Siegenburg and Garching/Alz), the rooting depth of understorey vegetation and mushrooms are under study, comparing the $^{137}Cs/^{134}Cs$ ratio in the different vegetation/mushroom species and in the different soil horizons. The influence of spatial variability of soil parameters on caesium migration and uptake by trees and understorey vegetation is investigated in the Clogheen, Sharahan, Weinsberger, Kobernausser, Tarvisio and Venzone forests. Six study sites have been selected in semi-natural meadow ecosystems, to identify critical transfer parameters and quantify their influence on the persistence of radiocaesium in these ecosystems. In the Cavan site radionuclides fluxes between different soil layers are under investigation. In the other sites the factors influencing the radionuclides fluxes from soil to vegetation (plant species, seasonality and soil erosion processes) are under investigation. All the methods and the experimental protocols used in the investigations were agreed and compared between the participants at the project.

2.2 DERIVATION OF FLUXES AND TRANSFER PARAMETERS

The following sets of data are being collected in the different experimental sites: soil parameters, biomasses in the different compartments, radionuclide content in the different compartments, radionuclide fluxes into and out the compartments. Spatial and temporal variability of these parameters are under investigation in order to estimate reliable rate coefficients to calibrate dynamic models. In all semi-natural ecosystems investigated, an high spatial variability in the radiocaesium content in the environmental compartments has been observed. In almost all sites, the coefficient of variation associated with the total caesium activity in the soil is higher than 50%. This high spatial variability makes it necessary to analyse many samples in order to obtain reliable data for model calibration and validation.

The observed depth profiles of radiocaesium in soils of natural ecosystems were used to assess the residence half-times of Chernobyl radiocaesium. The first results show that the residence half-times are in the order of few years per cm layer

and they vary spatialy and also with depth. This variability has to be considered in the prediction of the transfer of radiocaesium from soil to vegetation.

Radiocaesium inventory in trees has been investigated where the preliminary data indicate that tree canopy, in spite of the little biomass (17% of the tree weight), plays an important role in retention of radionuclides. The canopy stores 41% of total ^{137}Cs contained in the whole tree. In the different sampling sites, the amount of radiocaesium found in the tree biomasses ranges from 1 to 12% of soil inventory. Generally, mushrooms and understorey vegetation account for much less than 1% of the radiocaesium in the soil compartment. Aggregated transfer factors for mushrooms and understorey vegetation show a very high variability with coefficient of variation higher than 100%. The analysis of an extensive data set for a sampling site in Germany revealed that fungal mycelia and fine roots of understorey vegetation occupy distinct layers of forest soil. The temporal changes of radiocaesium activities in mushrooms and understorey vegetation reflect the time-dependent activity concentration of that soil layer, from which radiocaesium is predominantly taken up.

Aggregated transfer factors for meadow vegetation, investigated in Germany along a slope of an alpine pasture, range from 0.2 to 6.6 10^{-3} $m^2 kg^{-1}$ with a coefficient of variation higher than 60%, the high variability could be attributable to the changing plant community along the slope. Aggregated transfer factors assessed in Ireland for a meadow on peaty soils show values ranging from 5 10^{-3} to 2.6 10^{-1} m^2 kg^{-1}. This variability is attributable to the different plant species considered. The highest values were found for the shoots of *Colluna vulgaris*.

To identify the critical parameters and quantify their influence on the persistence and migration of caesium in semi-natural systems, laboratory experiments are in progress. The fate of radiocaesium in the weathering model is validated using a soil weathering sequence derived from sandstone. The soil sequence is: *acid brown soil -> ochreous brown soil -> ochreous podzolic soil -> podzol*. Clay contents are invariably low but the nature of clay minerals markedly differs according to soil type. The associated mineralogical sequence corresponds to the classical transformation process of micas in acid conditions: mica -> vermiculite and HIV -> smectite. The transformation process is not only identified in the various pedons, but also in the various horizons of each pedon, but at different extents. The weathering model is validated using a soil evolution sequence as described above, made of sandy soils with low clay contents (<8% clay), previously fully characterized. The first results show (i) a decrease in the net ^{137}Cs retention value of the Ah layer from the acid brown soil to the podzol, (ii) a "depth-dependance" of the maximum ^{137}Cs net retention value for each profile. The decrease in the net ^{137}Cs retention value in the Ah layers is associated with the transformation process from mica -> vermiculite (acid brown soil) -> from smectite (podzol). In each profile, the maximum value of the ^{137}Cs net retention is associated with the horizon in wich (i) vermiculite is dominant and (ii) Al-interlayering does not take place (complexing organic acids are present). The minimum value of the ^{137}Cs retention is invariably measured in Bw horizons in which Al-interlayering takes place (HIV minerals). The magnitude in ^{137}Cs net retention is directly related to the soil vermiculite content ($r = 0.948$) estimated by the rubidium assessment methodology.

These results are currently being validated with the soil samples collected in the various sites of the SEMINAT network.

2.3 MODELLING

As a first step in SEMINAT's development of dynamic models appropriate to forest ecosystems a screening model (RIFEQ) was developed which allowed preliminary data from each of the research groups' forest sites to be used in a probabilistic uncertainty analysis of radiocaesium distributions in major components of each ecosystem. The screening model developed uses a combination of aggregated transfer coefficients (T_{agg} values) and biomass estimates in a mass balance calculation. The results of such calculations allow an instantaneous 'snapshot' of the expected ranges of radiocaesium activity concentrations and distributions within forests and provide important insights into the relative importance of individual compartments with respect to the development of dynamic models (site-specific calculations of radiocaesium distributions using RIFEQ are intended to assist with the calibration of dynamic models).

While the RIFEQ screening model is useful in assisting with calibration of dynamic forest models, it does not itself provide a tool for interpreting and forecasting radionuclide behaviour in forests on a temporal basis. For this reason SEMINAT is developing a dynamic modelling capability using data from forest sites within five EU countries. At the outset of the project it was considered desirable to provide users with the ability to undertake model calculations either at a relatively simple or a more complex level, as appropriate to their needs. For this reason, the RIFE1 model, originally conceived and partially developed by the ECP5 [2] project, has been both retained and further developed by SEMINAT. Additionally, RIFE1 has been used as the basis for developing a more complex dynamic model, RIFE2. RIFE1 development into a fully probabilistic code is currently under way.

3. References

1. Belli, M Editors 1998, SEMINAT: Long-Term Dynamics of Radionuclides in Semi-Natural Environments: Derivation of Parameters and Modelling, Mid-Term Report 1996-997, European Commission, Nuclear Fission Safety Programme, Research Contract N° FI4P-CT95-0022, ISBN 88-448-0295-3, 46p.
2. Belli, M. & Tikhomirov F.A. Editors; 1996, Behaviour of Radionuclides in Natural and Semi-Natural Environments; Final Report of Experimental Collaboration Project N°5 (1991-1995), European Commission, Belarus, The Russian Federation, Ukraine. International Scientific Collaboration on The Consequences of the Chernobyl Accident (1991-1995), EUR 16531 EN, 1996, ISBN 92-827-5197-X, 147 P.