Contract No FIGE-CT-2000-00071

BORIS

Bioavailability Of Radionuclides In Soils : role of biological components and resulting improvement of prediction models.

SUMMARY OF FINAL REPORT

REPORTING PERIOD : FROM 2000-10-01 TO 2002-06-30

PROJECT START DATE : 2000-10-01 DURATION : 45 months

IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Fifth Framework Programme

1- PRESENTATION

BORIS is a European Project partly financed by the European Commission via the Fifth Framework Programme of the European Atomic energy Community (EURATOM) for Research and training in the field of Nuclear Energy (1998-2002)

This project is co-ordinated by the Institut de Radioprotection et de Sûreté Nucléaire (IRSN, co-ordinator Christian TAMPONNET) and is composed of the following partners:

Institut de Radioprotection et de Sûreté Nucléaire (IRSN) - Direction de l'Environnement et de l'Intervention (France)
Christian Tamponnet, Arnaud Martin-Garin, Marc-André Gonze

Centre for Ecology and Hydrology (CEH) - Lancaster Environment Centre (United Kingdom)
Brenda Howard, Nisha Parekh, Arthur Sanchez

Universita de Barcelona (UB) - Departament de Biologia Vegetal (Spain)
Ramon Vallejo, Teresa Sauras, Jaume Casadesus

Institut National de Recherche Agronomique (INRA) - Centre de Montpellier (France)
Siobhan Staunton, Claude Plassard

Statens stralskyddinstitut (SSI) - Department of Waste Management and Environmental Protection (Sweden)
Leif Moberg, Rodolfo Avila, Maria Norden

Imperial College of Science, Technology and Medicine (ICSTM) - T.H. Huxley School of Environment, Earth Science and Engineering (United Kingdom)
Graeme Shaw, Claire Wells

.1- Objectives

The general objectives of the proposal aim at:

- ✓ apprehending mainly the role of the biological elements (plants, mycorrhiza, microbes) involved in these transfers: radionuclides sorption/desorption in soils and radionuclides uptake/release by the plants.
- ✓ improving existing predictive models of radionuclides contamination of soils and plants, by incorporating knowledge acquired from the experimental results into two mechanistic models CHEMFAST and BIORUR specifically modelling radionuclides sorption/desorption from soil matrices and radionuclides uptake/release to/from plants and then by incorporating these mechanistic models into assessment models to enhance their prediction ability.

.2- Brief description of the research performed and methods/approach adopted.

To achieve the objectives of the BORIS project, the following research activities have been performed:

.2.1- Role of abiotic parameters in bioavailability of radionuclides in soils

Batch and column experiments have been performed both with complete soil and soil elements:

- ✓ to determine the effect of the main physico-chemical parameters of the soil solution (composition, pH, Eh) on the remobilisation of RN from the (artificially)contaminated natural soil,
- ✓ to analyse the influence of the water content and the kinetics of the processes on the efficiency of RN remobilisation,
- ✓ to provide experimental data allowing to determine the importance of microorganisms on the processes which control the fate of RN in the soil-soil solution system.

.2.2- Role of biotic parameters in bioavailability of radionuclides in soils

.2.2.1- potential role of soil microorganisms

Experimental devices have been developed to precise the role of microbiological processes in the bioavailability and cycling of radionuclides, to provide mechanistic information on various aspects of soil-microbe-radionuclide interactions, and to develop an innovative experimental approach, which incorporates microbial activity at optimal conditions of growth, in order to address the role of soil microorganisms on radionuclide sorption/desorption. The role of different groups of soil microorganisms on radionuclide uptake has been determined.

.2.2.2- role of plants

Research experiments have been developed to study the mechanisms of some plant-dependent processes affecting the soil-to-plant transfer of radionuclides and to clarify the variability of radionuclide concentration in soil solution at the root surface.

.2.2.3- potential role of mycorrhizal fungi
The aim of this investigation is to determine how ectomycorrhizal infection modifies the uptake and translocation of radionuclides (Caesium, Strontium and Technetium). The uptake of radionuclides by mycorrhizal and non-mycorrhizal tree seedlings from contaminated soils have been measured. Furthermore, the specially designed rhizospheric culture device enhanced chemical changes in the (myco-)rhizosphere and allow them to be quantified and their consequences on the availability of radionuclides to be assessed. A two-compartment rhizosphere device has been adapted, separating mycorrhizal roots and fungal hyphae, to determine separately uptake by these organs. The additional effect of non-radioactive metal pollution (Cu) on radionuclide uptake has been investigated. A screening of the fungi in pure culture allowed a pollutant metal (Cu) and a sensitive fungus to be selected for detailed studies in association with tree seedlings.

.2.2.4- global role of biological components
This work package aims to provide experimental results on radionuclide chemistry and uptake by plants as influenced by the physico-chemical conditions within soils and their biological activity.

.2.3- Improvement of assessment model in predicting the bioavailability of radionuclides in soils

The developed activities have been:

- ✓ the description of the model specifications in order to harmonise the needs of modellers with the constraints of experimenters.
- ✓ the development of a model describing how radionuclide chemistry and bioavailability is influenced by the physico-chemical conditions within soils and by their biological activity.
- ✓ the development of a mechanistic model of radionuclide bioavailability emphasizing on biological processes: root uptake, mycorrhizal transfer and mineralisation of organic matter.
- ✓ the development of assessment model integrating the outcome from the two models previously developed.

.3- Main achievements

The following achievements have been obtained.

.3.1- Role of abiotic parameters in bioavailability of radionuclides in soils

.3.1.1- specific role of soil components
Strontium sorption on soil may be expected to depend almost entirely on the clay mineral content. An important fraction of adsorbed Sr is not readily desorbable, and this should be taken into account in mathematical descriptions of its mobility in soils. The adsorption of Cs in soil is too complex to be simply described by the sum of adsorption on individual components. Furthermore, soil clay minerals may behave differently to reference minerals, because of long-term weathering and the presence of

inorganic and organic coatings. Adsorption is not completely and instantaneously reversible, even after a short contact period. This must be taken into account in mathematical descriptions of Cs mobility in soils.

Caesium adsorption on compost and compost-clay mixtures depends on the composition and proportion of electrolyte solution used to suspend the solid. Cs adsorption on the components is not simply additive because of interaction between the components, in particular potassium content. When clay has not been dispersed (by preparation of a dilute salt-free suspension) it presents a smaller adsorption surface and hence smaller Cs adsorption.

.3.1.2- adsorption on whole soil (INRA)

Technetium was added as pertechnate salt. A small fraction was detectable in soil solution in all samples the first day. After one week the activity concentration in extracts decreased strongly and in most cases no activity could be detected after 3 weeks. Tc was not extracted by any of the electrolyte solutions.

Changing soil properties, including pH and redox conditions may influence Sr adsorption. Adsorption appears to be largely reversible. The change in soil composition when soil is suspended may mask some of the changes in Sr distribution *in situ*, however the effects are rather small.

There are significant time dependent changes in soil affinity for ^{137}Cs and on the extent and strength of reaction between ^{137}Cs and soil. These changes depend upon the conditions of incubation, particularly aeration and the subsequent variations in ammonium content. The addition of organic matter had smaller and short-lived effects, probably linked to the release of ammonium and potassium. Some of these effects are lost when soil is suspended in a large volume of solution. However, Kd values calculated from soil solution activity concentration are underestimated at short contact times when the isotope has not fully penetrated soil aggregates. Drying causes considerable fixation of adsorbed Cs.

.3.1.3- adsorption on whole soil (IRSN)

For caesium, the range of K_d values determined in batch experiments mostly reproduced its behaviour in the column experiments for the first 120 days. Then, the apparent K_d value was 1 order of magnitude higher than predicted by the simple batch measurements. It has to be noted that only a small fraction of the sorbed Cs was available for remobilisation. That is, 95 % of Cs was irreversibly sorbed onto the soil even under conditions that theoretically increase its release.

For strontium, it was possible to approximately fit experimental column data, starting from the batch values without any other parameter adjustments. Thus, in a first estimate and in the context of our experimental conditions, the Sr sorption processes onto solid phases should be well represented by a simple K_d model.

Technetium migration is strongly controlled by kinetically limited processes leading to a large degree of irreversibility. These processes are directly linked to the oxidation and the microbial state of the porous media. That is particularly showed by the amended solution/sterilized soil conditions in which the γ-sterilised soil is not kept entirely abiotic during the first Tc migration soil and lead to a progressively microbial re-colonization. Some of these effects are lost in batch suspensions where solid/solution ratios are lower. In the case of Technetium, the K_d model alone could not reproduce at all the fate of that element in the soil.

.3.2- Role of biotic parameters in bioavailability of radionuclides in soils

.3.2.1- potential role of soil microorganisms

Based on a mineral-free organic soil prepared from collected leaf litter, experimental results showed that the distribution ratios for both radionuclides (Sr and Cs) are greater in biotic systems (with soil microorganisms) as compared to abiotic, sterile systems (without any soil microorganisms). Addition of clay minerals enhances radionuclide binding to soil in biotic systems but binding increase is very low in abiotic systems. The presence of microorganisms may increase radionuclide binding by increasing the surface area for binding in the organic matrix and clay minerals and/or by depleting the medium of competing ions.

Such increase in radionuclide binding is temperature-dependent with an optimum around 15-20°C for Cs and 20-25°C for Sr. This reinforces the role of soil microorganisms as the main responsible for radionuclide binding increase.

Such a role is devoted to soil and saprotrophic fungi (fusarium, penicillium, tricoloma, mycena)and not to soil bacteria (bacillus, pseudomonas, actinomycetes).

.3.2.2- role of plants

Starvation, variation in plant age, growth stimulation, growth inhibition, variation in plant transpiration affected both the radionuclide uptake by plants as well as the analogue nutrient uptake. All experiments show the existence of a linear relationship between absorbing power of K and Cs and of Ca and Sr. This sustains the hypothesis that radionuclide uptake follows the same pathway as analogue nutrient.

Such uptake ratio can be seen in the xylem ratio for the pair (Cs, K) but not for the pair (Sr, Ca) indicating the more important mobility of the former pair when compared with the latter.

.3.2.3- potential role of mycorrhizal fungi

Mycorrhizal symbiosis does not modify ^{85}Sr accumulation and transfer to the host plants, decreases the transfer of ^{95m}Tc to the shoots by increasing its immobilisation in roots, and increases the transfer of ^{137}Cs to the shoots, although this effect is fungal species-dependent.

The presence in soil concentration of additional non-radioactive pollutant (Cu) at concentration affecting the growth of mycorrhizal fungi decreases the effect of mycorrhizal fungi on the radionuclide uptake by plants.

.3.2.4- global role of biological components

For Caesium, ammonium, conductivity, potassium and soil water content (theta) are the principal variates influencing K_d. However, the degree of influence of each of these variates differs between soil types. Thus, water content is not a significant determinant of Cs sorption in the Batcombe soil and potassium is not significant in the Swinley soil.

For Srontium, stable (non-radioactive) Sr, water content, conductivity, pH , Ca and Mg all influence sorption to some degree in both soils. However, only stable Sr and water content are significant determinants of Sr Kd in the Batcombe soil and non of these variates were significant individually in the Swinley soil.

For Technetium, nitrate, ammonium, conductivity and soil water content (theta) are the principal variates influencing K_d. The influence of water content on Tc K_d is complicated by the fact that it controls redox potential, which, in turn, controls the interconversion of Tc^{VII} and Tc^{IV} and the balance between nitrate and ammonium concentrations.

.3.3- Improvement of assessment model in predicting the bioavailability of radionuclides in soils

One of the main achievements is the production of the CHEMFAST and BIORUR models specifically introducing the role of biological components along with other physical and chemical parameters both in the radionuclide sorption/desorption equilibrium in soil (CHEMFAST) and in radionuclide uptake/release by plants (BIORUR).

The other main achievement is the development of an assessment model incorporating the BIORUR and CHEMFAST models and introducing the quantification of the notion of bioavailability.

.4- Exploitation and dissemination

Dissemination of information revealed in the BORIS project will be multi-modal:

- ✓ Scientific outputs encompassing various partners will be published in a special issue of the *Journal of Environmental Radioactivity*.
- ✓ Scientific outputs from each partner will be published in various scientific publications.
- ✓ When considered non confidential, results of BORIS project are available on the following web site: http://www.irsn-dpre.com/boris/default.htm

Although it is considered that the BORIS project has little industrial and commercial applications, exploitation of results from BORIS can be found in the list of main results published in the technology implementation plan:

- ✓ Data base for Kd
- ✓ Data base for Root uptake
- ✓ CHEMFAST model
- ✓ BIORUR model
- ✓ Assessment model