

# Soil-radionuclides interaction and subsequent impact on the contamination of plant food products based on a simulated accidental source

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## Introduction

The Chernobyl accident, which resulted in a substantial release of radioactive materials in the atmosphere, demonstrated that large environmental areas may be contaminated by fall-out deposition of

radioactivity. In particular, contamination by  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  of agroecosystems where food production is taking place is most liable to contribute to population radiation dose (Strand *et al.*, 1996). Nuclear safety analysis shows that the possibility, although very small, of an accident occurring on a pressurized water reactor (PWR) cannot be completely ruled out. In such a situation, decision-making and management of the contaminated agricultural surfaces largely depends on our ability to predict how, and to which extent, the initial contamination may cause foodstuffs to be polluted. Furthermore, the efficiency of the prediction models relies on our level of understanding of the mechanisms governing the transfer of radionuclides in the soil-plant system.

Gaining an understanding of these mechanisms from *in situ* observations of environmental areas contaminated by past events is difficult due to the lack of knowledge and control on both, the contamination itself, arising from a critical situation, and the natural environment, which is highly variable, temporally and spatially. Such conditions prevent a clear identification of the most relevant parameters influencing the radionuclides transfer and thereby the prediction goal from being achieved. This is why IPSN developed a unique research facility capable of generating, in closed and controlled environmental conditions, a mini-accident with release of radioactive aerosols onto small-scale, but realistic, samples of crops. These crops are grown on undisturbed soil monoliths, featuring several soil types from various European countries, managed in advanced lysimeters, and placed in greenhouses where various climatic conditions can be reproduced artificially under computer control.

The PEACE Programme, gathering a European scientific collaboration around this facility, has been designed to tackle the consequences of an accidental release of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  on the soil-plant system of agricultural lands. The research conducted has focused on the interaction of radionuclides with soils with a view to improve our understanding of the mechanisms governing their transfer to plants via the roots, and their modelling for prediction.

## Materials and methods

A detailed description of the performances of the IPSN Controlled Lysimetric Environmental facility and its various technological components has already been published elsewhere (Bréchignac *et al.*, 1996; Bréchignac *et al.*, 1998; Madoz-Escande *et al.*, 1999). Only the essential features are briefly recalled in the following.

### *The climates and the soils in the controlled lysimetric environmental facility*

Located at Cadarache (Bouches-du-Rhône, France), the facility is a 2000 m<sup>2</sup> leak-proof (depressurized) laboratory consisting in four experimental greenhouses where artificial reproduction of predefined climatic and hydric conditions is made possible by means of computer based regulation. These greenhouses host various lysimeters made of soil monoliths which have been sampled in one block (12 tons) without disturbing the pedological stratification, and further fitted in metallic casings (1.5 m deep, 3.2 m<sup>2</sup> area). Sampled in Belgium (Mol), Germany (Jülich), France (Belleville), Spain (Barcelona) and the United Kingdom (Wellesbourne), they correspond to several soil types commonly encountered in western Europe. Prior to installing a soil monolith in its dedicated greenhouse, the bottom face of the lysimeter is fitted with a water reservoir interfacing the soil via a porous ceramic layer. This unique system allows the exchange of water which occurs naturally between the water table and the soil to be simulated artificially. Thus, the soil moisture can be mastered. The top side of the lysimeter to be cultivated is next brought to fit a hole in the greenhouse floor in such a way that the soil surface appears in the greenhouse where the climatic conditions prevailing at its sampling site are artificially reproduced. The underground parts of all the lysimeters are located in a common hall where temperature can be controlled

independently from that which is controlled above the soil surface, in the greenhouses. Climatic control and corresponding data acquisition (temperature, hygrometry, light and rainfall) are performed automatically. For the purpose of this study, three different climates have been reconstructed (temperate, Mediterranean and transition temperate-Mediterranean) within independent greenhouses by means of a dedicated software which reproduces the nyctemerial and seasonal climatic parameters variations in real time.

### *Contamination of the lysimeters for accident simulation*

Simulation of an accidental contamination is performed by using an induction furnace that produces aerosols similar to those which would be produced in the event of severe failure in a pressurized water reactor (PWR) with core fusion (about 3000°C). Aerosols are generated in a water-saturated atmosphere enclosed in a polyethylene envelope which can be made to communicate with a similar envelope covering the lysimeter surface to be contaminated. They are produced from a mixture of elements representative of the materials constituting a 900 MW reactor: the nuclear fuel (as uranium oxide), the structural materials of the reactor core, the components of the fuel sheath and the control bars. The proportions of elements within this mixture correspond to the relative quantities of the nuclear core inventory, reduced by  $10^7$ .  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , the two radioactive fission products most detrimental to the environment in a radiation protection perspective, are included in this experimental mixture. In order to produce such aerosols, the mixture is brought up, as powder, to a temperature of 2950 °C (reached within 30 minutes and maintained for 15 minutes) in a water-saturated atmosphere. A thorough physico-chemical characterization of these aerosols showed that they were produced as numerous oxides of combined elements, with a mean aerodynamic diameter of 3.5  $\mu\text{m}$ . The total dissolution rates (in rain water) of the  $^{137}\text{Cs}$  and the  $^{90}\text{Sr}$  contained in these aerosols amounted to 85-95 % after 1 day, and 75-85 % after 5 days, respectively.

## *Experimental protocol for radio-contamination follow up*

The general experimental protocol of lysimeter studies starts off with the aerosol deposition on the lysimeters soils and their crops, which took place in 1994 and 1995. Subsequent to contamination, several annual crops (barley, lettuce and beans) have been grown successively for several years, and in parallel on the various soils, with realistic rainfall cycles in order to simulate natural conditions. Two perennial crops (rye-grass and lucerne) have also been sown on some plots, either before or after the contamination event, and followed up to five successive years in order to simulate a cattle feeding semi-natural ecosystem. Migration profiles of radionuclides within untilled plots of the soils have been determined by sampling vertical soil cores (down to 50 cm), taking due care of potential compaction and in-depth contamination transport associated to this method. The same procedure has been applied to the tilled plots of the soils (down to 20-30 cm) in order to check for the vertical distribution homogeneity in this layer, and to determine the soil specific activity for each radionuclide. Plants have been sampled periodically, and annual crops harvested upon maturity, for determining their radio-contamination status.

## *Radionuclides and ionic determinations in soils and plants*

Measurements of radionuclides and major ions species (K, Ca, Mg) have been carried out on various sample types: bulk dry soil, soil interstitial water (soil solution) and plants (leaves and seeds, separately as appropriate). The soil solution has been extracted by high speed centrifugation of moist soil samples. Stable ionic species have been determined based on atomic absorption spectroscopy.  $^{137}\text{Cs}$  has been determined using a Germanium coaxial g emission detector, whilst  $^{90}\text{Sr}$  has been measured by liquid scintillation after rapid separation/purification as previously described (Tormos *et al.*, 1995).

## Results and discussion

### *Soils physico-chemical characterization*

The soils studied show contrasted properties with respect to texture, clay content and pH (Table 1). The soils texture range from sandy/sandy-loamy (Mol, Belleville, Wellesbourne), typical of alluvial and fluvio-glacial sediments, to loamy, characteristic of loess sediments present in Central Europe (Jülich), or derived from ancient pedogenesis of alluvial sediments typical of Mediterranean areas (Barcelona). The clay content varies from 4.1% (Mol) up to 13.9% (Barcelona), whereas the soils pH ranges from acidic (Mol and Belleville) up to about neutral (Wellesbourne, Jülich and Barcelona). The soils retention properties for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  have been quantified respectively by determining their RIP (Radiocaesium Interception Potential) and their CEC (Cationic

Geographical sampling site	Mol (Belgium)	Belleville (France)	Wellesbourne (U.K.)	Jülich (Germany)	Barcelona (Spain)
FAO classification	orthic podzol	fluvisol	eutric fluvisol	orthic luvisol	calcic luvisol
Soil texture	(Loamy) sand	Loamy sand	Sandy loam	Silt loam	Loam
Particle size					
% clay	4.1	5.8	9.5	11.2	13.9
% silt	14.6	13.7	19.7	78.8	28.8
% sand	78.4	79.0	66.6	8.4	47.7
Chemistry					
pH (KCl)	4.6	4.1	6.3	7.1	7.3
% organic matter	3	1.5	3.9	1.5	2.3
Total carbonates (%)	=0	=0	0.3	0.2	7.2
RIP ( $\mu\text{eq.g}^{-1}$ )	443	1126	2316	2328	2732
CEC ( $\text{cmol.kg}^{-1}$ )	11.7	5.8	17.8	13.4	16.4

Table 1  
Main physico-chemical characteristics  
of the various European soils in the plough layer.

Exchange Capacity). The Radiocaesium Interception Potential is the product of the capacity of the Frayed Edge Sites (FES) of clay minerals to adsorb radiocaesium, and the selectivity coefficient of caesium with respect to K for these exchange sites (Wauters *et al.*, 1994). Highly selective, and located at the interlayer edges of weathered micaceous clay minerals, such sites have been demonstrated to govern radiocaesium sorption (Maes *et al.*, 1998; Maes *et al.*, 1999). This approach allows to take due account of the competitive effect of K and Ca+Mg ions for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  sorption, respectively, which influence their respective solid-liquid partition coefficients ( $K_{\text{D}}\text{Cs}=\text{RIP}/[\text{K}]$  and  $K_{\text{D}}\text{Sr}=\text{CEC}/[\text{Ca}]+[\text{Mg}]$ , Sweeck *et al.*, 1990).

Another approach to identifying the degree of radionuclides fixation on soil matrices relies on the estimation of the exchangeable fractions as displaced by  $\text{NH}_4\text{Ac}$  (1 N), and expressed as a percentage of the total amount initially fixed. These have been measured on the various soils and amounted to 12-20 % for  $^{137}\text{Cs}$ , and to 35-60 % for  $^{90}\text{Sr}$  (Figure 1), therefore featuring a chemical availability of  $^{90}\text{Sr}$  in the soils larger than that of  $^{137}\text{Cs}$  which should reflect their respective mobilities in the soil-plant system with respect to both, migration and root uptake. More detailed analysis on the chemical

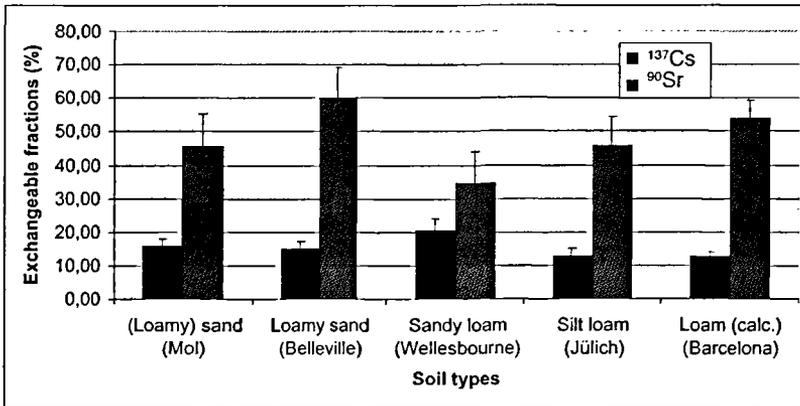


Figure 1  
Exchangeable fractions of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in various soils as measured from displacement by  $\text{NH}_4$  Acetate (1 N).

availabilities of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in these soils have already been reported (Sauras-Yera. *et al.*, 1999; Forsberg *et al.*, in press).

### *$^{137}\text{Cs}$ and $^{90}\text{Sr}$ migration in soils*

When attempting to understand the migration patterns, the first steps come from chemistry which indicates that the high potential mobility of radiocaesium, due to its significant solubility, is counteracted by its very specific and efficient sorption on clay particles (FES of illitic and vermiculitic clay, in particular). Conversely, the lower potential mobility of radiostrontium, due to its poorer solubility, is not counteracted to the same extent by specific fixation sites on the soil matrix. Overall, this results in radiostrontium being more mobile than radiocaesium with clearly distinct  $K_D$  values, as determined *in vitro*, from which a higher migration rate is expected for  $^{90}\text{Sr}$  than for  $^{137}\text{Cs}$  after several years. However, experimental observations do not fit this theoretical approach (Forsberg *et al.*, 2000).

On the one hand, the two radionuclides developed migration profiles, on a given agricultural soil (untilled plots), which still show similar shapes four years after the contamination deposition. Even the soil type, which yields various  $K_D$  coefficient values for the 5 soils of this study, does not significantly alter the observed profiles (Figure 2). On the other hand, the low rates of migration observed in the long term (around  $1 \text{ cm.yr}^{-1}$ , in the years following contamination) cannot explain the shapes of the profiles (Figure 3). In the shorter term, when experimentally forcing migration on soil columns with a high hydrological flux, the two radionuclides exhibit profiles which are established within one month, with very limited further evolution (Figure 4). Altogether, these observations strongly suggest that the migration profiles are established very early after contamination, under the influence of initial processes that do not depend primarily on the soils  $K_D$  coefficients, as determined *in vitro*. Their evolution is subsequently very slow. This indicates the importance of the early conditions prevailing immediately after contamination, such as soil moisture and first rain events, which may be paramount in determining the extent to which radionuclides will penetrate in depth.

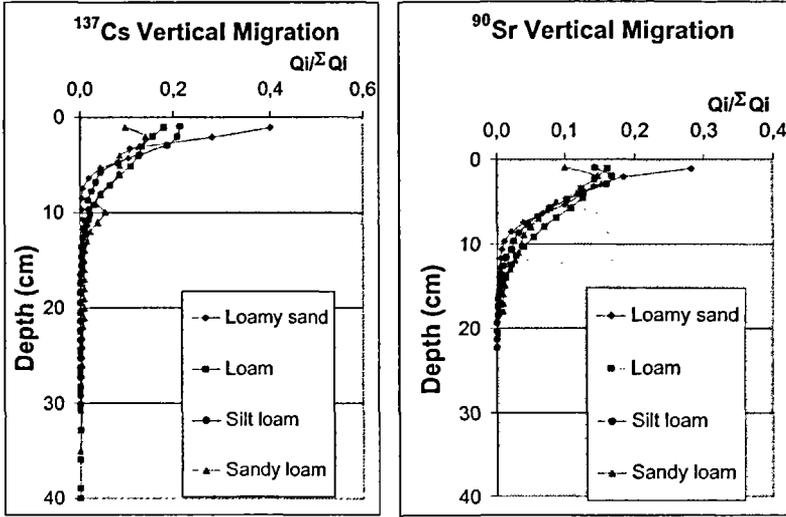


Figure 2  
<sup>137</sup>Cs and <sup>90</sup>Sr migration profiles observed four years after contamination on various agricultural soils.

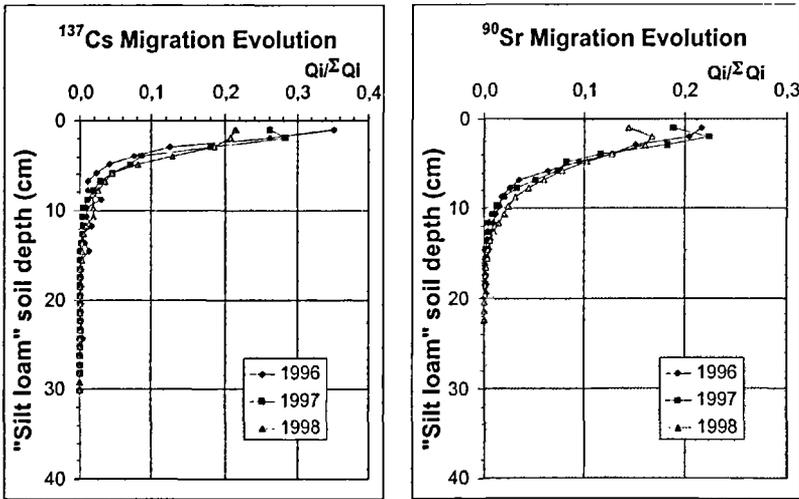


Figure 3  
<sup>137</sup>Cs and <sup>90</sup>Sr migration evolution during three successive years on an untilled silt loam soil.

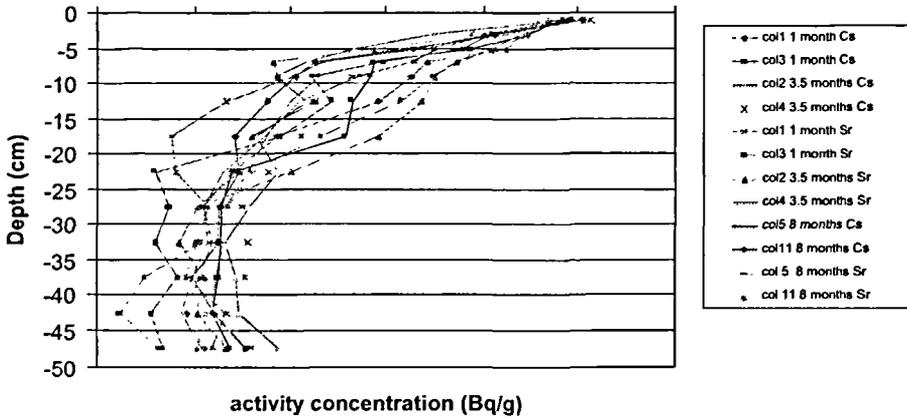


Figure 4

Cs and Sr profiles observed after 1, 3.5 and 8 months following contamination, in an undisturbed soil driven into experimental columns submitted to a high hydrological flux.

The simulation of the hourly-to-yearly vertical migration of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in a saturated/unsaturated soil column with vegetation has been undertaken based on a one-dimensional, dynamic physically-based model formulation (TRANSSOL model, Gonze, 1999). The major processes involved in water flow and transfer of radionuclides have been taken into account. Mechanisms such as downward migration enhancement due to macropores, water exchanges with surface atmospheric layer and time-dependent sorption of radionuclides onto the soil matrix are accounted for, as well as the slow initial dissolution of aerosols and the influence of soil moisture on the radionuclide concentrations in the soil solution (see below). Despite this accurate description effort, introducing the radionuclides' respective  $K_D$  coefficients, as derived from *in vitro* sorption/desorption experiments, produces calculated migration profiles which are underestimated for  $^{137}\text{Cs}$ , and overestimated for  $^{90}\text{Sr}$  (Figure 5). This suggests that migration is not governed by the soil traditional physico-chemical features only, as characterised by the *in vitro* radionuclides  $K_D$  coefficients. Additional soil properties are most probably of importance. The soil structure, which refers to the soil particles degree of aggregation, largely driven by the soil biological

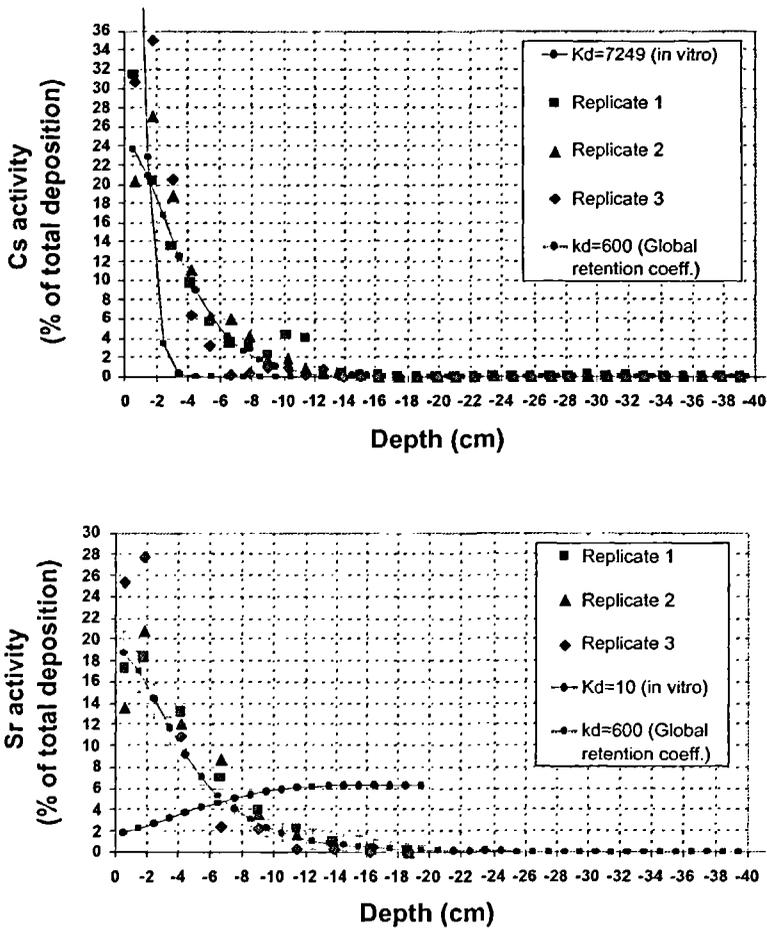


Figure 5  
 Model-based predictions of <sup>137</sup>Cs (up) and <sup>90</sup>Sr (down) migration against experimental data (three replicates).

activity present in the surface layer (microbial and root activity), is a good candidate. When taken into account in addition to processes inferred by the traditional  $K_D$  coefficients, the model predicts an “*in situ* global retention coefficient” of about  $600 \text{ l.kg}^{-1}$  (noted  $k_d$ ), which best fits the experimental profiles for both radionuclides in the first 15 cm soil layer (Gonze and Perrier, 1999).

## Soil influence on $^{137}\text{Cs}$ and $^{90}\text{Sr}$ root transfer to annual crops

Once contamination has reached the soil, long term pollution of plant food products occurs essentially via root uptake. Prediction models have first tackled this problem by applying a “transfer factor” coefficient, similar to a “concentration ratio” defined as the ratio between the specific activity in the plant and the specific activity in the soil. This empirical approach, however, suffers from high and poorly documented variabilities, which can largely be accounted for by the plant species considered, but also by the complexity and diversity of the soil compartment (IUR, 1989; Eriksson and Rosen, 1991). This is illustrated by the  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  concentration ratios (CR) which have been simultaneously measured on the five soil types for three different plant consumables: barley grains, bean pods and lettuce (Figure 6). For  $^{90}\text{Sr}$ , the classification of the observed Concentration Ratios (CR) lies in general agreement with the soils’ respective CEC values. For this radionuclide, the CR values are also 2 orders of magnitude larger than for  $^{137}\text{Cs}$ , irrespective of the plant considered, illustrating its larger mobility in agreement with the chemical availability studies previously mentioned. For  $^{137}\text{Cs}$ , however, the soils physico-chemical characteristics of relevance (such as RIP and % clay) do not fully explain the observed CR values. Whilst confirming previous observations (Frissel *et al.*, 1990; Smolders *et al.*, 1997), such characteristics are not sufficient to establish an accurate prediction of  $^{137}\text{Cs}$  transfer to plants. This lack of agreement results from complex variations of the soil solution composition which need to be elucidated. In particular, the soil water composition does not only depend on soil type, but also on a number of other parameters such as soil moisture, climate, fertilization (particularly with K and Ca) and root uptake activity.

Elucidating the parameters which influence the “soil solution” composition is of paramount importance since this is the very place where roots absorb radionuclides along with dissolved mineral nutrients. Current predictive assessment models usually involve the soil-specific  $K_D$  concept (soil solid/liquid partition coefficient) which reflects the radionuclide retention on the solid matrix (sorp-

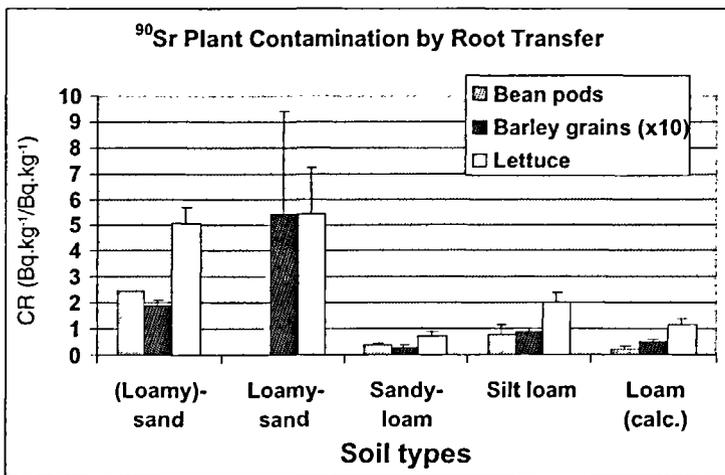
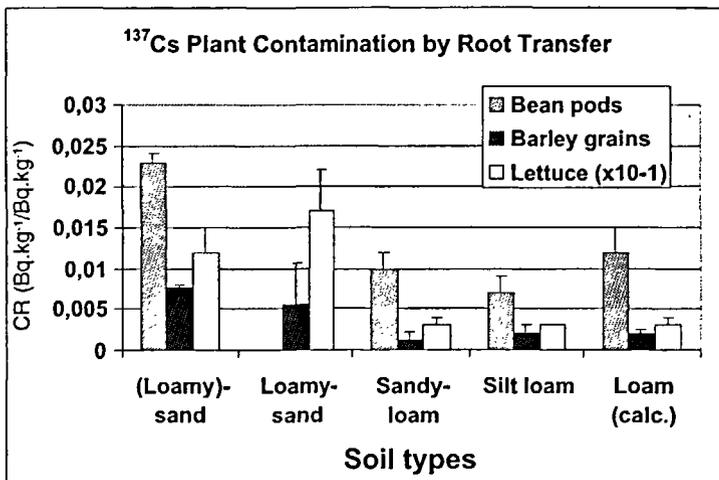


Figure 6  
<sup>137</sup>Cs and <sup>90</sup>Sr root transfer (as concentration ratios) obtained on five distinct agricultural soils for lettuce, bean pods and barley grains.

tion potential of a given soil) with respect to its solubility in interstitial water (potential availability to plant roots). The large sorption rate of  $^{137}\text{Cs}$  on most mineral soils determine trace concentrations in interstitial water. The radionuclides dissolved concentrations in soil water are further influenced to a large extent by the abundance of other ionic nutritive elements. As previously mentioned, this roots from the often reported competition occurring between Cs and K for adsorption on the FES of clay particles (Smolders *et al.*, 1997; Delvaux *et al.*, 2000; Thiry *et al.*, 2000), or between Sr and Ca+Mg for adsorption on the argilo-humic complex. However, inherent limits to adequately describing the soil water composition also arise from the equilibrium nature of the  $K_D$  coefficient, which is usually determined in water saturated conditions. Such conditions are rarely encountered in real nature, due, for example, to the continuous pumping activity of the roots or to climate-driven variations in soil moisture.

A previous investigation on pasture plants in northern Germany indicated that Cs and Sr root uptake were controlled by their concentrations in soil water which were reported to increase at reduced soil moistures (Kirchner and Ehlken, 1997). Based on a series of *in vitro* experiments carried out on samples from the soils considered in the present study, soil moisture is indeed demonstrated to markedly influence the radionuclides concentrations in soil water. For example, reducing soil moisture promotes an increase in Cs concentration, thereby reducing its  $K_D$  (Figure 7), an effect which in a first approach would tend to favour root uptake. However, the K concentration is also increased in parallel, to an extent proportionally even greater than for Cs (Cs/K lower at reduced soil moisture), hence promoting a larger competition with Cs for fixation on clay minerals. Similar observations have been obtained for  $^{90}\text{Sr}$  and Ca+Mg. When considering the soil matrix-liquid interface only, the radionuclides availability to root uptake will therefore result from the combination of both effects.

In turn, when considering the interface between the root surface and the soil water, an increased K concentration in the soil water (as measured upon plant harvest) promotes a reduction of  $^{137}\text{Cs}$  uptake expressed as a concentration factor (Figure 8, with  $\text{CF} = \text{specific activity in plants} / \text{specific activity in soil water}$ ). Similar

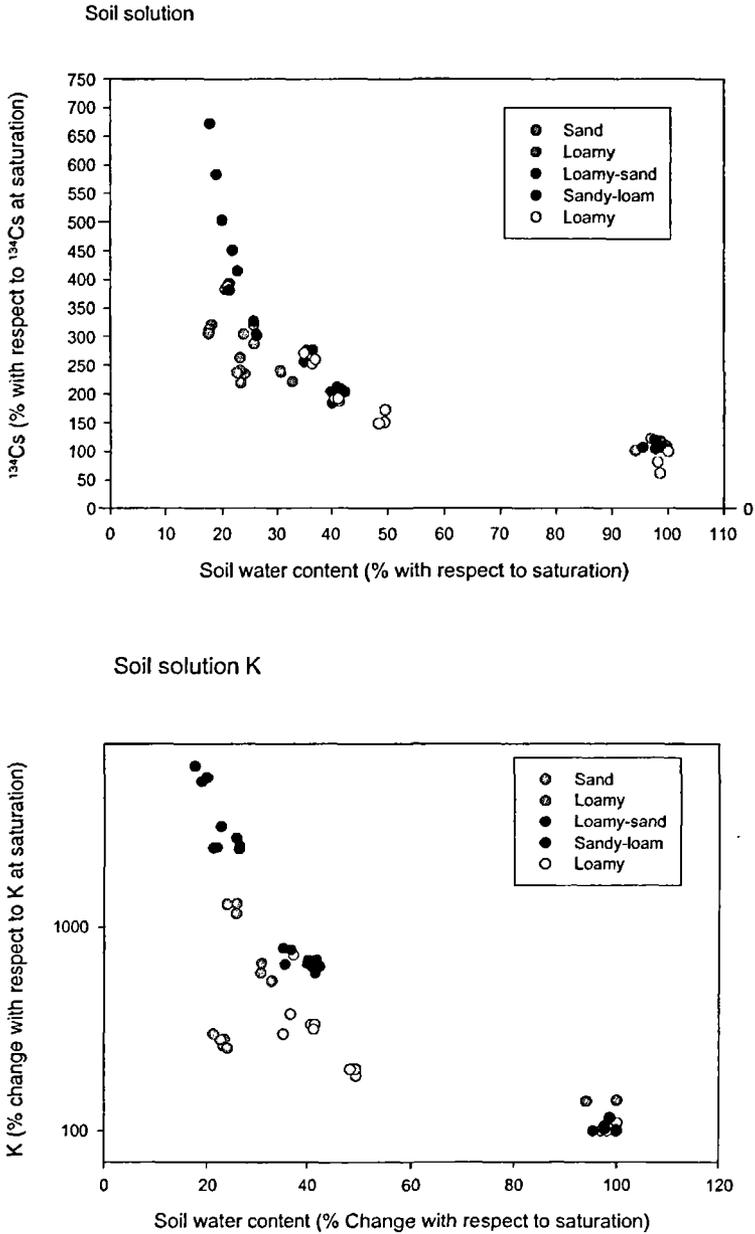


Figure 7  
 Influence of soil moisture on Cs (up) and K (down) concentrations in soil water.

observations have been obtained for  $^{90}\text{Sr}$  and  $\text{Ca}+\text{Mg}$ . The log-log plot of the  $^{137}\text{Cs}$  CF versus K concentration in soil water shows that this effect is more pronounced in the low range of K concentrations, most likely in soils with reduced fertility. Having been obtained from measurements on different soil types, the correlation between CF and the concentration in analogue ionic species suggests that plant contamination is primarily susceptible to the status of analogues in the soil water, irrespective of the soil type. Recent studies undertaken on a wide spectrum of different soil types, either mineral or organic, have established a similar competition promoted by K in soil water for root uptake of  $^{137}\text{Cs}$ , an effect most pronounced below 1mM K (Smolders *et al.*, 1997; Sanchez *et al.*, 1999). This agrees with the principle of radionuclide competition with its analogue ion occurring at the root level for membrane-based ionic transport systems.

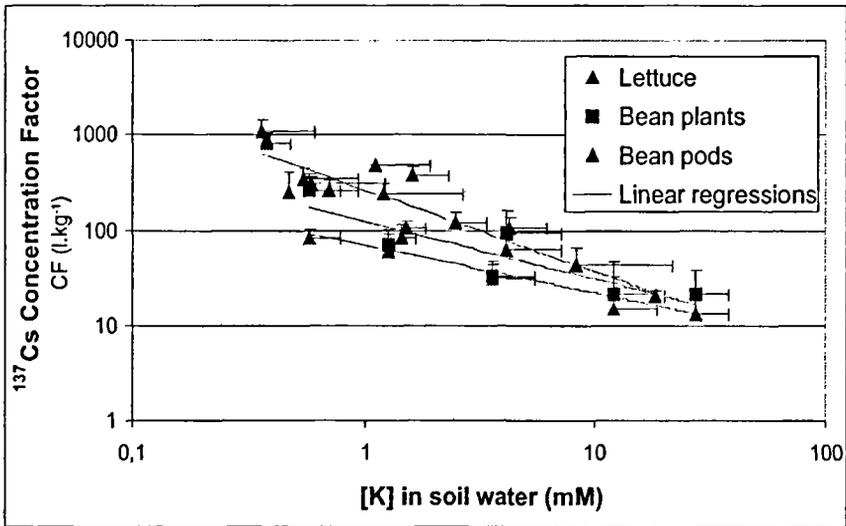
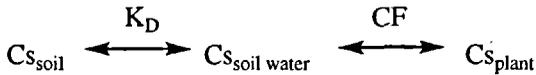


Figure 8  
Influence of the K concentration in soil water on the  $^{137}\text{Cs}$  Concentration Factor in lettuce and bean.

When considering finally the overall soil-plant system, it has been suggested that plant uptake could be described, provided some assumptions, by a series of two reactions (as for Cs, Smolders *et al.*, 1997):



From this model, the overall transfer factor (expressed as the concentration ratio  $CR = C_{s_{\text{plant}}} / C_{s_{\text{soil}}}$  [specific activity in plant] / [specific activity in soil]) can be derived:  $CR = CF / K_D$ . CF (plant root/soil water interface) and  $K_D$  (soil matrix/soil water interface) are both depending on the K status of soil water, but with opposite trends. In particular, when increasing K in soil water, the Cs availability at the soil matrix-liquid interface ( $K_D$ ) is increased (a feature which would favour root uptake) concurrently with a stimulation of the competition at the root level which tends to reduce root uptake. The ultimate plant contamination rate results therefore from the complex combination of these two interface-related processes, further explaining why CR is not easily linked to soil properties. This demonstrates the central role played by the soil water chemical composition, not only with respect to the trace amounts of radionuclides, but also with regard to their ionic analogues (K, Ca+Mg), as a key element contributing to the variability of the observed transfer factors. In other words, an accurate prediction of the soil water composition, as experienced by the roots of the plants during growth, will most probably resolve a large part of the transfer factors variability.

### *Kinetics of $^{137}\text{Cs}$ and $^{90}\text{Sr}$ transfer to crops via root uptake.*

Further to the influence of the soil parameters (such as texture, moisture, analogue ions availability, ...etc), the soil water composition is also influenced by the root uptake activity itself, as governed by the plant physiological requirements for homeostasis. Primarily designed to supply such nutrients as K and Ca to the plants in amounts suitable to support appropriate growth, the root uptake

activity is regulated by the plants physiological requirements. This can be illustrated by the variations during the growth cycle of the Cs accumulation rate in an annual crop (barley, Figure 9). Based on the finding that  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  are withdrawn respectively by the K- and Ca-membrane transport mechanisms, an analytical model describing Root Uptake of Radionuclides (RUR, Casadesus, 1999; Casadesus *et al.*, 1999) has been developed with particular emphasis on the depletion zone in the rhizosphere. This model accurately simulates the observed kinetics of radionuclides accumulation in plants. By integrating the physiology of plant growth, and in particular the regulatory mechanisms which overcome potential drastic depletion of nutrients in the root vicinity, this model accounts for the plant's ability to adjust the rate of supply to its demand.

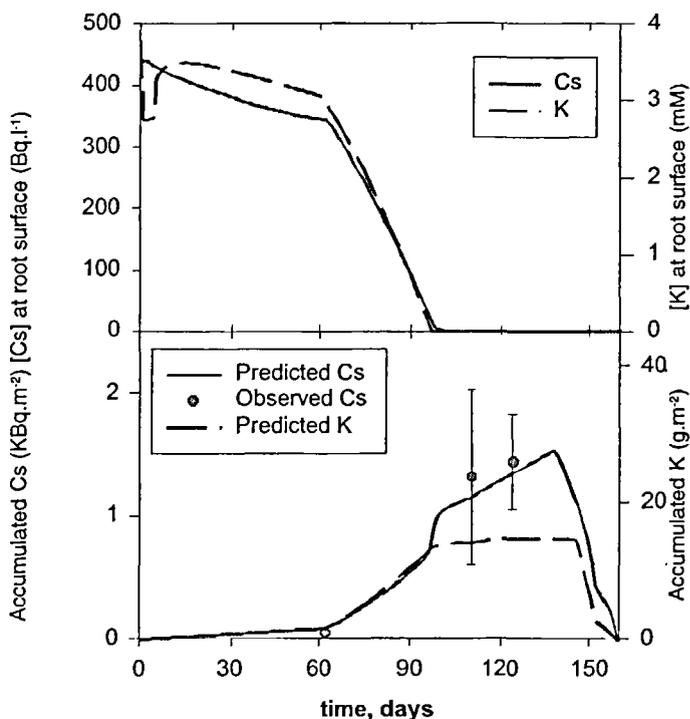


Figure 9  
Cs and K evolution at the root vicinity during a barley crop,  
and their observed /predicted accumulation in plants.

In addition to spatial variability, most often associated to soil type, such an in-year seasonality also contributes to puzzling the accurate determination of the longer-term contamination evolution within perennial vegetation, a prerequisite for valid long-term predictions. This is illustrated by the  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  activity concentrations in a rye-grass crop, grown on a silt-loam (Jülich) and a sandy loam soil (Wellesbourne), which have been followed up to 4.5 years after the contaminating deposition (Figure 10). Already present upon deposition, the contamination of the crop grown on the sandy loam (Wellesbourne) exhibits a rate of decline which can be decomposed into several successive phases with distinct characteristic times. For  $^{137}\text{Cs}$  contamination, the fast initial rate of decline during the first few months following deposition is attributed to foliar transfers as affected by rain wash-off and growth-mediated dilution. During this initial phase, the  $^{137}\text{Cs}$  contamination absorbed by the foliage is also translocated towards the roots. This root-accumulated pool will next be discharged to the rest of the plants, therefore promoting a slower rate of decline in the vegetation up to 2 years after contamination. Interestingly,  $^{90}\text{Sr}$  does not exhibit such an intermediate phase due to its reduced mobility within plants which prevents a similar pool

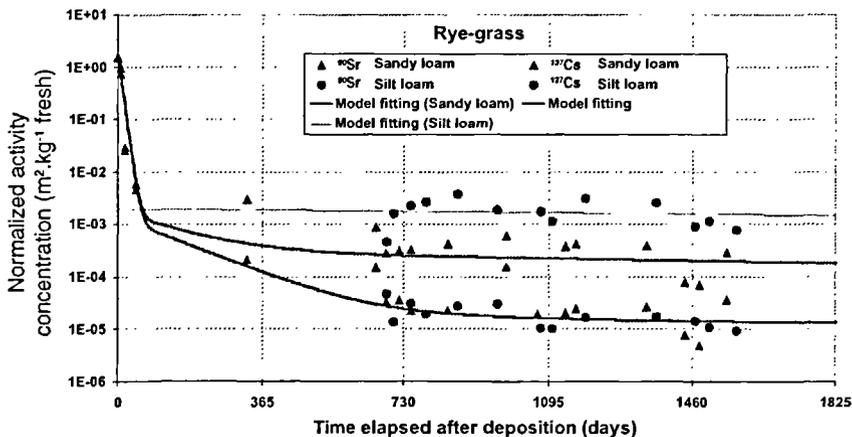


Figure 10  
 $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  contamination evolution in Rye-grass during 4.5 years after deposition, and current model fitting of experimental values (ASTRAL Model).

build up within the roots. The last phase, from 2 years after deposition and further on, shows a contamination evolution which only comes from root uptake, and whose very slow rate of decline is usually attributed to migration and gradual irreversible fixation (both on top of physical decay). During this phase, within-year seasonal variations are particularly clear for both,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , on the silt loam soil (Jülich), where rye-grass had been sown after deposition (therefore ruling out any contribution from foliar transfers). This illustrates the difficulty to experimentally determine the long-term kinetics of contamination decline in the vegetation.

## Conclusions and perspectives

PEACE constitutes the last piece of a series of international scientific programmes designed around the IPSN-controlled lysimetric environmental facility over the past decade. Focused on the behaviour of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , when accidentally released in the environment, these experimental investigations have generated a number of duly documented data (Bréchignac *et al.*, 1999). These are precious both, in enriching current radioecological data base and in providing a better understanding of mechanisms and processes, a key to making prediction models more accurate. The experimental designing of the soil-plant systems, as instrumented lysimetric soils in well-controlled climatic conditions, has allowed the use of a purely deterministic and mechanistic approach to modelling, because of both, the smaller space and time scales, and the reduced uncertainties on experimental data. Basically, such an approach has enabled the detailed study, with a relative accuracy, of the interactions between the various bio-physico-chemical processes of relevance, and their respective influence upon the contaminant behavior within such ecosystems. A sound scientific basis has now been reached, with prefiguration of appropriate research directions, in order to further elucidate the radionuclide "bioavailability" concept, which needs to move beyond the often misleading  $K_D$ -based formulation. Further work will address this particular issue.

## Bibliography

- BRÉCHIGNAC F., COLLE C., HUGON J., MADOZ-ESCANDE C., RONGIER P., SANCHEZ A., 1996 —  
 "RESSAC: A research facility for studying radionuclides behaviour within ecosystems". In *Proceedings of IRPA International Congress on Radiation Protection*, Vienna, Austria, April 14-19, 1996, 3: 137-139.
- BRÉCHIGNAC F., RONGIER P., HUGON J., DUBOIS E., COLLE C., MADOZ-ESCANDE C., 1998 —  
 "Controlled lysimetric environmental facility at IPSN: assessing accidental radioactive contamination". In *Proceedings of International Meeting on Influence of Climatic Characteristics upon Behavior of Radioactive Elements*, Rokkasho, Aomori, Japan, October 14-16, 1997: 35-42.
- BRÉCHIGNAC F., VALLEJO R.V., SAURAS YERA T., CASADESUS J., THIRY Y., WAEGENEERS N., FORSBERG S., SHAW G., MADOZ-ESCANDE C., GONZE M.-A., 1999 —  
*PEACE : Programme for Evaluating the impact of Accidents Contaminating the Environment-Soil-radionuclides processes of interaction and modelling of their impact on contamination of plant food products*. Final Report, European Atomic Energy Community Contract F14-CT96-0039a. (IPSN-DPRE/SERLAB Report 99-017(P)).
- CASADESUS J., 1999 —  
*Root uptake of radionuclides – Model description*. Technical Note, European Atomic Energy Community Contract F14-CT96-0039a.
- CASADESUS J., SAURAS YERA T., ROCA M. C., VALLEJO V. R., 1999 —  
*Root uptake model validation*. Technical Note, European Atomic Energy Community Contract F14-CT96-0039a.
- DELVAUX B., KRUYTZ N., CREMERS A., 2000 —  
 Rhizospheric mobilization of radiocaesium in soils. *Environmental Science and Technology*, 34: 1489-1493.
- ERIKSSON A., ROSEN K., 1991 —  
 "Transfer of caesium to hay grass and grain crops after Chernobyl". In Moberg L. (ed): *The Chernobyl fallout in Sweden – Results from a research programme on environmental radiology*, Stockholm, The Swedish Radiation Protection Institute: 291-304.
- FORSBERG S., ROSÉN K., FERNANDEZ V., JUHAN H., 2000 —  
 Migration of <sup>137</sup>Cs and <sup>90</sup>Sr in undisturbed soil profiles under controlled and close-to-real conditions. *Journal of Environmental Radioactivity*, 50 (3): 235-252.
- FORSBERG S., ROSÉN K., BRÉCHIGNAC F., 2000 —  
 Chemical availability of <sup>137</sup>Cs and <sup>90</sup>Sr in undisturbed lysimeter soils driven under controlled and close-to-real conditions. *Journal of Environmental Radioactivity*, in press.
- FRISSEL M. J., NOORDIJK H., VAN BERGELK K. E., 1990 —  
 "The impact of extreme environmental conditions, as occurring in natural ecosystems, on the soil-to-plant transfer of radionuclides". In Desmet G., Nassimbeni P., Belli M. (eds) : *Transfer of radionuclides in natural and semi-natural environments*, Brussels and Luxembourg, Elsevier Science Publishers: 40-47.
- GONZE M.-A., 1999 —  
*A physically-based model for simulating the soil-to-plant*

transfer of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  radionuclides: the TRANSOL code (updated release version 2.0), Technical Note, European Atomic Energy Community Contract F14-CT96-0039a.

GONZE M.-A., PERRIER T., 1999 — Numerical simulations of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  migration: TRANSOL versus lysimeter experiments. Technical Note, European Atomic Energy Community Contract F14-CT96-0039a.

IUR, 1989 — Workgroup of the International Union of Radioecologists on soil-to-plants Transfer Factors. Report VI. RIVM, Bilthoven, The Netherlands.

KIRCHNER G., EHLKEN S., 1997 — "Climatic influence on root uptake of Cs and Sr by pasture plants: evidence from field experiments in northern Germany". In Ohmomo Y., Sakurai N. (eds) : *Proceedings of International Meeting on Influence of Climatic Characteristics upon Behaviour of Radioactive Elements*, Aomori, Japan, Institute for Environmental Sciences: 83-90.

MADOZ-ESCANDE C., BRÉCHIGNAC F., COLLE C., DUBOIS E., HUGON J., JOUGLET H., MOUTIER M., RONGIER P., SANCHEZ A., SCHULTE E. H., ZANON R., 1999 — Experimental installation for radioecology research on defined ecosystems subjected to contamination in controlled conditions. *Nuclear Science and Engineering*, 133: 178-191.

MAES E., DELVAUX B., THIRY Y., 1998 — Fixation of radiocaesium in an acid brown forest soil. *European Journal of Soil Science*, 49: 133-140.

MAES E., ISERENTANT A., HERBAUTS J., DELVAUX B., 1999 — Influence of the nature of clay minerals on the fixation of radiocaesium traces in an acid brown earth-podzol weathering sequence. *European Journal of Soil Science*, 50: 117-125.

SANCHEZ A. L., WRIGHT S. M., SMOLDERS E., NAYLOR C., STEVENS P. A., KENNEDY V. H., DODD B. A., SINGLETON D. L., BARNETT C. L., 1999 — High plant uptake of radiocaesium from organic soils due to Cs mobility and low soil K content. *Environmental Science and Technology*, 33: 2752-2757.

SAURAS YERA T., VALLEJO V. R., VALCKE E., COLLE C., FÖRSTEL H., MILLAN R., JOUGLET H., 1999 —  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  root uptake under close to real controlled conditions. *Journal of Environmental Radioactivity*, 45(3): 191-218.

SMOLDERS E., VAN DEN BRANDE K., MERCKX R., 1997 — Concentrations of  $^{137}\text{Cs}$  and K in soil solution predict the plant availability of  $^{137}\text{Cs}$  in soils. *Environmental Science and Technology*, 31: 3432-3438.

STRAND P., BALONOV M., SKUTERUD L., HOVE L., HOWARD B., PRISTER B. S., TRAVNIKOVA I., RADNIKOV A., 1996 — "Exposures from consumption of agricultural and semi-natural products". In Karaoglou A., Desmet G., Kelly G.N., Menzel H.G. (eds): *The Radiological Consequences of the Chernobyl Accident*, EN-Luxembourg, Eur 16544: 261-269.

SWEECK L., WAUTERS J., VALCKE E., CREMERS A., 1990 — "The specific interception potential of soils for radiocaesium". In Desmet G., Nassimbeni P., Belli M. (eds): *Transfer of radionuclides in natural and semi-natural environments*, Brussels and Luxembourg, Elsevier Science Publishers: 249-258.

THIRY Y., KRUYTS N., DELVAUX B., 2000 — Respective horizon contributions to cesium-137 soil-to-plant transfer: a pot experiment approach. *Journal of Environmental Quality*, 29, in press.

TORMOS J., JOUVE A., REYV D., MILLAN-GOMEZ R., ZANON R., ERARIO M. J., 1995 —

A rapid method for determining strontium-90 in contaminated samples of soil and plant. *Journal of Environmental Radioactivity*, 27(3): 193-206.

WALTERS J., SWEECK L., VALCKE E., ELSEN A., CREMERS A., 1994 — Availability of radiocaesium in soils: a new methodology. *The Science of the Total Environment*, 157: 239-248.