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Effect of Industrial Pollution on the Distribution Dynamics of Radionuclides in Boreal Understorey Ecosystems (EPORA)

Final report

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Figure 3.3. *Map showing the locations of the sites for soil/plant studies and catchment areas used for runoff studies.*

sites were chosen in open areas in spruce forests (Piceetum fruticuloso-hylocomiosum) with an old tree stand (> 100 years).

Three catchment areas were chosen for the runoff studies. The distance of the most polluted catchment area (RI) is about 7 km from the Monchegorsk smelter. The catchment area (RII) represents an intermediate level of pollution and is situated 21 km from Monchegorsk. The reference area (RIII) for the catchment studies was selected at Naruska in Finland, about 137 km from Monchegorsk.

The geographical coordinates for the study areas are given in Table 3.1. A more detailed description of the study areas is given in the report Rahola et al. (1999).

Site descriptions

Quaternary deposits

The area, where the study sites are, is part of the glaciated terrain of Northern Europe. The lithology of the area is described by Rahola et al. (1999). The area was entirely covered by ice during the Pleistocene, which began 2 - 3 million years ago. The main quaternary deposit is till, which consists of an unsorted mixture of rock and mineral fragments from boulders to clay size, and the material in the till is mainly of local origin. At the catchment area the quaternary overburden on the site is dominantly till with some outcrops and boulder fields. In the mapping of Kola Ecogeochemistry project (Reimann et al. 1998) the nearest sampling site is some kilometres to the south, and there in the soil profile in E- and B-layers is some organic material and the layers are loose. The bottom part, parent material is till, stony and compact.

The predominant soils at the study area are podzolic Al-Fe soils. Podzolic Al-Fe-organic soils are characterised by high acidity and low base saturation. Organic horizons are the most acidic. In these horizons the cation exchange capacity is maximum.

At site A and catchment RI the overburden is meltout till, hummocky moraine. The area of the site is almost technogenic desert, almost all of the vegetation is destroyed and visible changes are found in drift, too.

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ABSTRACT

The project EPORA ‘Effects of Industrial Pollution on Distribution Dynamics of Radionuclides in Boreal Understorey Ecosystems’ is a part of the Nuclear Fission Safety Research programme of the European Union. A suitable environment for the study was found in the surroundings of the Cu-Ni smelter in Monchegorsk, in NW Russia where the huge atmospheric emissions from the smelter have polluted the environment since the 1930's. Samples of soil, litter, plants and runoff water were taken. Total concentrations of the main pollutants, Ni and Cu, in the organic soil increased from about 10 mg kg^{-1} at the reference site in Finland to about 5000 mg kg^{-1} at the most polluted site in Russia. Similar trends were observed for exchangeable fractions and plant concentrations of the same elements. Concentrations of exchangeable K, Ca, and Mg in the organic soil decreased strongly with increased input of chemical pollutants.

The radionuclides studied were ^{137}Cs, ^{90}Sr and $^{239+240}$Pu, mainly originating from the atmospheric nuclear weapons tests. The contribution of the Chernobyl derived ^{137}Cs deposition was about 10% but insignificant for the other nuclides. The activity distribution of all three radionuclides in the soil, their corresponding residence half-times as well as their aggregated transfer factors for various plants depended on the degree of pollution:

- *Activity distribution*: in the litter layer, the activity of all three radionuclides increased continually from the reference site to the most polluted site. This effect was most pronounced for $^{239+240}$Pu and least for

  ^{90}Sr and could, at least partly, be explained by the increase of the thickness of this layer. In the root zone, the opposite effect was observed: the largest fraction of all radionuclides was found at the reference site. In the organic layer, the exchangeable fractions of ^{137}Cs, ^{90}Sr and $^{239+240}$Pu decreased with increasing pollution.
- *Residence half-times*: in the root zone, the residence half-times of ^{90}Sr, but also of ^{137}Cs and $^{239+240}$Pu became considerably shorter when approaching the most polluted sites. Again, this effect was related to the thickness of the layer, which was significantly smaller at the most polluted site than at the other sites.
- *Aggregated transfer factors*: for two plant species, the aggregated transfer factors of ^{137}Cs and ^{90}Sr decreased with increasing chemical pollution, for one species it increased. Obviously, the soil-to-plant transfer of all three radionuclides can be significantly modified by the industrial pollution of the ecosystem, and these modifications are plant specific.

The studies of the transfer of ^{137}Cs and ^{90}Sr from the catchment soil to streamwater by runoff showed that the concentrations of these radionuclides depend more on the fraction of bogs in the catchment area than on the amount of pollution.

The external dose rate of ^{137}Cs as calculated from the depth profiles of its activity in the soil was approximately 1.5 nGy h^{-1} for all sites and did not depend on the chemical pollution. Related to the total activity per squaremeter, the dose rate of Chernobyl-derived ^{137}Cs was about 1.6 times higher than that of ^{137}Cs from global fallout, because the activity of Chernobyl-derived ^{137}Cs was concentrated closer to the soil surface than that of ^{137}Cs from the global fallout.

Summarising it can be concluded, that most of the radioecological quantities studied depended on the amount of pollution load at the various sites, even though the effects were related to the type of soil/plant ecosystem or soil/runoff water system. The methodology applied can also be used when investigating other polluted areas or evaluating the efficiency of restoration procedures applied to polluted areas.

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TIIVISTELMÄ

Tutkimusprojekti EPORA "Effects of Industrial Pollution on Distribution Dynamics of Radionuclides in Boreal Understorey Ecosystems" käsittelee teollisten saasteiden vaikutusta radioaktiivisten aineiden jakautumiseen ja kulkeutumiseen pohjoisten metsien aluskasvillisuuden muodostamassa kasvuympäristössä. Se kuuluu yhtenä osana Euroopan Unionin rahoittamaan Nuclear Fission Safety eli ydinturvallisuusohjelmaan. Tutkimukselle soveltuva alue löytyi luoteis-Venäjällä Montsegorskissa sijaitsevan kupari-nikkeli sulaton ympäristöstä, missä sulaton valtavat päästöt ilmaan ovat saastuttaneet ympäristöä 1930-luvulta lähtien. Näytteitä otettiin maaperästä, karikekerroksesta, kasveista ja valumavesistä. Tärkeimpien saasteiden, kupari ja nikkeli, kokonaispitoisuudet maaperän orgaanisessa kerroksessa lisääntyivät pitoisuudesta 10 mg kg^{-1} aina noin 5000 mg kg^{-1} siirryttäessä Suomesta valitulta vertailualueelta kaikkein saastuneimmalle alueelle Venäjälle. Sama suuntaus oli havaittavissa kuparin ja nikkelin helposti vaihtuvissa fraktioissa maaperässä ja niiden pitoisuuksissa kasveissa. Kaliumin, kalsiumin ja magnesiumin helposti vaihtuvat fraktiot maaperän orgaanisessa kerroksessa vähenivät voimakkaasti kemiallisten saasteiden määrän lisääntyessä.

Tutkittavat radionuklidit ^{137}Cs, ^{90}Sr ja $^{239+240}$Pu olivat pääosin peräisin ilmakehässä suoritettujen ydinasekokeiden aiheuttamasta laskeumasta. Tshernobylin onnettomuudesta peräisin olevan ^{137}Cs:n osuus kokonaislaskeumassa oli vain noin 10%. Muiden radionuklidien Tshernobyl-peräisten radionuklidein osuus oli merkityksetön. Kaikkien kolmen radionuklidien jakautuma maaperässä, niiden vii-

pymäpuoliintumisajat samoin kuin niiden kasvikohtaiset siirtokertoimet riippuivat alueen saastumisen asteesta:

- *Aktiivisuusjakautuma*: Karikekerroksessa kaikkien kolmen radionuklidien pitoisuudet kohosivat siirryttäessä vertailualueelta kaikkein saastuneimmalle alueelle. Selvimmin muutos näkyi $^{239+240}$Pu:n ja vähiten ^{90}Sr:n pitoisuuksissa. Karikekerroksen paksuuden lisääntyminen selittää ainakin osittain muutoksia. Päinvastainen vaikutus oli nähtävissä maaperän juurikerroksessa, jossa suurimmat pitoisuudet löytyivät vertailualueelta. Orgaanisessa kerroksessa ^{137}Cs:n, ^{90}Sr:n ja $^{239+240}$Pu:n helposti vaihtuvat osuudet kasvoivat saastumisasteen myötä.
- *Viipymän puoliintumisajat:* Erityisesti ^{90}Sr:n mutta myös ^{137}Cs:n ja $^{239+240}$Pu:n viipymän puoliintumisajat juurikerroksessa riippuivat saasteiden määrästä ja lyhenivät huomattavasti siirryttäessä saastuneempia alueita kohti. Myös tässä tapauksessa muutos riippui kerroksen paksuudesta, joka kaikkein saastuneimmalla alueella oli merkittävästi pienempi kuin muilla alueilla.
- *Siirtokertoimet:* Kahden kasvin ^{137}Cs:n ja ^{90}Sr:n siirtokertoimet pienenivät ja yhden kasvoi kemiallisten saasteiden määrän lisääntyessä. On ilmeistä, että teolliset saasteet voivat merkittävästi muuttaa kaikkien kolmen radionuklidin siirtymistä maasta kasveihin ja että muutos on lajikohtainen.

Valuma-alueilla tehdyt ^{137}Cs:n ja ^{90}Sr:n siirtymistä maaperästä jokiveteen koskevat tutkimukset osoittivat, että kyseisten radionuklidien pitoisuudet jokivedessä riippuvat enemmän valuma-alueella olevien soiden osuudesta kuin sillä olevien saasteiden määrästä.

Maaperän ^{137}Cs:n syvyysjakautuman perusteella laskettu ulkoinen säteilyannosnopeus oli kaikilla alueilla noin 1,5 nGy h^{-1} eikä kemiallisten saasteiden määrä vaikuttanut sen suuruuteen. Kun annosnopeus lasketaan neliömetrillä olevan aktiivisuuden kokonaismäärän suhteen, niin Tshernobyl-peräisen ^{137}Cs:n aiheuttama annosnopeus oli 1,6 kertaa suurempi kuin ydinasekokeista peräisin olevan ^{137}Cs:n, koska Tshernobylin laskeumasta peräisin oleva ^{137}Cs oli pääosin lähempänä maan pintaa kuin ydinasekokeiden laskeuman ^{137}Cs.

Yhteenvetona voidaan todeta, että suurin osa tutkittujen radioekologisten suureiden arvoista riippui alueella olevan saastekuormituksen

määrästä, vaikkakin havaitut vaikutukset olivat ominaisia kullekin maaperän ja kasvien muodostamalle eksosysteemille sekä kullekin maaperä/valumavesialueelle. Tutkimuksessa sovellettua metodiikkaa voidaan käyttää myös tutkittaessa muita saastuneita alueita tai arvioitaessa saastuneiden alueiden puhdistustoimien tehokkuutta.

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1. INTRODUCTION

Finnish, Norwegian and Swedish Lapland as well as the Kola Peninsula in North-West Russia belong to the arctic and northern boreal environment. Due to the climate and the soil properties of these ecosystems they are very sensitive to chemical pollutants, which may strongly affect the behaviour of radionuclides and essential nutrients.

The huge copper-nickel smelters in the Kola Peninsula have since the 1930's emitted enormous amounts of sulphur dioxide and heavy metals into the atmosphere. The effects of this airborne pollution on the local and regional scale have been extensively studied in recent years (e.g. Mäkinen 1994, Reimann et al. 1998, AMAP 1997). However, the possible impact of this chemical pollution on the turnover of artificial radionuclides present in these ecosystems has not been studied so far. The present project was therefore initiated to study the extent to which industrial pollution from these smelters affect the migration of key radionuclides in soil and their transfer from soil to plants and runoff water. The test areas for this study were selected along a decreasing pollution gradient to the south from the Monchegorsk Cu-Ni smelter in the Kola Peninsula. The annual emissions of this smelter were about 100 000 tons SO_2, 1600 tons Ni, 900 tons Cu and 80 tons Co in 1995 (Reimann et al. 1998). The reference areas were chosen in Finnish Lapland where the airborne pollution is very low compared to that in the test area.

The radioactive fallout in Northern Fenno-Scandia and in the Kola Peninsula originates mainly from the atmospheric nuclear weapons tests and to a minor extent of man-made radionuclides from other sources (STUK-A54, Saxén et al. 1987). However, the existence of nuclear power plants, stored liquid and solid nuclear waste, nuclear powered vessels and submarines as well as a nonspecified number of nuclear weapons in the Kola region enhance the risk of further radioactive pollution.

The radionuclides selected for investigation were the long-lived ^{137}Cs, ^{90}Sr and $^{239+240}$Pu. The aim of the study was to analyse to what extent the industrial pollution affects the dynamics of radionuclides in soil, transfer from soil to plants and runoff waters and influence radiation doses. After a potential accident at a nuclear facility where great amounts of radionuclides would be released into the vulnerable arctic environment, knowledge on the effect of pollutants on dynamics of these radionuclides and thus also on radiation risks is crucial.

2. OBJECTIVES

The general objective of the project was to study the effects of chemical pollution on the distribution and transfer of radionuclides in the northern boreal ecosystems. To reach the aims of the study the following objectives were set:

I General characterisation and selection of study area including sampling programmes

For this purpose the following task were to be performed:

a) assessment of the current information on industrial pollution and radioactive fallout on the study areas in Northern Finland and in the Kola Peninsula.

b) selection of study sites based on the background information. The reference sites were selected in Finnish Lapland in areas which were not polluted significantly more than what constitutes the general levels in the selected environments. The polluted sites were selected in surroundings of the Cu-Ni smelter in Monchegorsk, in the Kola Peninsula so that they represent different stages of damage caused by the pollution from the smelter.

c) definition and experimental protocols including standardisation, planning the sampling programmes and field expeditions.

II Determination of ecological effects of chemical pollutants in the study areas

The studies of the chemical pollution at the selected sites give basic information needed for analysis of the effects of industrial pollution on the distribution dynamics of radionuclides. For this purpose the following items were to be studied:

a) the atmospheric pollution load by analysing composition and amounts of chemical pollutants, especially the level of acidification and the concentrations of heavy metals (Cu, Ni etc.) in different soil layers and the association of heavy metals to different soil constituents.

b) the effect of pollution on the chemical properties of soil (e.g. pH, exchangeable cations, base cations, nutrients).

c) plant uptake of heavy metals (Cu, Ni) and nutrients (Ca, Mg, K etc).

III Effects of chemical pollution on the distribution dynamics of the radionuclides ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in soil and plants

For determination of the effects of chemical pollution on the distribution and dynamics of radionuclides soil and plant samples from the five plots at each of the five sites were analysed for:

a) concentration of deposited radionuclides ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in litter, organic, eluvial and illuvial layers..

b) association of the radionuclides ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in different soil constituents, especially in organic layers by sequential extraction.

c) residence half-times of the radionuclides ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in different soil layers.

d) concentrations of the radionuclides ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in plants and their transfer from soil to plants

IV Effects of industrial pollution on losses of the radionuclides ^{137}Cs and ^{90}Sr from the catchment areas

To estimate the losses the following items in the three catchment areas were to be studied:

a) concentration of the radionuclides ^{137}Cs and ^{90}Sr in organic and mineral soil and in streamwater.

b) characteristics of the catchment area (size, areal distribution of different soil types, biotopes etc.) for analysing parameters affecting the runoff.

V Conclusions on the effects of pollution on the distribution and dynamics of the studied radionuclides in the boreal environment.

As a summary of the whole study the following items were to be considered and discussed:

a) amount and composition of industrial pollution at the study sites and the effects of the industrial pollution on the contamination of soil and vegetation by the radionuclides.

b) effects of the industrial pollution on the residence half-times of the radionuclides in different soil layers.

c) effects on the industrial pollution on the losses of radionuclides from catchment areas by runoff.

d) effects of the industrial pollution on the external dose rate from the ^{137}Cs deposition.

e) estimation of the applicability of the methodology to other polluted areas and restoration of contaminated areas.

3. MATERIAL AND METHODS

Climate

Nearly all of the Kola Peninsula as well as Finnish Lapland belongs to the cool temperature zone where the mean annual temperature varies between – 1°C and + 1 °C. The air temperature is characterised by sharp seasonal variations. The temperature may drop below zero also during all months and the thermal growing season (day mean temperature is higher than + 5 °C) is short, only about 120 days. In the northern taiga sub-zone the total amount of precipitation is 450 - 500 mm throughout the year. The precipitation is highest in the summer and autumn period. The monthly rainfall in Monchegorsk varies from less than 20 mm in March to almost 70 mm in August. The duration of snow fall period is 6 - 6.5 months (from the middle of October to the end of May).

In winter the soil is frozen to the depth of 50 - 100 cm which characterises the podzolic sandy soil that prevails in coniferous forests in the Kola Peninsula. This results from mild winters and thick snow cover (60 - 100 cm). In the growing period the ”active” temperatures (higher than + 10 °C) are observed from the end of June -middle of July to the beginning or end of August, that is, the duration of the period of ”active” temperatures is 1 - 1.5 months. In relatively cold years the ”active” temperatures have been found only in the organic horizon (thickness of layer up to 10 cm). In moderate or relatively warm years (air temperature) the ”active” temperatures can be observed significantly deeper (15 - 20 cm and 40 - 50 cm respectively). The same climatic pattern can be seen in Finland. According to Reimann et al. (1998) winds from the south and south-west dominate in the Kola Peninsula during winter. During summer the wind directions are from east to west. In Finland with a rather flat topography the wind directions are almost the same in winter and summer without any dominant wind direction.

Industrial pollution

There are four important sources for chemical pollution: mining, energy, ore related industrial production and traffic. In the Kola Peninsula the main sources of pollution are the Cu-Ni smelters Severonikel at Monchegorsk and Pechenganikel at Nikel. The smelter at Nikel was built in 1932 and an even bigger one in Monchegorsk in1938. Since 1972 ore from Norilsk in Siberia was shipped via Murmansk to the smelters in the Kola Peninsula. Regarding SO_2 and heavy metal pollution the emissions from Nikel, Zapoljarnyij and Monchegorsk smelters place these sources amongst the largest world-wide. Figure 3.1 shows the variation of the emissions of SO_2, and Figure 3.2 changes in Cu, Ni and Co emissions in 1988 – 1996 (Lukina and Nikonov 1996). The ore from Norilsk contained more sulphur than the local ore, which resulted in increasing SO_2 emission after 1972. During last years the emission show a decreasing trend. In the area this study, most of the Ni and Cu originates from the smelter at Monchegorsk.

Radioactive pollution

There are two main sources for radioactive contamination, the atmospheric nuclear weapons tests in the 1950’s and 1960’s and the Chernobyl accident in 1986. The possible releases from other nuclear facilities are of minor importance. The long-lived radionuclide ^{137}Cs, ^{90}Sr and $^{239+240}$Pu are the most important when considering the long-term risks from internal radiation. In the case of external radiation the main contributor among the gamma emitting nuclides is ^{137}Cs. The radionuclide deposition originating in the nuclear weapons tests was rather evenly distributed in contrast to the deposition from Chernobyl. The accumulated deposition of ^{137}Cs before the end of 1985 was in Finland 1 800, of ^{90}Sr 1 100 and of $^{239+240}$Pu 60 Bq m^{-2}, respectively (STUK-A54, Hardy et al. 1973). The fallout from Chernobyl was areally unevenly deposited. The mean surface ^{137}Cs activity was 10 700 Bq m^{-2} on October 1, 1986 as mean for the whole Finland, but in northern Lapland only 1 000 to 2 000 Bq m^{-2}.

In all of Finland including Lapland the ^{90}Sr deposition from Chernobyl was much smaller only a few percent of that of ^{137}Cs and also small compared to the earlier deposition from the nuclear weapons tests. The $^{239+240}$Pu fallout follows the same patchy deposition pattern and was very low. The estimated range in Lapland varies from zero to a few becquerels. (Saxén et al. 1987, Arvela et al. 1990). A Finnish-Russian study showed that the concentration of ^{137}Cs in lichen samples collected in Inari, Finnish Lapland, and in Lovozero, Kola Peninsula, 1960 - 1994, was about the same; only after the Chernobyl accident somewhat more ^{137}Cs was found on the Finnish side (Doudarev et al. 1995, Rissanen et al. 1994, Saxén et al. 1987).

Selection of sites

The Severonikel smelter complex in Monchegorsk was studied as the release source producing a pollution gradient suitable for the aims of the EPORA project. The criteria for selection of sampling sites included investigation of visible damage of vegetation and availability of runoff at an adjacent catchment area. At distances from the release source

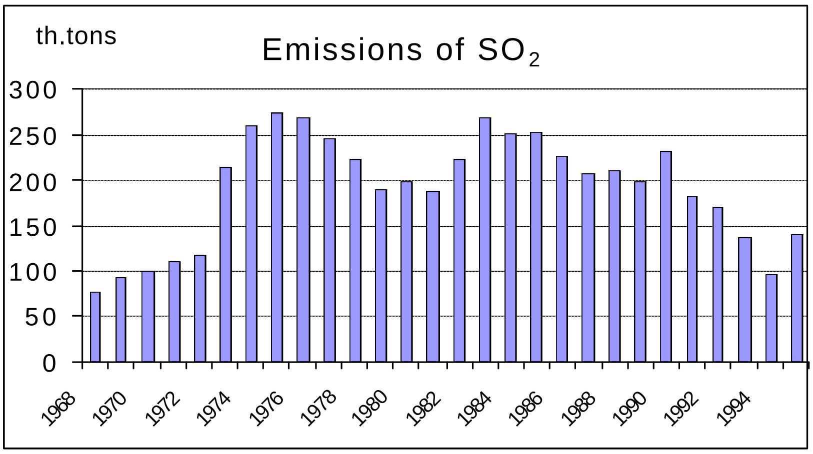


Figure 3.1. *Atmospheric emissions of SO_2 from the Monchegorsk smelter during 1984 – 1996 (Lukina and Nikonov 1996).*

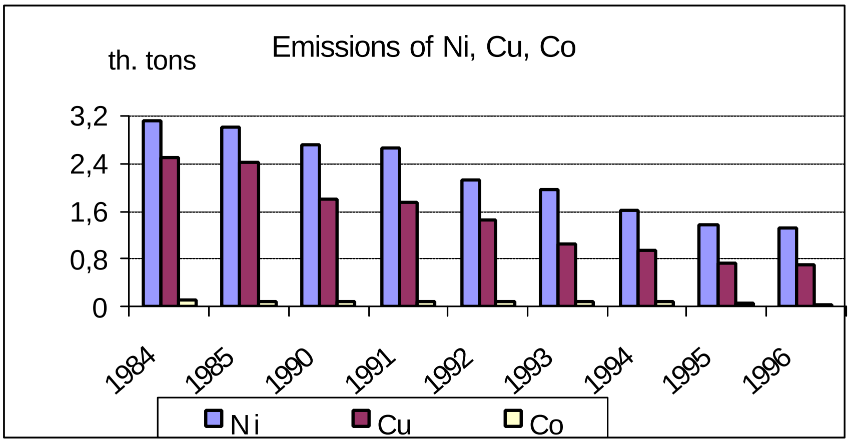


Figure 3.2. *Atmospheric emissions of Ni, Cu and Co from the Monchegorsk smelter during 1984 – 1996 (Lukina and Nikonov 1996).*

where the pollution level is small or insignificant the vegetation at the growing sites is still unchanged. When approaching the release source the original characteristics of the vegetation are gradually changing into an ultimate state that may be classified as an industrial desert. At some intermediate stage it is expected that there exists growing sites where the moss and lichen carpet is currently subjected to disintegration. At such sites the ground cover may have been substituted to a large extent by dwarf shrubs and grass.

The study areas chosen are shown in the map in Figure 3.3. They were selected along the decreasing pollution gradient from the Monchegorsk smelter. For the soil migration and soil/plant transfer studies, four sampling sites (A, B, C, D) were established to the south of the Severonikel smelter complex in Monchegorsk at points subjected to varying levels of pollution by sulphur, nickel and copper compounds. The distances varied from 7 to 28 km from the smelter. Site A represents the most polluted area and the level of pollution is decreasing towards site D which represents the less polluted area. The reference site (REF) was selected at Naruska in Northern Finland 152 km westwards from the smelter, where the chemical pollution level did not significantly deviate level of pollution of global origin. The sampling

Table 3.1. *Location of soil/plant sites and catchment areas.*

Sample site	Sampling area and the name of the place	Latitude	Longitude	Distance from smelter
Russia	Site A	67°51'	32°48'	7 km
	Site B	67°46'	32°48'	16 km
	Site C	67°44'	32°51'	21 km
	Site D	67°40'	32°47'	28 km
	Catchment RI	67°51'	32°50'	6 km
	Catchment RII	67°43'	32°49'	21 km
Finland	Reference site (REF)	67°26'	29°27'	152 km
	Reference catchment RIII	67°29'	29°46'	137 km

The organic layer is almost totally absent and the samples of organic layer are litter samples in practice. The E-layers, eluvial layers, are very thin, instead the B-layers, illuvial layers, are thick. The parent soil is yellowish green till.

The area of site B is also almost totally destroyed. The overburden is very stony, the organic layer is thin and includes a lot of roots and is poorly decomposed. The thickness of the E-layers is variable. The B-layers are thick, compact and partly cemented. The parent soil is grey till.

At site C and catchment RII the organic layer is thin, the thickness of the E-layers varies greatly and the B-layers are very stony. The parent soil is grey till.

At site D the thickness of the organic layer varies in general from 3 to 4 cm. Also the E-layers are very thick, from 4 to 10 cm, though the variability is great. The thickness of B-layers is more than 10 cm. The parent soil is grey till.

At the reference site (REF) and catchment RIII, in Finland, the podzol profile is well developed. The organic top layer is not well decomposed, whereas the lower parts represents the more decomposed organic material. The E layer is from 4 to 7 cm thick and the B-layer quite thick. The parent soil material is brownish till.

Vegetation in soil/plant sites

According to the detailed descriptions done in 1930's the pollution-transformed spruce forests in Monchegorsk were originally of the *Piceetum fruticuloso* - *hylocomiosum* type. In these forests mosses *Hylocomium splendens* and *Pleurosium schreberi* were the dominant among understorey species and *Dicranum spp.* were also found. The proportion of green mosses out of the total aboveground biomass was approximately equal to that of *Empetrum nigrum, Vaccinium myrtillus* and *Vaccinium vitis-idaea* taken together. Solitary specimens of *Vaccinium uliginosum, Lycopodium annotinum, Cornus suecicum, Linnea borealis, Solidago lapponica,* and of the lichens *Nephroma arcticum, Cladina stellaris, Cladina mitis* and *Cladina rangiferina* were found. In 1997 the forest in the Naruska soil/plant reference site in Finland was still in this pristine state.

It should be noted that the spatial distribution of biomass of the understorey species in spruce forests is far from even. The trees regulate light, temperature, and hydrological and nutrient regimes in the forests. They also regulate intensity of element fluxes, including pollutants and acid forming substances, in forest ecosystems.

On the basis of primary productivity parameters for four significantly different degradation stages of the *Piceetum fruticuloso* - *hylocomiosum* subjected to air pollution in the Kola Peninsula were identified: 1) *Piceetum fruticulosum*, 2) *Piceetum graminoso-fruticulosum*, 3) sparse *Piceetum empetrosum*, 4) industrial barrens. One of the four soil/plant sites in Monchegorsk presents the first stage of degradation - *Piceetum fruticulosum* (site D), two sampling sites (C, B) present the second stage of degradation - *Piceetum graminoso-fruticulosum* and sample site A presents the third stage - sparse *Piceetum empetrosum*.

At site D, the spatial distribution of ground vegetation is much more heterogeneous than in the reference area. This heterogeneity results from lack of green mosses and from the beginning of colonisation of the forest hair grass (*Deschampsia flexuosa*) and crowberry (*Empetrum nigrum*). The important reason is also that the state of trees, which are responsible for distribution of understorey species, is very different: from defoliating (with different extent of defoliation and crown thinning) to dead.

At sample sites C and B the total biomass of ground vegetation is lower than in the reference area but significantly higher than at site D. The main reason is an increase in biomass of crowberry and hair grass. Crowberry formed individual microgoups on fallen trees and stumps. Hair grass also formed microgroups.

Sampling site A represents the third stage of spruce forest degradation - sparse *Piceetum empetrosum*. In this stage the area not covered by vegetation, so called industrial barrens, is increasing. The major part of the area is occupied by crowberry. Hair grass formed individual microgroups. The biomass of crowberry and hair grass reached maximal values as also the amount of litter.

Catchment areas

Catchment area RI is the most affected area close (7 km) to Monchegorsk. The area appears to have been of the North Taiga type with a mixed spruce and pine forest before the occurrence of the drastic changes in the biotope during the last decades, as a consequence of the airborne releases from the nearby Monchegorsk smelters. The pollution effects are now very severe and the present state may be classified as a technogenic desert. Old trees are lacking. The stumps indicate that tree felling has occurred. There are also traces of forest fire. The loss of living understorey carpet has led to considerable erosion of the organic layer, probably mainly mediated by wind. In several places organic material was no longer present. At other spots organic material had accumulated to considerable thickness.

The stream runs approximately from NW to SE and ultimately discharges into Lake Imandra. It runs in a ravine 5 - 10 m deep and 100 m wide, somewhat meandering. Banks are steep and soft. Debris and organic material have accumulated in the ravine, which was overgrown by bushes, grass and sedge. Tussocks of sedge are present along the stream.

Catchment area RII was on the border of the ”Lapland natural park”, and was much less affected by acid fallout than catchment area RI. The forest consisted of an aged, mixed forest (spruce, pine, birch) with normal understorey vegetation including berries (blueberry, lingonberry, cloudberry) and mushrooms. The ground rises on both sides of the stream, more steeply to the east with the bedrock visible between the stream and the road bank. A road runs west from the main road M10 into the natural park. It fords the stream and water samples were taken just upstream of the ford.

The stream runs roughly from north to south, parallel to the M10 main road and about 500 m west of it. Below the ford the stream runs into a peat area, but upstream the terrain was less peaty with firm ground along the stream and tussocks of sedge along it. 800 m upstream from the ford there was a ridge of rock across the stream, about 1-meter high, and above this ridge was a small circular swamp, which could not be crossed on foot.

The landscape on the Finnish side at catchment area RIII was hillier than on the Russian sites, and not significantly affected by acid fallout from any regional sources. The stream is running from west to east. On the south side of the stream, up to the hill there has been extensive logging, and furrows have been plowed downhill. Spruce has been planted along the furrows but has not yet grown over 50 cm. This concerns a limited area adjacent to the water sampling position. Upstream (ca 300m) of the water sampling position, a similar-but much older area exists where soil preparation and spruce planting had occurred in an about 100m wide band up along the hill on the south side. The number of branch whorls on these pine plants indicated a tree age of about 30 years. The forest consists of an aged, mixed forest with understorey vegetation typical for a morainic site of intermediate productivity. The ground is very stony gravel with a thin organic cover.

Sampling

Vegetation from the soil/plant sites

The sampling sites were chosen in open areas between the trees in a typical spruce forest. The 5 plots were chosen at the corners of an approximately 10 m x10 m square with one plot in the middle of the square. The size of the sampling plots were 1 m x 1 m at the reference site in Finland and 1 m x 1.5 m at sites A, B, C and D in Russia. The aboveground vegetation was carefully cut using scissors and later sorted according to the different species. Then the litter layer above the organic layer was quantitatively collected from the five plots at each site. The quantitative sampling of all plants and litter was carried out June 25, 1997 in Finland and August 12 - 15, 1997 in Russia. The plants chosen for analyses in this study were crowberry (*Empetrum nigrum*), blueberry (*Vaccinium myrtillus*), lingonberry (*Vaccinium vitis-idaea*) and forest hair grass (*Deschampsia flexuosa*). The state of growth was not exactly the same for these different sampling periods. Because of the late spring the Finnish vegetation was unusually delayed in June.

The amounts of the above mentioned plants growing on the plots were rather small. To enable the radiochemical analyses of the presumably very low concentrations of ^{90}Sr and ^{239,240}Pu, additional large volume samples of the four species were collected around the quantitative sampling plots (marked as P-samples) during the sampling expeditions in Finland and Russia. An additional collection of P-samples was performed in August 1997 after the expedition in Russia. Blueberries and crowberries were also collected on all five sampling sites.

The vegetation and litter samples collected at the Finnish site were transported in two days to STUK's laboratory in Rovaniemi and stored in + 4 ^{o}C temperature. All the vegetation and litter samples collected from Monchegorsk sites D and C and the vegetation bags from sites B and A were transported from Russia to Rovaniemi in a few days after sampling and stored in + 4^{o}C. The plastic bags containing the litter fraction from sites C and D had to be stored in Monchegorsk and were transported to Rovaniemi late in December. Further details on sampling procedures are given in (Rahola et al. 1999).

Soil from the soil/plant sites

To carry out soil sampling, the plot area was divided into two parts. The first one (two strips of about 25 cm x 100 cm at the peripheral of the plot area at the reference site in Finland and 50 cm x 100 cm in the Russian sites A - D) was used to sample the organic layer, the second one (a 50 cm x 100 cm area in the middle) was needed to collect the samples from the mineral horizons.

The organic layer was divided in pieces of about 25 cm x 25 cm using a sharp spade and then carefully put down on a plastic film. Attached mineral soil was scraped off from the organic layer using a knife. After this procedure, the organic layer was divided into two parts (Of and Oh) by hand according to their natural constitution. The top layer (Of) consists of poorly decomposed material, fibric soil material, whereas the second layer (Oh, about 1.5 to 2 cm thick) represents the more decomposed organic material (sapric soil material). Both layers can be well separated from each other because of a weak zone between them. Most part of the roots of the understorey vegetation was in the organic layer.

To sample the mineral horizons, the organic layer was removed without destroying the first cm of the E-horizon. Samples were taken from a defined area (about 35 cm x 35 cm) inside the plot. The E-horizon was divided into two layers: The first 2 cm were collected separately, whereas the second layer was taken according to there natural thickness down to the B-horizon (about 2 - 5 cm). The same procedure was also carried out with the B-horizon, but in contrast the first layer consists of 5 cm.

In conclusion, samples representing six different soil layers were collected: two (Of and Oh) from the organic layer, two (E1 and E2) from the E-horizon (eluvial) and two (B1 and B2) from the B-horizon (illuvial).

Sampling and preconcentration of runoff water

It was necessary to take water samples of 200 litres from each of the three catchment areas (RI, RII and RIII), since concentrations of ^{137}Cs and ^{90}Sr are only of the order of 1 Bq m^{-3}. To avoid transporting these large samples preconcentration of the radionuclides was necessary. In Finland the samples were preconcentrated on the sampling area but in

Russia the water samples were taken to a laboratory in Monchegorsk. Two samples from the reference area RIII were also acidified and taken to the laboratory for evaporation. Preconcentration was done by running the sample through a 400-ml column of the exchange medium by hydrostatic pressure at a flow rate of 15 l/h. The water was not pretreated, but 500 Bq of ^{85}Sr was added to some samples for determination of recovery. For most samples clinoptilolite, a natural zeolite, was used, but two samples from the reference area were preconcentrated with a strongly acidic cation resin.

Sampling of soil from the catchment areas

The primary objective of the sampling of soil is to get an estimation of the mean level of ^{137}Cs and ^{90}Sr remaining in the catchment area and the variability of the activity levels. For that purpose area defined samples were taken of a) the whole organic layer from the area of 25 cm x 25 cm and b) 15 cm of the underlying mineral soil from the area of 12.5 cm x 25 cm. From each runoff area ten sets of organic and mineral soil samples were taken from the river banks, five on each side upstream of the water sampling point within a distance of 2 km.

Analytical methods

Chemical pollution

The methods used for determination of chemical properties of soil, heavy metals (Ni, Cu), nutrients (Ca, Mg, K, Zn, Mn) and stable Sr were standard methods. Speciation of heavy metals by sequential extractions is done according to the same procedure (Bunzl et al. 1997), which was used for radionuclides. The method is described more closely in the report by Thørring et al. (1999).

Radionuclides

The measurement methods used for determination of ^{90}Sr and ^{137}Cs in soil, plant and water samples are described in two STUK reports (Rantavaara et al. 1993 and Ikäheimonen et al. 1995). The corresponding $^{239+240}$Pu analyses were done according to the description in the publication of Hakanen and Jaakkola (1977). For the studies of the chemical association of ^{90}Sr, ^{137}Cs and $^{239+240}$Pu the sequential extraction method (Tessier et al. 1979) as modified by Bunzl et al. (1997) was used.

The concentration of ^{137}Cs in runoff water samples was determined by direct gammaspectrometric measurement of the clinoptilolite, ion exchanger or evaporation concentrate. Some measurements were verified by duplicate measurements. ^{90}Sr was eluted from clinoptilolite with 4 bed volumes of 2M ammonium chloride and from cation exchanger with 8M nitric acid. Strontium was radiochemically separated from the eluate or evaporation concentrate. After ingrowth of ^{90}Y the activity was determined by measuring the Cerenkov radiation with a low-level liquid scintillation counter.

Quality assurance

All participating partners work at well-established institutions with quality assurance programmes of their own. The quality assurance programmes are specially created for each establishment. During the experimental part of the EPORA project the quality assurance programmes of the partners institutes will guarantee the quality.

The sampling methods were routine methods used in the participating laboratories, slightly modified to fit the EPORA project. Only the method for treatment of the water samples was developed for this project. Also the analysis methods were well established in the organisations of the partners. All methods have been intercompared in a number of international intercomparison exercises. Since the same methods were not used in all the laboratories intercomparison between partner laboratories were done to a limited extent.

4. RESULTS AND DISCUSSION

4.1. Distribution of nutrients and heavy metals in the soil/plant system along the pollution gradient

Effects of pollutants from copper-nickel smelters on coniferous forest ecosystems may be evident in several different ways:

- Direct effects of sulphur dioxide on the forest canopy.

- Toxic effects by heavy metals on soil microflora and sensitive plant species.

- Indirect effects of acidic deposition via soil acidification which may have several different effects on plant growth, the most important of which presumably being displacement of nutrients on soil colloid surfaces by cation exchange and subsequent loss by leaching.

- Indirect effects of heavy metals by cation exchange displacement of nutrients.

The two first points were considered to be outside the scope of EPORA. In the chemical studies of soil and plant samples from the four sites (A-D) along the pollution gradient from the Monchegorsk smelter and the reference site (REF) the following groups of parameters were studied:

- Properties related to soil acidification: Soil pH (in H_2O and $CaCl_2$), cations exchangeable with 1M ammonium nitrate (K, Mg, Ca, Mn, Fe, Al, Cu, Ni), and exchangeable acidity. From these parameters the cation exchange capacity (CEC) and base saturation factor were calculated. Ammonium nitrate was selected because this extractant has no buffer capacity in the pH range considered and hence the extraction takes place near the natural soil pH.

- Parameters related to heavy metal pollution, limited to the major metal contaminants copper and nickel. These were primarily nitric acid soluble concentrations in soil and total concentrations in plant tissue (also after nitric acid digestion). Moreover the soil samples were subjected to sequential extractions according to the procedure by Bunzl et al. (1997) and Cu and Ni determined in the various fractions.

- Parameters related to plant nutrients: In addition to the above “base” cations (K, Mg, Ca and Mn), zinc was included. These nutrients were determined in plant tissue in addition to the ammonium nitrate extract.

- In addition some investigations on the behaviour of stable Sr and Cs in the soil/plant system were performed.

In the following selected examples of the results are shown, and the most important conclusions regarding the changes in soil chemistry and plant concentrations along the pollution gradient are summarised. A full account of the data is given in the publication by Thørring et al. (1999).

Properties related to the soil acidification status

In Tables 4.1.1 – 4.1.3 data for pH(H_2O), cation exchange capacity, and base saturation factor in the various soil horizons at the five sites are shown. The values listed in the tables are mean values for the 5 plots at each site.

From Table 4.1.1 it is apparent that there was no significant trend in soil pH along the pollution gradient in any of the soil layers sampled. The pH levels observed and the differences observed between different horizons are normal for nutrient-poor boreal forest soils (e.g. Steinnes et al. 1993). Very similar results were obtained for pH($CaCl_2$). Also values for cation exchange capacity (Table 4.1.2) showed only small variations between the sites. Even for base saturation, the factor normally exhibiting soil acidification most clearly, the apparent effects are only moderate. Only at site A the base saturation factor in the O horizon is significantly low compared to the other plots. This is mainly due to replacement of base cations (K, Mg, and to some extent Ca) with other metal cations. From Figure 4.1.1 it is evident that Al is the exchangeable cation showing the greatest increase at site A.

Table 4.1.1. *Mean values ± SD for pH(H_2O) in different soil layers at the various sites.*

Layer	A	B	C	D	REF
Of	3.79 ± 0.08	3.94 ± 0.17	3.79 ± 0.05	3.76 ± 0.15	3.72 ± 0.11
Oh	-	3.75 ± 0.11	3.82 ± 0.07	3.87 ± 0.07	3.73 ± 0.06
E1	4.13 ± 0.16	4.37 ± 0.14	4.13 ± 0.12	4.28 ± 0.15	4.14 ± 0.05
E2	4.27	4.48 ± 0.10	4.23 ± 0.05	4.39 ± 0.08	4.24 ± 0.08
B1	4.58 ± 0.19	4.67 ± 0.15	4.44 ± 0.13	4.61 ± 0.10	4.47 ± 0.11
B2	4.84 ± 0.25	4.88 ± 0.15	4.69 ± 0.14	4.77 ± 0.11	4.72 ± 0.05

Table 4.1.2. *Mean values ± SD for cation exchange capacity (meq kg^{-1}) in different soil layers at the various sites.*

Layer	A	B	C	D	REF
Of	173 ± 42	208 ± 41	242 ± 27	243 ± 22	227 ± 19
Oh	-	235 ± 33	183 ± 28	162 ± 22	168 ± 31
E1	46.4 ± 12.4	31.8 ± 12.6	36.7 ± 12.9	25.2 ± 7.6	20.8 ± 3.7
E2	33.3	20.6 ± 4.4	25.1 ± 3.6	19.7 ± 5.4	16.6 ± 5.5
B1	45.4 ± 6.9	43.1 ± 14.0	45.8 ± 9.5	34.7 ± 6.1	22.1 ± 5.4
B2	17.8 ± 6.3	15.5 ± 8.7	24.1 ± 12.2	23.2 ± 5.2	15.5 ± 5.0

Table 4.1.3. *Mean values ± SD for base saturation factor (%) in different soil layers at the various sites.*

Layer	A	B	C	D	REF
Of	42 ± 15	62 ± 3	69 ± 3	68 ± 1	69 ± 4
Oh	-	65 ± 9	65 ± 5	61 ± 5	55 ± 11
E1	22 ± 8	30 ± 20	40 ± 13	32 ± 12	17.9 ± 7.1
E2	6.8	21 ± 13	21 ± 7	13.3 ± 4.6	9.3 ± 3.3
B1	13.8 ± 2.5	12.1 ± 3.4	9.9 ± 1.4	6.6 ± 1.5	6.6 ± 3.4
B2	26 ± 8	17.0 ± 5.6	10.9 ± 2.4	8.0 ± 2.7	6.1 ± 2.2

On the contrary, in the B-horizon the base saturation factor is higher at sites A and B than at the other sites. Most probably that is due to exchange of acidic cations (H, Al) with base cations released from the surface horizons by acidification and recovered in the B-horizon.

Heavy metals from the smelter

The concentrations of copper and nickel in the litter and O horizon (Figure 4.1.2) show a very strong increase with decreasing distance from the smelter. Concentrations in the mineral horizons are also higher at site A indicating some leakage from the surface layers, but the observed trends indicate that only a very small fraction of the metal has been leached below the maximum sampling depth. The total amount of nickel or copper per area unit at a given site may therefore be used to represent the integrated pollution effect at that site relative to the other sites.

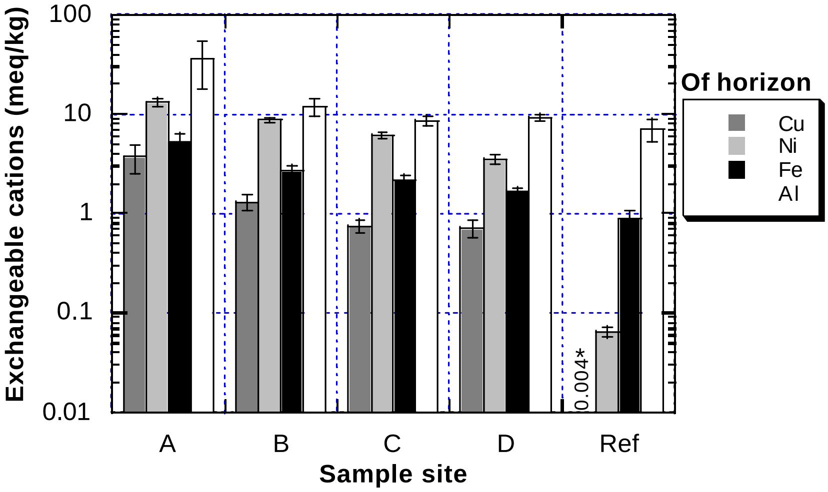


Figure 4.1.1. *Exchangeable concentrations of Al, Fe, Ni, and Cu in the Of horizon at different sites. * Mean values calculated using ½ DL for samples below detection limit (DL).*

The exchangeable concentrations of the two metals in the O horizon (Figure 4.1.3) and their concentrations in the four selected understorey plant species (Figure 4.1.4) show trends that are very similar to those in the litter and surface soil. It may be noted that although exchangeable concentration of Ni is about 5 times higher than that of Cu, their concentrations in vegetation are not much different. *Deschampsia flexuosa* generally shows the highest and *Vaccinium vitis-idaea* the lowest metal concentrations.

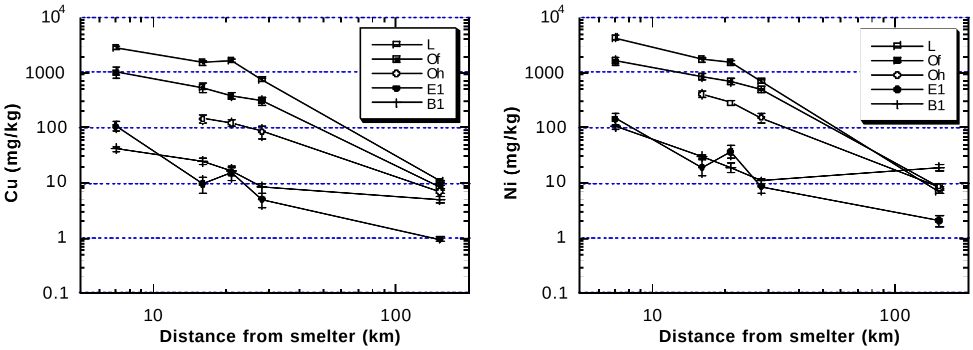


Figure 4.1.2. *Concentrations of Cu and Ni in different soil layers. The points at the distance of 7 km represent results from site A, those at 16, 21, 28 and 152 km sites B, C, D, Reference sites, respectively.*

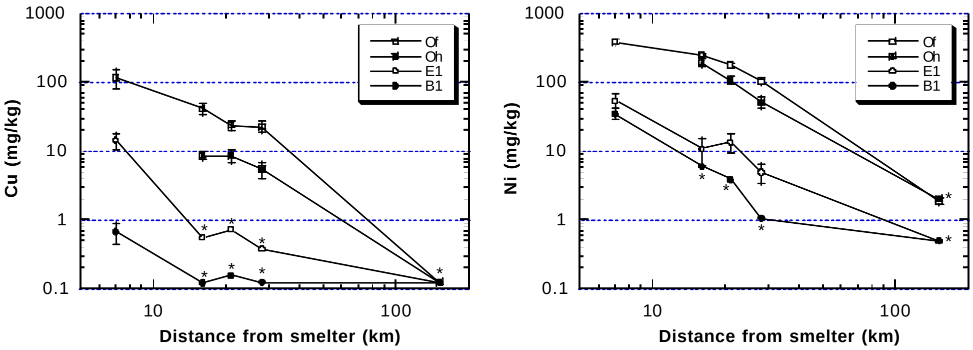


Figure 4.1.3. *Exchangeable concentrations of Cu and Ni in different soil layers. For explanations cf. Figure 4.1.2. * Mean values calculated using ½ DL for samples below detection limit (DL).*

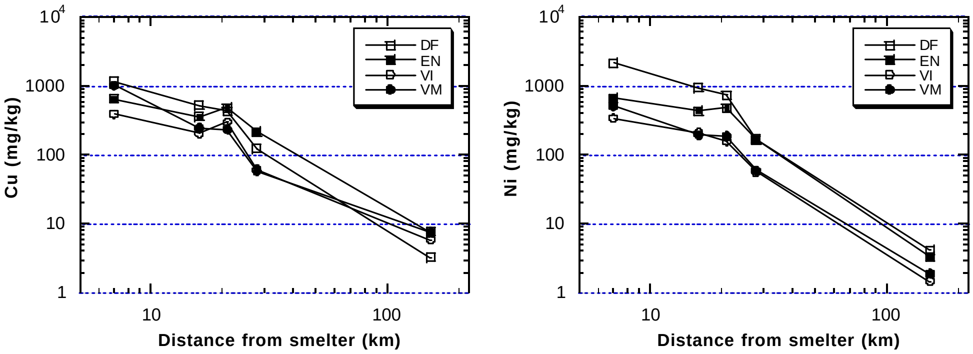


Figure 4.1.4. *Concentrations of Cu and Ni in understorey plant species*:
DF = *Deschampsia flexuosa*, EN = *Empetrum nigrum*
VI = *Vaccinium vitis-idaea*, VM = *Vaccinium myrtillus.*

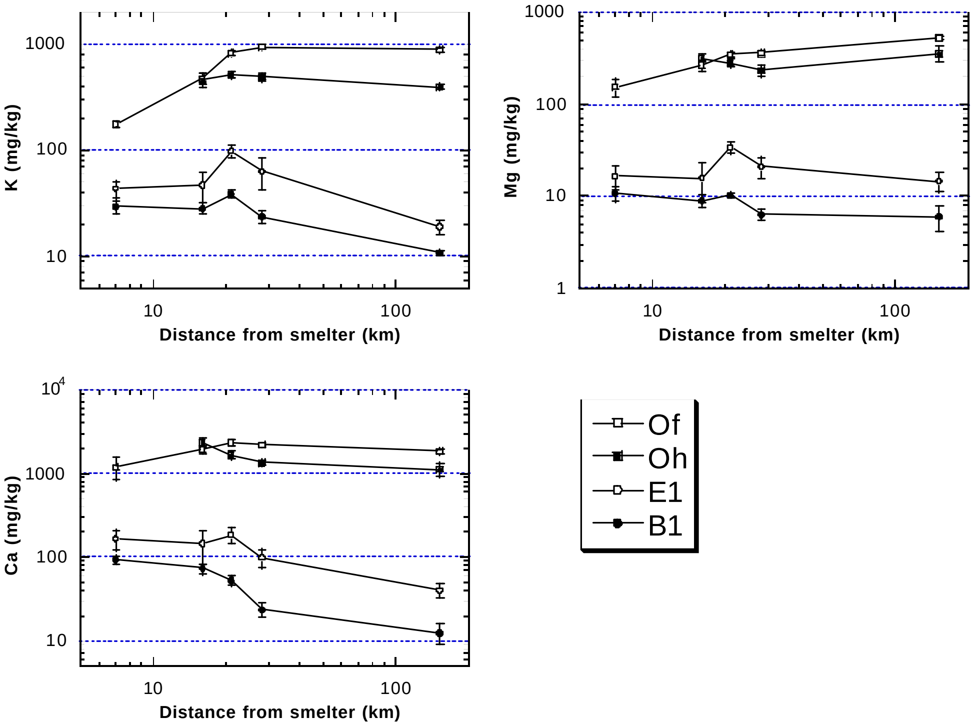


Figure 4.1.5. *Exchangeable concentrations of K, Mg and Ca in selected soil layers.*

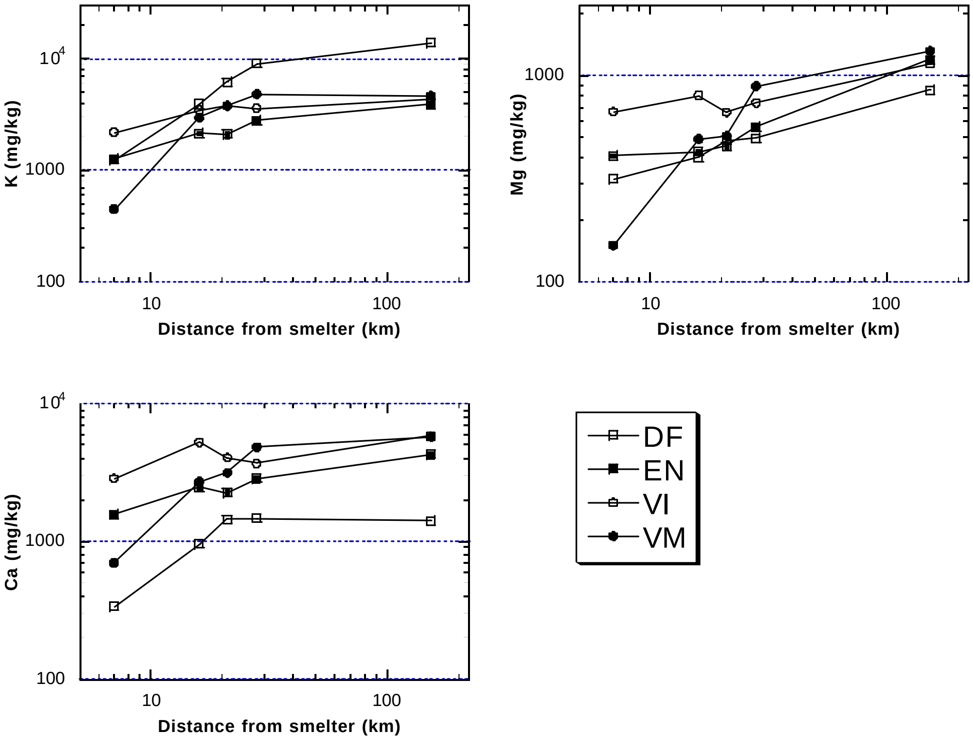


Figure 4.1.6. *Concentrations of K, Mg and Ca in understorey plant species:*
DF = *Deschampsia flexuosa*, EN = *Empetrum nigrum*
VI = *Vaccinium vitis-idaea*, VM = *Vaccinium myrtillus.*

Nutrient elements

Concentrations of exchangeable K, Mg, and Ca in selected soil layers are shown in Figure 4.1.5. For all three major nutrient cations there is a strong depletion in the Of layer with increasing pollution levels, whereas in the lower horizons a similar trend is not evident. In fact there is an increase of all three cations in the B-horizon, as discussed above. The concentrations of these elements in plants, shown in Figure 4.1.6, follow a trend similar to that observed for the exchangeable fraction in the Of horizon with strongly depleted concentrations near the smelter. Manganese shows a similar behaviour as the other major cations with respect to exchangeable fraction and plant contents.

Stable cesium and strontium

The data available for stable Cs and Sr are less extensive than for other elements due to low concentration levels. Total Cs concentrations tend to decrease towards the smelter, but in the litter and Of horizon an increase is observed again at site A. Similar trends are observed in vegetation, probably as a result of extensive resuspension in the strongly damaged areas near the smelter. Concentrations of Sr in the exchangeable fraction and in plants show only a small tendency of decrease towards the smelter, in contrast to K, Mg, and Ca.

Sequential extractions

Samples from all sites and soil layers were subjected to sequential extractions according to the procedure described by Bunzl et al. (1997), and the distributions of Cu and Ni between the four fractions were determined. In the O horizon both metals were found predominantly in the fraction bound to organic matter, and the distribution between fractions did not change much with distance from the smelter. In the E- and B-horizons the organic bound fraction increased and the persistently bound decreased on the most polluted sites. Cu showed a stronger association to organic matter than did Ni.

Summary of main trends

The main trends observed in soil chemistry and plant uptake along the pollution gradient may be briefly described as follows:

- Total concentrations of Ni and Cu in the organic layer increase regularly from about 10 mg kg^{-1} at the reference site to about 5000 mg kg^{-1} at the most polluted site. Similar trends are observed for exchangeable fractions and plant concentrations of the same elements.

- Concentrations of exchangeable K, Ca, and Mg in the Of horizon decrease strongly with increased input of chemical pollutants. A similar strong decrease is not observed in the lower soil horizons. Plant contents generally follow the trends in the O horizon, with the largest relative decrease observed for *Vaccinium myrtillus*.

- For stable Sr no clear trend is observed with distance from the smelter either in soil or plants.
- Only minor changes are observed in soil pH, cation exchange capacity, and exchangeable acidity with distance from the smelter. The vertical distribution of these properties in the soil is normal for podzols.
- Soil acidification at sites near the smelter, as evident from the base saturation, is less pronounced than what might have been expected considering the acidic emissions. This may be explained by the following two facts:

    1. The dry deposition of SO_2 from Monchegorsk is much more widespread than the deposition of particles, and the corresponding deposition gradient therefore much less steep than e.g. for metals. A major part of the SO_2 emitted from the smelter is distributed over rather large areas before oxidised to sulphate and deposited.
    2. There are considerable emissions of alkaline ash particles from the industrial processes which may partly neutralise the effects of acidic deposition at sites near Monchegorsk.

- The main changes in the distribution of exchangeable cations in the organic layer is a higher content of Al, Fe, Ni, and Cu and a lower content of H^+ and base cations near Monchegorsk.

4.2. Fallout radionuclides in the soil/plant system

To investigate the effect of industrial pollution on the transfer of the fallout radionuclides within the soil and from the soil to plants, the following approaches were used:

i) to determine vertical distribution of the radionuclides in the soil at the sites selected in different distances from the smelter in Monchegorsk.

The aim was to examine partitioning of the radionuclides in the various soil horizons and especially to quantify their inventory in the root zone as a function of industrial pollution load at the site. The vertical distribution of the radionuclides in the soil was also used to evaluate the mean residence half-times of the radionuclides at the various sites in the root layer and in the soil layer below the root layer.

ii) to examine the association of the radionuclides with the various soil constituents and to determine the easily exchangeable (also called: readily available) fraction of a radionuclide, a sequential extraction procedure was used. In addition, because it was not possible to apply the sequential extraction to all fresh samples immediately (which would be the ideal case), for the first time in a separate study, the effect of air drying and subsequent sample storage on the fractions extracted for a forest soil (reference site in Finland) and a grassland site in Germany was examined

iii) to detect the effect of industrial pollution on the soil-to-plant transfer of fallout radionuclides, the activity concentrations of the radionuclides in selected plants were determined, and the resulting aggregated transfer factors T_{ag} were obtained for each plant species as a function of the distance from the smelters.

Distribution in the soil profiles and residence half-times

Total inventory of ^{137}Cs, ^{90}Sr and $^{239+240}$Pu.

The total inventories (cumulative activities per unit area in Bq m^{-2}) of these radionuclides at each site are shown in Figure 4.2.1. The amount of radiocesium below the B2 horizon (25 cm depth) was below the detection limit. With help of the observed ratio ^{137}Cs / ^{134}Cs in each soil layer, the inventories were obtained separately for ^{137}Cs from the global fallout and from the Chernobyl-derived fallout. As evident from Figure 4.2.1, the total inventory of ^{137}Cs from the global fallout varies for all plots from for all sites between 820 and 1900 Bq m^{-2}, that of Chernobyl derived ^{137}Cs between 60 and 200 Bq m^{-2}. On average, Chernobyl-derived ^{137}Cs contributes by about 10% to the total ^{137}Cs inventory. A continuous trend of the mean ^{137}Cs inventories from the global fallout within the sites A to D is not detectable (Spearman trend test, $p>0.95$), but the inventory at the reference site is about 20% smaller than that at the other sites. This difference is also statistically significant (U-test by Mann and Whitney, $p < 0,05$, two-tailed). For Chernobyl derived ^{137}Cs, such a difference is not detectable. From the variability of the ^{137}Cs inventories at each site, a mean coefficient of variation of 23% is obtained for the intra-site variability of ^{137}Cs from the global fallout, and of 27% for Chernobyl-derived ^{137}Cs.

^{90}Sr is usually more mobile in the soil than radiocesium. Therefore, it could not be expected that all fallout radiostrontium deposited would still be present in the upper 25 cm layer. The total inventory of this radionuclide at each site was, therefore, calculated with help of global fallout-derived ^{137}Cs inventories and the known ratio of ^{137}Cs / ^{90}Sr = 1.6 in the global fallout (Saxén et al.1987). The contribution of Chernobyl-derived ^{90}Sr to the total ^{90}Sr inventory is negligible in this area (Saxén et al. 1994). The resulting total ^{90}Sr inventories at the various sites (not given here separately) can be compared with the corresponding cumulative inventories, obtained by summing up the ^{90}Sr inventories in all soil layers above the B2 horizon (the deepest soil layer sampled). These results are shown quantitatively in Figure 4.2.1(bottom, right), where the percentage of ^{90}Sr in the soil above the B2 horizon with respect to the total deposition of ^{90}Sr are shown for all sites. Because this fraction is at most plots less than 100%, this demonstrates that indeed a fraction of ^{90}Sr must have been transported to soil layers deeper than sampled by us. The extent of this loss to deeper soil layers is, however, site dependent. While at the reference site at most plots 100% of the deposited ^{90}Sr are still present above the B2 horizon, this amount decreases continually, the closer the sites are located near the smelters. Thus, at site A, where the soil is most strongly polluted, only about 40% of the deposited ^{90}Sr is still found above the B2 horizon. The decreasing trend of the ^{90}Sr in the soil above the B2 horizon is also statistically significant (Spearman trend test, $p < 0.01$). This enhanced loss of ^{90}Sr activity from the upper soil layers at the polluted sites will be discussed in more detail below.

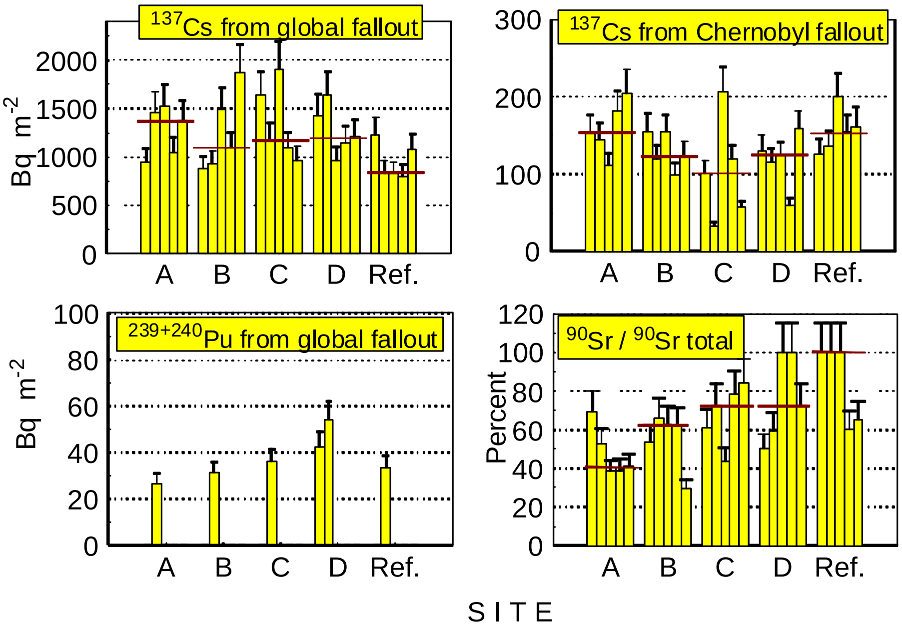


Figure 4.2.1. *Total inventories of ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu from the global fallout of weapons testing, and ^{137}Cs from the Chernobyl fallout in the soil (0 - 25 cm, including the litter layer) at the five soil/plant sites, located at various distances from the smelter (A: 7 km; B: 16 km; C: 21; D: 28 km; reference: 152 km). Because not all ^{90}Sr deposited was found in the soil layer sampled, the inventories of this radionuclide are given with respect to the total amount of ^{90}Sr deposited at each plot. The five bars in each group correspond always to the individual values, determined separately at the five plots at each site; the horizontal lines are the respective medians. The errors shown are one sigma counting errors.*

The cumulative inventory of $^{239+240}$Pu in the soil above the B2 horizon is also shown in Figure 4.2.1. However, because these values are not the means of five plots from each site but were obtained only for one single plot (plot #2) at each site, no information on their variability at an individual site is available. For this reason, no attempt was made to use these data to detect a trend of $^{239+240}$Pu inventory along the pollution gradient from the reference site to the polluted sites. By calculating, however, at each site for plot #2 the ratio of the inventories $^{239+240}$Pu / ^{137}Cs from the global fallout, one obtains for the mean ratio $^{239+240}$Pu /^{137}Cs of all sites a value of 0.032 ± 0.004. This value is rather similar to the corresponding ratio reported during 1962 –1967 for ground level air in Southern Finland, which is 0.018±0.002 (reference date 1997) (Miettinen, 1979). ^{238}Pu/$^{239+240}$Pu ratio in top soil of sampling sites in Russia and Finland was on average 0.029±0.003 indicating that all plutonium present originates from global fallout. According to literature the Chernobyl-derived $^{239+240}$Pu/^{137}Cs ratio observed in 1986 in Southern Finland was also small: 1.8 x 10^{-4} (range 9.1 x 10^{-7}- 8.8 x 10^{-4}) (Reponen et al, 1993). This further proves that the Chernobyl-derived $^{239+240}$Pu in the top soil is negligible.

Vertical distribution of ^{137}Cs, ^{90}Sr and $^{239+240}$Pu

The vertical distribution of the radionuclides as observed in the various soil horizons is illustrated in Figure 4.2.2. They reveal a strong site dependence for the litter layer and the organic horizon: Especially the inventory of total ^{137}Cs and $^{239+240}$Pu in the litter layer decreases with increasing distance from the smelters, while for all radionuclides the inventory in the organic layer increases with increasing distance from the smelters indicating a clear effect of industrial pollution.

These values are needed below for the evaluation of the residence halftimes, but for an understanding of radioecological consequences, it is more relevant to use these values to calculate first the mean inventories of each radionuclide (in Bq m^{-2}) at a given site for the following 3 compartments: i) the litter layer, ii) the root zone (i.e. activities in the O- and E horizon, where essentially all roots we present), and iii) in all combined layers below the root zone (i.e. the B1 and B2 horizons). Dividing the activities in each compartment by the corresponding total inventory (in Bq m^{-2}), the percentage contribution of each compartment to the total inventory is obtained (see Figure 4.2.3). The total inventory for ^{90}Sr was again calculated with help of the corresponding global fallout - derived ^{137}Cs inventories and the known ratio of ^{137}Cs / ^{90}Sr = 1.6 in the global fallout.

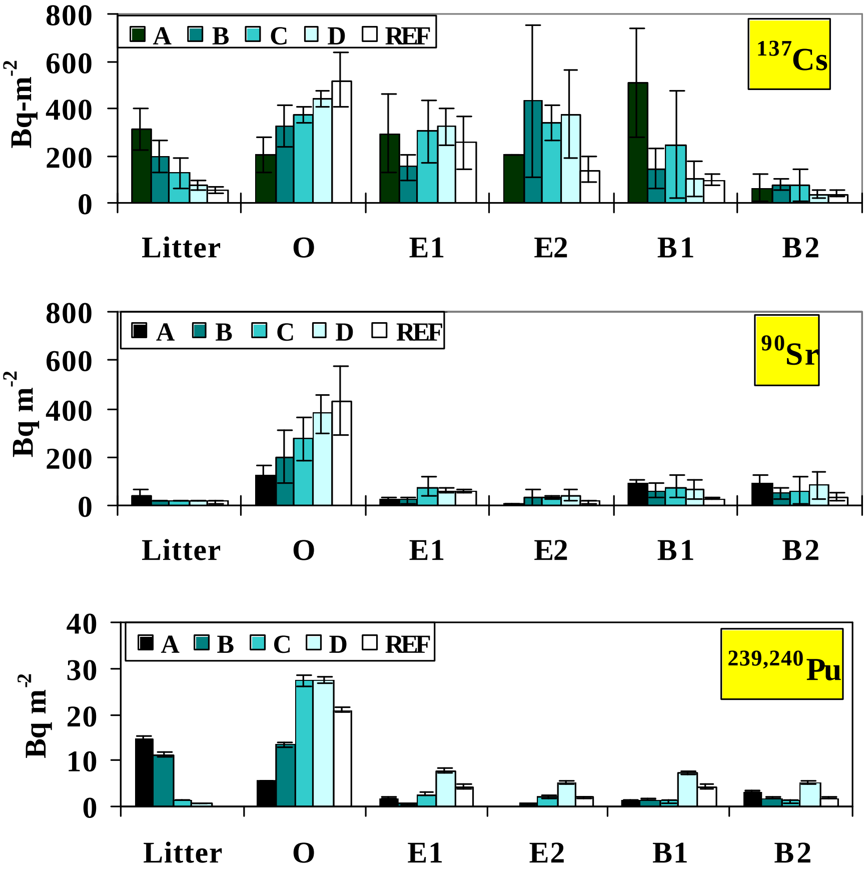
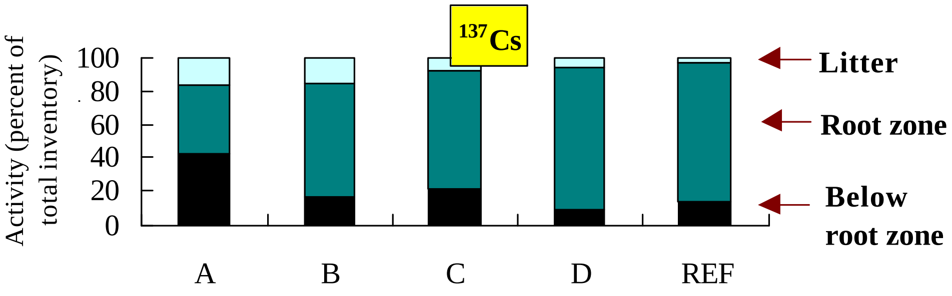


Figure 4.2.2. *Activity concentrations of total fallout ^{137}Cs, ^{90}Sr and $^{239+240}$Put in the different soil horizons at the various sites. The values shown are for each site the means and standard deviation, as obtained by evaluating separately 5 plots each. In the case of $^{239+240}$Pu the values shown were obtained from only one plot at each site (plot #2).*

Figure 4.2.3 reveals clearly the effect of industrial pollution on the distribution of the radionuclides from the global fallout among these three compartments: Thus, in the litter layer, the contribution of ^{137}Cs from global fallout to the total inventory increases continually from the reference site (3%) to the most polluted site A (17%). Similarly, while ^{137}Cs in the root zone contributes to the total inventory by about 80% at the less contaminated sites (reference site and site D) it contributes only to 40% at the site A. As a consequence, one finds at the reference site only 14% of the ^{137}Cs inventory in the layers below the root zone, but 43% at the site A. Most likely, the main reason for this observations is that in the vicinity of the polluted sites the thickness of the litter layer increases while that of the organic layers, i.e. the root zone, decreases. Obviously, this is the result of the inhibited conversion of litter into humic substances at the polluted sites.

In the case of radiostrontium, this trend is even more conspicuous. While about 74% of the total ^{90}Sr inventory is found in the root zone at the reference site, this value decreases continually to 18% at the site A. In the litter layer, only a very small fraction is found: about 2% at the lesser polluted sites, and about 5% at site A. Again, as a consequence, about 25% of the inventory of ^{90}Sr is in the layers below the root zone at the reference site, but as much as 76% at the site A.

A somewhat different pattern is observed for $^{239+240}$Pu. Similar to ^{137}Cs, but much more striking, the fraction of this radionuclide in the litter layer increases from about 1% at the reference site continuously to 56% at the site A. As a consequence, the $^{239+240}$Pu fraction in the root zone decreases from about 80% to 30% at the site A. Rather little change is noticeable in the layers below the root zone: In this compartment, about 20% of the total $^{239+240}$Pu inventory is found at all sites.



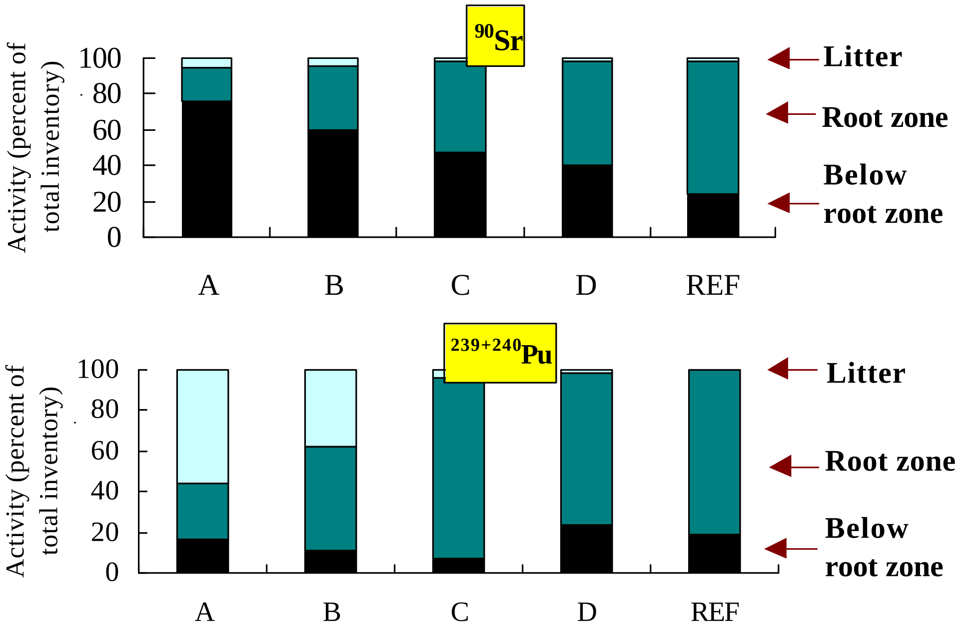


Figure 4.2.3. Partitioning of ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu from the global fallout among the litter layer, the root zone, and the soil below the root zone at the five soil/plant sites. The values given are always arithmetic means as observed at the five plots at each site.

Residence half-times of ^{137}Cs, ^{90}Sr and $^{239+240}$Pu

From the observed vertical distribution of a radionuclide in a soil, residence half-times were evaluated using a multilayer compartment model. Only the mean residence half-time of a radionuclide for the period between deposition and sampling date were calculated, because the changes in the depth profile of a radionuclide over time were not available in the present study. Thus a possible time dependence of the residence half-time could not been determined. Because the thickness of the horizons differed considerably (vertically and horizontally from plot to plot and site to site) the various horizons were subdivided into several compartments with about the same thickness of 1.5 ± 0.5 cm for all horizons at all plots and sites (i.e. the number of compartments varied between 10 and 20) In this model, the transfer of activity A_i (Bq m^{-2}) of a radionuclide in the compartment i in a small time interval Δt (day) is given as

$$\Delta A_i / \Delta t = K_{i-1} A_{i-1} - K_i A_i - \lambda A_i \qquad (1)$$

where K_i (day^{-1}) is the fractional rate of transfer from compartment i-1 to compartment i, and λ is the disintegration constant of the radionuclide. For the first compartment Eq. (1) has to be written as

$$\Delta A_i / \Delta t = RACD - K_i A_i - \lambda A_i \qquad (2)$$

where RACD is the rate of activity deposited per unit area (in Bq cm^{-2} day^{-1}), which must be known as a function of time. To obtain the values of K_i for a given radionuclide in a given soil layer, the differential equation system of Eq. (1) and (2) is integrated numerically. The K_i are varied until the compartment activity matches the observed activity in this compartment. The residence half-time of a radionuclide in layer i is finally obtained as $\tau_i = 0.693/K_i$. For details see e.g..Bunzl et al. (1992). Residence half-times given here do not include the radioactive decay of the radionuclide. For this reason, this kind of residence half-time is sometimes called the ecological residence half-time, because it characterises the change of the activity in each compartment as resulting only from migration processes but does not take into account radioactive decay.

The residence half-times of the three radionuclides from the global fallout were evaluated for the various soil horizons separately for each of the five plots at all sites as described above (with the exception of $^{239+240}$Pu, where only data for the plots #2 were available). These data are shown for the two most important soil compartments of interest: the root zone (Figure 4.2.4) and the soil layer below the root zone; i.e. the B-horizon (Figure 4.2.5).

The residence half-times of ^{137}Cs in the root zone (Figure 4.2.4, top, left) are for the reference and the lesser polluted sites D and C around 30 to

50 years, but they decrease significantly for the most polluted site A, where they are only around 15 years. Application of the Spearman trend test to the data from all sites also confirms the presence of a negative trend at the 0.01 level. However, if we exclude the data from site A, a significant trend is no longer detectable ($p > 0.05$). The absence of significant trend of the residence half-times within the sites: reference, D, C and B thus shows that essentially only the residence half-times of ^{137}Cs at site A are different from those at the other sites. This can be confirmed also by application of the U-test by Mann and Whitney for the comparison of medians ($p < 0.01$).

The reduction of the residence half-times, as one approaches the most polluted sites, is even more conspicuous for ^{90}Sr, which decreases continually from about 35 years at the reference site to about 10 years at site A. For this radionuclide, this trend is statistically significant at the $p < 0.001$ level (Spearman trend test) not only if all sites are considered, but even if site A is excluded.

For $^{239+240}$Pu, a similar trend seems to exist, even though not sufficient data are available for each site to justify the application of a statistical test.

As mentioned, the thickness of the root zone is not constant but is much smaller at site A (only about 2 - 5 cm) than at the other sites (7 - 15 cm). This, and the considerable variability of the thickness of the root zone at each site, is also illustrated in Figure 4.2.4. Because in a compartment model the residence half-time of a radionuclide in a given compartment i will always decrease with decreasing thickness of the compartment, the comparatively short residence half-times at site A of all radionuclides studied here are largely due to the small thickness of the root layer at this site. This can be demonstrated by calculating the residence half-time in years per cm layer of the root zone. Significant differences between the resulting medians at each site are then no longer detectable (U-test by Mann and Whitney, and Spearman trend test at the $p = 0.05$ level), regardless whether we consider ^{137}Cs or ^{90}Sr from the global fall-out. If these residence half-times from all sites are pooled, the mean residence half-times for the root zone are: for ^{137}Cs 4.5 y cm^{-1} (95% confidence limits: 3.5 - 5.6 y cm^{-1}) and for ^{90}Sr 2.2 y cm^{-1} (95% confidence limits: 1.9 - 2.5 y cm^{-1}).

The reduced thickness of the root layer at the most polluted site is an effect of the inhibited conversion of the litter layer to humic matter by the soil micro-organisms. In addition, however, it is conceivable that the chemical composition of the humic substances in the root zone might also be different from that of the lesser-polluted sites. Not only because the type of the micro-organisms will probably depend on the degree of soil pollution, but also because the composition of the litter is different at the polluted sites due to the modified plant communities there. A different chemical composition of the soil organic matter might, however, also have an effect on its interactions with the radionuclides, and thus also on the corresponding residence half-times (when reported in years per cm layer). However, such an effect was not detectable here.

The residence half-times of radionuclides in the soil layer below the root zone (B-horizon) are illustrated in Figure 4.2.5. The residence half-times were evaluated per cm layer, because the B-horizon extended below the sampling depth. For this reason the values given are representative only for the upper part of the B-horizon.

As evident from Figure 4.2.5, the residence half-times of ^{137}Cs are at the reference and the lesser polluted sites about 2 - 5 y per cm soil layer, but about two times higher at the site A. If all values at the sites reference, D, C, and B are pooled and its median compared with that at site A, it is evident that they are also statistically different ($p < 0.01$; U-test by Mann and Whitney).

For ^{90}Sr, the residence half-times are on average (medians) for all sites between about 0.5 to 1 year per cm soil layer. A continuous trend is not detectable. If the residence half-times for ^{90}Sr at site A are again compared with the pooled residence half-times at all other sites, it can again be shown that this difference is also statistically significant ($p < 0.05$; U-test by Mann and Whitney).
The residence half-times of $^{239+240}$Pu are about 1 year per cm layer. It seems that they decrease with increasing pollution of the sites, but because no information on the variability of the residence half-times at each site is available, such dependence cannot be established unambiguously

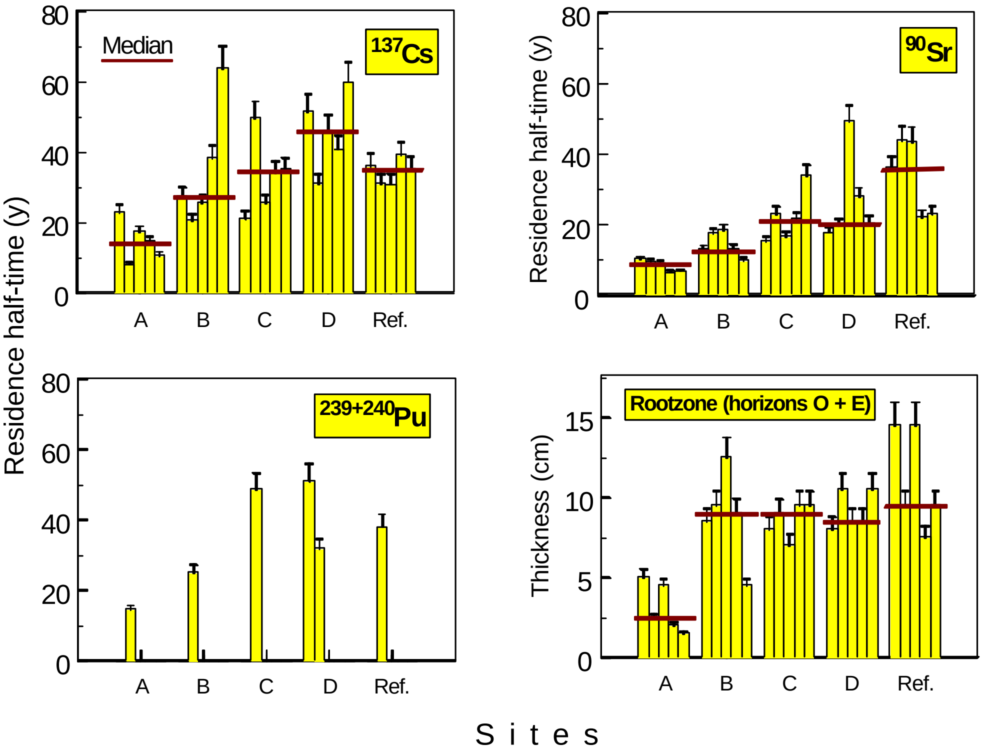


Figure 4.2.4. *Residence half-times (in years) of ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu from the global fallout in the root zone at the five sites The five bars in each group correspond always to the individual values, determined separately at the five plots at each site. The errors given are one sigma errors. Also shown (bottom, right) is the thickness of the root zone (which corresponds to the combined O+E horizons) at the various sites.*

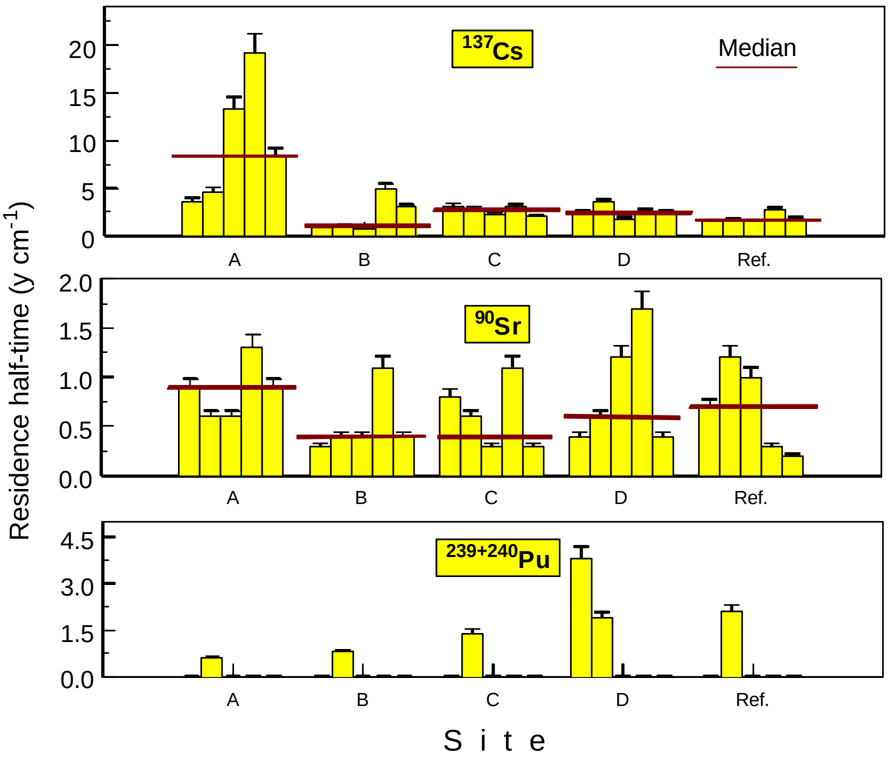


Figure 4.2.5 *Residence half-times (in years per cm soil layer) of ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu from the global fallout in the soil below the root zone (B-horizon) at the five sites. The five bars in each group correspond always to the individual values, determined separately at the five plots at each site. The errors given are one sigma errors.*

No residence half times were evaluated for Chernobyl-derived ^{137}Cs, because the depth profiles of this radionuclide immediately after the deposition are not available for the sites studied. This information is, however, required, unless one can exclude that during this single deposition event, a considerable portion of the total inventory was deposited with a heavy rain shower and, thus, infiltrated already initially rapidly in the soil by by-pass fluxes (Schimmack et al., 1998).

Association of radionuclides with soil constituents

Effect of air drying and storage time

Sequential extraction techniques have been used since many years to obtain information on the form of trace metals in sediments and soils. It was, however, early realised that already during sampling and sample storage a transformation of the species may occur. Bartlett and James (1980) showed e.g. that air- or oven drying might cause instant and major changes in overall sediment and soil equilibrium by converting fractions relevant to trace metal binding into highly unstable and reactive forms. They observed an enhanced solubility of organic matter and manganese as a result of soil drying. In addition, they found that the manganese extractability by pH 4.8 NH_4Ac continued to increase with storage time. As noted by Kersten and Förstner (1995), air drying, sieving and mixing the soil in order to obtain a representative sample for bioavailability analysis may lead to precise but inaccurate results.

In spite of the substantial evidence that sample drying and storage may have an effect on the extractability of trace metals from soils, no analogous information seems to be available for the extraction of radionuclides from these samples. This is somewhat surprising, because in the recent years sequential extraction techniques are used increasingly for investigating also the forms of either artificial radionuclides in the soil, resulting from fallout, such as ^{137}Cs, ^{90}Sr, or $^{239+240}$Pu**.** There is, however, no reason why sample drying may not also have an effect on the extractability of radiocesium or, even more likely, for plutonium, which is known to exist in several oxidation states.

For this reason it was decided to examine within this project the effect of air drying and storage of soil samples on the percentage amounts of fallout ^{137}Cs and $^{239+240}$Pu extracted. Both of these radionuclides originated from the global fallout of nuclear weapons testing and, in the case of ^{137}Cs, also due to the fallout following the accident at the nuclear power plant in Chernobyl. For comparison, also the corresponding natural stable element Cs was determined in all soil samples and extracts. Because, as mentioned above, the effect of drying and storing has been observed for Mn in soils, the existence of this effect was also studied here.

The effect of drying and storage might depend on the form of the radionuclides in the soil. Because this form can again depend on the physicochemical properties of the soil, two different soils types were chosen: The organic layer of a podzolic Al-Fe soil from the reference site in Finland, and a typical grassland soil (Parabrown earth soil, Alfisol) in Germany as an example of a mineral soil.

The sequential extraction procedure used was that of Tessier et al (1979) as modified by Bunzl et al. (1997). It was used to extract the fractions: I) Easily exchangeable: Extraction with1M NH_4Ac; II) Acid reducible (Fe-Mn oxides-bound): Extraction with 0.04 M $NH_2OH{\bullet}HCl$ in 25% (v/v) HAc; III) Acid oxidizable (organic-bound). Extraction with 30% H_2O_2; IV) Residual. Because carbonates were not present in the above soil samples, this fraction was not extracted separately. Fresh soil samples, taken in the field without any pretreatment and storage were used as reference. To study the effect of air drying and storage time, sequential extraction of the above radionuclides and stable elements was applied to air dried soil samples after various time periods, starting from one week up to 30 weeks. To estimate also the reproducibility of the results, duplicate extraction profiles of the above radionuclides and stable elements were always performed after each storage period.

The results obtained are shown in Figure 4.2.6 for ^{137}Cs and stable Cs and in Figure 4.2.7 for $^{239+240}$Pu and stable Mn. For a detailed discussion the reader is referred to the corresponding publication (Bunzl et al. 1999c). The main conclusion of this investigation are:

- A substantial effect of air-drying and subsequent sample storage up to 30 weeks was not detectable for the fallout radionuclides ^{137}Cs and $^{239+240}$Pu, and for stable Cs. Nevertheless, due to a considerable scattering of the extraction profiles, the presence of small trends cannot be excluded. Considering, however, that the natural variability of the extraction profiles, small storage effects should be of no practical importance.

- A clear effect of drying and subsequent sample storage was observable for stable Mn extracted from the mineral soil into fraction I, where its concentration increased continually with storage time. This supports an earlier observation of Bartlett and James (1980) for a similar soil. Nevertheless, because such an effect was not detectable for the soil sample from the organic soil layer of the forest soil, one should be careful to generalise this result.

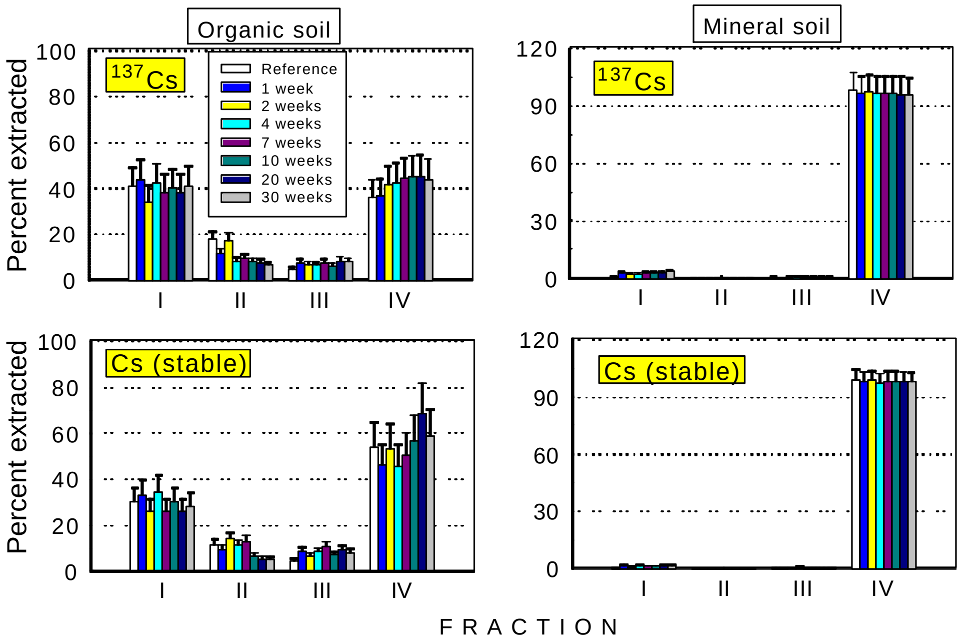


Figure 4.2.6. *Percent distribution of fallout* ^{137}*Cs and stable Cs among the 4 fractions extracted as a function of air-drying and subsequent storage for the two soils. All values are means and errors (2 sigma) from duplicate experiments. The 'organic soil' is the top layer (Of + Oh) from a forest podzol, the 'mineral soil' is the layer 2 - 5 cm from the* A_h *-horizon of an Alfisol. The four fractions extracted were: I, easily exchangeable; II, bound to iron-manganese oxides; III, bound to organic matter; IV residual. The fraction 'bound to carbonates' was not extracted because these soils did not contain any carbonates.*

- It is remarkable that quite similar extraction profiles were found for fallout ^{137}Cs and stable Cs, although these profiles were substantially different in each soiltype. This suggests that isotopic equilibrium was obviously attained in both soils and might also imply that the availability of ^{137}Cs for root uptake can be estimated from the uptake of their stable isotopes, as suggested recently by Rühm et al. (1999) for fallout ^{137}Cs several years after deposition.

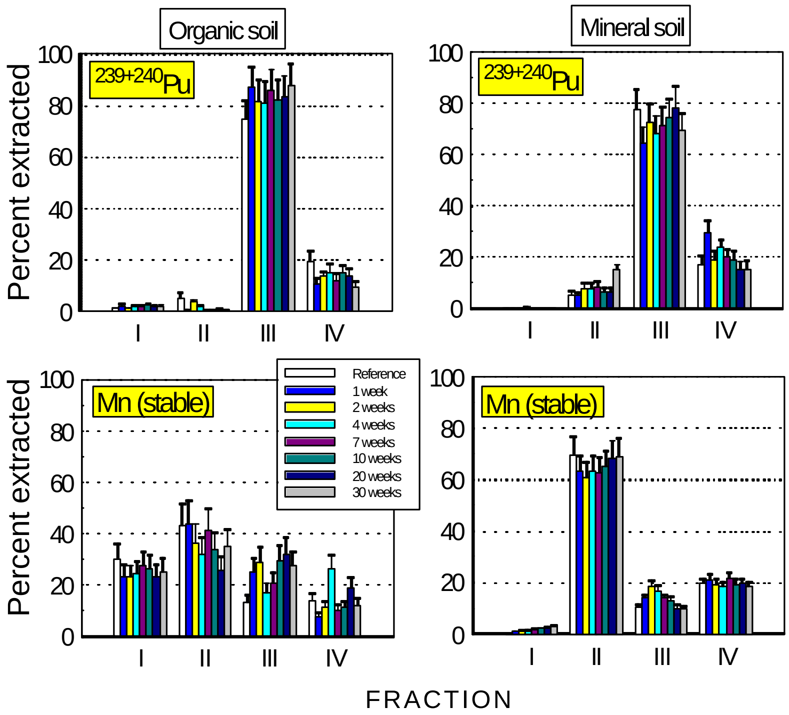


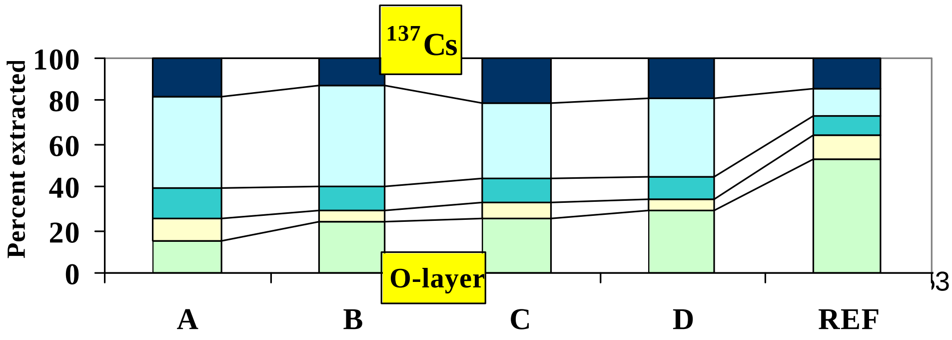
Figure 4.2.7. *Percent distribution of fallout* $^{239+240}Pu$ *and stable Mn among the 4 fractions extracted as a function of air-drying and subsequent storage for the two soils. All values are means and errors (2 sigma) from duplicate experiments. The 'organic soil' is the top layer (Of + Oh) from a forest podzol, the 'mineral soil' is the layer 2 - 5 cm from the* A_h *- horizon of an Alfisol. The four fractions extracted were: I, easily exchangeable; II, bound to iron-manganese oxides; III, bound to organic matter; IV residual. The fraction 'bound to carbonates' was not extracted because these soils did not contain any carbonates.*

Effect of pollution on the association of radionuclides with soil constituents

To investigate the speciation of ^{137}Cs, ^{90}Sr and $^{239,240}Pu$ in soil profiles sequential extraction analyses were performed. Extraction was made using the Tessier method modified by Bunzl et al. (1997). Sequential extraction analyses of ^{137}Cs were performed for the organic (Of, Oh) and first mineral E1 layers at 5 sites at 5 plots each. Sequential extraction of ^{90}Sr and ^{239,240}Pu were performed for the uppermost organic layer (Of) at sites A, B, D and REF. For each site the samples from the five plots were combined before extraction.

The results of sequential extraction of ^{137}Cs in organic and mineral soil are illustrated in Figure 4.2.8. The readily exchangeable fraction of ^{137}Cs increases from about 15 % at site A to 25 % at site D with decreasing pollution load in the organic layer at the sites in the Kola Peninsula. Most of the ^{137}Cs (35 % – 45 %) was persistently bound at sites A-D and no significant variation due to the different pollution load was observed. At the reference site in Finland about half of the ^{137}Cs was in readily exchangeable form and the persistently bound fraction was 15 %. The high percentage of ^{137}Cs in organic layer of the reference site compared to those in Russian sites could be explained by high loss of ignition in this layer (97 %) compared to that at site A (67 %) indicating low presence of mineral constituents. In the mineral E1 layers most of ^{137}Cs (39 % - 50 %) was in the persistently bound fraction at all sites including the reference site.

To examine whether there is statistically significant trend for the percent extracted of ^{137}Cs with increasing distance from the smelter, the Spearman correlation coefficient was calculated. There is a clear positive correlation between the distance from the smelter and the extracted percent of ^{137}Cs in readily exchangeable fraction ($r_{sp}=0.7805$, $p=0.0001$) and an inverse correlation in the persistently bound fraction ($r_{sp}=-0.7650$, $p=0.0001$) and a lower correlation in fraction ’bound to organic matter’ ($r_{sp}=-0.3707$, $p=0.0681$) in organic layers. In E1 layers no correlation was noticed between the ^{137}Cs percentage in different extraction steps and the distance from the smelter



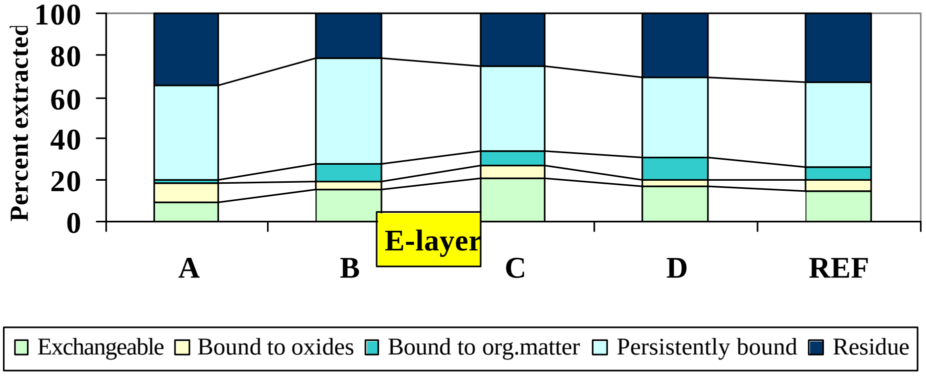
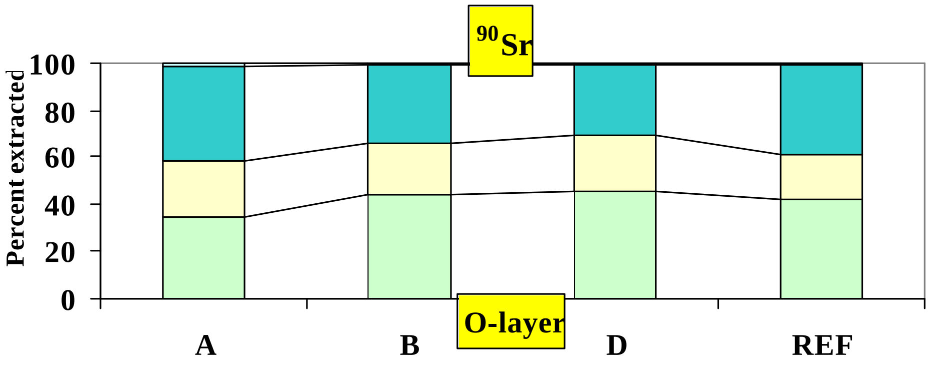


Figure 4.2.8. *Relative distribution of ^{137}Cs in the different steps of sequential extraction of the O- and E1-layers from the Russian sites (A-D) and the reference site (REF).*

The sequential extraction of ^{90}Sr and ^{239,240}Pu in pooled uppermost organic layers along the pollution gradient are shown in Figure 4.2.9. Most of ^{90}Sr was distributed between to the first three fractions: exchangeable, bound to oxides and bound to organic matter. Only 1-2 % of ^{90}Sr was persistently bound. The association of ^{90}Sr in soil particles at the site A differs from the other sites in Russia: less ^{90}Sr was in exchangeable form and more ^{90}Sr was bound to organic fraction compared to the other sites in Russia. In the fraction 'bound to oxide' and 'persistently bound' no effect of pollution was seen.



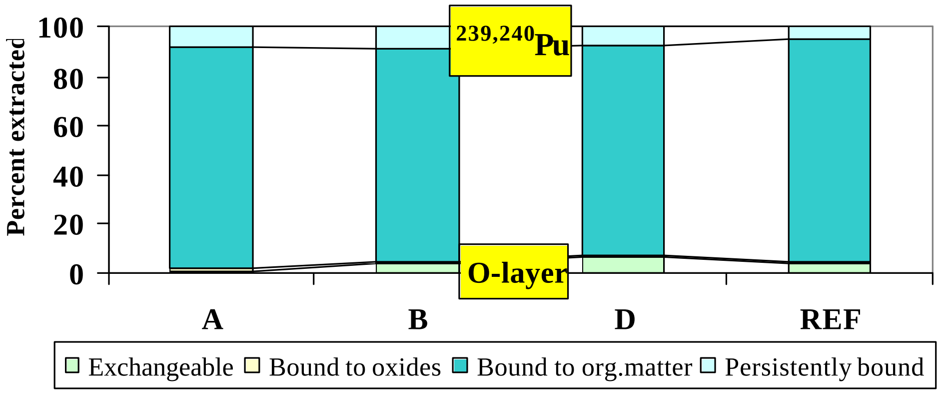


Figure 4.2.9. *Relative distribution of* ^{90}*Sr and* ^{239,240}*Pu in the different steps of sequential extraction of the O-layers from the Russian sites (A, B, D) and the reference site (REF).*

At each site most of ^{239,240}Pu, 85-90 %, was bound in organic fraction. At the site A there was less exchangeable ^{239,240}Pu (1%) compared to the other sites (4-6%). In the fractions ’bound to oxides’, ’bound to organic matter’ and ’persistently bound’ no effect of pollution on extracted ^{239,240}Pu percentages was found.

The explanation for the lower percentages of exchangeable ^{239,240}Pu, ^{90}Sr and ^{137}Cs at site A compared to the other sites is the different composition of organic layer. At this site the organic layer was very thin. The loss of ignition was only 67 % while at the other sites the loss of ignition was over 80 %. It means that there was more mineral material in organic layer at site A, which may result in the decrease of exchangeable radionuclides in the organic layer at this site.

Concentration in plant species and transfer factors

Concentrations in plants

The activity concentrations of a radionuclide in a given plant exhibit frequently a strong seasonal dependence. Thus, for a comparison of values obtained at different sites, it is important that all vegetation samples were collected at the same time. In the present study, this was possible for the sites A to D along the pollution gradient, but not for the reference site. For this reason, we will report in the following only the activity concentrations observed in the various plant species collected at the sites A to D.

The total ^{137}Cs activity concentrations per mass unit in the four plant species collected at each site are shown in Figure 4.2.10. The individual values observed at each of the 5 plots from each site are given there. In general, the activity concentrations for this radionuclide per mass unit are between about 10 and 70 Bq kg^{-1} dry weight. A considerable intra-site variability of these data is apparent. In addition, a clear trend of the ^{137}Cs plant activity concentration with increasing distance from the smelter is present for *Vaccinium myrtillus* and *Empetrum nigrum* (positive trend) or for *Deschampsia flexuosa* (negative trend). This behaviour will be discussed in more detail below in context with the corresponding transfer factors.

In addition to the activity concentrations in whole plants (see above), ^{137}Cs was also determined in the berries of *Vaccinium myrtillus* and *Empetrum nigrum* at all sites. The values found for these samples were rather similar to those observed for the whole plants. An enrichment of ^{137}Cs in these berries with respect to the whole plants at the more polluted sites was not detectable

For ^{90}Sr (Figure 4.2.11) the activity concentrations are between about 2 and 20 Bq kg^{-1} dry weight. Considerable differences between the four plant species are present, the smallest concentrations are mainly found in *Deschampsia flexuosa*. Comparing the values found at the various sites, Figure 4.2.11 shows that, with the exception of *Vaccinium vitis-idaea*, one finds at site A always smaller values for ^{90}Sr in the plants than at the other sites.

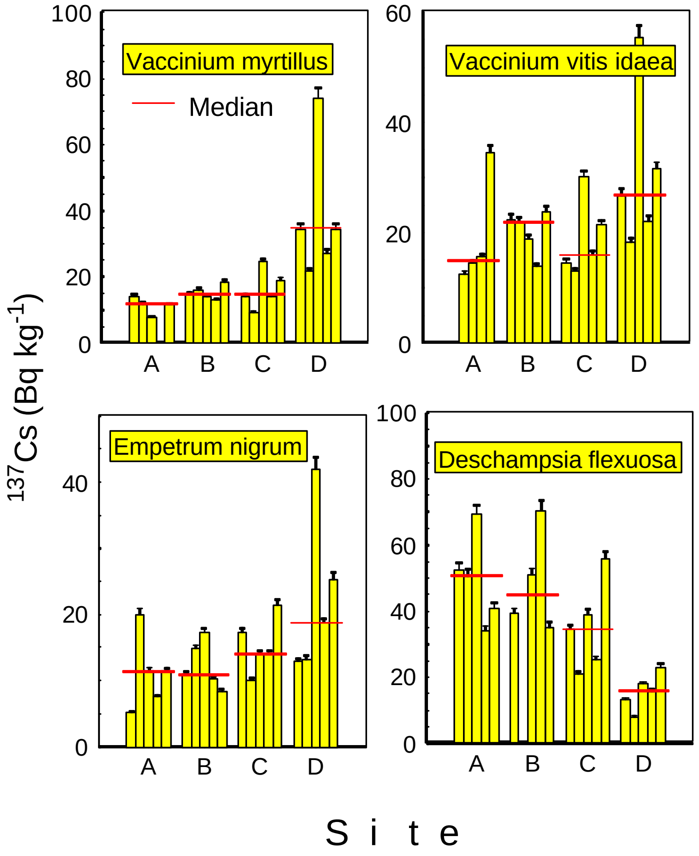


Figure 4.2.10. *Total ^{137}Cs (in Bq per kg dry plant material) in four plant species collected at sites A - D. The five bars in each group correspond to the individual values determined separately at the five plots at each site. The errors shown are one sigma counting errors.*

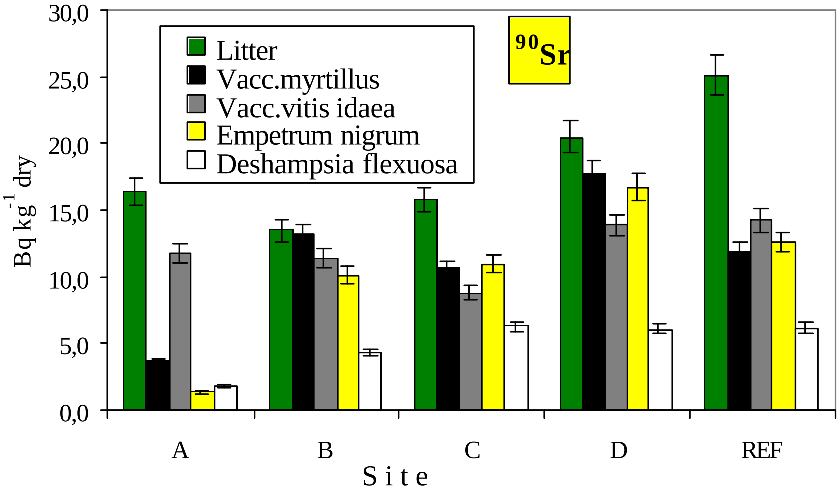


Figure 4.2.11. *Total ^{90}Sr (in Bq per kg dry plant material) in four plant species. To obtain sufficient material for the analysis, these plants were collected around the plots at each site (P-samples). The errors shown are one sigma counting errors.*

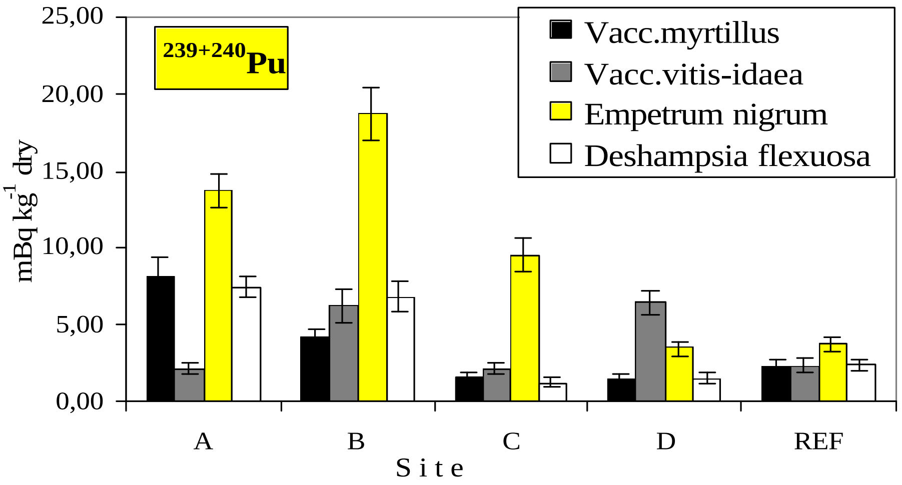


Figure 4.2.12*. Total $^{239+240}$Pu (in mBq per kg dry plant material) in four plant species. To obtain sufficient material for the analysis, these plants were collected around the plots at each site (P-samples). The errors shown are one sigma counting errors.*

For $^{239+240}$Pu (see Figure 4.2.12), the activity concentrations vary between about 2 and 20 mBq kg^{-1}. With the exception of *Vaccinium vitis-idaea*, the concentrations of this radionuclide are higher at the A and B sites than at the more remote sites C and D. Because, however, the soil-to-plant transfer factors of Pu are much lower than those observed for ^{137}Cs and ^{90}Sr (see below), it cannot be excluded that the higher $^{239+240}$Pu concentrations at the highly polluted sites are not due to a larger Pu-rootuptake, but rather to a superficial contamination of the plants with litter. As shown above, this radionuclide is enriched considerably in the litter at the more polluted sites (see Figure 4.2.3), and already a small plant contamination with this material might produce substantial higher $^{239+240}$Pu plant concentrations.

Transfer factors

The inventories and activity concentrations per mass unit of total ^{137}Cs in soils and plants can be used to calculate the resulting aggregated transfer factors T_{ag}, defined as {Bq per kg dry plant material} divided by {Bq per m^2 soil in the sampled soil profile}. These values are shown in Figure 4.2.13 for the four plant species as collected from the five plots at each site. At each site a considerable intra-site variability of the T_{ag} is observable. This variability is, however, to some extent correlated for the different plants. For example, at site D (Figure 4.2.13) T_{ag} increases or decreases at the five plots according to the same pattern for *Vaccinium myrtillus*, *Vaccinium vitis-idaea*, and *Empetrum nigrum*. This is most evident for the 3rd plot, where maximum T_{ag} values are observed for each of these species. Similarly, at site B the highest T_{ag} values for *Vaccinium myrtillus*, *Vaccinium vitis-idaea* and *Empetrum nigrum* are always observed at the first two plots. Another example is site C, where T_{ag} increases rather regularly for the two Vaccinium species when proceeding from the first to the 5th plot. At site A, the T_{ag} at the first plot always exhibits the smallest value for *Vaccinium myrtillus*, *Vaccinium vitis-idaea* and *Empetrum nigrum*. This suggests that if at a given plot of a site the availability for ^{137}Cs is comparatively high, elevated T_{ag} values are likely to be observed for several plants growing there. However, the observation that this coincident behaviour is predominantly observed between *Vaccinium myrtillus*, *Vaccinium vitis-idaea* and *Empetrum nigrum* but not for *Deschampsia flexuosa* indicates the ^{137}Cs uptake for Deschampsia is obviously different from that of the other three species.

As evident from Figure 4.2.13, the median values for the T_{ag} of ^{137}Cs in the four plant species studied are generally between about 0.01 and 0.03 m^2 kg^{-1}. These values are within the relatively large range reported in the literature for the transfer factors of understorey forest vegetation, even though considerably higher values have also been reported (see e.g. Lindner et al., 1994; Wirth et al., 1994; Pietrzak-Flis et al., 1996; McGee et al., 1996; Strebl et al., 1995; Farawis and Johanson, 1995).

The median ^{137}Cs T_{ag} values found for the plants at each plot indicate a trend with increasing distance from the smelter (see Figure 4.2.13). To examine whether this trend is also statistically significant, the Spearman correlation coefficient and the linear correlation coefficient were calculated for the association between the T_{ag} of each plant species and the distance of the site from the smelter. The resulting values demonstrate that the T_{ag} of ^{137}Cs increases significantly ($p < 0.05$) with increasing distance from the smelter for *Vaccinium myrtillus* and *Empetrum nigrum*, but decreases for *Deschampsia flexuosa*. This result is obtained from the linear correlation coefficient as well as from the Spearman correlation coefficient. For *Vaccinium vitis-idaea*, a positive correlation is indicated, but not significant at the $p < 0.05$ level. For a more detailed discussion, which includes also the behaviour of ^{40}K in soil and plants see Bunzl et al. (1999a).

The aggregated transfer factors for ^{90}Sr in the plants from the sites along the pollution gradient are illustrated in Figure 4.2.14. These data show that for all plants from sites B, C and D no significant differences are observable. Only the T_{ag} values from site A are significant smaller (by about a factor of two) than those from the other sites, but with the exception of *Vaccinium vitis-idaea*, which exhibits similar values for all sites.

For $^{239+240}Pu$ (see Figure 4.2.15) the transfer factors increase as one approaches the most polluted site significantly for *Vaccinium myrtillus*, *Empetrum nigrum*, and *Deschampsia flexuosa*. For *Vaccinium vitis-idaea* no trend is detectable. However, as discussed above, because of the very low values of the T_{ag} and the high amount of $^{239+240}Pu$ in the litter layer as one approaches the site A, it cannot be excluded that the above trend are due to a superficial contamination of the plants with this material.

Figure 4.2.13. *Aggregated transfer factor T_{ag} of ^{137}Cs from the global fallout for four plant species collected at sites A - D. The five bars in each group correspond to the individual values determined separately at the five plots at each site. The errors shown are the one-sigma counting errors.*

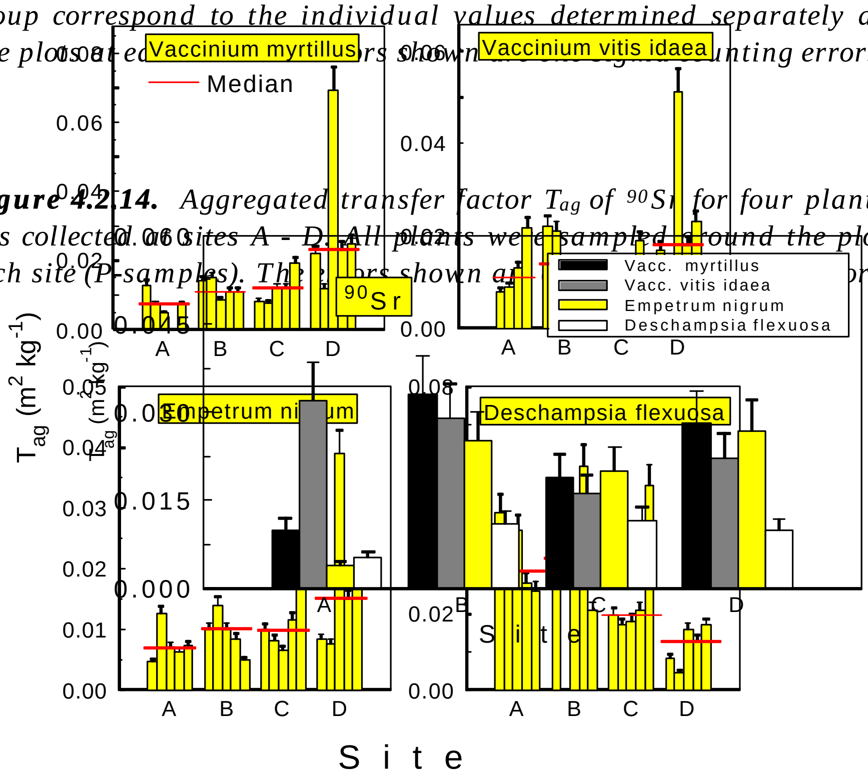


Figure 4.2.14. *Aggregated transfer factor T_{ag} of ^{90}Sr for four plant species collected at sites A - D. All plants were sampled around the plots at each site (P-samples). The errors shown are one-sigma counting errors.*

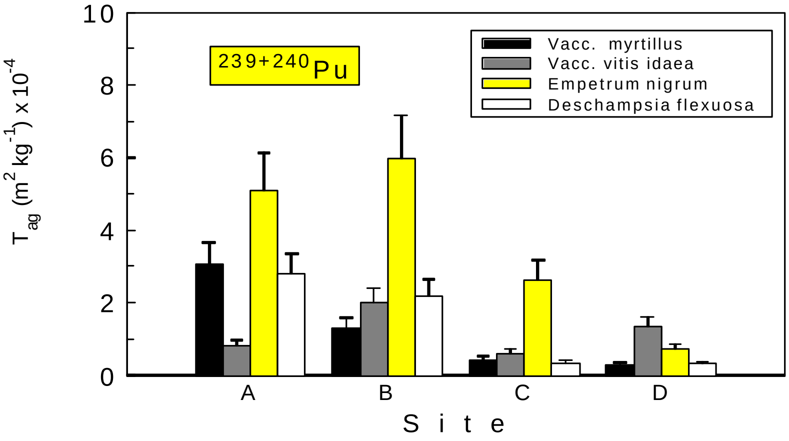


Figure 4.2.15. *Aggregated transfer factor* T_{ag} *of* $^{239+240}$*Pu for four plant species collected at 4 sites A - D. All plants were sampled around the plots at each site (P-samples). The errors shown are one sigma counting errors.*

4.3. Discharge of ^{90}Sr and ^{137}Cs via runoff from Boreal catchments

Distribution in soil and estimated total deposition

The distributions of ^{137}Cs and ^{90}Sr in the two Russian sites RI and RII and in the Finnish reference site RIII are illustrated in the figure 4.3.1a and 4.3.1b. The amounts of ^{137}Cs and ^{90}Sr in the ten samples of organic and mineral soil taken from the river banks (five on each side of the stream at each catchment area) are shown separately. The levels of industrial pollution at the catchments RI and RII correspond the levels at the soil/plant sites A and C and that of the reference catchment RIII the level at the reference soil/plant site (Ref).

The activity distribution of ^{137}Cs and ^{90}Sr

Horizontal distribution

The mean concentrations of ^{137}Cs calculated using the ten samples of organic and mineral soil are for the Russian sites RI, RII and the reference site RIII 1.54, 1.42 and 1.86 kBqm^{-2}, respectively. Also the respective coefficients of variation for each of the three sites RI, RII and RIII are almost the same: 0.35, 0.38 and 0.36. Furthermore, there is no statistically significant difference between these average levels, which imply that the effect from industrial pollution on the total ^{137}Cs inventory is of no significance in the studied areas.

For ^{90}Sr the average activity concentration differ notably between most polluted site RI (131 Bq m^{-2}) and the two other sites RII (447 Bq m^{-2}) and RIII (487 Bq m^{-2}). The coefficients of variation for ^{90}Sr: RI=0.73, RII=0.37 and RIII=0.24, are strongly related to position in the pollution gradient. The trend in the coefficient of variation indicates a substantial effect of the chemical pollution on the horizontal distribution pattern. Furthermore, the analysis made below under Vertical distribution indicates that this is likely to be concomitant to a progressive loss of ^{90}Sr with increasing pollution.

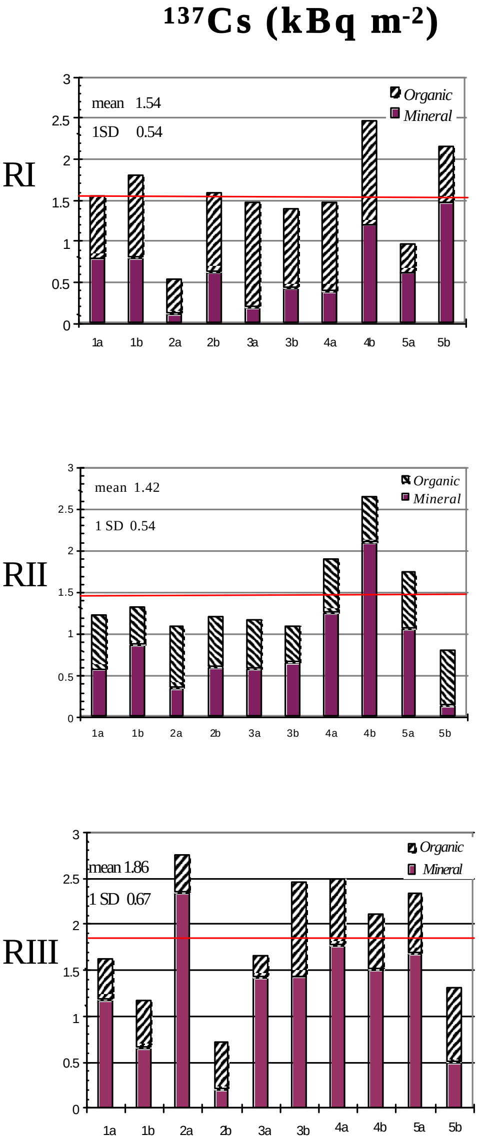


Figure 4.3.1a

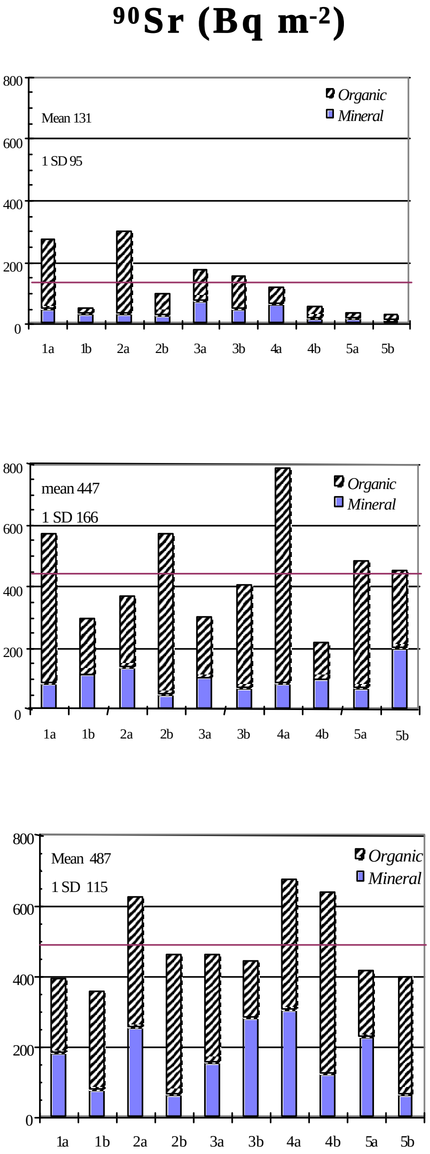


Figure 4.3.1b

Figure 4.3.1. *The soil inventories of ^{137}Cs, kBq m^{-2}, (Figure 4.3.1a) and ^{90}Sr, Bq m^{-2}, (Fig 4.3.1b) in the three studied catchments. The fractions residing in the organic layer and mineral soil are indicated. Suffixes a and b indicate parallel samples at each side of the stream.*

Vertical distribution

The fraction of ^{137}Cs residing in the organic layer about 30 years after the peak of nuclear weapons caesium deposition is on average 50 ± 10% for the studied sites. A trend towards higher ratios in polluted areas is noticed (c.f. Ch. 4.2). This relatively high amount in mineral soil has not been reported for ^{137}Cs of Chernobyl origin in the boreal ecosystems. The present results imply that a progressive net transfer of ^{137}Cs of Chernobyl origin may be expected from the organic to the mineral layers in boreal soils.

For ^{90}Sr 70 ± 5% was found in the organic layer. The ratio exhibit no dependence on site despite the relatively strong influence of the level of pollution on the variability and average remaining amount of activity in the soil horizons.

According to the results of the measurements of ^{134}Cs and ^{137}Cs (cf. Ch.4.2), the deposition associated with the Chernobyl accident contributes about 10% of the total inventory of ^{137}Cs in soil. In areas where nuclear weapons fallout dominate so strongly as at the present sampling sites in the Kola Peninsula and Northern Finland, the contribution of ^{90}Sr from Chernobyl is so small that it can be neglected. The mean ratio of ^{137}Cs to ^{90}Sr in nuclear weapons fallout was 1.6 in the end of 1985 (Saxén et al.1987). Based on this ratio for ^{137}Cs from the nuclear weapons fallout – and neglecting the very small increment of ^{90}Sr by the contribution from Chernobyl (Saxén et al. 1994) – the deposited activity of ^{90}Sr has been estimated for each of the three catchment areas. The ratio of measured activity of ^{90}Sr in the soil column to the estimated total deposition in Figure 4.3.2 indicates the fraction remaining in the soil horizon covered by the sampling, i.e. the organic layer and 15 cm of the mineral soil underneath.

The most polluted catchment area RI exhibits a loss of about 85% of the initial ^{90}Sr deposition. To a large extent this may be due to vertical migration. However, wind erosion is obvious at this site, giving rise to considerable loss of the original organic horizon. Therefore, airborne transport also contributes to the loss at the most affected site. For the other two sites about half of the ^{90}Sr deposition remains in the investigated soil layers.

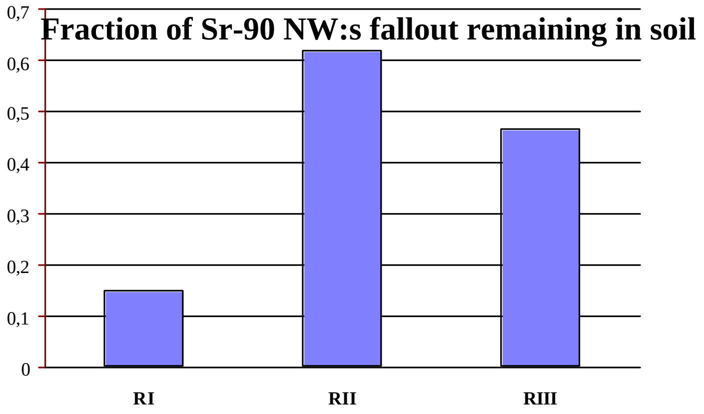


Figure 4.3.2. The fraction of ^{90}Sr in nuclear weapons fallout remaining in soil (the organic layer and 15 cm of the mineral soil beneath) in 1996.

Table 4.3.1 The inventories of ^{137}Cs and ^{90}Sr in kBq m-^{2}: measured values and values calculated for the nuclear weapons fallout (NW). For ^{137}Cs the measured inventory is multiplied by 0.9 to get the NW fraction , and this value is in turn divided by 1.6 to get the NW deposition of ^{90}Sr.

Area	^{137}Cs measured	^{137}Cs/^{90}Sr ratio	^{137}Cs NW	^{90}Sr measured	^{90}Sr NW
RI	1.54 ± 0.54	9.8 ± 7.0	1.39 ± 0.54	0.141 ± 0.095	0.85 ± 0.33
RII	1.42 ± 0.54	2.86 ± 2.95	1.28 ± 0.54	0.447 ± 0.166	0.80 ± 0.33
RIII	1.86 ± 0.67	3.44 ± 1.64	1.67 ± 0.67	0.487 ± 0.115	1.04 ± 0.40

^{137}Cs and ^{90}Sr concentration in runoff-water

The concentration of ^{137}Cs and ^{90}Sr in water sampled at various occasions in 1997 and 1998 (Riekkinen et al. 1998) are shown in table 4.3.2. For most of the sampling occasions at the reference site RIII ^{137}Cs was measured in two separate samples (a and b). ^{90}Sr has been measured in single samples.

Table 4.3.2. The activity concentrations of ^{137}Cs and ^{90}Sr in water from the three catchments. The variation is given as one standard deviation.

Sample	Sampling day	^{137}Cs (Bq m^{-3})	^{90}Sr (Bq m^{-3})	Concentration method
RIa	14.8.1997	0.72 ± 0.14	-	Clinoptilolite
RIb	14.8.1997	0.69 ± 0.14	2.81 ± 0.10	Clinoptilolite
RIIa	13.8.1997	1.76 ± 0.16	-	Clinoptilolite
RIIb	13.8.1997	1.68 ± 0.17	4.28 ± 0.16	Clinoptilolite
RIII1a	20.8.1997	0.42 ± 0.13	-	Clinoptilolite
RIII1b	20.8.1997	0.37 ± 0.13	0.77 ± 0.03	Clinoptilolite
RIII2a	22.6.1998	0.36 ± 0.07	-	Clinoptilolite
RIII2b	22.6.1998	0.29 ± 0.06	-	Clinoptilolite
RIII3	10.8.1998	0.60 ± 0,08	1.31 ± 0.05	Clinoptilolite
RIII4a	23.6.1998	0.37 ± 0.04	-	Organic resin
RIII4b	23.61998	0.57 ± 0.04	-	Organic resin
RIII5	10.8.1998	0.66 ± 0.04	1.1 ± 0.07	Evaporation

Site-specific loss of ^{137}Cs by runoff

The activity ratio, R, defined as the ratio between the mean activity concentration (Bq m^{-3}) in water and the measured average remaining deposition in the catchment (Bq m^{-2}) was calculated for ^{137}Cs and ^{90}Sr, and shown in the table 4.3.3 for the three catchments. The R-norm describes the concentration levels normalised to that for the reference site RIII.

The activity ratio, R, allows for comparisons with findings from other areas in Fenno-Scandia. In figure 4.3.3 this activity ratio in water from the EPORA sites: RI, RII and RIII are compared to corresponding ratios in: a catchment of mainly pasture land, (Arable land Central Sweden), an alpine site in southern Norway (Alpine Norway), boreal forest catchments in Sweden (Northern and Southern Boreal Sweden) and in Finland (Southern Boreal Finland). These results illustrates the strong dependence of the relative concentration of ^{137}Cs in runoff water on fraction of bog, or other wet areas with deep organic soils upstream of the water sampling sites.

Table 4.3.3. The activity concentration ratio for ^{137}Cs and ^{90}Sr . R = the mean activity concentration (Bq m^{-3}) in water and the measured average remaining deposition in the catchment (Bq m^{-2}). The values R-norm are concentration ratios relative to the concentration ratio at the reference area RIII.

The activity concentration ratio, R in runoff water for ^{137}Cs			The activity concentration ratio, R in runoff water for ^{90}Sr	
Catch-ment	R±SD [m^{-1}]	R-norm±SD	R±SD [m^{-1}]	R-norm±SD
RIII 1*	0.00021 ± 0.0017	1 ± 0.74	0.0016 ± 0.00038	1 ± 0.34
RIII 2**	0.00017 ± 0.0001	0.82 ± 0.77		
RIII 3***	0.00032 ± 0.00012	1.5 ± 1.24	0.0027 ± 0.0065	1.7 ± 0.7
RII#	0.0012 ± 0.00042	5.7 ± 4.7	0.0086 ± 0.0032	5.5 ± 2.9
RI##	0.00043 ± 0.00017	2.2 ± 1.8	0.021 ± 0.00079	13.5 ± 10
RIII 13	0.0002 ± 0.000076	0.95 ± 0.79		
RIII 14	0.00031 ± 0.00012	1.4 ± 1.24	0.0016 ± 0.0004	1 ± 0.42
RIII 21			0.0023 ± 0.0005	1.43 ±0.57
RIII 22	0.00044 ± 0.00017	2.1 ± 1.79		

*Sampling date : *)20.8.1997; **)22.6.1998; ***) 10.8.1998; #) 13.8.1997; ##) 14.8.1997

The activity ratios (the R levels in table 4.3.3) for RI fall within the range of the Nordic observations. The other two EPORA sites RII and RIII fall within the lower fraction of the interval for the variability indicated as a general pattern by the results from the other sites in Figure 4.3.3. However, the effective discharge of ^{137}Cs from bogs (2-40% annually, Nylén 1996, 5% annually Hilton et al. 1993) implies a depletion of the inventory of ^{137}Cs in bogs while the inventory in mineral soils is more or less unaffected by time. As an effect of this the R-value will decrease at about the same rate as the activity concentration in water. This process is expected to lead to relatively low levels for the three EPORA sites since they are dominated by "old" caesium (constituting about 90% of the present level). Levels of RI-corr, RII-corr and RIII-corr in Figure 4.3.3 are the corresponding ratios for the three EPORA catchments when corrected for an annual depletion rate 0.5 %. These values represent a predicted situation after about 30 years from the measurements. So they can be compared with the values obtained in the other Nordic studies, which were performed after approximately the same number of years from the main fallout period of the nuclear weapons tests. The corrected values are higher than the values calculated directly from the measurements. Thus, with regard to the uncertainties involved in this assessments there seems to be no notable effect of industrial pollution on the discharge of ^{137}Cs in the studied catchments.

Site-specific loss of ^{90}Sr by runoff

The inventory of ^{90}Sr from the samples of organic and underlying 15 cm of mineral soil is much lower in the RI than in the other studied catchments (cf. Fig 4.3.2). This is probably not explained by differences in deposition in the catchments, since no significant variation is observed between the three catchment areas concerning the measured ^{137}Cs deposition. The loss may rather to a large extent be ascribed to catchments (cf. Fig 4.3.2). This is probably not explained by differences in deposition in the catchments, since no significant variation is observed between the three catchment areas concerning the measured ^{137}Cs deposition. The loss may rather to a large extent be ascribed to the increased mobility of ^{90}Sr under the influence of industrial pollution, allowing penetration to depths in soil below those covered by the sampling. The substantial wind erosion at the most affected site (RI) gives evidence of the possible loss at catchment area also by the atmospheric pathway.

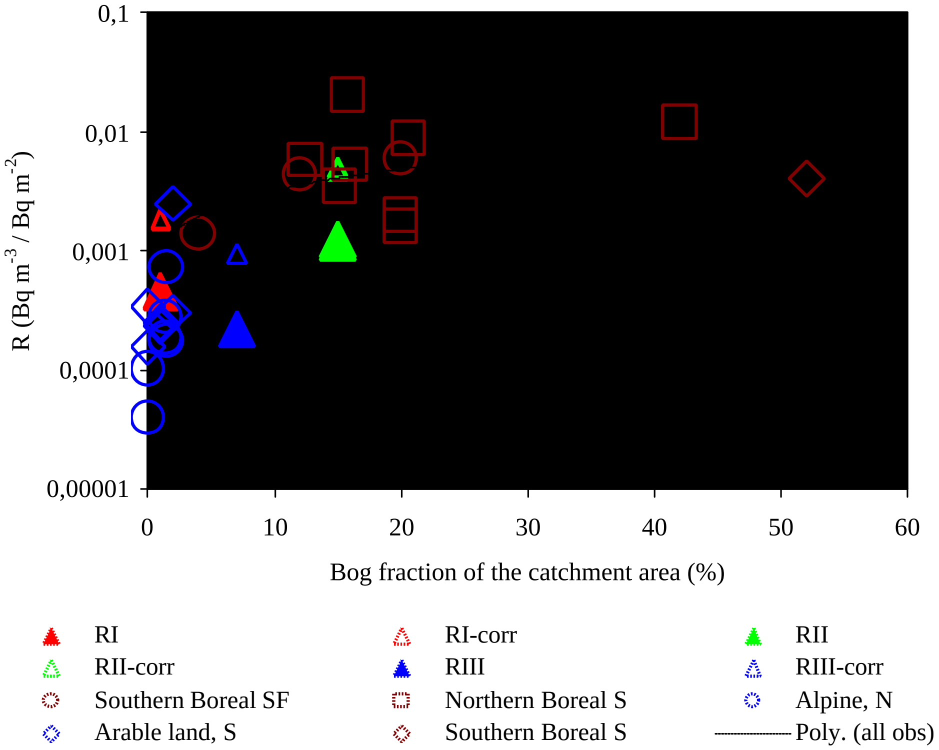


Figure 4.3.3. The ratios of ^{137}Cs activity concentration in water (Bq m^{-3}) to the average deposition (Bq m^{-2}) in the catchment. Results for the Finnish reference catchment area RIII and the two Russian catchment areas (RI and RII) are illustrated in relation to the fraction of the catchment area consisting of bog. Corresponding results for some catchments in Norway (N), Sweden (S) and Finland (SF) are included for comparison. For explanation of the points RI-corr, RII-corr and RIII-corr are given in the text.

A ratio of ^{90}Sr and ^{137}Cs concentration in runoff water at each catchment (cf. Table 4.3.4) has been used for analysing the site dependent behaviour of ^{90}Sr in relation to that of ^{137}Cs. This ratio – illustrated in Figure 4.3.3 – varied between 1.3 and 2.2 for the reference area in the samples from 1997 and 1998, i.e. reflecting the expected higher mobility for strontium in comparison with that for cesium. Despite the much higher content of upstream peat area at the Russian catchment RII – known to strongly influence the loss of ^{137}Cs – the ratio 2.55 of ^{90}Sr/^{137}Cs, although significantly different from that for the reference site RIII, did not deviate much from the ratio for RIII. This implies that the loss of ^{90}Sr and ^{137}Cs in these cases depends rather similarly on the fraction of the catchment area covered by bog.

In contrast, the about two times higher ^{90}Sr/^{137}Cs ratio for RI implies a much greater loss of ^{90}Sr by runoff at that catchment area than expected from the corresponding behaviour of ^{137}Cs. This about two times higher relative loss of ^{90}Sr from the most polluted catchment corroborates with the coinciding loss of about 85% of the initial deposition from the sampled soil columns according to Figure 4.3.2, i.e. a much greater loss than from the other catchments.

The observed strong wind erosion at the most affected catchment area, giving rise to a substantial transfer of organic soil to the stream and streambed, probably contributes to the loss by runoff. However, the ratio ^{90}Sr/^{137}Cs in runoff water appears to be only weakly dependent on the presence of deep organic soil or peat, according to the similar results for the reference site and the Russian catchment RII; the latter with relatively high peat content. This means that any loss of ^{137}Cs from the organic soil percolated by water in the stream is expected to

Table 4.3.4. The Means of the quotient ^{90}Sr/^{137}Cs in water for catchments RI, RII and RIII with standard deviations (SD), coefficient of variation (%) and number of sampling occasions (n).

Catchment	Quotient mean value	SD.	[%]	n
RI*	4.07	0.84	20	1
RII**	2.55	0.28	11	1
RIII***	1.71	0.86	50	4

Sampling dates:
*) 14.8.1997; **) 13.8.1997;
***) 20.8.1997, 23.6.1998, 16 .8.1998 and 10.8.1998.

be accompanied by a two times higher loss of ^{90}Sr. Furthermore, the loss of ^{37}Cs according to Fig 4.3.3 does not reveal any influence beside that likely to be related to a very small peat area. This indicates that the contributions of ^{137}Cs and ^{90}Sr from the atmospheric transfer of organic matter to the stream are not the major cause to the notably changed ratio ^{90}Sr/^{137}Cs in water. A more probable explanation for the relatively high loss of ^{90}Sr from the most polluted catchment is, therefore, the transfer by shallow groundwater in the steep slopes of the stream.

5. MAIN ACHIEVEMENTS

Simultaneous investigations of the extent and composition of industrial pollution and of the distribution and the dynamics of fallout radionuclides in the same area formed the basis for the main achievements of the project. The polluted study area was selected in the surroundings of the huge Cu-Ni smelter in Monchegorsk, in the Kola Peninsula and the reference areas in Northern Finland. The prevailing soil type in the selected study area is a podzol with a low level of nutrients. For this reason, it could be expected that the ecosystem present there should be especially vulnerable to any type of pollution.

Background data on the amount of industrial pollution, on the observable damage of the vegetation, and on the level of fallout of radionuclides in the study area were used to select several sites along a pollution gradient in different distances from the smelter. The main part of sampling was carried out in the Kola Peninsula with the essential contribution of Russian scientists. The details are given in the publication Rahola et al. (1999)

A large amount of valuable and new information concerning the existence and behaviour of heavy metals and sulphur was obtained in the project. The results and the corresponding conclusions are given in the publication of Thørring et al. (1999). They corroborate the level of pollution in the study sites, describe its soil chemical characteristics as well as the amounts of heavy metals and nutrients in soil and plants. The results also showed, for example, that when the concentrations of Cu and Ni in organic soil layer increased, a similar trend was found in plants, but an opposite trend was noticed in concentrations of nutrients in soil. Especially interesting new information was obtained from the measured concentrations of stable Cs (Publication Haugland 1999) and Sr and their relations to the corresponding radioactive analogues.

The radionuclides studied in the project were fallout ^{137}Cs, ^{90}Sr and $^{239+240}$Pu. They are the most important radionuclides when the long-term risks of nuclear fallout are considered. The analysis of these radionuclides in different soil layers and plants yielded for the first time information on their behaviour in the boreal ecosystems as a function of a chemical pollution load. The results indicated that in the organic layer as well as in the whole root zone (organic and eluvial horizons), the fraction of the activity decreased when the amount of pollution on the site increased. The opposite behaviour, however, was found in the litter layer. Main part of the total amounts deposited of ^{137}Cs and $^{239+240}$Pu were found in the litter and soil layers sampled. However, at the polluted sites, a rather large fraction of ^{90}Sr had percolated to deeper soil layers, and this fraction seemed to be related to the pollution load (Publication Bunzl et al. 1999b).

The results also showed that the residence half-times of the radionuclides in the root zone decreased when the pollution load at the site increased. With respect to the external dose rate in 1 m height from ^{137}Cs in the soil, the study revealed no significant differences between the reference site and the polluted sites. Due to the importance of the organic layer for retaining the ^{137}Cs deposition and the special features of the soil, the external of gamma radiation was, however, higher than observed for typical grasslands, e.g. in Bavaria, Germany, even when the data were normalised to the same amount of radiocesium deposited. This should be kept in mind when future risks of external radiation in northern boreal environment will be predicted after a possible new deposition. A more detailed description is given in publication (Publication Bunzl et al. 1999b).

Sequential extraction studies and radiochemical determinations of radiostrontium and plutonium are usually complicated and time consuming. As a consequence of the great number of samples to be analysed, the time between soil sampling and pre-treatment before analysis can sometimes be up to several months. Previous experience has shown that this might affect, however, the results of extraction studies of some trace metals from soils. Therefore, the effect of air drying and storage time was also tested for the radionuclides ^{137}Cs and $^{239+240}$Pu analysed in this study. Fortunately, the results indicated that this effect was not observable in the sequential extractions of the radionuclides (Bunzl et al. 1999c). The association studies of radionuclides with soil constituents indicated that the exchangeable fraction of ^{137}Cs in the organic soil decreased with pollution load, but that for ^{90}Sr and $^{239+240}$Pu the decrease was not as apparent as for ^{137}Cs. These sequential extraction studies of the soil samples gave for the first time information of speciation of radionuclides in the arctic boreal environment (Puhakainen et al 1999).

The effect of industrial pollution on the soil to plant transfer factors of radionuclides proved to be dependent on both radionuclide and plant species studied. For crowberry *(Empetrum nigrum)* and blueberry *(Vaccinium myrtillus)* the aggregated transfer factors decreased for ^{137}Cs (Publication Bunzl et al. 1999a) and ^{90}Sr at most polluted sites but increased in the case of $^{239+240}$Pu. For the forest hair grass (*Deschampsia flexuosa)* the transfer factors decreased with pollution load for ^{90}Sr but increased for ^{137}Cs and $^{239+240}$Pu. For lingonberry *(Vaccinium vitis-idaea)* no trend was detectable for any of the radionuclides studied

Results from the runoff study supported the soil profile data. The total ^{137}Cs inventory in the 0- 25 cm layer is not dependent on pollution load. For ^{90}Sr, however, there is a progressive loss from the upper soil with increasing pollution, with 85% of the initially deposited ^{90}Sr lost from the catchment closest to the pollution source. Comparison of the activity ratios of these nuclides in catchment soil samples and in runoff water shows that ^{90}Sr is transferred by shallow groundwater to the streams and not removed by wind erosion. To enable analysis of the very low concentrations of ^{90}Sr and ^{137}Cs in the streamwater a new preconcentration method was developed. The method uses the natural zeolite *clinoptilolite* as an ion exchanger. It was tested in the laboratory and used in field conditions.

6. CONCLUSIONS ON EFFECTS OF INDUSTRIAL POLLUTION ON THE DYNAMICS OF RADIONUCLIDES

Effect of pollution on the distribution, migration and association of radionuclides in soil

Main part of the ^{137}Cs and $^{239+240}$Pu deposited by the global fallout was found at all sites to be still present in the upper soil layers sampled (0 - 25 cm). For ^{90}Sr, however, this is not the case. A considerable fraction of this radionuclide has been meanwhile transported to soil layers deeper than sampled for the project. The extent of this loss to deeper soil layers is site dependent. While at most plots at the reference site 100% of the deposited ^{90}Sr are still present above the B2 horizon, this amount decreases continually, the closer to the smelter the sites are located. Thus, at site A, where the soil is most strongly polluted, only about 40% of the deposited ^{90}Sr is still found in the 0 - 25 cm layer.

A strong dependence on the extent of industrial pollution is also found for the partitioning of the radionuclides among the following three soil compartments: the litter layer, the root zone, and the soil below the root zone: The contribution of all three radionuclides in the litter layer to the corresponding total inventory increases continually from the reference site to the most polluted sites. This effect is most pronounced for $^{239+240}$Pu and least for ^{90}Sr. For the root zone the opposite effect is observed: In this compartment the largest fraction of all radionuclides are found at the reference site, and only rather small fractions at the more polluted sites. As a consequence, only a small fractions of ^{137}Cs but especially of ^{90}Sr of the total inventory are found in the layers below the root zone at the reference site, but rather high fractions at the polluted sites. Most likely, the main reason for these observations is that in the vicinity of the polluted sites the thickness of the litter layer increases while that of the organic layers and the E-horizon, i.e. the root zone, decreases. Obviously, this is the result of the inhibited conversion of litter into humic substances at the polluted sites.

Using a sequential extraction method to study the association of the radionuclides with the soil constituents, the following conclusions can be drawn:

- A substantial effect of air-drying and subsequent sample storage up to 30 weeks was not detectable for the sequential extraction of the fallout radionuclides ^{137}Cs and $^{239+240}$Pu, and for stable Cs as demonstrated for the first time for the organic forest soil from the study area and a typical mineral grassland soil in Germany.

- It is remarkable that quite similar extraction profiles were found for fallout ^{137}Cs and stable Cs, even thought, for a given element, these profiles differed substantially in the studied and German soils. This suggests that isotopic equilibrium was obviously attained in both soils and might also imply that the availability of ^{137}Cs for root uptake can be estimated from the uptake of the stable isotope, as suggested recently by Rühm et al. 1999 for fallout ^{137}Cs several years after deposition.

- Association of radionuclides in soil constituents was found to be influenced by industrial pollution. In organic layer the amount of exchangeable ^{137}Cs decreased with pollution. In mineral layer pollution did not have any significant effect on association of ^{137}Cs. The amount of exchangeable ^{90}Sr and $^{239+240}$Pu in organic layer also decreased with pollution, but the phenomenon was not as significant as in the case of ^{137}Cs. The explanation for the lower percentages of exchangeable ^{137}Cs, ^{90}Sr and ^{242}Pu might be the different composition of organic layer. At most polluted sites the organic layer was very thin and contained more mineral material compared to the other sites.

Effect of pollution on the residence half-times of radionuclides in the soil

The residence half-times of the radionuclides in the root zone of each site depend especially for ^{90}Sr, but also for ^{137}Cs and $^{239+240}$Pu significantly on the industrial pollution. For all three radionuclides a considerable reduction of the residence half-times was found as one approaches the more polluted sites. Again, this effect is essentially due to the analogous decrease of the thickness of the root zone at the more polluted sites.

In the soil layer below the root zone, no effect of the industrial pollution on the residence half-times was detectable for ^{90}Sr and $^{239+240}$Pu, only the residence half-time of ^{137}Cs was somewhat longer at the most polluted site as compared to all other sites.

Effect of pollution on the transfers of radionuclides from soils to plants

The effect of an industrial pollution on the transfer of fallout radionuclides from soil to plant depends not only on the radionuclide considered but also on the plant species studied:

- The root-uptake of ^{137}Cs, as characterised by the median aggregated transfer-factor T_{ag}, decreased significantly with decreasing distance from the smelter for the plants Vaccinium myrtillus and Empetrum nigrum, but increased for Deschampsia flexuosa. For Vaccinium vitis-idaea a significant trend for the T_{ag} was not observable. The median ^{40}K activity concentrations in these plants decreased also significantly with decreasing distance from the smelter for Vaccinium myrtillus, Empetrum nigrum, and also for Deschampsia flexuosa. For Vaccinium vitis-idaea such a continuos significant trend was not observable. The results for the Cu-Ni polluted soils thus show i) that the soil-to-plant transfer of radiocesium can be significantly modified, ii) that these modifications are quite plant specific, and iii) that modifications of the uptake of potassium do not always correspond to those of radiocesium.

- The aggregated transfer factors for ^{90}Sr from the sites along the pollution gradient show that for all plants from the lesser polluted sites no significant differences are observable. Only the values from the most polluted site are significantly smaller (by about a factor of two) than those from the other sites, but with the exception of Vaccinium vitis-idaea, which exhibits similar values for all sites.

- For $^{239+240}$Pu the transfer factors increase as one approaches the most polluted site significantly for Vaccinium myrtillus, Empetrum nigrum, and Deschampsia flexuosa. For Vaccinium vitis-idaea no trend is detectable. However, because of the very low values of the transfer factor for this element, and the high amount of $^{239+240}$Pu in the litter layer as one approaches the more polluted sites, it cannot be excluded that the above trends are due to a superficial contamination of the plants with this material.

It is interesting to examine the trends observed above for the radionuclides along the pollution gradient with those found for the distribution of nutrients and heavy metals. A comparison reveals that not only the transfer factors of e.g. ^{137}Cs and ^{90}Sr for some plants decrease with an increased input of chemical pollutants but also the concentrations of exchangeable nutrient cations K, Ca, and Mg in the Of horizon. Also, at the more polluted sites, one observes in the organic layer higher contents of exchangeable cations Al, Fe, Ni, and Cu and a lower content of H^+ and base cations. One should note, however, that the analogous trends in the transfer factors mentioned are not observed for the all plants and not for all radionuclides. This demonstrates that the trends found for the nutrients and other exchangeable cations can produce quite different trends for some other plants and other radionuclides. At present, it is not clear why some plants exhibit so different trends along the pollution gradient with respect to their root uptake, even for the same radionuclide. Plant specific effects are obviously of considerable importance, and demonstrate that a generalisation of the present results to other plants investigated here is hardly possible.

Effect of pollution on the loss from radionuclides from the area due to run-off

The average deposition of ^{137}Cs in the two catchments close to Monchegorsk in Russia and in the reference site in Finland are not significantly different according to results obtained and the major contribution (about 90%) stems from the atmospheric nuclear weapons tests. The small contribution from the Chernobyl accident means that the average deposition of ^{90}Sr should also have been similar in all the studied catchments, and that the ratio of ^{137}Cs to ^{90}Sr deposition should be close to 1.6 for the about 90% of nuclear weapons origin.

Of the total ^{90}Sr deposition only about half is still present in the organic horizons and the upper 15 cm of the underlying mineral soil in the Finnish reference site (RIII) and the Russian catchment (RII) subjected to an intermediate level of chemical pollution. In the most affected catchment (RI) close to Monchegorsk only 15% of the estimated total deposition of ^{90}Sr remains at these depths in soil. 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 streamwater corroborate with a relationship based on the fraction of the upstream catchment area containing deep organic soil – found to be valid at several Nordic sites lacking significant 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. On the contrary, in the most polluted Russian catchment the activity concentration of ^{90}Sr in the streamwater is about twice as high as expected from the activity ratios ^{90}Sr/^{137}Cs in the Finnish reference area and the Russian catchment subjected to an intermediate level of chemical pollution. Furthermore, this coincides with a loss of about 85% of the initial deposition from the sampled soil columns in the most affected Russian site, i.e. a much greater loss than from the other catchments.

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

The increased rate of ^{90}Sr transfer to the streamwater in the heavily polluted catchment points to the importance of considering the concomitant input to downstream aquatic recipients in landscapes affected by chemical pollution and the potentially increased contamination in fresh water food-chains over fish to man.

Effect of pollution on radiation doses

Radiation exposure of man and animals due to fallout radionuclides in soil and plants can result from inhalation of resuspended soil, ingestion of food, and - in the case of radiocesium - also from an external radiation exposure. While a calculation of the doses resulting from inhalation or ingestion was not within the scope of this project, it was studied whether the industrial pollution affected the external gamma dose-rate in the study area.

The external gamma radiation due to ^{137}Cs in the soil depends on the vertical distribution of this radionuclide in the soil as well as on the wet bulk density of the various soil layers. Because these data were determined at all plots from all sites, we calculated the resulting gamma dose-rate in 1 m height above ground according to Jacob et al. (1994). The resulting values are shown in Fig. 6.1 separately for ^{137}Cs from the global fallout and from the Chernobyl fallout. For comparison, they are given for each site with respect to a total deposition of 1 kBq ^{137}Cs per square meter. The values shown are for each site the means and standard deviations as obtained by calculating the dose-rates separately for each of the five plots at each site.

A comparison of the external dose-rates calculated for the four polluted sites A to D and the reference site shows:

- At each site the dose rate from Chernobyl-derived ^{137}Cs (normalised to a deposition of 1 kBq per m^2) is about 1.6 times higher than that from ^{137}Cs from global fallout. This is not surprising, because Chernobyl-derived ^{137}Cs is closer to the soil surface than ^{137}Cs from the global fallout. Because, however, the total deposition of Chernobyl-derived ^{137}Cs is only about 10% of the total ^{137}Cs inventory of the soil the total external dose-rate due to ^{137}Cs in the soil results mainly from ^{137}Cs that has been deposited during the global fallout. Using the values determined for the depositions of ^{137}Cs from both sources and the normalised dose-rates from Fig 6.1, a total external dose rate of about 1.5 nGy h^{-1} is obtained for all sites.

- Within local variations at each site, no significant differences for the external dose-rates are present at the various sites, although the vertical distribution of ^{137}Cs in the soil is site dependent. However, because of the very low wet bulk densities of the litter and the organic horizons in the study area (about 0.1 g cm^{-3}) the attenuation of the gamma rays is quite small and different depth profiles of ^{137}Cs in the soil produce hardly any changes in the dose-rates at 1 m height above ground. To demonstrate the effect of the wet bulk density of a soil on the external gamma-dose rate, thewe show, for comparison, in Fig.6.1. (right side) also the corresponding mean dose rate observed for typical grassland (mineral) soils in Bavaria/Germany (Schimmack et al., 1998), where the wet bulk densities are about 1g mL^{-1}, and ^{137}Cs is also found essentially only in the upper 25 cm layer. At these sites, the external gamma-dose rate (again with respect to a total ^{137}Cs deposition of 1 kBq per m^{2}) is only about half as high as observed at the boreal forest sites in the study area

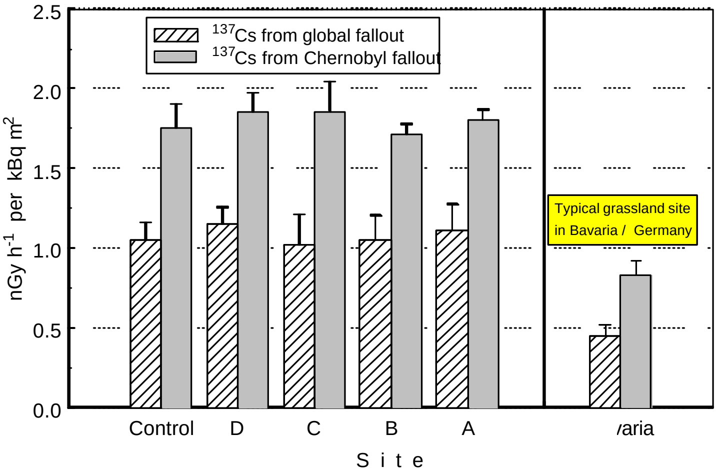


Fig. 6.1. *Gamma dose-rate in 1 m height above ground due to ^{137}Cs in the soil the five sites, located at various distances from the smelter (A: 7 km; B: 16 km; C: 21; D: 28 km; reference: 152 km). For comparison, the dose rates resulting presently from fallout ^{137}Cs in the soils at typical grassland sites in Bavaria Germany are also given. All values are normalised with respect to a total deposition of 1 kBq ^{137}Cs per m^{2}.*

Applicability of the methodology to other polluted areas

The purpose of the project was to estimate the effects of industrial pollution load on the dynamics of radionuclides in boreal understorey ecosystems. The study area was chosen from the Kola Peninsula where the enormous atmospheric emissions of the smelter in Monchegorsk had continued for decades and thus polluted the environment to a long distance. The polluted study areas were chosen at different sites along the decreasing pollution gradient and thus offered a possibility to study the effects as a function of pollution load.

The radioecologicalstudies included determination of migration of radionuclides in soil, their association into different soil constituents, transfer from soil to plants and runoff water as well as effects on radiation exposure rates. The long-lived radionuclides ^{137}Cs, ^{90}Sr and ^{239,240}Pu from the global fallout from the nuclear weapons testing offered the possibility to study long-term changes in the ecosystem and as a consequence effects on the long-term radiation risks and are, in principle, as such applicable to other polluted environment. However, although the result showed the effect of pollution on the radioecological quantities studied, they also showed that the results are dependent on the type soil/plant ecosystem or soil/runoff water system.

Nevertheless, the methodology (i.e. the experimental approach and the data evaluation) used or developed in thisstudy should be applicable also for other polluted areas.. In addition, the methodology should be also useful when evaluating the efficiency of restoration procedures applied to areas which are polluted not only by radionuclides but also by chemicals.

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