INSTITUT DE PROTECTION ET DE SÛRETÉ NUCLÉAIRE

LONG-TERM ENVIRONMENTAL BEHAVIOUR OF RADIONUCLIDES

François Bréchignac, Leif Moberg, Matti Suomela

RECENT ADVANCES IN EUROPE

CEC-IPSN Association Final Report

Statens strålskyddsinstitut
Swedish Radiation Protection Institute
STUK

In December 1996, the Institut de Protection et de Sûreté Nucléaire (IPSN) signed an Association contract (F14P-CT96-0039) with the European Commission (EC), Directorate General Research.

Within the framework of the Association Contract, research in radiation protection was carried out in the fields covered by the Fourth Framework Programme on Nuclear Fission Safety. The contract running over a period of 36 months from January 1, 1997 to December 31, 1999, contained 3 projects, which were executed by a total of 18 partners from all over Europe including Norway as a non EU member state funded by its own government. The contract value amounted to 4,2 MECU, with the contribution from the commission of up to 2,3 MECU.

IPSN has born the overall responsibility for coordination of the work within three multinational projects. Each project was managed by a scientific project leader coordinating the scientific work ; Dr François Bréchignac, IPSN, FR, for the PEACE project, Dr Leif Moberg, SSI, SE, for the LANDSCAPE project and Dr Matti Suomela, STUK, SF, for the EPORA project. The implementation of the association contract was supervised by a steering committee consisting of EC and IPSN representatives. IPSN was providing the secretariat and administrative support to the committee. Chairmanship was assumed by Mr. Ezio Andreta, then Mr. Hans Forsström for EC and Pr. Henri Métivier for IPSN.

The association contract covered the following topic "Evaluation of radiation risks: quantification of parameters which determine the fluxes of radionuclides in ecosystems, and long-term consequences of accidental contamination in semi-natural environments".

The overall objectives of the PEACE project aimed at assessing the impact of the radioactive contamination on agricultural environments by accidental releases of caesium and strontium radioisotopes from a nuclear power plant. Research efforts focused on studies of the soil-to-plant interface processes with special emphasis on the quantification of key parameters which influence and govern the fate of Cs and Sr in soils and their transfer to plant crops.

The objectives of the LANDSCAPE project were to obtain a basis for reliable assessments of the radiation exposure to man from radionuclides in plant and animal systems of semi-natural (forest) ecosystems in Europe under different time scales. The work included studies of the seasonal and annual variations of the radionuclide content in the main above-ground biological

components of different representative forest ecosystems, the estimation of radioactive caesium flow between these biological components and the soil, and quantification of the impact of classical forest management on the radionuclide contents and fluxes within the vegetation.

The objective of the EPORA project was to evaluate the potential effect of industrial pollution on migration of radionuclides in soil, on radionuclides association in different soil constituents and on transfer of radionuclides from soil to plants. The potential impact of industrial pollution on runoff of radionuclides as well as radiation doses has also been studied. Therefore, an assessment was made of the current pollution status in the Kola peninsula with particular emphasis on the Monchegorsk area in Northern Fennoscandia.

The integration of these three projects within this association created a unique opportunity to link research efforts together, by bringing about experimental data under laboratory conditions and research concerned with the production of field data bases, therefore allowing for a confrontation between the different approaches with their underlying concepts for modelling. This report shows the permanent effort of integration of the three projects and provides a good illustration of the european synergy. In this report an overview of the output results from this synergy is given. For more information on the three projects readers should refer to the final project reports listed at the end of this document.

The success of this integration is mainly due to the constant efforts of the three projects leaders, François Bréchignac, Leif Moberg and Matti Suomela. Working with them was a constant pleasure and a permanent enrichment.We wish to thank them for their European spirit along the three years and thanks to such scientists we can be confident in the future of the scientific Europe.

The association chairmen wish to gratefully acknowledge the three project leaders but also, the members of the association, Jean-Claude Barescut, Martine Contini, Gilbert Desmet, Laurent Le Yanc, Jack Sinnaeve, Martine Wauters, Ernst H. Schulte, Remi Seydoux, and more especially our secretary Stéphane Lorthioir and Martine Jacquard who prepared all the association meetings with her usual efficacy.

Pr. Henri MÉTIVIER
Director of Research IPSN

Mr Hans FORSSTRÖM
Head of Unit DG RTD-DII-3 EC

What have we learnt?

- It is of paramount importance to know the soil solution composition in order to understand radionuclide transfer to plants.
- On untilled mineral agricultural soils, the migration of ^{137}Cs and of ^{90}Sr remain similar, and the depth profiles are established early on, with very slow further evolution.
- On the forest podzolic soils studied, migration of ^{137}Cs is pronounced, with increased amounts in the mineral layer ten years after an aerial deposition.
- The classically used K_D, as determined *in vitro*, does not allow an acceptable prediction of migration. The initial soil structure and hydric conditions upon contamination are important.
- During the past decade, there is a general decrease of 60-80 % in ^{137}Cs concentrations in ground vegetation, fungi, mosses and lichens at both temperate and boreal forest sites studied.
- More than a decade after Chernobyl, the total inventory of ^{137}Cs is still rising in pine trees of boreal forests.
- The forest models currently developed yield agreeing estimates of the ^{137}Cs concentrations in the different compartments of the ecosystem, taking natural variability into account.
- An increasing load of heavy-metal pollution in boreal forests modifies the radionuclides status in podzolic soils and their subsequent transfer to plants.
- Fertilisation can be a beneficial tool for restoring contaminated forests, as well as for the remediation of the state of agricultural lands in severe fallout situations.
- There is almost no ^{137}Cs loss via runoff water from boreal forest ecosystems except from the wetter portions of bogs.
- Spatial variations in radionuclide deposition are dominant and largely cancel out evolution over time and the influence of climatic intermittence.
- Variability of ^{137}Cs uptake by moose is caused more by spatial variation in deposition and the normal movements of the animal than by variations in diet selection or intake.
- The ^{90}Sr contamination of plant food products is important for internal dose assessment and may be affected by additional non-radioactive pollution.


EPORA
Norway
PEACE
LANDSCAPE
United
Kingdom
PEACE
LANDSCAPE
Belgium
EPORA
Germany
France
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Spain
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Kola Peninsula

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Italy

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Leif Moberg: Swedish Radiation Protection Institute, SSI, Sweden.
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Moose photo on cover page: Per Claesson, Bildhuset

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The widespread radioactive environmental pollution which resulted from atmospheric nuclear weapons testing during the mid-years of this century prompted the first concern among the scientific community about its potential impact on human health. Although the development of the civilian nuclear industry has evolved with a parallel effort of mastering the associated risks, the Chernobyl accident in 1986 revealed, on a global scale, the radiological impact of radioactive pollution of the environment. Assessing the resulting radiation doses that humans might receive, either by exposure from radionuclides on the ground or by internal ingestion (via food), requires a good understanding of the long-term behaviour of radionuclides in the environment. This knowledge constitutes a basis for the prevention goals of radiation protection.

This document reports on a joint European effort to advance this understanding, placing particular emphasis on the soil-plant system of semi-natural ecosystems and agricultural land. The problem has been tackled through various converging and innovative approaches to the long-term behaviour of the radionuclides ^{137}Cs, ^{90}Sr and $^{239/240}Pu$. The approaches range from *in situ* experiments, designed to exploit past contamination events, to laboratory simulations in controlled conditions of accidental contamination, in view of mastering the wide variability of the natural environment. Particular attention has been devoted to the philosophy of experimental data acquisition in order to secure the various modelling developments undertaken with appropriate validations. A broad scope of different ecosystems, ranging from arctic and boreal regions down to Mediterranean ones, has been considered, with emphasis on forests and agricultural land, where food production takes place. Up to 22 different site locations scattered throughout this territory have been investigated, reflecting a wide, and representative, variety of European soils and climatic conditions. A brief description of the various ecosystems and soils considered is given in Table 1. Table 1

Long-term behaviour of radionuclides in these ecosystems are most susceptible to their vertical distribution and migration in the soils, as well as to their availability and uptake by plants and animals, processes which have been examined particularly closely, within time-frames ranging between a few years to several decades. Finally, the occurrence of additional non-radioactive pollutants in many ecosystems, which could invalidate our current prediction pertinence, has been questioned for the first time with respect to their potential influence on radionuclide movements.

This report is focused on six different themes, each of which having been studied at least in two of the three projects: *redistribution in the soil-plant system, modelling, countermeasures, runoff, spatial variations, and dose assessment*. More extensive information on the project results can be found in the final reports of the individual projects (see short reference list).

Table 1 : Main soil characteristics of the various ecosystem/site locations investigated.

Geographical Site	Ecosystem Type	Soil type Soil texture	Soil layer	Particle size % clay	% silt	% sand	pH	CEC (cmol/kg)	Organic matter %
Mol (Belgium)	Agricultural	Podzol Sand	P[1]	4.1	14.6	78.3	4.6[a]	11.7	3
Jülich (Germany)	Agricultural	Luvisol Silt loam	P[1]	11.2	78.8	8.4	7.1[a]	13.4	1.5
Barcelona (Spain)	Agricultural	Luvisol Loam calc.	P[1]	13.9	28.8	47.7	7.3[a]	16.4	2.3
Belleville (France)	Agricultural	Fluvisol Loamy sand	P[1]	5.8	13.7	79.0	4.1[a]	5.8	1.5
Wellesbourne (U.K.)	Agricultural	Fluvisol Sandy loam	P[1]	9.5	19.7	66.9	6.3[a]	17.8	3.9
Hille (Sweden)	Forest	Podzol	O[2]	n.d.	n.d.	n.d.	3.5[b]	n.d.	58.2
		Ferric	M[3]	1.9	8.9	89.2	4.2[b]		2.0
Långsjön (Sweden)	Forest	Podzol	O[2]	n.d.	n.d.	n.d.	3.6[b]	n.d.	43.8
			M[3]	3.7	28.1	67.5	4.1[b]		5.0
Prylen (Sweden)	Forest	Podzol	O[2]	n.d.	n.d.	n.d.	3.7[b]	n.d.	53.6
			M[3]	3.4	17.1	79.5	3.9[b]		5.1
Vindeln (Sweden)	Forest	Regosol	O[2]	n.d.	n.d.	n.d.	3.4[b]	n.d.	66.8
			M[3]	<11	26.1	69.7	4.4[b]		5.2
Liesineva (Finland)	Forest	Peatland	O[2]	n.d.	n.d.	n.d.	3.2[b]	n.d.	95.2
			M[3]						
Siikakangas (Finland)	Forest	Podzol	O[2]	n.d.	n.d.	n.d.	3.1[b]	n.d.	59.1
		Ferric	M[3]	2.6	17.5	66.3	4.1[b]		6.8
Lapland (Finland)	Forest	Podzol	O[2]	n.d.	n.d.	n.d.	3.0[b]	193	79
			M[3]	2.4	7.5	90.1	3.7[b]	24	3.2
Monchegorsk (Russia)	Forest	Podzol	O[2]	n.d.	n.d.	n.d.	3.0[b]	206	76
			M[3]	1.3	8.2	90.5	3.8[b]	30	8.6

[1] Tilled layer
[2] Organic layer
[3] Mineral layer

[a] pH (KCl)
[b] pH ($CaCl_2$)
n.d. Not determined

It should be borne in mind that forest podzols are fundamentally different from agricultural soils. Forest podzols present a clear multilayered vertical structure characterised mainly by a clay-poor mineral layer (sand) which supports an organic layer rich in organic matter (>50 %). In contrast, agricultural soils present reduced amounts of organic matter (a few %) associated with higher clay contents.

1 REDISTRIBUTION IN THE SOIL-PLANT SYSTEM

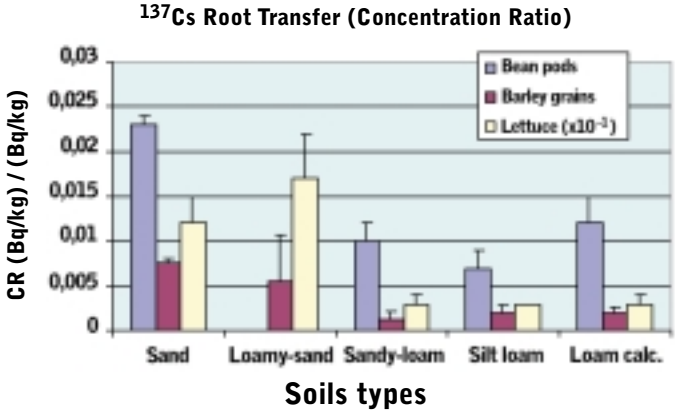
- *Agricultural ecosystems*
- *Forest ecosystems (podzols)*
- *Forest ecosystems submitted to additional pollution by heavy metals*

Agricultural ecosystems

On a contaminated soil, the long-term pollution of plant food products occurs via root uptake. Prediction models have first tackled this problem by applying a "transfer factor" coefficient 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. This is illustrated by the ^{137}Cs and ^{90}Sr transfer factors which have been measured simultaneously on five distinct soil types, with variable respective contents of clay, loam and sand, for three different plant consumables: barley grains, bean pods and lettuce. figure 1

These data have been obtained from long-term experiments carried out in a dedicated facility designed for simulating, in fully controlled conditions, an accidental deposition of radioactive aerosol contaminants. The facility hosts various soil monoliths sampled throughout Europe, which are monitored in lysimeters with advanced control over hydrological fluxes, and conducted under artificially reconstructed and computer-controlled climatic conditions.

The classification of the observed transfer factors does not always agree with soil physico-chemical characteristics alone. For ^{137}Cs in particular, this is due to complex variations of the soil solution composition, which is a function, not only of soil type, but also of a number of other parameters such as soil moisture, climate, fertilisation and root uptake activity. The "soil solution" composition is of paramount importance, since this is the very place where roots absorb radionuclides along with dissolved mineral nutrients. Predictions have been refined by introducing the "K_D" concept, defined as the radionuclide partition coefficient between the soil liquid and solid phases, reflecting either the immobilization potential of a given soil for a radionuclide (retention on the solid matrix), or its availability to plants (presence as solute in the liquid



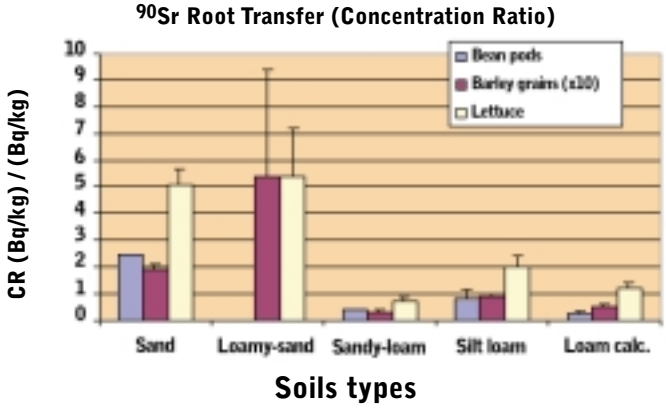


figure 1

^{137}Cs and ^{90}Sr root transfer (as concentration ratios, Bq.kg^{-1} in plants /Bq.kg^{-1} in soil) obtained on five distinct agricultural soils for lettuce, bean pods and barley grains.
PEACE

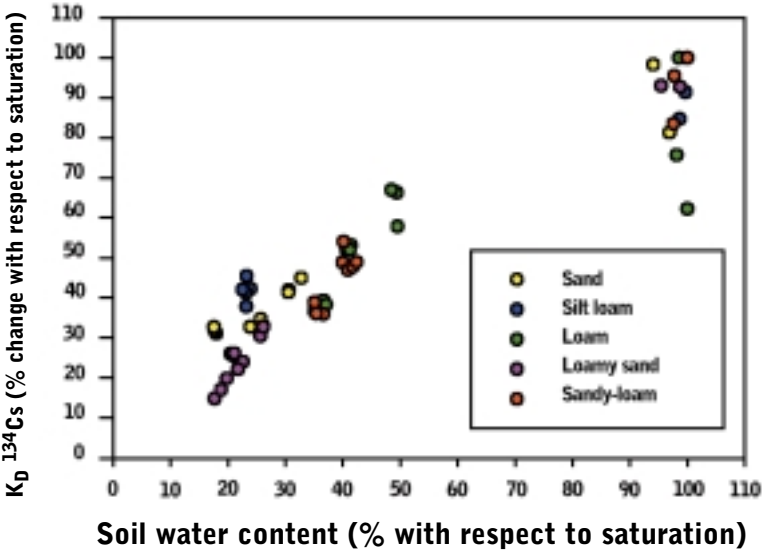


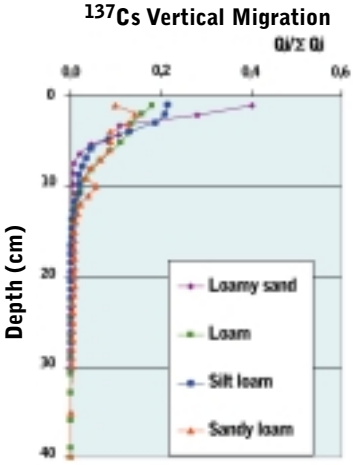
figure 2

Influence of soil moisture on K_DCs.
PEACE

phase). This approach has made it possible to take into account the well-known competition occurring between Cs and K for fixation on clays, or between Sr and Ca for adsorption on the argilo-humic complex, which both influence the soil solution content. Limits are imposed on this concept by the equilibrium nature of the K_D coefficient, which is further often determined in water-saturated conditions. Such conditions are rarely actually encountered in nature, due for example to the continuous pumping activity of the roots or climate-driven variations in soil moisture.

Based on a series of *in vitro* experiments, soil moisture is indeed demonstrated to have a marked influence on the radionuclides concentration in the soil solution. For example, reducing soil moisture promotes an increase in Cs concentration, thereby reducing its K_D, tending to favour root uptake. figure 2

In parallel, the K concentration is also increased, to an extent proportionally even greater than for Cs (Cs/K lower for reduced soil moisture). Furthermore, considering the root uptake, it has been confirmed that increasing K in the soil solution reduces ^{137}Cs uptake due to competition for the transport systems. The ultimate ^{137}Cs root uptake, and subsequent plant contamination rate, results from a combination of these effects. This demonstrates the central role played by the soil solution composition as a key element contributing to the variability of transfer factors. This is further supported by modelling



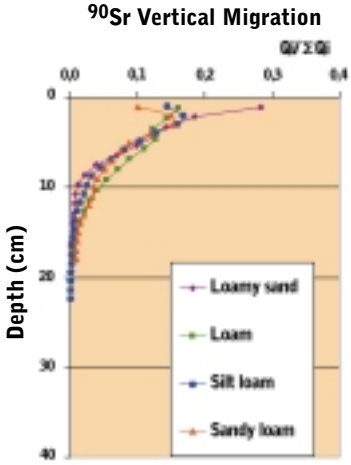


figure 3

^{137}Cs and ^{90}Sr migration profiles observed 4 years after contamination on various agricultural soils.
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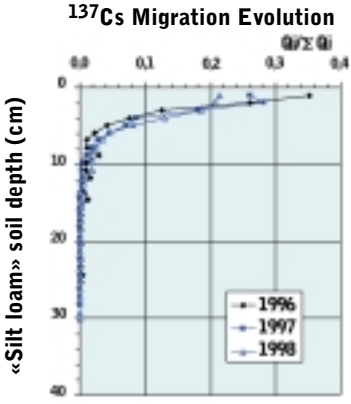


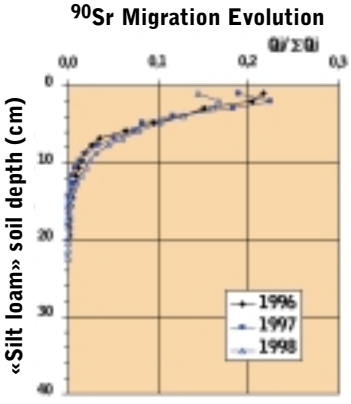


figure 4

^{137}Cs and ^{90}Sr migration evolution during three successive years on an untilled silt loam soil installed in an advanced lysimeter facility with hydrological and climatic control. Experimental contamination by aerosols was performed in 1994.
PEACE

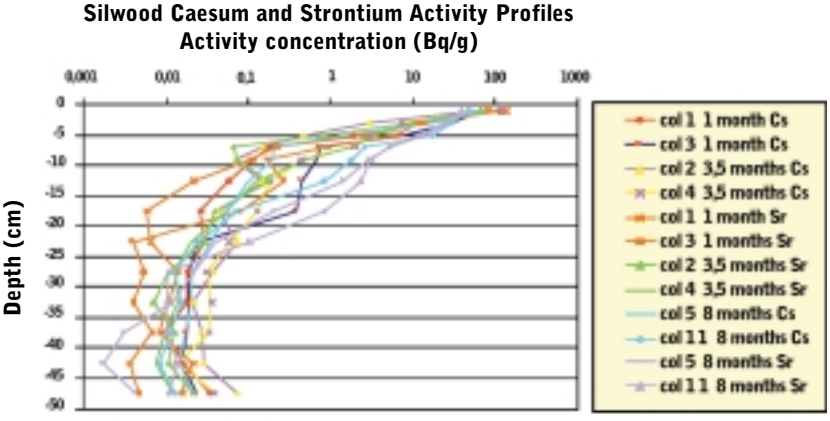


figure 5

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

developments of root uptake, which reveal that soil type and climatic conditions act on the soil-to-plant transfer essentially via the chemical composition of the soil solution and the physiological features of the plant: K and plant growth for ^{137}Cs transfer, Ca and transpiration for ^{90}Sr transfer.

When attempting to understand migration patterns, the first elements are furnished by chemistry, which indicates that the high potential mobility of ^{137}Cs, due to its significant solubility, is counteracted by its very specific and efficient capture within clay particles (illite, in particular). Conversely, the lower potential mobility of ^{90}Sr, due to its lower solubility, is not counteracted to the same extent by specific fixation sites on the soil matrix. This results in clearly distinct K_D values for the two radionuclides, as determined *in vitro*, from which a higher migration rate is expected for ^{90}Sr than for ^{137}Cs after several years. However, experimental observations do not fit this theoretical approach.

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 is traditionally characterized by a radionuclide-specific K_D coefficient, does not significantly alter the observed profiles. figure 3

On the other hand, the low rates of migration observed in the long term (around 1 cm/year in the years following contamination) cannot explain the shape of the profiles. figure 4

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 5

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. This aspect particularly deserves further consideration.

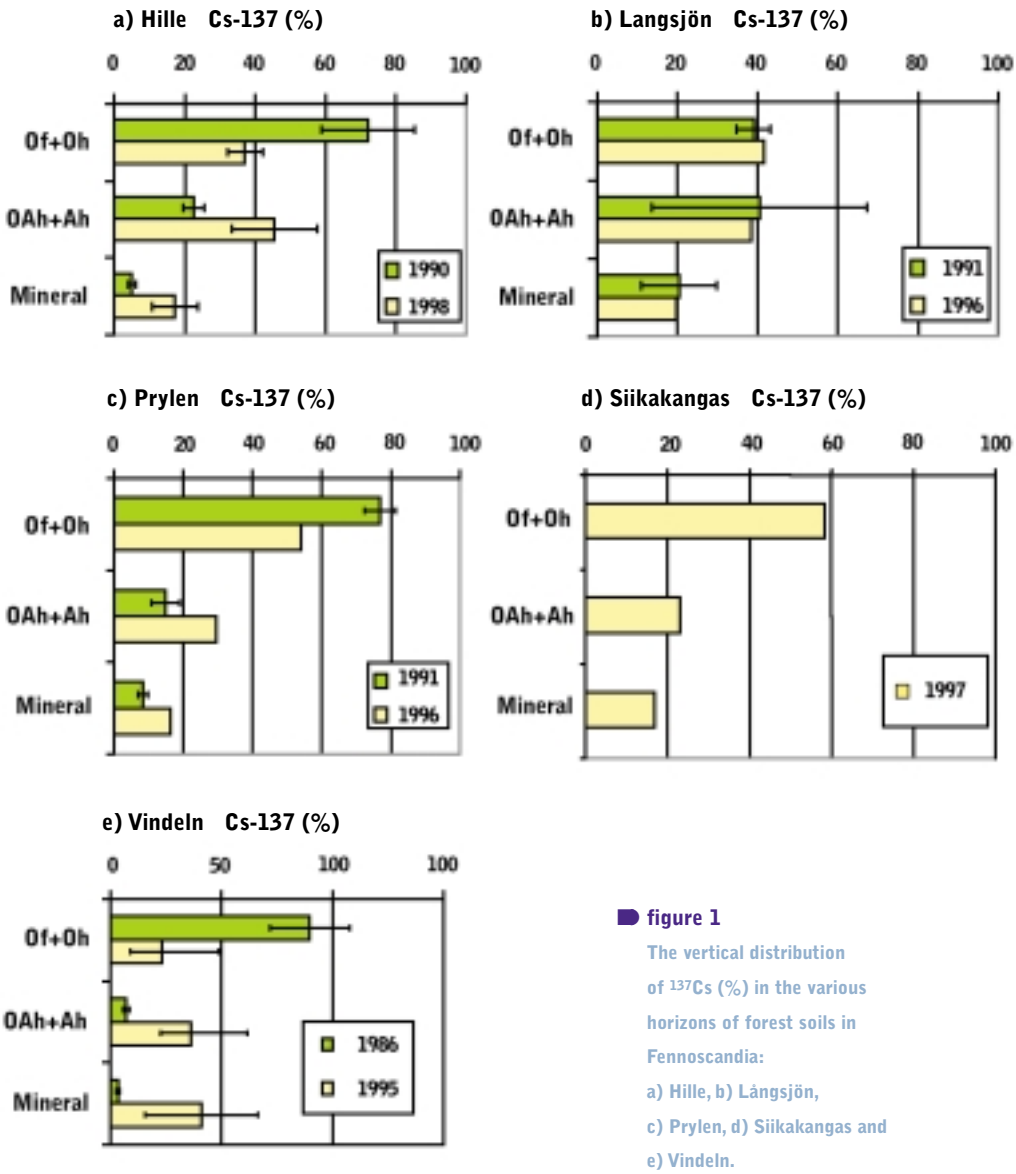


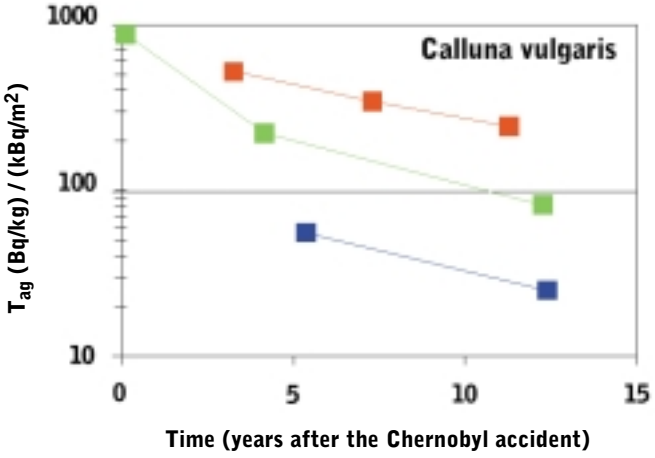
figure 1

The vertical distribution of ^{137}Cs (%) in the various horizons of forest soils in Fennoscandia:
a) Hille, b) Långsjön,
c) Prylen, d) Siikakangas and
e) Vindeln.
LANDSCAPE

Forest ecosystems (podzols)

In contrast to agricultural soils, the forest podzol soils are characterized by a multi-layer structure, or horizons: organic horizons (Ol=non-fragmented litter, Of=fragmented organic matter, Oh=humified organic matter and the OAh and Ah horizons which are organic-rich mineral horizons), and mineral horizons (A and B). From studies after the Chernobyl accident, it is known that most of the caesium during the first years was located in the raw humus layers. The vertical distributions of ^{137}Cs in podzol soils in the regions of this study show a trend towards redistribution from the vegetation and raw humus layers down to the mineral layer. figure 1 By 1995 to 1998 after deposition, between 20 and 40% of the original deposit had reached the mineral layer. Although there is a general trend to downward migration, the depth distribution varies between different locations. The soil particle distribution affects the hydraulic conductivity and capillarity. Dense soil with low hydraulic conductivity and high capillarity will probably be more effective in retaining caesium in shallow layers due to slow percolation, high ion-exchange capacity and an upward water flow during warm periods. Biotic factors may also lead to variations in soil microflora which may influence the caesium distribution. The depth distribution is further affected by differences in clay content in the mineral layer which, in areas with high contents, may trap caesium in the mineral fraction.

The vertical redistribution of radiocaesium in podzol soils is also reflected as a decreasing rate in photon fluence as measured by *in situ* gamma spectrometry. Between 1986 and the late 1990´s, the photon fluency at 662 keV decreased faster by a factor of at least two than what would have been expected from the physical half life of ^{137}Cs. Except for the exponential decrease, temporal fluctuations up to a factor of two were observed. These are due to variations in soil moisture and lead to lower external doses during winter, spring and autumn compared to the summer.



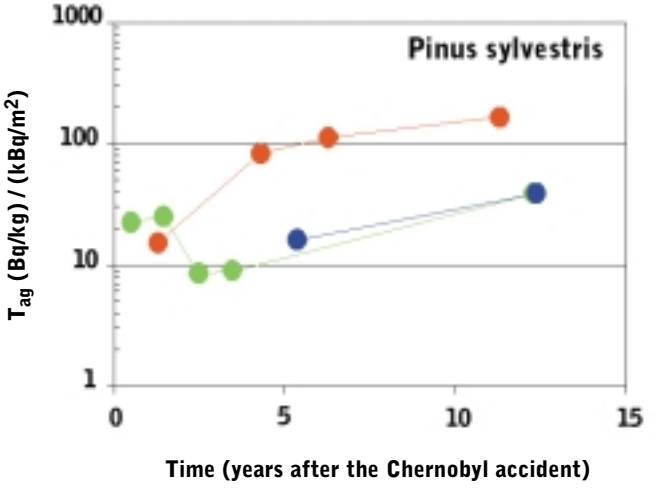


figure 2

Temporal changes of ^{137}Cs (Tag) in green parts of heather and current shoots of young Scots pine for three regions: Saint-Léger, Belgium (red); Hille, Sweden (blue) and Vindeln, Sweden (green).

LANDSCAPE

The changes in the vertical distribution of ^{137}Cs in soil have implications for plant root uptake, which, depending on species, can cause either an increase or a decrease in the uptake of caesium. At both boreal (Hille) and temperate sites (Belgium and Luxembourg), plants on average lost 70% of their activity over the past decade. In relative terms, however, major differences were observed in connection with this loss, which surprisingly tallied with the taxonomic families. For instance, most of the plants in the *Asteraceae, Fabaceae* and *Liliaceae* families were characterised by a loss exceeding 70%. In contrast, the loss in most of the grasses and *Rubus* species was considerably less than 70%. The loss in *Ericales* and ferns was in the middle range.

The relationship between rooting depth and radiocaesium migration may partly explain the differences in the contamination trends of plants. This is undoubtedly the most likely explanation in the case of the shallow-rooting *Oxalis acetosella* and in pine, which has a deep tap root. Furthermore, the contribution of successive cohorts of ever deeper-rooting mycorrhizal fungi in this species cannot be excluded. Most of the radiocaesium stored in the *Liliaceae* runners has probably been lost to the soil surface horizon, in which there has been a general decrease of ^{137}Cs due to migration. On the other hand, the fibrous rootsystem of grasses enables them to maintain firm anchorage in deeper humiferous horizons that are still relatively heavily contaminated. *Ericales* and ferns often have running roots. However, they also produce numerous, more or less fibrous, adventitious roots that explore deep soil layers. One should also bear in mind that these plants are mycotrophic, and therefore the buffer role of mycorrhizal fungi in the decontamination of these species cannot be precluded.

Aggregated transfer factors, T_{ag} ($Bq.kg^{-1}$ per $kBq.m^{-2}$) can be used to compare data from different sites. Such a comparison shows that the T_{ag} values in vegetation ranged from 0.2 to 241 (10^{-3} $m^2.kg^{-1}$). The lowest values were found in young plants of deciduous trees and in herbs, while the highest values were found in dwarf shrubs. On average, the general trend for ^{137}Cs in the vascular plants investigated from seven studied sites was: *Salix spp* < *Betula spp* < *Epilobium angustifolium* < *Deschampsia flexuosa* < *Pinus sylvestris* < *Vaccinium spp* < *Calluna vulgaris,* with the same variations between the sites.

A comparison of the long-term annual changes in aggregated transfer factors shows that the activity in heather *(Calluna vulgaris)* changed in a very similar way over time for the sites studied. figure 2

For Scots pine *(Pinus sylvestris)*, the T_{ag} values have increased dramatically since a few years after the deposit. Although the T_{ag} amplitudes are different, the rate of increase is equal between the sites, indicating (as in the case of *Calluna vulgaris*) that a general process is at work. This can be a protracted redistribution of Cs in the forest floor from the mosses and litter layers down to the humus and mineral soil, where pine roots are found. The monotonously increasing T_{ag} values in young pines need further investigation, since this may be of significance for external doses, internal doses via meat from moose and gallinaceous birds, for forestry in general (biofuels) and the steady-state levels for the different compounds in the forest ecosystem.

Mushrooms are the most heavily contaminated organisms in forest ecosystems and many of them play a direct and significant role in the contamination of food chains by radioactivity. The contamination and decontamination curves of mushrooms reflect the evolution of the activities in the various soil horizons. This characteristic has to be considered in the light of the specific use of soil horizons by the mycelium of these organisms. The data available indicate an overall reduction of ^{137}Cs concentrations in mushrooms between 1991 and 1998. When comparing the average activity, during this period, of obligate mycorrhizal species (usually characterised by a deep mycelium) with that of facultative mycorrhizal species (which have a more superficial mycelium), a reduction of 60% and 80% is observed, respectively.

A rough assessment of the proportion of caesium retained in fruitbodies in their growing area shows that the average value in *Cortinarius alboviolaceusis* was about 5% of the deposition, whereas it varied from 1.3 to 1.7% in other mushrooms. In mushrooms that have lost 70% of their contamination over the past seven years, one can estimate that at least 5% of the total deposition had been retained at the time of maximum fruitbody contamination. There is a need for continuing the investigations on the role of mycorrhizae in the active radiocaesium transfer from the soil to host plants.

Trees are often the most significant component in a forest ecosystem from an economical point of view. Concentrations of radiocaesium in different parts of trees vary, and are changing with time. The use of trees in forest industry and the question of access of people to heavily contaminated forests motivate analysing the distribution of man-made radionuclides in trees in the short and long term.
Root uptake adds to the radionuclide content of the tree roughly in proportion to growth intensity and thus increases the radiocaesium content of the tree which originates from metabolised, initially deposited, radiocaesium. This caesium will be found in both above-ground parts and in roots, and varies with the season of the year.

Growth dynamics cause fluctuation in radiocaesium contents in conifer needles of different ages during the growth period. Different growth conditions add to the variation between sites.

There is an overall increase in ^{137}Cs concentration in the stem wood of pine between 1991 and 1997. The concentration of ^{137}Cs in the new growth of twigs and needles of these trees figure 2 has also increased during the period between the two samplings, due to root uptake. The caesium concentrations are highest in the youngest twigs, which is consistent with other measurements. The total activity, in percentage of the ground deposition, stored in the above-ground parts of the trees of the older stand has increased from 1.1% in 1991 to 2.0% in 1997.

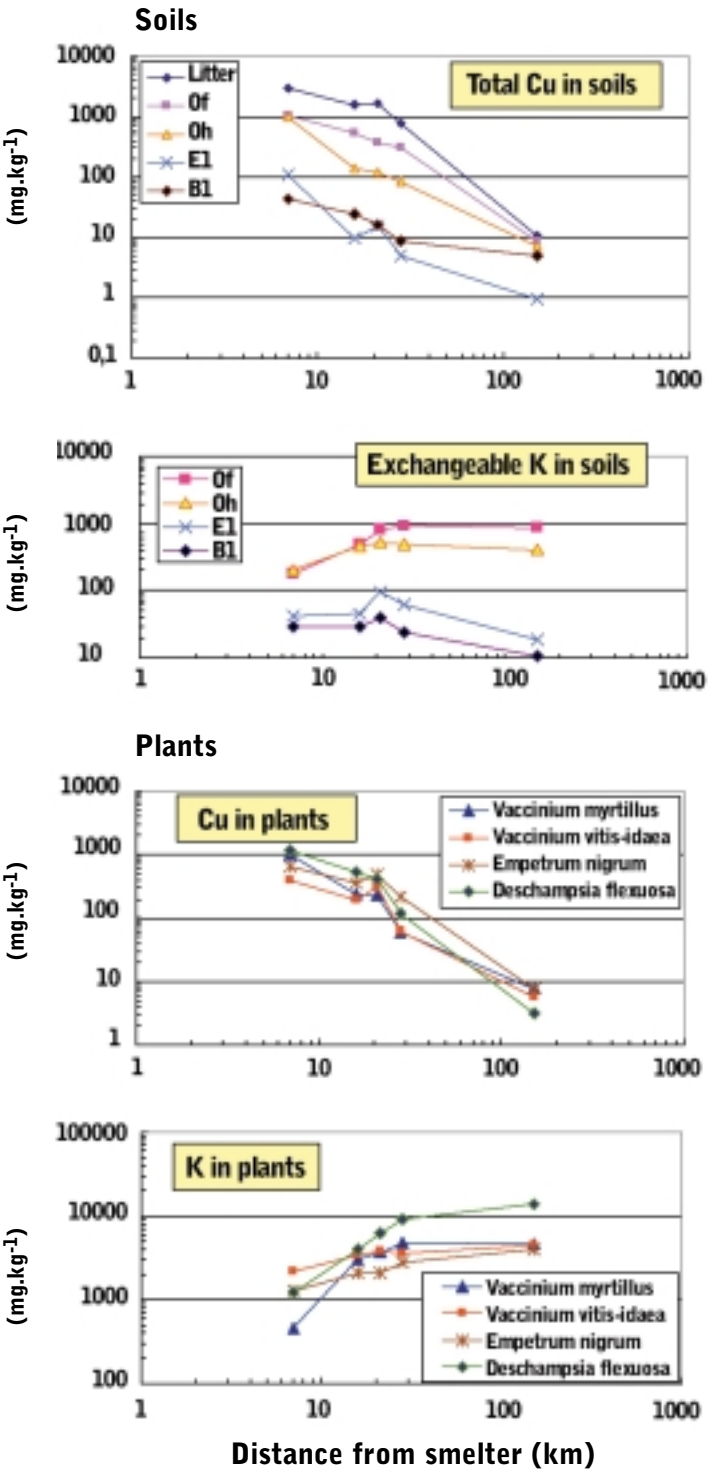


figure 1

Characterisation of the heavy metal pollution gradient generated around the smelter. Illustration of the soil contents in Cu and exchangeable K observed at 7 up to 152 km from the smelter, and the corresponding plant content in Cu and K as measured within 4 understorey plant species. EPORA

Forest ecosystems submitted to additional pollution by heavy metals

The Severonikel smelter at Monchegorsk, Kola Peninsula, has released huge amounts of SO_2, Ni and Cu into the atmosphere since its construction in 1938. As an example the annual emissions released in 1984 amounted to about 270,000 tons for SO_2, 3,200 tons for Ni and 2,400 tons for Cu. Annual emissions had been reduced by about one half in 1994 but although decreasing, considerable pollution of the surrounding environment has been going on. This pollution has caused a dramatic alteration in the vegetation status as observed in the vicinity of the plant.

A strong pollution gradient has been generated around the smelter, as illustrated in figure 1. The concentrations of heavy metals in the upper soil horizons (litter and O horizons) show a very sharp increase as distance from the smelter decreases (the reference site was located 152 km from the smelter). The total concentrations of Ni and Cu in the organic layer increase regularly, from about 10 $mg.kg^{-1}$ at the reference site up to about 5,000 $mg.kg^{-1}$ at the most polluted site. Near the smelter (7 km), concentrations in the underlying mineral horizons are also higher, indicating some vertical migration from the surface layers, but the trends observed indicate that only a very small fraction of the metals has leached to levels lower than the maximum sampling depth. The total amounts of nickel or copper at a given site may therefore be used to represent the integrated pollution effect at that site, which can be compared to the other sites. Only minor changes are observed in soil pH and cation-exchange capacity with distance from the smelter, their respective vertical distribution being usual for such podzols. The heavy-metal accumulation in plants displays a similar trend, with

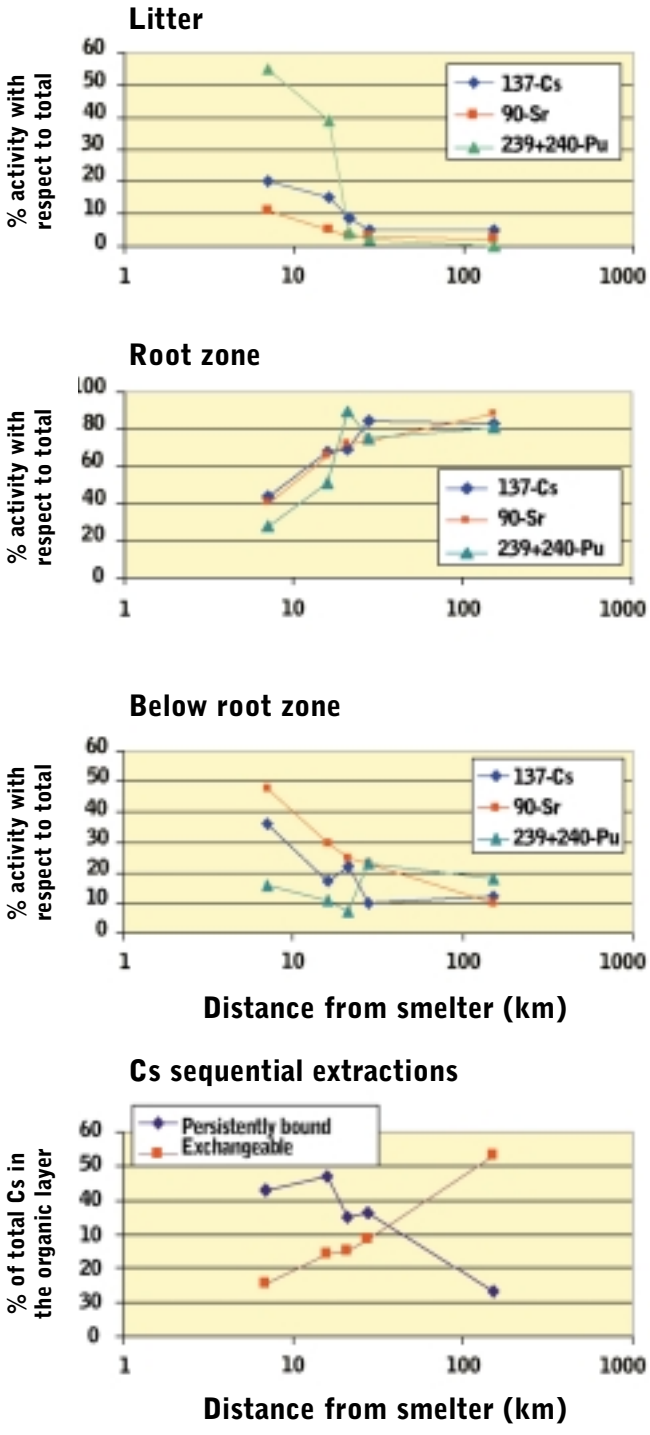


figure 2

Influence of heavy metal loads on the distribution of ^{137}Cs, ^{90}Sr and $^{239+240}Pu$ in the litter, the root zone (O and E horizons) and below the root zone, expressed as percentages relative to the total inventories in the overall soil profile.

EPORA

figure 3

Influence of heavy metal loads in the soil organic layer on the ^{137}Cs exchangeable and persistently bound fractions as determined through sequential extractions.

EPORA

higher concentrations in the vicinity of the smelter.

In view of this pollution gradient, it is of great interest to examine how the presence of heavy metals is affecting the soil nutrients status and their uptake by plants. Concentrations of exchangeable K, Mg, and Ca all exhibit a strong depletion in the O-layer (organic) with increasing pollution levels, whereas there is an increase in the lower horizons (E and B).

The corresponding contents of these nutrients within plants follow a trend similar to that observed for the exchangeable fraction in the O horizon, with highly depleted concentrations near the smelter. figure 1

The additional pollution by heavy metals (Cu and Ni) clearly affects the distribution of radionuclides (^{137}Cs, ^{90}Sr and $^{239+240}$Pu) in the various soil horizons. In the litter layer, the radionuclide content (expressed as a percentage of the total inventory in the overall soil profile) increases with decreasing distance from the smelter (higher load of heavy-metal pollution). This increase is especially sharp for $^{239+240}$Pu, which rises from 1% at the reference site up to 56% at the most polluted site. Conversely, in the root zone (O and E horizons), the radionuclide content decreases versus decreasing distance from the smelter. figure 2

These observations most likely result from an increased thickness of the litter layer at the most polluted sites (vicinity of the smelter), with a parallel thickness reduction in the organic layers (root zone), both being promoted by an inhibition (by heavy metals) of the microbial activity which converts litter into humic substances. Below the root zone, ^{90}Sr and ^{137}Cs are also affected. This is most evident for ^{90}Sr, the content of which increases in the B-layer from about 10 % at the reference site (152 km from the smelter) up to 50 %

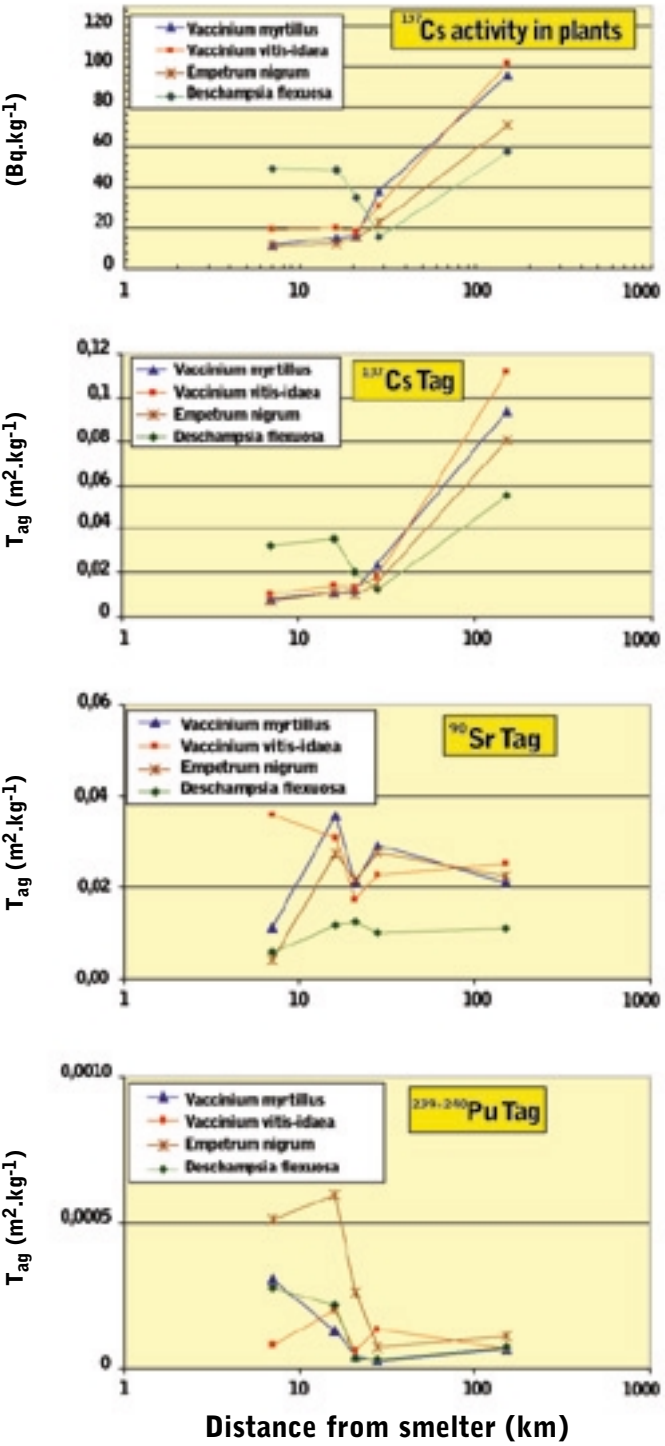


figure 4

Concentrations of ^{137}Cs (Bq.kg^{-1} dry weight) and aggregated transfer factors (T_{ag}, m^2.kg^{-1}) of ^{137}Cs, ^{90}Sr and $^{239+240}Pu$ from the global fall-out for four plant species as affected by the heavy metal pollution load at various distances from the Cu-Ni smelter.
EPORA

at the most contaminated site. No effect, however, can be observed for $^{239+240}$Pu. It is to be stressed that these results are also due to differences in the solubility of the radionuclides in the soils studied.

A clear influence of this additional pollution by heavy metals has also been observed on the availability of radionuclides in the soil's organic layer (O horizons), as examined through sequential extractions of radionuclides from the soils. The effect is most marked for ^{137}Cs, the exchangeable fraction of which is significantly reduced for increased heavy-metal loads. figure 3 Also, for ^{90}Sr and $^{239+240}$Pu, the exchangeable fractions were lower at the most polluted site (35 % and 1 %, respectively) as compared to those found at the other sites (46 % and 4-6 %, respectively). At each site, ^{90}Sr was essentially distributed between the exchangeable, bound-to-oxides and bound-to-organic matter fractions, with only 1-2 % persistently bound, whilst 85-90 % of $^{239+240}$Pu was bound to the organic fraction.

Radionuclide behaviour in plants along the heavy-metal pollution gradient was observed by measuring the ^{137}Cs, ^{90}Sr and $^{239+240}$Pu content of four understorey species at the five sites, ranging from 7 up to 152 km from the smelter, each site consisting of five distinct plots. The plant activity concentrations ranged from 10 to 100 Bq.kg^{-1} (dry weight) for ^{137}Cs, from 1 to 17 Bq.kg^{-1} for ^{90}Sr and from 0.001 to 0.019 Bq.kg^{-1} for $^{239+240}$Pu, with considerable intra-site variability.

Vaccinium myrtillus, *Empetrum nigrum* and *Vaccinium vitis-idaea* (to a lesser extent) all displayed reduced ^{137}Cs contamination levels at higher pollution loads, whereas *Deschampsia flexuosa*, the only grass-like species, exhibited higher contamination levels. figure 4

Except for *Vaccinium vitis-idaea*, which tended to remain unaffected, the ^{90}Sr activity concentration also decreased in all plants at higher pollution loads in contrast to $^{239+240}$Pu, which increased.

When taking into account the total radionuclide inventory in the soil profile considered, the aggregated transfer factors can be calculated for each plant (T_{ag}, m^2.kg^{-1}) (Figure 4). Ranging from 0.01 to 0.11 m^2.kg^{-1}, the ^{137}Cs T_{ag} values agreed with the relatively large range reported in the literature for understorey forest vegetation. Except for *Deschampsia flexuosa* again, the median T_{ag} values for ^{137}Cs were observed to decrease at higher pollution loads (reduced distances from the smelter). When excluding the reference site and only considering the four polluted sites, the effect was statistically significant, except for *Vaccinium vitis-idaea*. These results

agree with the data on the easily exchangeable ^{137}Cs fractions in the root zone, previously described, which also decrease at higher pollution loads (Figure 3). It is interesting to point out that this is quite similar to the K behaviour.

In contrast, transfer factors of *Deschampsia flexuosa* tended to increase at higher pollution loads, but it is important to note that this plant exhibited considerably clearer physiological alterations than did the others, especially at the most polluted site. The aggregated ^{90}Sr transfer factors do not appear to be as sensitive to the pollution load as ^{137}Cs is. Only *Vaccinium myrtillus* and *Empetrum nigrum* from the two most polluted sites show a trend similar to that observed for ^{137}Cs. These results broadly agree with the easily exchangeable ^{90}Sr fractions in the organic horizon, as reported above. In contrast to the general trends observed for ^{137}Cs and ^{90}Sr, the aggregated $^{239+240}Pu$ transfer factors increased significantly at higher pollution loads. This seems particularly clear for *Vaccinium myrtillus*, *Empetrum nigrum*, and *Deschampsia flexuosa*, no effect being detectable for *Vaccinium vitis-idaea*. However, it should be stressed that this could be an artefact resulting from the higher amounts of $^{239+240}Pu$ in the litter layer at the most polluted sites, which might have led to superficial contamination of the plants. (Due to its extremely small soil-to-plant transfer, the T_{ag} values for Pu are especially sensitive to superficial contamination of plants by soil).

It is of great interest to compare the trends observed for radionuclide behaviour along the pollution gradient with those observed for the distribution of nutrients and heavy metals in the soil-plant system. It appears that higher inputs of additional pollutants promote a reduction of ^{137}Cs and ^{90}Sr transfer factors (at least for some plants), which parallels a concomitant decrease in nutrient cations, such as K, Ca, and Mg, in the organic horizon. Conversely, the most polluted sites also exhibit higher contents in exchangeable metallic cations (Al, Fe, Ni, Cu) in the organic layer, where root colonisation is most pronounced. However, it should be stressed that the effects observed are not similar for all plants and all radionuclides considered, demonstrating that some specific plant and radionuclide features are involved which are still unresolved.

2 MODELLING

- *Root uptake in mineral agricultural soils*
- *Migration within mineral agricultural soils*
- *Redistribution of radiocaesium in forest ecosystems*

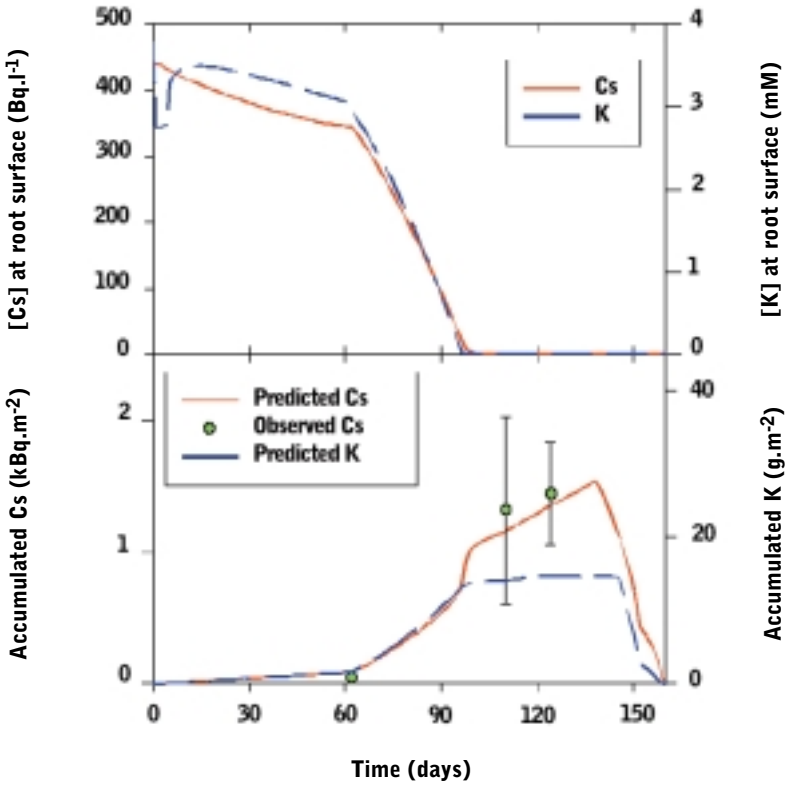


figure 1

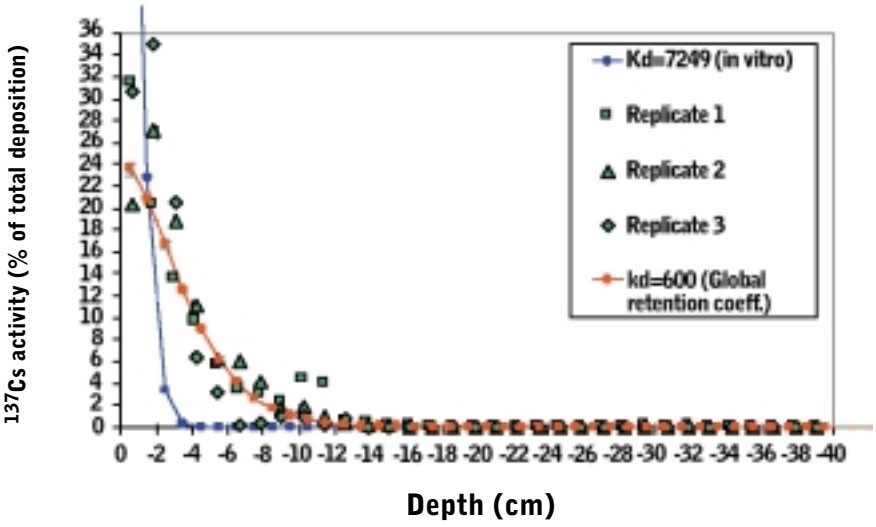
Soil solution ^{137}Cs and K evolution in the root vicinity during a barley crop, and their observed/predicted accumulation in plants. PEACE

Assessment models currently used for prediction of radionuclide behaviour have generally been derived empirically from given contamination situations against which they have been calibrated. Largely ignoring the actual processes involved, they often lack a more generic nature, which would allow their broader and more secure application. This is where analytical model development can provide substantial improvements, since they provide a better understanding of what is actually occurring. This strategy is illustrated by model-based clarification of the relevant mechanisms involved in plant contamination via root uptake and migration through confrontation with experimental observations.

Root uptake in mineral agricultural soils

If the soil solution composition is affected by soil parameters (such as texture, moisture content, counterions availability, etc…), it is also influenced by the root uptake activity itself, which is primarily designed to supply such nutrients as K and Ca to the plant in amounts suitable to support appropriate growth. Based on the finding, derived from recent advances in plant biology, that ^{137}Cs and ^{90}Sr are withdrawn respectively by the K- and Ca-transport mechanisms, the analytical model developed (RUR: Root Uptake of Radionuclides) takes the physiology of plant growth into account in that it reproduces the plant's ability to adjust the rate of supply to its demand. By this means, the dominant process leading to ^{137}Cs accumulation in plants can vary according to soil type, and even change during crop growth on certain soils. Large temporal variations of radionuclide accumulation in plants have indeed been observed during growth, and these are accurately predicted by the model. figure 1

But also, in the event of drastic K depletion that roots may promote in their vicinity under certain soil conditions, plants can regulate the efficiency of their transport systems in such a way that soils with high initial ^{137}Cs and K concentrations in the solution may lead to ^{137}Cs accumulations in plants that are similar to those found in soils with lower ^{137}Cs and K. The empirical relationship, often reported, between the uptake of a radionuclide and its nutrient analogue concentration is now reaching an appropriate resolution, which clarifies the influence of soil type, agricultural practices and time. The main conclusion to which the RUR model leads is that radionuclide bioavailability is not a constant for a given soil, but rather is variable depending on crop type and on time.



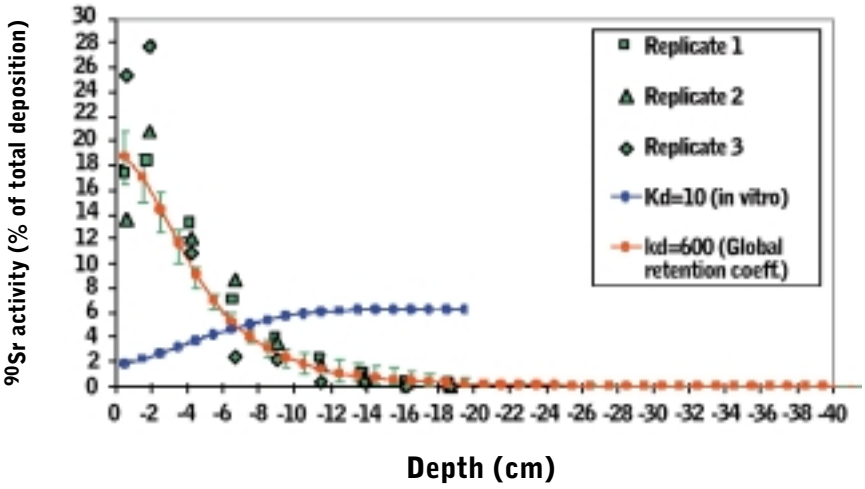


figure 2

Model-based predictions of ^{137}Cs and ^{90}Sr migration against experimental data obtained in a «silt loam» agricultural soil (three replicates).
PEACE

Migration within mineral agricultural soils

The simulation of the hourly-to-yearly vertical migration of ^{137}Cs and ^{90}Sr in a saturated/unsaturated soil column with vegetation is undertaken based on a one-dimensional, dynamic physically-based model formulation (TRANSOL model). 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. Despite this accurate description effort, introducing the radionuclides' respective K_D coefficients, as derived from *in vitro* sorption/desorption experiments, shows migration profiles which are underestimated for ^{137}Cs, and overestimated for ^{90}Sr.

figure 2

However, the model also demonstrates that prediction of the observed profiles is improved within the first 15 cm soil layer for both radionuclides when a partition coefficient of about 600 is introduced. This suggests that migration is governed not only by physico-chemical features accounted for by the *in vitro* radionuclide K_D, but also by additional soil properties which, when taken into account, result in a more appropriate overall "global retention coefficient". In addition to soil type, as described by the respective K_D values of the radionuclides, migration also probably depends on the soil structure (particle aggregation) and the associated biological activity of the soil (microbial and root activity), which can be accounted for by a "global retention coefficient".

Variables	K_{m-m}	K_{m-d}
All variables	-	2.1
Litter	1.9	-
Litter/Soil	1.9	1.15
Needles	2.2	1.8
Tree	1.9	3.0
Wood	2.9	3.2
Berries	1.6	1.5
Moose	1.4	-

Table 1

Reliability Indices (RI), showing the level of agreement between the model predictions themselves (K_{m-m}) and between the models predictions and the experimental data (K_{m-d}). A RI of 2 corresponds to an agreement within a factor of 2. The smaller the RI, the better the agreement.

LANDSCAPE

1- Swedish Defence Research Establishment
2 - University of Trieste
3 - Swedish Radiation Protection Institute. The development of FORESTLAND was performed in close co-operation with the Russian Institute of Agricultural Radiology and Agroecology, Obninsk, Russia (SSI Project RYS 6.15).
4 - It should be noted that part of the data used for parameterisation of FORESTLAND was collected in experimental plots located in the Bryansk area.

Redistribution of radiocaesium in forest ecosystems

The modelling of radionuclide migration and transfer in forest ecosystems has been an area of scientific investigation since the advent of radioecology in the late 1950's and early 1960's. The Chernobyl accident led to radioactive contamination of vast forested areas. The knowledge obtained from studies carried out in these forests has been used in the development of several new models.

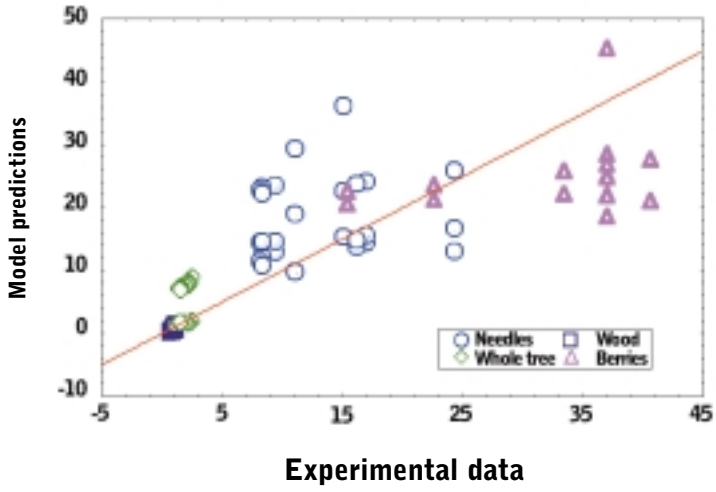
Based on a general conceptual model and the results of a review of existing models, three models were developed: the FOA model [1], the LOGNAT model [2] (LGN) and FORESTLAND [3] (FRLD). These three models are based on the compartment model principles and first-order kinetics for the turnover of caesium in the boreal forest. However, owing to the individual strategies chosen for describing and structuring the ecosystem and the different emphasis put on certain transfer and turnover processes, the resulting models exhibit, in essence, different characteristics.

The three models are also based on experimental data obtained in different geographical areas. This has resulted in differences in the model structures and in the values assigned to different model parameters. To what extent do these differences lead to disagreement in the predictions made with the models? To what degree would model predictions agree with experimental observations made independently in other areas, i.e. not used for calibrating the models? To answer these questions, a two-step inter-comparison exercise was carried out with the following specific objectives:

1. To determine the level of agreement between the predictions of radiocaesium levels in different forest components after an aerial deposition (model-model comparison).
2. To assess the agreement of the model predictions with experimental data obtained in field studies (model-data comparison).

The experimental data for step 2 were obtained from pine forests in the Bryansk [4] region of Russia that were contaminated by the Chernobyl accident, and these data were collected independently from 1986 to 1994. The experimental values were not disclosed until after the results of the calculations had been submitted.

The model predictions were made on a yearly basis for the first 50 years following the contamination event. The results were presented for the variables/endpoints: *Litter, Litter/soil, Needles, Tree, Wood, Berries, Moose.* Table 1 shows the level of agreement between the predictions for both steps 1 and 2 of the inter-comparison. Table 1



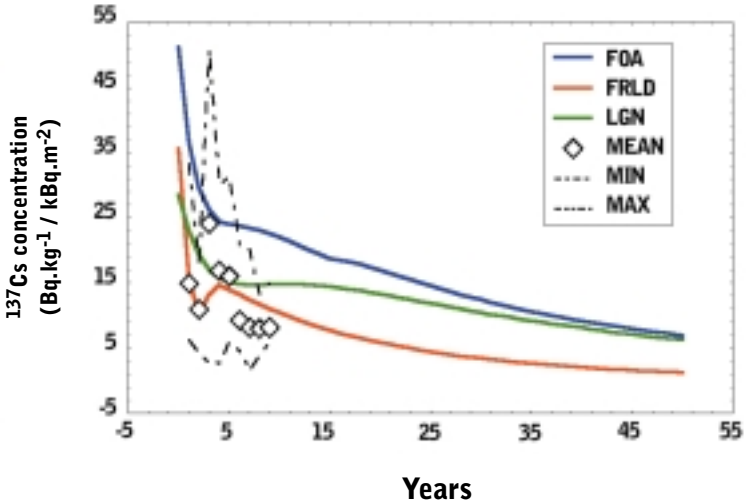


figure 1

Experimental values versus model predictions for the variables Needles, Wood, Tree and Berries and for all three models (up). Model predictions and observed values of the radiocaesium concentration in needles (down). The dotted lines correspond to the interval of variation of the experimental values. LANDSCAPE

Model predictions for all three models versus the experimental data for needles, wood, tree and berries, are shown in Figure 1 (up). Figure 1 (down) also shows the results for needles including the 50-year predictions, the experimental values and the natural variability in the experimental data. figure 1

The predictions made with the models for the scenario studied were in relatively good agreement (within a factor of 1.4 to 2.9) for all the variables considered. There was also good agreement between the model predictions and the experimental data (within a factor of 1.15 to 3.2). There were no significant differences between the models studied as to how they reproduced the experimental data. This suggests that, at least for the scenario concerned and for the first ten years after the deposition, all three models are equally serviceable if the final aim is to estimate absolute concentrations in different forest components. The agreement between the models, however, decreases with time, and there were differences in the form of the time dependencies predicted by the models, especially for wood. This may lead to larger differences, although probably within a factor of 10, between the model predictions and the experimental data for time intervals in excess of the period for which data were available for comparison. From the experimental data (due to the high variability) it was not possible to discern which model predicts best the kinetics.

The comparison with the experimental data confirms the fact that a major question remaining to be solved is whether the models correctly predict the kinetics of radiocaesium levels in different forest components. The experimental data used in the comparison did not allow this question to be answered due to the high spatial variability as compared to the observed time variations.
Any similar data set collected for validation purposes will probably encounter the same problem. To answer this question, longer time series need to be used. Alternatively, field or laboratory experiments can be carried out to test the hypothesis underlying the models, and from them conclusions drawn about the accuracy of the time dynamics predicted.

3 COUNTERMEASURES (FERTILISATION)

- *On agricultural soils*
- *On forest soils*

On agricultural soils

Fertilisation of agricultural crops has often been reported as a potential means for reducing the contamination of plant food products. From initial, sometimes confusing, experimental results, a consensus has evolved that this can be expected to be most effective in soils with low retention capacity (low K_D or CEC) and reduced fertility (especially, low K and Ca concentrations in the soil solution). Focusing on the soil solution composition provides an efficient approach to understanding this feature. The ^{137}Cs and ^{90}Sr concentration factors in lettuce and beans (CF, expressed with respect to the soil solution) decrease logarithmically with increasing concentration of their respective analogue ions, K and Ca+Mg. figure 1 figure 2

The influence of analogue ions is therefore stronger in the low range of concentrations-most likely in soils with reduced fertility. Measured on various soil types, the degree of correlation between CF and the concentration in analogues remains high, suggesting that plant contamination is primarily susceptible to the status of analogues in the soil solution, irrespective of the soil type. This agrees with the principle of radionuclide competition with its analogue ion occurring at the root level for membrane-based ionic transport systems.

It is of interest to note that concurrent with this competition effect at the root level where plant radionuclide uptake (CF) is reduced by increasing concentrations of analogue ions, the radionuclide availability at the soil matrix/liquid interface (K_D) is increased. The resulting plant contamination is therefore a combination of both effects, CF being more sensitive to analogues than K_D, especially in the low concentration range. This is why the fertilisation effect on CR (expressed with respect to the bulk soil) is most often limited on traditionally fertilized agricultural soils. figure 3

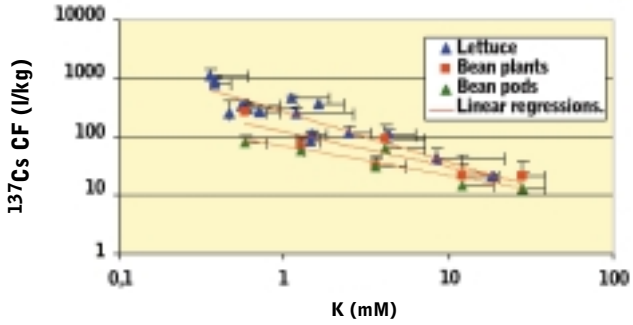


figure 1

Influence of the K concentration on the ^{137}Cs plant contamination (CF: concentration factor, Bq.kg^{-1} in plants / Bq.l^{-1} in soil solution) for lettuce and beans. PEACE

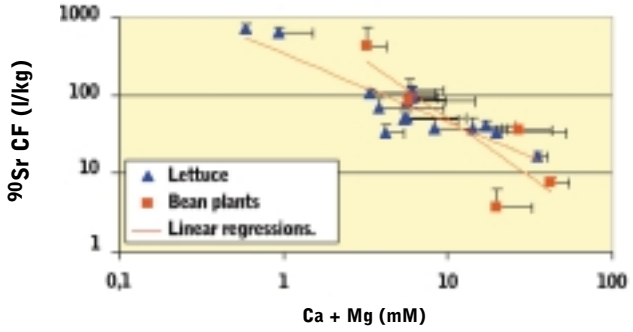


figure 2

Influence of the Ca+Mg concentration on the ^{90}Sr plant contamination (CF: concentration factor, Bq.kg^{-1} in plants / Bq.l^{-1} in soil solution) for lettuce and beans. PEACE

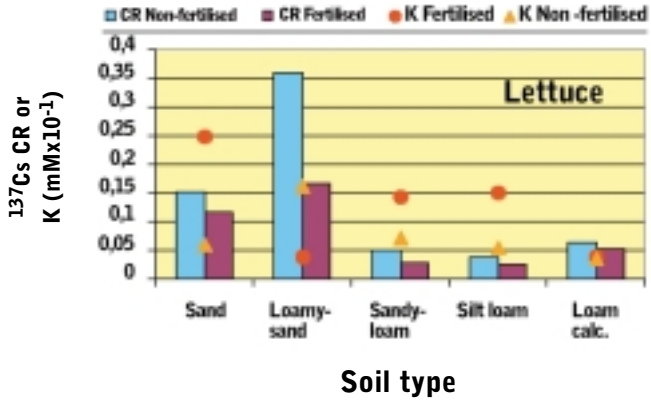


figure 3

Fertilisation effect on the ^{137}Cs Concentration Ratio (CR, Bq.kg^{-1} in plants / Bq.kg^{-1} in soil) in lettuce observed on various soils. PEACE

On forest soils

When assessing the influence of the different forest ecosystems on the exposure of man to radiation, it is also necessary to understand how radionuclides circulate in forest ecosystems that have been subjected to various management measures. The most common management measures are fertilisation, liming and site preparation. Potassium is probably the most interesting fertiliser affecting the distribution of radiocaesium in forest ecosystems. Generally, peat soils contain quite small amounts of potassium compared to the other main nutrients and compared to the amounts bound in the tree stands. Mineral soils, on the other hand, normally contain enough potassium for the tree growth. Instead, nitrogen is quite often a growth-limiting factor on podzol soils.

It is therefore of interest to investigate the effects of potassium fertilisation on the radiocaesium distribution in Scots pine stands on peat and mineral soils and to assess whether fertilisation can be a possible countermeasure or restoration technique in a radioactive fallout situation. Such fertilisation experiments were located in western Finland, where the radioactive fallout resulting from the Chernobyl accident was relatively high.

The ^{137}Cs concentrations in bark, wood and needles of Scots pine, as well as in many samples of branches of different ages, were found to be lower in the fertilised plots than in the control plots, both on mineral soil and on peat-land. For dead branches and dead portions of living branches, some samples of branches of different ages and of needles more than four years old, ^{137}Cs concentrations did not differ between treatments.

On the mineral soil site, the ^{137}Cs concentration was lower in *Deschampsia flexuosa* and *Vaccinium myrtillus* on the fertilised plots compared to the control plots. For grasses, herbs, lichens, mosses and other dwarf shrubs (*Empetrum nigrum* and *Calluna vulgaris*) no statistically significant difference in the ^{137}Cs concentration was observed between the treatments. However, at the fertilised plots, the concentrations were only 15-50 % of those at the control plots in these species.

On the peat-land site, *Vaccinium myrtillus,* tree seedlings, lichens, mosses and other dwarf shrubs (*Empetrum nigrum* and *Calluna vulgaris*) had lower concentrations of ^{137}Cs than on fertilised plots. In herbs, grasses, dead vegetation, *Dryopteris carthusiana* and *Vaccinium vitis-idaea,* the concentrations at fertilised plots were 38–68 % of those at control plots, although the differences were not statistically significant.

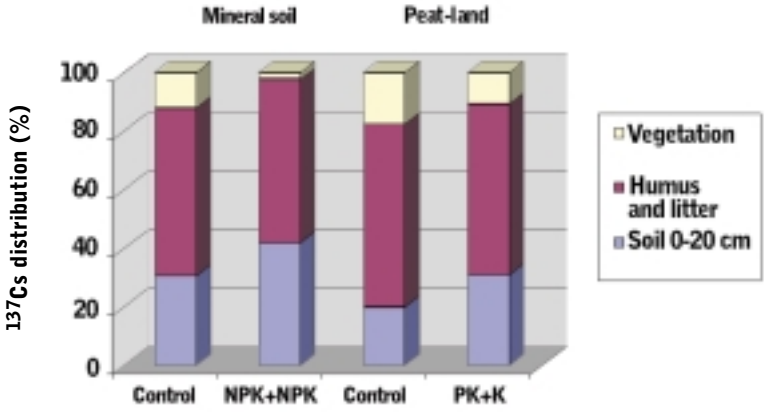


figure 1

The relative distribution of ^{137}Cs in pine-dominated forests on mineral soil and on peat-land (up), and the distribution of ^{137}Cs in vegetation (down) as a percent of the ^{137}Cs budget of pine-dominated forests on mineral soil and on peat-land.

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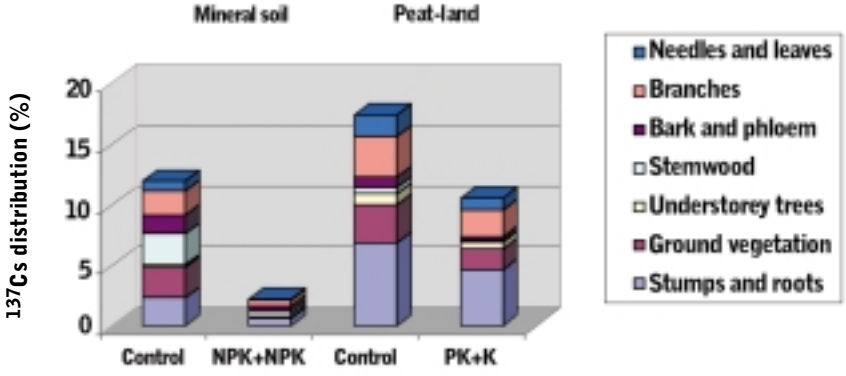


The ^{137}Cs distributions on fertilised and control plots on the mineral soil site and on the peat-land site (Figure 1) show significant changes due to the treatments. The activity fraction in vegetation decreased and the activity in the soil increased on fertilised plots. On the mineral soil, fertilised vegetation contained less than 20 % of the ^{137}Cs activity found in vegetation on control plots; on peat-land, the corresponding figure was 60 %. In above-ground parts of vegetation the reduction in ^{137}Cs activity in vegetation was slightly more than when roots and stumps were included in vegetation. figure 1

In general the results show that radiocaesium uptake in vegetation can be reduced by site-specific nutrient fertilisation on nutrient-poor sites. The more efficient reduction of ^{137}Cs content in vegetation on fertilised mineral soil compared to peat-land may reflect the excess of nutrients. On peat-land, the dose of K-fertiliser was less abundant in relation to the prevailing nutrient status of the stand. Dry-mass accumulation can be increased by fertilisation, especially in stem-wood. The results on mineral soil site show clearly that the reduction in Cs uptake cannot be explained by growth response alone, but that is also related to root uptake.

The growth response of trees to fertilisation is normally observed in the growing season immediately following that of the treatment. As to the timing of treatments in relation to Chernobyl fallout, the observed reduction in uptake of ^{137}Cs on the mineral soil site may represent nearly the maximum reduction that can be achieved with the amounts of fertiliser used. However, the effect of fertilisation on the dry-mass accumulation of vegetation will last only a few years on sub-dry mineral soil sites, and 15-20 years on peat-land sites poor in mineral nutrients.

In summary, the results indicate the benefits of fertilisation for restoring contaminated forests in a severe fallout situation. The availability of timber to the forest industry can be increased considerably by long-term treatments of forests. Through multiple use of forests, wild berry and mushroom gatherers, as well as hunters, receive less radiocaesium through foodstuffs from fertilised forests than from unfertilised ones.

The results of the study motivate further research on the applicability of forest management methods as countermeasures in a fallout situation. Such methods offer possibilities for respecting the principles of sustainable forestry, as the treatments do not radically alter the ecosystem.

4 RUNOFF

- *From forest ecosystems*
- *From forest ecosystems submitted to additional pollution by heavy metals*

From forest ecosystems

In northern Sweden, snowmelt and runoff had achieved their maximum annual intensity at the time of the main caesium deposition of Chernobyl, which occurred on April 29, 1986. The amount of ^{137}Cs discharged during this period from a 0.5 km catchment that was studied was about 600 MBq, corresponding to 5 % of the total deposition in the area. In the following years only relatively low levels of radioactive caesium were detected in the stream water, and in total about 10 % has been lost over the period 1986-1994. figure 1

New and more detailed calculations show that the initial loss during snowmelt in 1986 essentially occurred from the peat areas and amounted to about 40 % of the total deposition in these areas. No significant loss from the pine and spruce forests occurred during the same period. From autumn 1986 onwards, the annual loss amounted to 30% from the "wetter" part of the peat bog-i.e., the part often contributing to the surface runoff-while it is about 2% from the "drier" part of the peat area. The loss from areas of unsaturated mineral soil type is less than 0.03%. The corresponding ecological half-life (T_{eco}, not taking into account radioactive decay) for the boreal type ecosystem was estimated from measurements on groundwater and exceeded 4000 years (T_{eff} = 30 years, including decay), while the T_{eco} for the drier fractions of bogs was about 34 years (T_{eff} = 16 years). On the wet fractions of bogs,

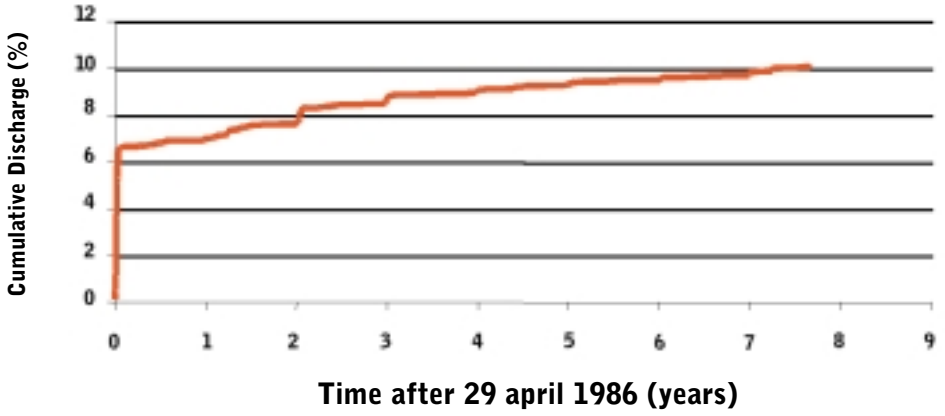


figure 1

The cumulative amount (%) of ^{137}Cs in stream water discharged from a catchment.

LANDSCAPE

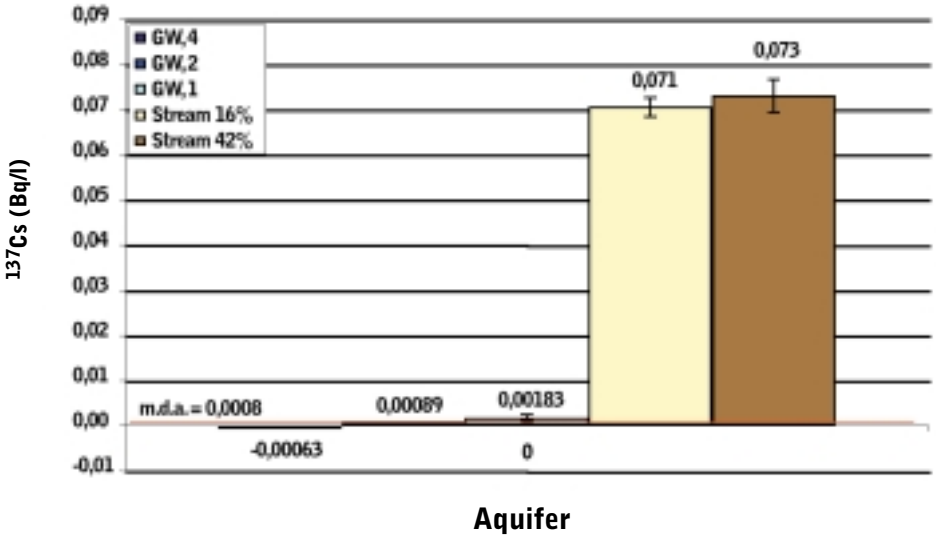


figure 2

The activity concentration (Bq/l) of ^{137}Cs in stream- and ground-water (GW) in January 1999 at the Vindeln site (the error bars represent 1 σ).

LANDSCAPE

which often contribute to saturated surface discharge, T_{eco}, was about two to three years.

The measurements on surface and groundwater show that a small fraction of the ^{137}Cs deposition has reached the groundwater at a depth of about 1 m. Assuming that the total annual discharge of 330 mm exceeds this depth, with a constant activity of 0.0018 $Bq.l^{-1}$ (Figure 2), the corresponding discharge via groundwater would be 6 x 10^{-5}% per year, which can be neglected compared to the discharge from the bog, which on average has been 1-2% per year over a ten-year period. figure 2

In conclusion, more than a decade after the Chernobyl deposition, the amount of ^{137}Cs leaving the forest ecosystem via runoff is negligible except from the wetter fraction of bogs. The caesium has now reached the groundwater at a depth of 1 m, but the discharge from the system remains negligible.

From forest ecosystems submitted to additional pollution by heavy metals

In addition to soil type, vegetation, geography and climate, a chemical industrial pollution of the catchment area may also affect the runoff process. This question has been addressed through investigations on two catchment areas located at 7 km (heavily pollution load) and 21 km (moderately pollution load) from the Monchegorsk Cu-Ni smelter. The reference catchment area was selected in northern Finland, about 137 km from the smelter.

Of the total ^{90}Sr deposition, only about one half is still present in the organic horizons and the upper 15 cm

of the underlying mineral soil, both at the reference and moderately-polluted catchments.
Conversely, at the most polluted catchment, close to Monchegorsk, only 15 % of the estimated total deposition of ^{90}Sr remains in soil at these depths. Yet, the fraction in the organic horizon of the total in the sampled soil is very similar at all three sites, indicating that loss from the organic fraction probably governs the residence time and distribution between organic and mineral fractions of the sampled soil.

The activity concentrations of ^{137}Cs in stream water correlate well with a relationship based on the fraction of the upstream catchment area containing deep organic soil found to be valid at several Scandinavian sites free of notable impact from regional sources of chemical pollution. Thus, no significant influence of chemical pollution on the loss of ^{137}Cs by runoff can be seen. In contrast, the activity concentration of ^{90}Sr in the stream water at the most polluted catchment is about twice as high as expected from the $^{90}Sr/^{137}Cs$ activity ratios at the two other catchment areas (reference and moderately-polluted). This coincides, moreover, with a loss of about 85 % of the initial deposit from the sampled soil columns at the most polluted catchment (i.e., a much greater loss than from the others).

Severe wind erosion of the organic soil is obvious at the most polluted catchment, giving rise to an airborne transfer to the stream water. However, the loss of ^{137}Cs in this catchment does not agree, apart from that likely to be related to a very small peat area. This indicates that the contributions of ^{137}Cs and ^{90}Sr to the transfer of organic matter from the atmosphere to the stream water are not the major cause of the notably changed $^{90}Sr/^{137}Cs$ ratio in water. A more probable explanation for the relatively high loss of ^{90}Sr from the most polluted catchment is therefore transfer by shallow groundwater in the steep slopes of the stream.

The increased rate of ^{90}Sr transfer to stream water in the heavily polluted catchment emphasises the importance of considering concomitant input to downstream aquatic recipients in landscapes affected by chemical pollution, and the potentially increased contamination in freshwater food chains leading to man.

5 SPATIAL VARIATIONS

- *On agricultural soils*
- *In forest ecosystems-radiocaesium in moose*

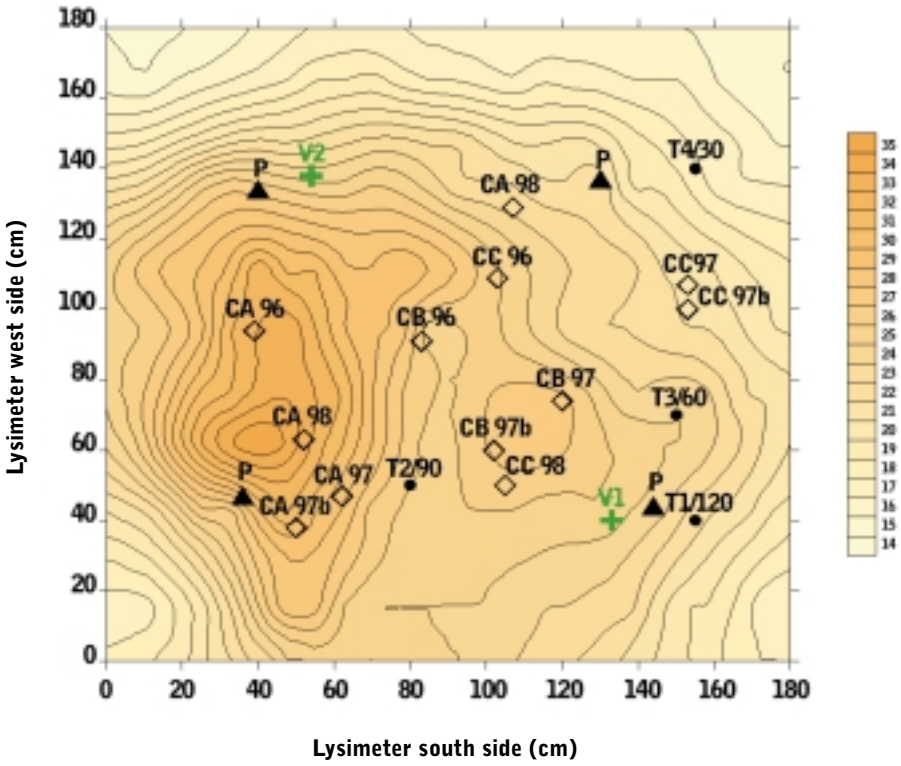


figure 1
Gamma emission mapping (10^3 counts/s) over a lysimeter soil surface two years after contamination. PEACE

On agricultural soils

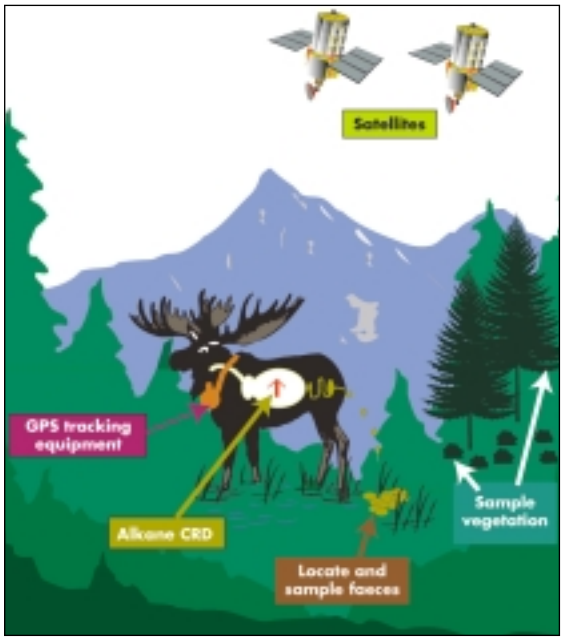
After the Chernobyl accidental release in the atmosphere, large spatial variations in radionuclide deposition were observed due to rain events over certain areas, that washed the radioactive cloud to a varying extent. On a smaller scale, large spatial variations resulted from the deposition of hot particles (essentially in the vicinity of the plant), and also, in the longer term, from horizontal redistribution by runoff. It should be noted that large spatial variations are still present on a small scale. For example, records show the metric to decimetric surface deposition of calibrated aerosols (3 µm on average) simulated under controlled conditions (no wind, no rain) to be heterogeneous. The subsequent influence of rain in washing off the radionuclides intercepted by the canopies further contributes to this heterogeneity, as illustrated by gamma emission mapping of the lysimeter soils' surface. figure 1

Large spatial variations in radionuclide migration have also been observed in the soil profiles, which may even exceed that caused by soil type or climatic intermittence. The soil structure (particle aggregation resulting from the soil's biological activity), already cited to explain the patterns of migration observed, most likely contributes to this variability. Soil microfauna (earthworms, in particular) and microflora (decomposition and turnover of organic matter) are key elements governing the soil structure.

In forest ecosystems-radiocaesium in moose

The semi-natural ecosystems of northern Europe have abundant large wild herbivores that are a significant food source for man. After the Chernobyl accident, large variations in radiocaesium levels were found in the tissues of free-ranging herbivores culled from the affected areas. Although in herbivores there is relatively little scope for variation in the degree of absorption and metabolism of pollutant radionuclides, the ability to predict the intake of radionuclides by herbivores living in heterogeneous environments has been poor. The radionuclide intake by herbivores was expected to depend critically on diet composition, in terms of plant parts and plant species, and the overall levels of environmental contamination, as it varies over a range of spatial scales. The need to quantify diet composition is especially acute where herbivores ingest fungal fruiting bodies, which can contain very high concentrations of radiocaesium and other pollutants. These are most abundant and may be ingested in late summer and autumn, prior to the culling seasons for large ruminants, whose radionuclide contamination levels may hence be strongly affected. In this project, methods were developed and applied (i) to measure the intake and diet composition of moose in the Boreal forest, and (ii) to assess the use of space by moose, and to consider these in relation to likely patterns of deposition and distribution of radionuclides in vegetation. This facilitates examination of the likely contributions of these factors to variability in radionuclide intake and contamination levels in moose, against the wider background of variability due to deposition pattern.

A method had been previously developed to quantify diet composition and intake by domestic ruminants grazing on temperate grassland. This non-invasive technique relies on comparing faecal concentrations of natural plant hydrocarbons with the similar compounds-usually long-chain n-alkanes which are orally dosed at known rates. The method had not heretofore been applied to wild ruminants because of the requirement to administer a daily oral dose, nor had it been applied to ruminants foraging on diets of woody browse or mixtures of woody and non-woody plants, typical of wild boreal herbivores. The availability of intra-ruminal, controlled release devices (CRD's), which release the n-alkane marker over a period of several weeks, provided the opportunity to apply the method to wild herbivores. Experiments were conducted to test and validate the method using CRD's under conditions relevant to foraging by boreal forest ruminants before it was applied to wild moose.



Previous results had suggested that the behaviour of the dosed hydrocarbons in the digestive tract is similar for all species of ruminant, and is similar to that of natural plant wax constituents. The expectation that the method would apply equally well to both moose *(Alces alces)* and domestic ruminants was borne out in the experiment, but further work is needed on the quantification of the exact faecal recoveries of all the markers used, in order to obtain accurate measurements of food intake. Validation experiments conducted with red deer (*Cervus elaphus*) and captive moose suggested that the methodology is equally applicable to ruminants ingesting diets of woody browse plants, in both summer and winter seasons. There was good quantitative agreement between the results of the hydrocarbon marker method and measurements of diet composition by weighing, in the case of moose. There was also good agreement between the hydrocarbon marker method of intake estimation with detailed calibration of mass intake via browsing damage, in the case of red deer.

The experiments further showed that n-alkanes are suitable dietary markers for birch in leaf, and, although the concentrations in twig material are low, they are sufficient to be used as markers in winter, when leaves are absent. Scots pine did not contain significant amounts of n-alkanes. Hence the use of long-chain fatty alcohols is suggested when this is a likely dietary constituent. In a separate experiment, using housed goats, the common fungal sterol, ergosterol, was tested as a potential faecal marker for dietary fungi. Because of the negligible recovery of ergosterol itself in the faeces of goats, it is unlikely that it can be used as a suitable marker for dietary fungi in ruminants.

figure 1
Trajectories of two female moose in mid-March 1997.
LANDSCAPE

figure 2
Home ranges for five moose for the period late April to late June 1998.
LANDSCAPE

The plant hydrocarbon marker method was applied to wild moose in northern Sweden, with the objective of estimating the intake and diet composition of moose in two seasons, winter and summer. The moose were located from helicopter and shot with an anaesthetic dart. The immobilization, the CRD insertion and the attachment of GPS (Geographical Position System) collars were carried out in 1997 and 1998. Inall, CRD's were inserted into eight animals and 20 animals were equipped with GPS-collars. Location of the alkane-dosed moose was carried out using the radio telemetry facility of the GPS collars in the winter of 1997, whereas in winter and summer 1998, the GPS system was called on to locate the dosed moose for the collection of faeces.

Samples of the vegetation available to the moose were collected in all three seasons in order to provide information on the range of hydrocarbons and long-chain fatty alcohols likely to be ingested by moose.
The conclusion on the basis of the results for the intensively studied moose in northern Sweden is that the winter diet of both moose bulls and cows was dominated by Scots pine. In contrast the summer diet was dominated by broad-leaved tree species, primarily birch *(Betula spp.)*. Some samples showed evidence that heather (*Calluna vulgaris*) was present in the diet, but the levels were such that it was unlikely to have significant radioecological consequences at the population level.

Overall, however, it is unlikely that variation in diet composition of moose in late summer and autumn, prior to and during the hunting season, is a major determinant of variation in radionuclide uptake. The exception to this remains the ingestion of fungal fruiting bodies, which can lead to significant uptake of radionuclides, but the ingestion of which is unpredictable due to their ephemeral temporal and spatial distribution.

It seems likely that the greatest variability in radionuclide uptake by moose is brought about by spatial variation in deposition, in conjunction with normal movements of the animals, rather than being due to variations in diet selection or intake *per se*. figure 1 figure 2

6 DOSE ASSESSMENT IMPLICATIONS

- *Forest ecosystems*
- *Forest ecosystems submitted to additional pollution by heavy metals*
- *Agricultural lands*

Forest ecosystems

Semi-natural ecosystems can make substantial contributions to the radiation exposure of man. This fact became known among radioecologists as a result of research performed following

the atmospheric nuclear weapons tests in the 1950's and 1960's. This knowledge was broadened after the Chernobyl accident, and today it is also well known to the general public living in contaminated regions. In particular, it has been shown that forests can deliver large radiation doses through the consumption of berries, mushrooms and game, but also through the industrial use of forest products. The LANDSCAPE project focussed on creating a basis for reliable assessments of the radiation exposure of humans. Knowledge about the distribution and concentrations of radionuclides (^{137}Cs) in the forest ecosystem and changes taking place over time is necessary for estimating the doses. However, in addition, assumptions also have to be made concerning consumption habits and time spent in the contaminated environment in order to calculate the internal and external doses. The results of the project give ^{137}Cs evolution over time in important compartments of the forest, as well as the change in the radiation field expressed as photon fluence, and necessary migration models have been tested.

One particular exposure case was investigated: the radiological consequences following energy production using radioactively contaminated biofuels from forests in Sweden. The dose estimates used the available data concerning the transfer of ^{137}Cs from soil to forest flora and fauna. Several exposure pathways were considered, including the enrichment of ^{137}Cs in the waste products and their recycling back to the forest as fertilizer. With an activity concentration of 5 kBq.kg^{-1} ^{137}Cs in the ashes, which is the recommended limit for ash recycling in Sweden, the highest doses among the public, about 0.02 mSv.y^{-1}, are received by a critical group composed of hunters and gatherers in forests. The average Swedish population in the same forest conditions receives a tenth of that dose. Considering the occupational doses, the highest doses-on the order of 0.1 to 0.5 mSv.y^{-1}-are received by persons working on an ash deposit.

Forest ecosystems submitted to additional pollution by heavy metals

The extensive data generated on ^{137}Cs redistribution in the podzolic soils of boreal forest ecosystems have been exploited to assess the potential influence of heavy-metal pollution on external radiation doses. The external gamma radiation due to ^{137}Cs in the soil depends on the vertical distribution of this radionuclide as well as on the wet-bulk density of the various soil layers. The gamma dose rates were calculated at all plots from each site along the pollution gradient.

The contributions of ^{137}Cs from nuclear bomb and Chernobyl fallouts to the external dose rate are different. When normalising the data on a deposition of 1 $kBq.m^{-2}$, the ^{137}Cs-generated external dose rate from Chernobyl is about 1.6 times higher than that from the global fallout. In these podzolic soils, it is true, the Chernobyl-derived ^{137}Cs remains closer to the soil surface than that from the global fallout. However, since total deposition of Chernobyl ^{137}Cs is only about 10 % of the total inventory in the soil, the total external dose rate from ^{137}Cs in soils is mainly due to the global fallout. Although the pollution load was clearly affecting the vertical ^{137}Cs distribution in the soils, the total external dose rate was not affected, with an estimated amount of about 1.5 $nGy.h^{-1}$. This might be due to the very low wet bulk densities of the litter and the organic horizons of these podzolic soils (0.1 $g.cm^{-3}$). As a consequence, the attenuation of the gamma rays is quite small, and different depth profiles of ^{137}Cs in the soil produce very small changes in the dose rates at a height of 1 m above the ground.

It must be stressed, however, that considering the influence of heavy metals on the radionuclide uptake of four understorey plant species, a pioneer aspect examined in EPORA, an influence of the pollution load on external and internal dose rates from the vegetation is not at all precluded. This is a question which particularly deserves further attention.

Agricultural lands

Agricultural lands, which support human food production, are central for assessing the potential radiological impact to humans of radioactive contaminants. The improved understanding of their behaviour in the soil-plant system, as derived from the PEACE project achievements, is such as to contribute to refining radiation protection issues, with a particular contribution made to improving prediction performances. As an example, in-depth soil migration profiles of both ^{137}Cs and ^{90}Sr appear to be most strongly influenced by the initial climatic conditions prevailing upon contamination-such as soil humidity and the first rain events. ^{90}Sr does not penetrate in depth as fast and deeply as existing prediction models would indicate; it therefore remains in the root zone of most crops, with potential impact on their further contamination. Indeed, ^{90}Sr is shown to be transferred to food crops in higher amounts than ^{137}Cs, irrespective of the plants considered or of soil type. Such features illustrate how the better understanding of radionuclide movements thus procured is conducive to the improvement of assessment models, whilst providing useful information for optimising the current philosophy on post-accidental intervention.

What is still needed?

- Reaching a better understanding of the concept of "bioavailability" and its applicability in different ecosystems.
- Clarifying the importance of climatic conditions immediately after radioactive deposition.
- Identifying what is needed for the assessment of radiation doses and where basic research can afford support.
- Investigating the scientific pertinence of dose to ecosystems.
- Distinguishing dose distribution over time from different ecosystems.
- Unravelling the influence of chemical industrial pollutants on the behaviour of radionuclides in the environment.
- Further model development and testing, including other lines of enquiry related to waste repositories and other radionuclides, for example.
- Practical countermeasures in different time perspectives and different terrestrial ecosystems.
- Clarifying current knowledge and identifying research needs through a systematic expert evaluation of all experimental data.
- Collection of strategic data series for model validation.

Bréchignac F. *et al.* 1999 PEACE Final Report. 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. CEC-IPSN Association contract n° F14P-CT96-0039a within the 4th EURATOM Framework Programme on Nuclear Fission Safety. IPSN Report Series DPRE/SERLAB/99-017(P), Paris, France.

Moberg L. *et al.* 1999 LANDSCAPE Final Report. An integrated approach to radionuclide flow in semi-natural ecosystems underlying exposure pathways to man. CEC-IPSN Association contract n° F14P-CT96-0039b within the 4th EURATOM Framework Programme on Nuclear Fission Safety. SSI Report 99:19, Stockholm, Sweden. ISSN 0282-4434.

Suomela M. *et al.* 1999 EPORA Final Report. Effect of industrial pollution on the dynamics of radionuclides in boreal understorey ecosystems. CEC-IPSN Association contract n° F14P-CT96-0039c within the 4th EURATOM Framework Programme on Nuclear Fission Safety. STUK Report Series A168, Helsinki, Finland. ISSN 0781-1705

This document provides an overview on the recent advances in Europe concerning the long-term environmental behaviour of radionuclides. It describes and illustrates the most significant scientific achievements gathered through three European projects, PEACE, LANDSCAPE and EPORA, which have been co-funded by the European Commission (DGXII) under the Nuclear Fission Safety Programme.

The improvement of radiation protection issues requires an accurate understanding of the radionuclide contaminants behaviour within ecosystems. The three projects have therefore been focused on the soil-plant system of agricultural lands and semi-natural forest ecosystems based on both, experimental approaches in controlled conditions and in the field. A particular emphasis is devoted to unravelling intra-compartments loads and inter-compartments movements of ^{137}Cs, ^{90}Sr and $^{239+240}Pu$, addressing also for the first time the potential influence of non-radioactive additional pollutants (heavy metals). The variety of conditions encountered in Europe is emcompassed through a wide selection of experimental sites ranging from Mediterranean up to Boreal ecosystems. Experimental data acquisition and mechanistic modelling are synergistically developed in order to promote access to improved assessment predictions in an accidental situation.

The knowledge generated first includes the redistribution of radionuclides in the soil-plant system with emphasis on vertical migration in soils and root uptake by plants. It provides next recent advances into the mechanistic modelling of these features. From the new data gathered through three years of investigations, essential conclusions on contamination discharge via run-off water are drawn, and insights discussed with respect to counter-measure, and spatial variations. Preliminary conclusions are finally derived in the perspective of dose assessment, and areas deserving further research identified.

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Under the joint authority of the Ministries of Industry and of the Environment, IPSN has, since 1990, been granted a specific status as well as its own budget. IPSN's staff numbers about 1300, two-thirds of whom are engineers and research personnel specialized in a variety of branches (physics, chemistry, biology, medicine, agronomy, etc.). Within the framework of an ongoing reflection concerning the French nuclear inspection system, IPSN has the vocation to become a fully independent public establishment in its own right.

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