

Proceedings of the International Symposium on Radioecology 1996

Ten Years Terrestrial Radioecological Research Following the Chernobyl Accident

22nd-24th April 1996, Vienna

Austrian Soil Science Society

in cooperation with the

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INHALTSVERZEICHNIS

G. DESMET: OVERVIEW OF EC COORDINATED RADIOECOLOGICAL RESEARCH AFTER THE CHERNOBYL ACCIDENT	1
W. SCHIMMACK, K. BUNZL: MOBILITY OF CHERNOBYL-DERIVED RADIOCESIUM IN THE SOIL	11
C. MURITH, A. GURTNER: IN SITU SPECTROMETRY TO FOLLOW THE BEHAVIOUR OF THE CHERNOBYL RADIONUCLIDES IN THE SOIL	19
G. SHAW, X. WANG: CAESIUM & PLUTONIUM MIGRATION IN FOREST SOILS OF THE CHERNOBYL 30km ZONE	27
S. STAUNTON, P. R. DARRAH: APPLICATIONS AND LIMITATIONS OF MATHEMATICAL MODELS IN RADIOECOLOGY WITH PARTICULAR EMPHASIS IN RADIOCAESIUM IN SOIL	35
G. KIRCHNER, G. NAGELDINGER: SORPTION/DESORPTION PROCESSES OF CESIUM AND STRONTIUM IN SOIL: IS THE K_d -CONCEPT ADEQUATE?	43
N. LEWYCKYJ, C. M. VANDECASTEELE, A. CREMERS: LABORATORY STUDY OF THE CAESIUM MIGRATION IN A PODSOLIC SANDY SOIL AS A FUNCTION OF THE IONIC COMPOSITION OF THE SOIL SOLUTION	51
A. KONOPLEV, A. BULGAKOV: KINETICS OF RADIONUCLIDE LEACHING FROM FUEL PARTICLES IN THE SOIL AROUND THE CHERNOBYL NUCLEAR POWER PLANT	61
A. B. HIRD, D. L. RIMMER, F. R. LIVENS: FACTORS AFFECTING CAESIUM FIXATION IN UPLAND ORGANIC SOILS	69
F. STREBL, M. GERZABEK, V. KARG: TIME DEPENDENT VERTICAL DISTRIBUTION OF ^{137}Cs IN AN ACID FOREST SOIL	77
E. VALCKE, C. M. VANDECASTEELE, M. VIDAL, A. CREMERS: THE USE OF MINERAL AND ORGANIC ADSORBENTS AS COUNTERMEASURES IN CONTAMINATED SOILS: A SOIL CHEMICAL APPROACH	85
Y. THIRY, C. M. VANDECASTEELE, B. DELVAUX: ABILITY OF SPECIMEN VERMICULITIC MINERALS TO FIX RADIOCAESIUM: EFFECT OF THE CHEMICAL ENVIRONMENT	93
K. ROSEN, Å. ERIKSSON, E. HAAK: TRANSFER OF RADIOCAESIUM IN SENSITIVE AGRICULTURAL ENVIRONMENTS 1986 - 1994 AFTER THE CHERNOBYL FALLOUT IN SWEDEN	101
M. GERZABEK: SOIL-TO-PLANT TRANSFER OF CS AND SR IN AUSTRIA AFTER THE CHERNOBYL ACCIDENT	111
H. LÖNSJÖ, E. HAAK: SOIL FACTORS INFLUENCING THE LONG-TERM TRANSFER OF ^{90}Sr AND ^{137}Cs TO ARABLE CROPS	119
E. HAAK, H. LÖNSJÖ: LONG-TERM TRANSFER OF ^{137}Cs AND ^{90}Sr TO GRASS ON CONTRASTING TYPES OF SWEDISH PASTURES	129

C. A. SALT, J. W. JAMES, K. E. JARVIS: SEASONAL CHANGES IN THE DISTRIBUTION OF ^{137}Cs , ^{133}Cs AND K IN BENT-GRASS (<i>AGROSTIS CAPILLARIS</i>)	137
H. VANDENHOVE, M. VAN HEES, S. DE BROUWER, C. M. VANDECASTEELE: EFFECT OF AFCF ON THE SOIL-PLANT TRANSFER OF ^{134}Cs	145
H.-J. ZEHNDER, P. KROPP, J. EIKENBERG, U. FELLER, J. JOERTLI: UPTAKE AND TRANSPORT OF RADIOACTIVE CESIUM AND STRONTIUM INTO STRAWBERRY PLANTS AND GRAPEVINES AFTER LEAF CONTAMINATION	155
F. CARINI, I. ANGUISSOLA SCOTTI, M. MONTRUCCOLI, S. SILVA: ^{134}Cs FOLIAR CONTAMINATION OF VINE: TRANSLOCATION TO GRAPES AND TRANSFER TO WINE	163
P. BOSSEW: ANALYTICAL MODELS OF THE VERTICAL DISTRIBUTION OF RADIONUCLIDES IN SOIL	171
P. BOSSEW, H. LETTNER, A. K. HUBMER: SPATIAL VARIABILITY OF FALL-OUT ^{137}Cs	179
K. KIENZL, E. HENRICH, P. BOSSEW, T. FALKNER: CONTAMINATION OF AUSTRIAN SOIL BY CAESIUM-137	187
H. LETTNER, P. BOSSEW, A. HUBMER, M. GASTBERGER: VARIABILITY OF THE DEPTH-PROFILES OF ^{137}Cs IN SOILS OF THE PROVINCE OF SALZBURG	191
K. MÜCK, M. H. GERZABEK: LONG-TERM REDUCTION OF ROOT UPTAKE OF CS-ISOTOPES AFTER NUCLEAR FALLOUT	199
Y. MURAMATSU, S. YOSHIDA: BEHAVIOR OF IODINE-129 IN THE SOIL-PLANT SYSTEM	207
P. SCHULLER, A. ELLIES: INFLUENCE OF SOIL PROPERTIES AND CLIMATIC CONDITIONS ON ^{137}Cs VERTICAL DISTRIBUTION IN SOME CHILEAN SOILS	215
F. STREBL, E. HENRICH, K. KIENZL, M. GERZABEK: ASSESSMENT OF RADIOCAESIUM BEHAVIOUR IN AN AUSTRIAN FOREST ECOSYSTEM	223
M. TSCHURLOVITS: RADIOECOLOGY: DEVELOPMENT AND OBJECTIVES	229
A. WENISCH, G. MRAZ, E. HIESEL, P. BOSSEW: RADIOCESIUM IN AN AGRICULTURAL ECOSYSTEM RESULTS OF A FIELD STUDY IN THE WALDVIERTEL, AUSTRIA	237
G. HEINRICH, K. REMELE: ^{137}Cs ; ^{90}Sr , K^+ , AND Ca^{++} IN LICHENS; MOSSES; AND VASCULAR PLANTS OF A MOUNTAIN AREA IN STYRIA, AUSTRIA	243
S. YOSHIDA, Y. MURAMATSU, T. BAN-NAI: ACCUMULATION OF RADIOCESIUM AND TRACE ELEMENTS IN MUSHROOMS COLLECTED FROM JAPANESE FORESTS	251
K. J. JOHANSON, I. NIKOLOVA: THE ROLE OF FUNGI IN THE TRANSFER OF ^{137}Cs IN THE FOREST ECOSYSTEM	259
E. KLEMT, J. DRISSNER, V. FLÜGEL, S. KAMINSKI, G. LINDNER, M. WALSER, G. ZIBOLD: BIOAVAILABILITY OF CESIUM RADIONUCLIDES IN PREALPINE FORESTS AND LAKES	267
B. H. EL-FAWARIS, K. J. JOHANSON: MONITORING OF CHERNOBYL FALLOUT ^{137}Cs IN SEMINATURAL CONIFEROUS FOREST OF CENTRAL SWEDEN	275
F. TATARUCH, F. SCHÖNHOFER, E. KLANSEK: RADIOCESIUM LEVELS IN ROE DEER AND WILD BOAR IN TWO LARGE FOREST AREAS IN AUSTRIA	285

OVERVIEW OF EC COORDINATED RADIOECOLOGICAL RESEARCH AFTER THE CHERNOBYL ACCIDENT

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Abstract.

The uncontrolled release of radionuclides coming up after the Chernobyl accident has led to a large number of scientific and political activities to assess the contamination of the environment and the consequences for the population. A large scale of measures were deployed attempting to mitigate the consequences and initiatives were launched to follow the fate of the radionuclides in and around the Chernobyl area. Some of these efforts are described in this paper, summarizing which way radioecologists had chosen to evaluate the problem, to compare the scientific culture existing in East and West, to sharpen their views on the fundamentals of radioecology and to test their knowledge in the real field.

1. Introduction

In April 1986 Chernobyl 4 exploded and large areas around the nuclear power station were contaminated. The cloud of contamination spread further on largely over Europe and elsewhere. A few million people were potentially affected by the deposition of radioactivity from this accident. The environment was influenced by the short-lived radionuclides, equally though longer lived radionuclides were deposited such as Caesium-137, Strontium-90 and some even longer lived ones such as Plutonium, Ruthenium and others.

Agricultural land, water catchments with their tributaries and rivers, as expected, became contaminated; the deposition of radionuclides had no preferences though, and forests as well as extensively used pasture and semi-natural land were touched by the deposits.

Around the reactor zone, later on called "exclusion zone", radionuclides were deposited as simple chemical ionic entities as well as in particulate form with a more complicated chemistry. The more distant from the nearby deposition area, the more the radionuclides deposited were in the normal ionic form.

Radioecology which is the scientific discipline studying problems of "interaction between the ecosphere and radioactivity and how to deal with it" [2] was confronted with a real problem and an amazing wealth of "all true" statements saw the light during "scientific" parleys!

Right after the Chernobyl quite some contention arose as regards the behaviour of radionuclides and primarily Caesium in the environment. Terms like "new" Caesium and "old" caesium were "in the air" everywhere especially when "Caesium in soil" was discussed! "Caesium is ionic, Caesium is in "condensed" or trapped in "hot particles, Caesium is fixed for ever and no longer available, Caesium is dynamically bonded and its bioavailability is more persisted than expected", is a selection of the affluence of statements about this element. The discussion is less vehement nowadays; Caesium unfortunately though has persisted. The future is now to come for strontium; as it is being eluted from "hot particles", some more "true and undiscussable" statements are expected to come!

Another selection of "uncontestable" statements are about the "state of knowledge" radioecology had reached before "Chernobyl" and now "ten years after Chernobyl". Expressions are used like "what have we learned what we did not know already" and "we have done all this in the sixties", which has not stopped the quest for more research funds though! The reality forces to accept however the simple fact that 10 years after Chernobyl a number of problems still have persevered, and that a decent environmental planning still remains difficult, notwithstanding large and costly efforts by scientists and organisations! (ref. Post-Chernobyl activities by EC and other organisations)!

2. Implementation of radioecological research after the Chernobyl accident.

Roughly 65% of the contamination was deposited over Belarus, 20 % over Russia and 10% over Ukraine. Belarus contains most of the highly contaminated areas ($>40 \text{ Ci km}^{-2}$ = $>1480 \text{ K bq m}^{-2}$). The inhomogeneous distribution can be demonstrated by the fact

activities inside the 30 km zone are sometimes as low as 1 Ci km⁻², whereas at 50 km southwest of Chernobyl and 300 km north-east soil activities of more than 40 Ci km⁻² have been measured.

The European Commission, immediately after the accident, took several initiatives to assess the burden of the event for the population and the environment. These initiatives finally resulted in an Agreement between the three Republics affected, Ukraine, Belarus and Russia and the European Commission. A group of Joint Study Projects (JSP) and Experimental Collaborative Projects (ECP) was set up.

They were related to environmental issues, dosimetry, and health problems.

In the environmental area, problems of contamination of land used for intensive agriculture, of zones of extensive agriculture, of zones of consumption of natural foodstuffs and problems of contamination of surface water and their food products were investigated through four directed Experimental Collaborative Projects (ECP). In the area of radioecology four ECP's were aiming at a basic understanding of environmental mechanisms, governing the fate of radionuclides.

In relation to this inhomogeneity, the work of the ECP's 2, 3, 5 and 9 have not been restricted to the Chernobyl exclusion zone. Investigations have been carried out in areas where there was little change in the occupation of the land by the population, and where the traditional living habits were maintained.

The contamination being dispersed over various sorts of landscapes of economic use, research was thence conducted by four teams looking at the effect of contamination on agricultural land and on pasture land (meadows) (ECP2 and 5), on forested areas (ECP5) and on drainage areas of rivers and lakes (ECP3). A global study on the impact of land use and food consumption habits was carried out by ECP9. Such included the comparison between the effect of the consumption of food products from private (small-scale) farming and those of industrial (collective) farms.

Research activities were conducted in the exclusion zone and in the large periphery where either the activity was high or where mainly the population makes intensive use of the local products. The necessities of life consist of provisions for food, water,

clothing, housing etc. They come mainly from farming and general husbandry, water reservoirs and deducted water supplies, manufacturing and so forth.

Another criterion for the selection of research location is the soil type. Soil type indeed does play a dominant role in the behaviour and fate of radionuclides, and their (bio)-availability too a large extent depends on the way and intensity the soil binds and releases the deposited radionuclides for further integration in the foodchain or for deeper migration.

The places of research are located:

- in Ukraine in the 30 km zone at Chistogalovka and Kopachi, outside the zone at Poleskoe, the Rovno region (Sarny), etc
- In Belarus at Bragin, Vietka, Mogilev, Gomel, etc
- In Russia in the Bryansk, Kaluga and Tula region, etc
- The drainage area of the Pripjat-Dnieper river-reservoir system

3. Evaluation of the results of the radioecological research projects for a long-term assessment of the persistence of environmental consequences of the accident.

Beyond basic radioecological research, there was striven simultaneously after the application of the basic findings, and equally after the obtention of insights and a grip on the influence of features of general ecological nature including agricultural practices and living habits of the concerned population.

Important phenomena came to the attention through the radioecological research after the Chernobyl accident.

Research on plain geographical distribution and redistribution as well as on the physico-chemistry and bioavailability of the deposits gave the main following results:

- The deposition of the radionuclides after the accident was very heterogeneous, not only from a geographical point of view but also from a physico-chemical point of view. In the exclusion zone around the exploded reactor a considerable amount of the radioactive material was deposited as "hot particle", it means complex amorphous particles containing variable amounts of a spectrum of elements. The

further away from the location of the accident the more though the form of the deposits became simpler and more ionic.

- "Hot particles" have such features that they have a low bio-availability and are less swiftly assimilated by the vegetation or animals. In the course of time though "hot particles" are beginning to disintegrate and the availability of the composing elements such as Sr increases, and affects its radio-ecological half-life.

- Soil types and their concurring chemistry is paramount for the long-term behaviour (bio-availability and migration) of radionuclides through its control of their absorption/desorption features. The availability of Cs in arable soils, with a high clay content is rather low when at least perturbations are under control or envisageable. The problem of availability of Cs in soils with high organic content is however still considerable such as in meadow pastures with low quality soil. The availability remains considerable as these soils hardly can benefit of the strong binding properties of clay particles, if present. The high proportion of organic matter confuses the clay capacities, and the radioactive materials shows a persistent bioavailability. Moreover, there always remains the problem of some "reversibility" of the bonded radioactive nuclear material, especially Caesium; it is clear from the research done that a great deal of attention still will have to be paid to kinetic aspects of radionuclides fixation in soils!

The interaction between competitive ions (fertilisation!) for the reduction of the dose depends very much on the soil type and soil condition. Potassium (K) can for instance be successfully applied where no K saturation condition prevails, i.e. on poor or poorly fertilised land. Otherwise the effect would be marginal! For the assessment of the effect of other soil ameliorants, processes and systems such as application of some zeolites or mulching of soils, that work well under laboratory or pilot conditions are still in a stage of circumstantial knowledge when applied in the field. From a scientific-technical point of view, a better "STRUCTURE" of the available and new knowledge has been attained. Loose statements about the use of fertilizers or manure, etc, can be tightened now, and all sorts of amendments to soils and animals such as mulching or of a chemical nature (Prussian Blue) can be better assessed and quantified. It is still true though that more data are

available than there have been properly evaluated. Too many "random" measures have been tried and performed, which have not been profoundly (statistically) tested against the insights as regards mechanisms of behaviour of radionuclides in the environment, recently acquired! Efficiency could be enhanced if a consistent correlation exercise would be carried out further on, based on the affluence of data obtained. This is definite not only for soil and vegetation but true for all compartments of the environment.

- Seminatural ecosystems, especially coniferous forests intercepted considerable amounts of radioactive material and litter fall take it to the soil surface. The cycling of matter in semi-natural environments is a well-known phenomenon. The matter flows as well through migration as through native soil processes of soil horizon production below the upper surface. From there on they become available for further migration to deeper layers if the horizon profile is fit for it and could from there on seep through to the underground water table; they become also available for tree root uptake and further cycling in the tree; it takes up to four years before the RN's deposited on forest canopies fully enters the biogeochemical cycling of a dynamic ecosystem. "Root" uptake thence prevails. The radionuclides are mainly delayed in Oh (Organic humic layer zone) horizon. The depth depends on the soil type, and microbial life can perturb profiles. Mushrooms also are part of the cycle as their mycelium (fungal roots) are "grazing" their nutrients at different soil depths and thence accumulate the radionuclides in a species dependent way.

- In zones with a complicated hydrological system of vague river tributaries and canalization such as in north-west Ukraine, flooding of river foreland may result in increases in radionuclide concentrations especially Caesium; this leads to important redistribution on the soils of the original distribution of the deposition.

- Two main sources of long-term contamination of water prevail:

- annual flooding of the Pripjat flood plain
- catchment transport, meaning that water reservoirs can become a long-lasting sink for contaminants through releases of radioactivity from peat bogs; the water pathway is thus a very important pathway for eventual

transfer of contamination in the long-term to uncontaminated areas. The sediments of fresh water systems (rivers and lakes) can act as sinks but are subject to the surrounding chemical conditions for the further fate of the radionuclides; clays are important compounds of these sediments and the absorption/desorption dynamics depends on competitive ions such as potassium and ammonia. Models including straightforward "distribution factors" (K_d) ought to be considered with caution.

In the course of the years a number of somewhat fortuitous measures were taken as regards hydrological measures to reduce or to avert the risk that could be caused by the flooding of the Chernobyl area. The years have taught to treat these floodings with caution in order to avoid undesirable side-effects and unexpected contamination redistributions. Fluvial planning and flood averting dike construction has thence been considered carefully and eventually became based on computer-simulated hydrological management.

- The economic structure of the ex-Soviet Union is special in the sense that the main provision of food stuffs is still provided by so-called Collective Farms, controlled by governmental regulations. Therefore arable land in the "exclusion zone" as well as outside on Collective Farm lands was studied in the three Republics, and the mechanisms exerting an influence on the dose to man assessed. Consumption though of seasonally collected or privately grown food products is becoming an important common practice on large rural areas. The most salient result of these studies is the revelation food products from seminatural zones are playing a major role in determining intake to several particular groups and possibly also to the entire population. The zones of extensive agriculture production are private farms, semi-natural ecosystems and forests. This means herding cattle grazing on semi-permanent pastures and forested land, picking of mushrooms in the season, or producing food on the own garden. Rural populations including town habitants rely on subsistence farming and the use of nearby forest products, mainly mushrooms and are exposed to higher doses than in "collective farm" systems. Assessments of doses have to be comprehensive and

include all pathways, since general knowledge of important sources may not be sufficient to describe radionuclide intake.

- Wild animals are a potentially important source of radioactivity; the transfer is high but very variable, so predictions of mean contamination levels are difficult. Wild animal contamination is considerable but being though connected mainly to the "exclusion zone" also a revival of the wildlife is noticed.

4. General conclusions

4.1. Conclusions regarding progress in methodology, some examples:

Questions such as "how deep should one muster in order to take a representative soil sample" are false ones if one does not consider horizon formation in natural soils!

The lessons learned especially demonstrate how essential it is to be remain constantly aware that random sampling is to be carefully interpreted in the light of the realization of the complexity of the habitat of animal and man; it means test samples are to be representative for the total habitat being a society of life living in and feeding from simple private farming and collection of foodstuffs in small villages, over industrially organised and exploited farming up to collecting provisions in urban areas.

4.2 Conclusions regarding the progress of radioecological science:

The complexity of the post-Chernobyl situation is immense and the need for an integrated scientific approach has been clearly shown. Each of the different ecosystems all representing different production systems are tightly connected, and is pointless to do just some isolated measurements or monitoring of contamination levels if these data are not put in a frame where urban zones, use of agricultural land and forest and finally water supply are looked upon as complex dynamically interacting environment. It is realised now that this complexity of contamination necessary leads to investigations of total inventory changes which are best defined as fluxes of matter and energy between different ecosystems.

Notwithstanding the progress made we still know not enough about *long-term effects of the contamination nor of the mitigative measures*, or in other words what will be the ultimate ecological quality and socio-economic usefulness of the treated zones?

To which extent and for how long is the population of especially rural areas going to be effected by the fact that their land has a reduced nutritional quality and an economic value, and which will be the health impact on people living constantly under the pressure of a low exposure?

4.3. Conclusions regarding the progress in environmental management:

The necessity to do the research in a thrust of attempting to comprehend the complexity of the problem in an integrated way which lead at least to a "wake up" and to the awareness of the need for integrated environmental management. There not only plain ecological features of chemical and biological nature are to be investigated, but where also features of human habitats and human industrial and agricultural activities have to be taken into account. The consideration of the latter features and activities have now become an integral part of the knowledge structuring of radioecology!

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MOBILITY OF CHERNOBYL-DERIVED RADIOCESIUM IN THE SOIL

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ABSTRACT

The mobility of Chernobyl-derived radiocesium was investigated in a grassland soil and a near-by forest soil in Bavaria/Germany. Both sites were sampled at several times after the Chernobyl accident. The initial mobility of radiocesium during wet deposition was very high due to high rain-fall intensities. After a more complete sorption by soil components, which occurred within a few days, the mobility was reduced by several orders of magnitude. During the first years after the Chernobyl accident, the mobility was not constant, but decreased even further. However, at the end of the observation period, Chernobyl-derived radiocesium was still more mobile than radiocesium from global fallout. The time dependence of the mobility after deposition is discussed with respect to the fixation processes in clay minerals, which may also explain the increase of the mobility with soil depth found in the mineral soil at both sites investigated.

1. INTRODUCTION

The mobility of radiocesium in the soil is one of the most important factors controlling its long-term transfer to man via terrestrial pathways: plant uptake, resuspension into the air and contamination of groundwater. Therefore, the behaviour of ^{137}Cs in the soil has been investigated since the beginning of the global fallout from nuclear weapons tests in the fifties. Radiocesium deposited by the Chernobyl fallout in 1986, however, in some aspects showed a different behaviour as compared to presently observed ^{137}Cs from global fallout (see e.g. Livens and Baxter, 1988; Bunzl et al., 1992 and 1994). Obviously, in the fifties and sixties not so much attention has been paid to the initial phase of migration of ^{137}Cs into the soil.

In this paper some results about the mobility of Chernobyl-derived radiocesium will be presented, as obtained for the soil of a grassland und a near-by spruce stand in Bavaria. Both sites were sampled at various times after the Chernobyl accident in order to elaborate the changes with time of the mobility of radiocesium in the soil. For this reason, the residence half-times of radiocesium in the various soil layers at each site were evaluated from its vertical distribution as a function of time by using a multilayer compartment model.

2. MATERIAL AND METHODS

2.1. Sites, Soils and Sampling

Grassland and forest investigated are situated about 40 km northwest of Munich, 545 m above sea level. The mean annual precipitation is 800 mm, the mean annual temperature 7.3 °C. The forest consisted of Norway spruces (*Picea abies*), ca. 80 years old. The grassland is located ca. 100 m west of the forest border and was undisturbed by tillage since the fifties. All sampling locations were selected on a flat area to exclude precipitation run-off.

The soil of the grassland was classified as a Parabrown earth pseudogley (US soil taxonomy: slightly wet Alfisol), the forest soil as a podzolic Parabrown earth (Hapludult). Several properties of the various horizons of both soils are given in Table 1. For further details see Bunzl et al. (1992, 1994). All soil samples were taken with a frame (50 cm x 50 cm) and air dried. The samples from the mineral soils were sieved to 2 mm and carefully mixed.

Table 1: Several physico-chemical properties of the soils at the sampling sites

	Forest soil under spruce					Grassland soil		
	Horizon					Horizon		
	LOf1	Of2	Oh	Aeh	Alh	Ah	Al	Bt
Depth (cm)	7-4.5	4.5-2	2-0	0-5	5-10	0-10	10-20	20-30
pH (CaCl ₂)	3.2	3.2	2.9	3.2	3.6	4.4	4.3	4.2
Clay (%)	-	-	-	19	21	32	30	37
Silt (%)	-	-	-	46	43	52	55	55
Organic matter (%)	48	48	37	2.8	1.3	7.5	3.0	2.4
CEC (mmol _c Kg ⁻¹)	207	196	202	87	55	84	71	66

2.2. Determination of Radiocesium

Cesium-134 and ^{137}Cs were determined by direct gamma spectrometry, using a high-purity germanium detector and a multichannel analyser. With ^{134}Cs , losses by sum-coincidences during counting were taken into account. The experimental errors were $\leq 10\%$.

2.3. Evaluation of the Residence Half-times

The residence half-time τ of radiocesium in the various soil layers was evaluated with a multilayer compartment model (Bunzl et al., 1992). The equation for the transfer of activity A_i (Bq m^{-2}) of ^{134}Cs or ^{137}Cs in the compartment i in a small time intervall Δt (days) is:

$$\frac{\Delta A_i}{\Delta t} = K_{i-1} A_{i-1} - K_i A_i \quad (1)$$

where K_i (day^{-1}) is the fractional rate of transfer from compartment $i-1$ to compartment i . The residence half-time τ_i of A_i in compartment i is $(1/K_i) \ln 2$. Since all A_i are corrected with respect to 1.5.1986, radioactive decay need not to be considered in Eqn (1). The rate of deposition ($\text{Bq m}^{-2} \text{ day}^{-1}$) to the first compartment has to be known as a function of time in order to solve Eqn (1). For further details see Bunzl et al. (1992, 1994).

3. RESULTS AND DISCUSSION

3.1. Initial infiltration of Chernobyl-derived radiocesium into the soil

In Bavaria, the main activity deposition of the Chernobyl fallout on the soil surface occurred on April 30, 1986, during a heavy thundershower (Hötzl et al., 1987). At May 7, the first depth profile was sampled in the forest. Due to the lack of precipitation between deposition and sampling, the observed vertical distribution of radiocesium was essentially the result of the initial migration into the soil during deposition. The same holds for the first depth profile of radiocesium in the grassland (Schimmack et al., 1989).

At both sites, Chernobyl-derived radiocesium had partially penetrated the soil to depths between 5 and 10 cm below the soil surface. In the grassland, one percent of the total activity was found below 10 cm. From these depth profiles the initial residence half-times of ^{134}Cs could be estimated to 3 h/cm in the forest and 5 h/cm in the grassland, assuming an activity transport in the soil during the deposition only (Schimmack et al., 1989). In contrast to

these very small values, the long-term residence half-times of radiocesium from global fall-out were about 2-3 y/cm in the upper soil layers at both sites (Bunzl et al., 1992, 1994).

The high mobility of radiocesium infiltrating with high rainfall intensities was also demonstrated by column experiments in the laboratory (Schimmack et al., 1994). Although ^{137}Cs exhibits a low mobility in long-term migration (global fallout and column experiments with low rainfall intensities of 2 mm/h), the radionuclide was not completely retained in the first layer of the forest soil investigated when the activity was supplied with high rainfall intensities (30 mm/hour). About 30 % of the activity penetrated into the mineral soil down to a depth of 14 cm. These high migration rates are probably related to the phenomenon of macropore water flow (see e.g. Beven and Germann, 1982).

The high mobility of the infiltrating Chernobyl-derived radiocesium was drastically reduced, as soon as the nuclide was sorbed to soil components. This was shown by the determination of its depth profiles and residence half-times at several times after deposition.

3.2. Mobility of Chernobyl-derived radiocesium in the forest soil

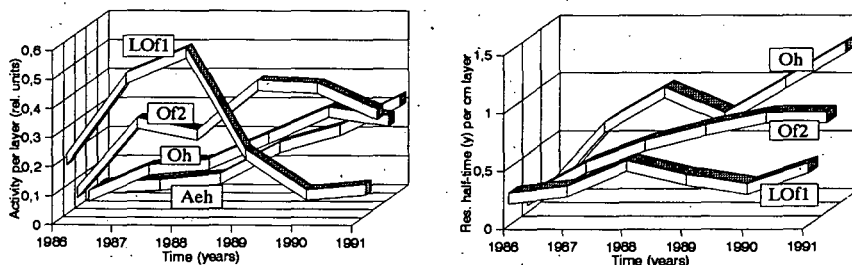


Figure 1: Time dependence of the activity of Chernobyl-derived radiocesium (left) and its residence half-time (right) in the soil for various layers of a podzolic parabrown earth under spruce. The activity is given as activity per sqm per layer with respect to the total activity deposited at the site per sqm. The residence half-time is given with respect to the thickness of each soil layer.

Due to interception of Chernobyl-derived radiocesium in the canopy of the spruce stand, its activity per sqm in the soil, determined in the first week after the accident, was only about 30 % of the activity deposited in the near-by grassland (Bunzl et al., 1989). As a consequence of weathering (rain, wind, litter fall), the activity in the canopy was removed to the forest floor within two years after deposition. Therefore, the activity in the first horizon LOf1 increased to more than 50 % within one year. Subsequently, it decreased rapidly due to leaching, decomposition of the spruce needles and bioturbation to values < 10 % in 1990 and later on. In the deeper organic horizons, the activity increased not so fast as in LOf1, the maximum was attained later and not so high as compared to LOf1.

In the first time after deposition, the residence half-times (years) per cm layer of Chernobyl-derived radiocesium in the three organic horizons were very small: 0.1 - 0.2 y/cm (Fig. 1 right). Subsequently they increased continually till the end of the observation period of five years to 0.5 y/cm in the LOf1, 0.8 y/cm in the Of2, and 1.3 y/cm in the Oh horizon. For the Aeh horizon of the mineral soil, residence half-times of 2 y/cm in the layer 0-2 cm and 0.4 y/cm in the layer 2-5 cm were determined by Bunzl et al. (1992) in the same spruce stand. These values were considerably smaller as compared to those of radiocesium from global fallout (Bunzl et al., 1992). A possible explanation for the time dependence of the mobility of radiocesium is given in the next section.

3.3 Mobility of Chernobyl-derived radiocesium in the grassland soil

In the case of grasslands in Bavaria, the wet deposition of Chernobyl-derived activity mainly occurred in a single rain event at April 30, 1986. As a consequence, the activity in the top layer (0-2 cm) did not increase as observed in the forest soil, but decreased continually from about 80 % in 1986 to about 20 % in 1994 (Fig. 2, left). In all other layers the activity increased within the first years after the deposition. With time, when radiocesium continued to migrate to the deeper soil layers, the activities will decrease again. In the second layer (2-4 cm), the maximum may have been passed around 1992.

The residence half-times of radiocesium in all layers of the grassland soil increased significantly with time (Fig. 2, right). Even after 8 years of observation a constant value of the residence half-time was not yet attained in the soil layers, with the possible exception of the

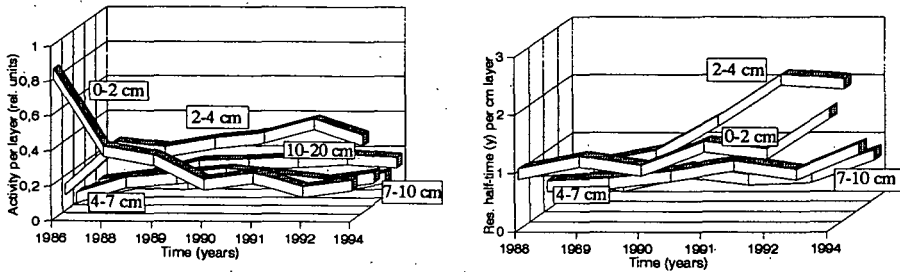


Figure 2: Time dependence of the activity of Chernobyl-derived radiocesium (left) and its residence half-time (right) in the soil for various layers of a parabrown earth pseudogley of a grassland. The activity is given as activity per sqm per layer with respect to the total activity deposited at the site per sqm. The residence half-time is given with respect to the thickness of each soil layer.

second one (2-4 cm). In addition, the residence half-times in the top soil layers (0-4 cm) were always higher as observed in the deeper layers (4-10 cm). In 1994, the residence half-time in the top soil was about 2 y/cm, as found in the layer 0-2 cm of the mineral soil in the spruce stand (see above), in the deeper soil < 1 y/cm. Again, these values were still smaller than those of radiocesium from global fallout by a factor of 2-5 (Bunzl et al, 1994).

The decrease of the mobility of Chernobyl-derived radiocesium with time, found in all soil layers at both sampling sites, may be explained by a gradual 'locking up' of cesium in the soil over the years, e.g. by diffusion and subsequent fixation in clay minerals (Livens and Baxter, 1988). Evidence for the time dependence of cesium fixation was already provided in the sixties by Squire and Middleton (1966). From long-term lysimeter experiments they concluded, that the fixation "appeared to be largely complete 3 years after contamination" of the soil. For soils of the Chernobyl area, Sanzharova et al. (1994) found, that the fraction of radiocesium, extractable by 1 N ammoniumacetat ("mobile" or exchangeable fraction) from the soil, decreased by a factor of 2-3 from 1986 to 1991.

The mobility of Chernobyl-derived radiocesium increased significantly with depth at a given sampling date (Fig. 2, right), which was also observed for radiocesium and other radionuclides like plutonium from global fallout in the same grassland soil (Bunzl et al., 1994) as well as for the mineral soil of the spruce stand (Bunzl et al., 1992). This may be explained by the time dependence of the cesium fixation by clay minerals: Cesium found in deeper soil layers arrived there comparatively late and thus did not have as much time for fixation as Cs ions in the upper layers.

In contrast to that, in the organic layer of the spruce stand, the mobility decreased with depth (Fig 1, right). This may be explained by the uptake of cesium by soil fungi (Bakken and Olsen, 1990), which will be more abundant in the deeper horizons of the organic layer.

4. CONCLUSIONS

From the results presented about the mobility of Chernobyl-derived radiocesium in the soil three conclusions of general importance may be drawn.

- During the deposition phase, the mobility of radiocesium can be extremely high, depending on meteorological conditions, state of the soil, water flow, chemical form and sorption kinetics of the radionuclide.
- Shortly after deposition, the mobility in a given soil layer is reduced by several orders of magnitude. Even then, however, the mobility does not remain constant, but decreases furthermore. This is due to a progressive fixation of cesium by clay minerals and proceeds for many years.
- In the mineral horizons, the mobility may increase with depth. This is explained by the time dependence of the fixation process. In forest soils, the effect of soil fungi on the behaviour of radiocesium has to be considered in addition.

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IN SITU SPECTROMETRY TO FOLLOW THE BEHAVIOUR OF THE CHERNOBYL RADIONUCLIDES IN THE SOIL

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Abstract

This paper presents a global evaluation of the knowledge relative to the behaviour of the Chernobyl fallout in swiss soils, especially based on in situ measurements. After the contamination phase most of the activity initially limited to the soil surface gradually migrates into deeper layers. This progressive transfer depends on the properties of the involved radionuclides and on local characteristics, which can considerably affect the depth distribution of the activity in the terrestrial environment. The developed in situ procedure allows a sensitive control throughout the years of the radioactive inventory of the gamma emitters in the soil and of their individual contribution to the ambient dose rate equivalent 1 meter above it. The method also provides an effective characterization of a site, especially for the most important still remaining detectable long-lived caesium isotope, by expressing its in situ apparent decrease as an evolution of the relaxation mass-per-area ρ/α ($\text{g}\cdot\text{cm}^{-2}$), which describes its transfer in deeper layers according to an exponential depth distribution. In practice this characterization often involves in situ spectrometry in conjunction with soil sampling. For most of the undisturbed swiss sites, it results 10 years after the Chernobyl deposit an apparent half-live of round 10 years instead of 30 years due to the vertical transfer of caesium. This additional diminution is lower in particular sites by which caesium is very efficiently retained in the upper layer by clay or organic materials (forest soils) and higher in those by which the natural transfer is modified by erosional processes or human activities such as plowing.

1 Introduction

For world-wide contamination caused by major weapon tests in the late 50s and early 60s and for the large scale contamination after Chernobyl in 1986, the predominant contamination path is via the atmosphere. According to the differences in meteorological conditions, the radioactivity deposited by rain and dry fallout onto the ground is very inhomogeneous, Völkle and Murith (1989). The influence of these contaminations on the activity of the air at Fribourg is given in Figure 1. For both contaminations the main long-lived artificial gamma emitter still remaining detectable in the environment these days is caesium-137. A significant difference between them has been observed in Switzerland for the isotopic ratio $^{137}\text{Cs}/^{90}\text{Sr}$: ~ 2 for the weapon tests fallout resp. ~ 100 for the Chernobyl fallout. The beta emitter strontium-90 mainly from the weapon tests shows a mean apparent half live of round 10 years instead of 28 years due to its transfer in deeper layers. According to its properties, strontium-90 reveals a higher transfer from the soil into grass, agricultural products and milk than ^{137}Cs .

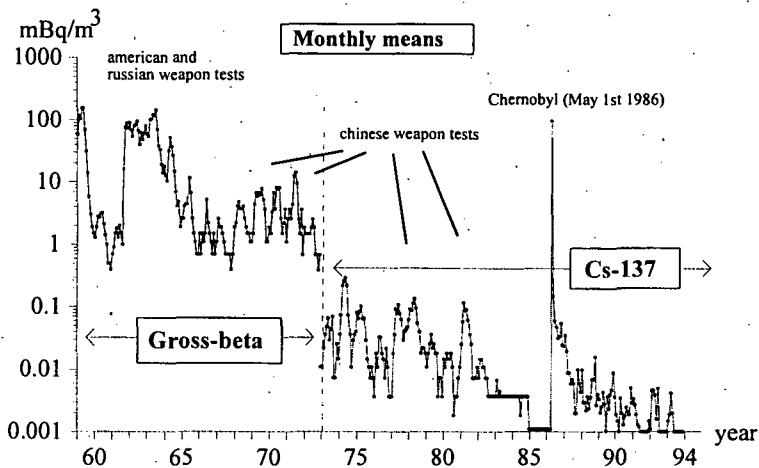


Figure 1: Air-activity at Fribourg

2 In situ procedure

In order to obtain accurate measurements of radionuclides in the soil, the Ge(HP)-detector is generally placed 1m above a relative even and open area. The power in the in situ technique described in Beck et al. (1972) lies in the fact that the detector measures gamma radiation from sources over an area of several hundred square meters, with the advantage to average out small-scale inhomogeneities of the source geometry, that invariably are present. In most undisturbed land, one generally can assume an uniform distribution with depth for natural emitters and a negative exponential distribution with depth for fallout radionuclides. If there is almost no problem to determine the specific activity for natural emitters (Bq/kg), the Chernobyl wet deposit in Switzerland and Germany, Winkelmann (1986) has shown that an initial assumption of p/α on the order of 1.0 g.cm^{-2} ($\pm 30\%$) would be a realistic compromise. For aged fallout well defined site criteria can be difficult to meet, especially when erosional processes or human activity such as plowing have disturbed the site. Therefore to describe the behaviour of radionuclides in the soil we use following in situ procedure: assuming an uniform distribution, we calculate the permanent activities of natural gamma emitters, which provide a quality control of the measurements and of the corresponding artificial ones, which show a higher decrease than expected from their half live. This additional decrease is attributed to their transfer in deeper layers and then expressed as an evolution of the relaxation mass-per-area p/α , that characterizes the behaviour of the individual radionuclides according to an exponential depth distribution. The not detector dependent values of the flux and ambient dose rate equivalent at various energy from our calculation are given in Table 1 resp. Table 2 per unit γ emission in the soil. A flux resp. ambient dose rate equivalent value for a γ energy of any arbitrary nuclide with a given distribution can be computed by interpolation from Table 1 resp. Table 2 at this particular energy and then by multiplying by the yield for that γ ray. To obtain the total contribution of a nuclide to the ambient dose rate equivalent, the sum of its individual γ rays must be considered.

Table 1: \emptyset - unscattered flux ($\gamma \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$) at 1m above ground per unit source strength *

Source energy (MeV)	$\rho/\alpha - \text{g} \cdot \text{cm}^{-2}$					
	∞ (uniform)	0 (plane)	0.3 dry deposition	1.0 wet deposition	10 aged fallout	30 aged fallout
0.10	2.63E-3	1.71E-4	1.19E-4	7.88E-5	2.14E-5	8.50E-6
0.15	3.20E-3	1.77E-4	1.29E-4	8.77E-5	2.61E-5	1.25E-5
0.20	3.62E-3	1.82E-4	1.35E-4	9.30E-5	2.90E-5	1.29E-5
0.30	4.26E-3	1.89E-4	1.42E-4	9.98E-5	3.23E-5	1.32E-5
0.40	4.81E-3	1.95E-4	1.47E-4	1.04E-4	3.45E-5	1.31E-5
0.50	5.31E-3	1.99E-4	1.50E-4	1.08E-4	3.62E-5	1.32E-5
0.60	5.76E-3	2.03E-4	1.54E-4	1.11E-4	3.80E-5	1.37E-5
0.70	6.15E-3	2.06E-4	1.57E-4	1.14E-4	4.01E-5	1.49E-5
0.80	6.59E-3	2.09E-4	1.59E-4	1.16E-4	4.17E-5	1.55E-5
0.90	6.96E-3	2.12E-4	1.61E-4	1.18E-4	4.25E-5	1.60E-5
1.00	7.37E-3	2.15E-4	1.64E-4	1.21E-4	4.40E-5	1.74E-5
1.50	9.11E-3	2.25E-4	1.74E-4	1.31E-4	4.92E-5	2.26E-5
2.00	1.06E-2	2.33E-4	1.82E-4	1.38E-4	5.37E-5	2.58E-5

Table 2: $H^*(10)$ - Ambient dose rate equivalent ($\text{nSv} \cdot \text{h}^{-1}$) at 1m per unit source strength*

Source energy (MeV)	$\rho/\alpha - \text{g} \cdot \text{cm}^{-2}$					
	∞ (uniform)	0 (plane)	0.3 dry deposition	1.0 wet deposition	10 aged fallout	30 aged fallout
0.10	2.70E-2	5.36E-4	4.04E-4	3.16E-4	1.27E-4	6.30E-5
0.15	5.00E-2	7.74E-4	6.08E-4	4.78E-4	2.01E-4	1.06E-4
0.20	6.80E-2	1.03E-3	8.18E-4	6.43E-4	2.74E-4	1.45E-4
0.30	1.00E-1	1.55E-3	1.23E-3	9.66E-4	4.11E-4	2.18E-4
0.40	1.30E-1	2.04E-3	1.63E-3	1.27E-3	5.41E-4	2.87E-4
0.50	1.59E-1	2.50E-3	1.99E-3	1.55E-3	6.62E-4	3.52E-4
0.60	1.86E-1	2.92E-3	2.33E-3	1.82E-3	7.75E-4	4.12E-4
0.70	2.15E-1	3.33E-3	2.66E-3	2.08E-3	8.88E-4	4.72E-4
0.80	2.44E-1	3.71E-3	2.98E-3	2.33E-3	1.00E-3	5.33E-4
0.90	2.73E-1	4.10E-3	3.29E-3	2.58E-3	1.11E-3	5.92E-4
1.00	3.00E-1	4.45E-3	3.57E-3	2.80E-3	1.21E-3	6.45E-4
1.50	4.46E-1	6.20E-3	4.97E-3	3.91E-3	1.72E-3	9.20E-4
2.00	5.88E-1	7.72E-3	6.22E-3	4.92E-3	2.20E-3	1.20E-3

* For $\rho/\alpha = \infty$ (uniform profile, natural emitters), the source strength is $1 \gamma \cdot \text{kg}^{-1} \cdot \text{s}^{-1}$ for soil at all depth. For $0 \leq \rho/\alpha \leq 30$ (exponential profile, fallout emitters), the source strength is $1 \gamma \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ emitted from an infinite column of soil, where the activity exponentially decrease with depth at relaxation length of $1/\alpha$ at soil density ρ .

As it appears from Table 1 and Table 2, the unscattered flux and the ambient dose rate equivalent for a given inventory vary much more significantly with the source depth distribution than the ratio of these two quantities. In consequence different assumptions for the depth distribution would considerably affect the inventory, however the corresponding ambient dose rate equivalent still would be reasonably close to the true value.

3 Chernobyl fallout in Switzerland

The in situ results of the nuclide specific soil contamination measurements for one of the most affected Swiss sites by the Chernobyl fallout are shown in Figure 2. In the first few days of May, the short-lived isotopes of tellurium and iodine formed the main contribution ($\sim 80\%$) to the contamination. Their rapid decrease resulted in an increase of the contribution of the caesium isotopes from about 6% on May 1st to 80% on August 1st, showing up to now a progressive predominance of ^{137}Cs .

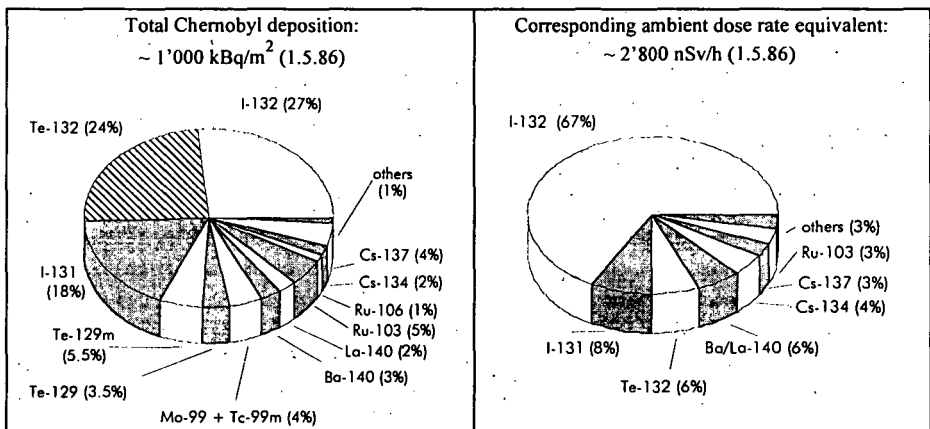


Figure 2: Contributions to the inventory and ambient dose rate equivalent at Caslano (TI).

As caesium-137 remains the main long term contribution, it permits to estimate the degree of contamination on May 1st 1986, according to the proportions per cent given in Figure 2 from the about 400 in situ measurements performed till now in Switzerland. The regionally large differences in the caesium deposit calculated for May 1st 1986 and shown in Figure 3 confirm the heterogeneity of the contamination observed in Switzerland by ambient dose rate measurements after the Chernobyl fallout. In comparison the deposit of caesium from atmospheric nuclear weapons testing determined from earlier measurements or by taking into account the Chernobyl ratio $^{137}\text{Cs}/^{134}\text{Cs}$ (1.8 on May 1st 1986) for the post Chernobyl measurements show less heterogeneity. This is mainly due to the fact that for the instantaneous high atmospheric Chernobyl contamination, very inhomogeneous rain conditions induced regionally larger differences in the degree of soil contamination than for the weapon tests one by which the deposits follow chronically over years.

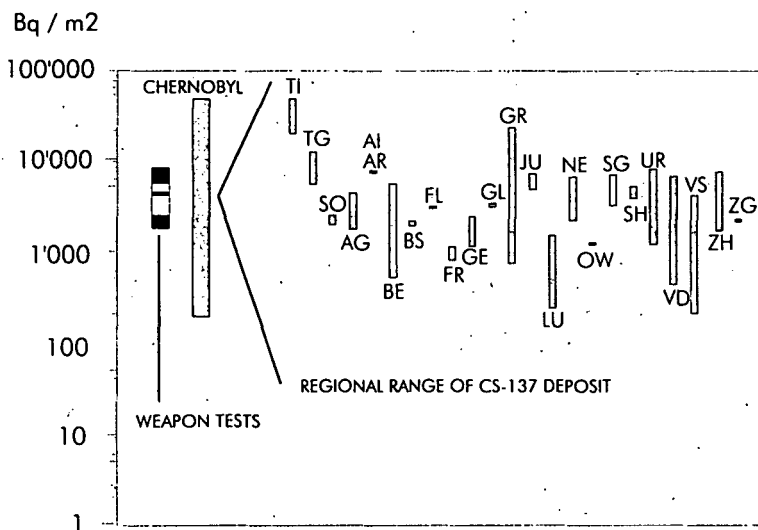


Figure 3: Regional caesium-137 in situ inventory (Bq/m^2) in Switzerland on May 1st 1986

4. Rates of the vertical transfer of radiocaesium

To obtain an indication of the rate of the vertical transfer of caesium in the soil, we compare its in situ activity evolution assuming throughout the years uniform depth distribution to the expected activity evolution caused by caesium half live. We attribute then the observed additional activity decrease to the transfer of caesium in deeper layers by estimating which increase of the relaxation mass-per-area p/α (g.cm^{-2}) would induce the same activity diminution. As shown in the table annexed to Fig. 4, this increase can also be expressed as a corresponding increase of the conversion factor $\text{Bq.m}^{-2} / \text{Bq.kg}^{-1}$, which allows to determine the inventory in Bq.m^{-2} from the measured Bq.kg^{-1} . For most of the undisturbed sites, in situ measurements show a mean decrease of the caesium activity during the 10 last years down to 27% of its value in 1986 (Fig. 4). It corresponds to an activity diminution due to the transfer into the soil about 3 times higher then expected by the half live decrease, that means an apparent half live of round 10 years instead of 30 years. So we can estimate an evolution of the relaxation mass-per-area of the Chernobyl caesium from 1 g.cm^{-2} for the measurements performed in summer 1986 to about 10 g.cm^{-2} in 1996. Particular sites such as Hinterrhein by which caesium is very efficiently retained in the upper layer by clay or organic materials (forest) can show practically the expected half live decrease. Otherwise disturbed sites by which the natural transfer is modified, e.g. by plowing, show an activity diminution up to 7 times higher than expected by the half live decrease. These considerations make it clear that knowledge of the type of soil and its history are very helpful to interpret the evolution of the caesium activity and its contribution to the ambient dose rate equivalent.

137Cs	ρ/α g.cm ⁻²	Bq.m ⁻² Bq.kg ⁻¹
Ideal Plane	0	30
Dry deposit	0.3	40
Wet deposit 1986	1	53
Gradual transfer 1987	2	66
1988	3	79
Swiss soils 1989	4	90
~ 1.g.cm ⁻² per year 1990	5	101
1991	6	112
1992	7	124
1993	8	134
1994	9	144
1995	10	153
1996	11	161
1997	12	175
1998	13	189
1999	14	202
2000	15	216
2001	16	230
2002	17	244
2003	18	258
2004	19	271
2005	20	285
2010	25	351
~ homogeneous 2015	30	415

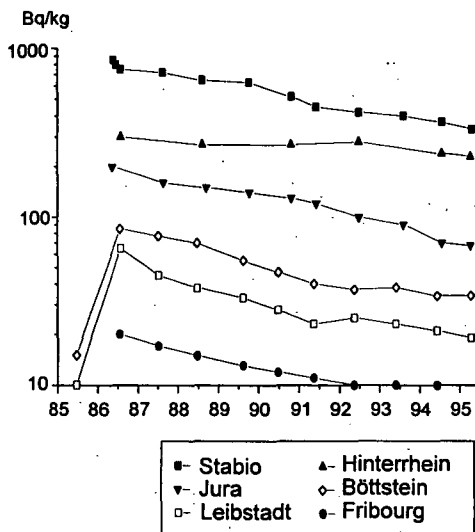


Figure 4: Evolution of in situ caesium-137 inventory (Bq/kg) at several swiss sites.

5. Soil profiles

Soil profiles are also taken in Switzerland to determine the radioactive inventory of the soil and to verify in situ assessments relative to the distribution of the radionuclides in it. For natural gamma emitters, the assumed uniform depth distribution is generally confirmed by soil profiles measurements and also by reasonable agreement between in situ spectrometry results and undried soil sample analysis. Some caution is needed for the ²³⁸U - ²²⁶Ra series since the emanation of ²²²Rn results in a disequilibrium between radium and its daughters on the typical order of 10 to 20%. For in situ measurements performed during rain, particular assumptions taking into account the radon daughters deposit must be achieved to avoid an overestimation of their concentration in soil. The potential inhomogeneity in the horizontal distribution of fallout activity which may vary from one year to the other (Fig. 5) complicates the comparison between in situ spectrometry and soil sample analysis. The first example shows a good agreement between the caesium inventory obtained by fresh and dried samples. For the fresh ones we determine a mean conversion factor of 70 kg/m² about 30% higher than for the dried ones according to the difference in moisture content. The same inventory would be restored by in situ measurements assuming $\rho/\alpha = 17 \text{ g.cm}^{-2}$ for ¹³⁷Cs resp. $\rho/\alpha = 14 \text{ g.cm}^{-2}$ for ¹³⁴Cs. It is interesting to observe that the mean value of the relaxation mass-per-area of ¹³⁷Cs is generally higher than the corresponding value for ¹³⁴Cs.

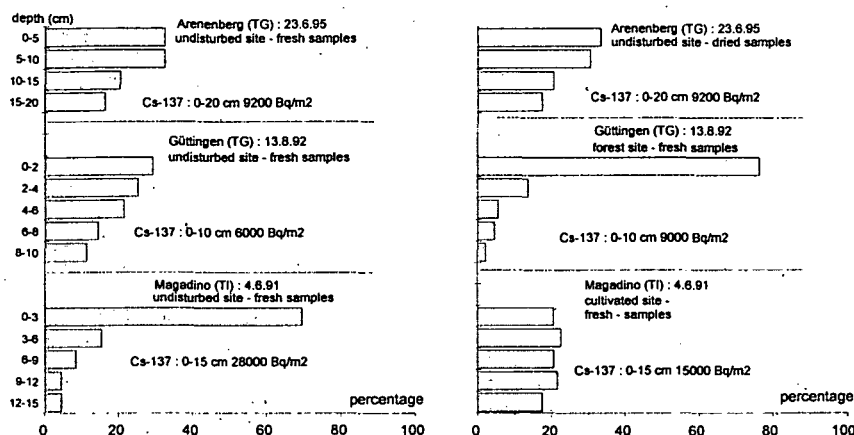


Figure 5: Percentage ¹³⁷Cs in soil profiles

This difference is due to the ¹³⁷Cs contribution from the weapons fallout. In the forest soil at Göttingen, the sampling down to 10 cm yields the global inventory of caesium, which is mainly retained in the upper layer. The adjacent undisturbed soil yields only about 70% of that inventory indicating that a deeply sampling would be required to determine the total inventory. The relaxation mass per area values which would restore the global ¹³⁴Cs resp. ¹³⁷Cs inventory for the corresponding in situ measurements are 1 g cm⁻² resp. 3 g cm⁻² at the forest site and 8 g cm⁻² resp. 11 g cm⁻² at the undisturbed site. A similar observation appears in Magadino between the undisturbed site and the adjacent cultivated one that was plowed deeply. The last one yields only about 50% of the effective inventory, which would be restored from the corresponding in situ measurements by assuming $p/\alpha = 4 \text{ g.cm}^{-2}$ for ¹³⁴Cs resp. $p/\alpha = 6 \text{ g.cm}^{-2}$ for ¹³⁷Cs at the undisturbed site and $p/\alpha = 25 \text{ g.cm}^{-2}$ for both caesium isotopes at the cultivated one. So an important precaution to investigate the soil inventory by sampling is to sample to a great enough depth taking care not to spill soil from a layer into another.

6. Ambient dose rate equivalent

The total ambient dose rate equivalent from penetrating radiation (gamma and cosmic) in the environment can be measured accurately with a pressurized ionization chamber. This can be compared with the sum of the individual contribution of each nuclide from spectrometric in situ determinations with the cosmic component added in. The last depends on the altitude H (km): $n\text{Sv.h}^{-1} (\text{cosmic}) = 37 \exp(0.38.H)$, Murith (1987). Agreement to within $\pm 5\%$ is a quality assurance for the detector calibration and the assumed source distribution. Disagreement by more than 10% points to a calibration problem or a flawed assumption for the source distribution. Figure 6 shows the overestimation inherent to the assumption of uniform depth distribution for artificial radionuclides in soil in comparison with the measured ionization chamber values. The transfer of the artificial radionuclides in deeper layers induces the observed decrease of the overestimation from initially about a factor 2 down to $\sim 20\%$ up to now, at Stabio in the most affected swiss region Tessin by the Chernobyl fallout.

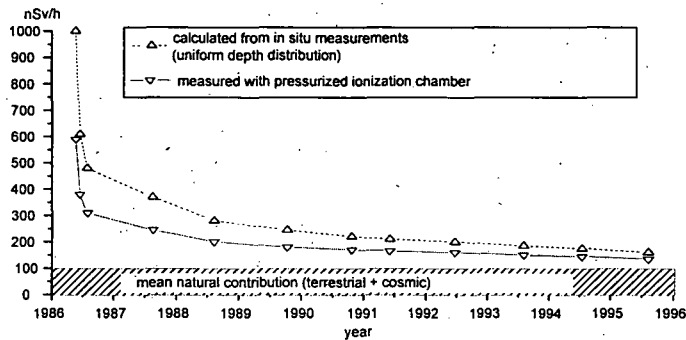


Figure 6: Evolution of the ambient dose rate equivalent at Stabio (Tessin)

A reasonable concordance between calculated and measured values can be achieved for this site by assuming an exponential depth distribution for the artificial radionuclides with a mean evolution of the relaxation mass-per-area of round 1 g.cm^{-2} per year.

7. Conclusion

In situ spectrometry based on uniform depth distribution of the radionuclides in soil is well appropriate to follow the behaviour of artificial deposits. Its especially allows a direct comparison in Bq/kg between the natural and artificial contributions of the gamma emitters, with the advantage of a high sensitivity for routine checking of low level contamination in the terrestrial environment. In order to obtain an accurate assessment of deposited activity with in situ spectrometry, an estimate or actual measurement of ρ/α can be useful. The evolution of the contamination in soil can be followed with sufficient approximation by using the developed in situ procedure that describes the corresponding evolution of the relaxation mass-per-area which would restore the expected inventory. To confirm or interpret in situ informations it is recommended to take into account laboratory spectroscopic measurements of soil profiles and knowledges relative to the soil properties and history. Furthermore a quality control of the in situ results can be achieved by comparison with the measured ambient dose rate equivalent with the advantage to allow an improved investigation of the radiation exposure to the population in providing or even predicting its individual contributions.

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CAESIUM & PLUTONIUM MIGRATION IN FOREST SOILS OF THE CHERNOBYL 30km ZONE

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ABSTRACT

Vertical distributions of radiocaesium and plutonium inventories and uranium ratios (^{238}U : ^{235}U) are presented for forest soils at three sites within the Chernobyl 30km exclusion zone. The uranium ratios indicate the presence of reactor fuel fragments ('hot particles') at all sites, but with declining significance with distance from the ChNPP. Caesium and plutonium distributions in soil profiles were evaluated using a compartmental model to derive migration half times for each horizon. Migration rates at each site were lowest for the AoH horizon. The AoH horizon at the site with the largest apparent population of 'hot particles' exhibited the longest caesium and plutonium migration half times, which generally decreased with distance from the ChNPP (ie. with decreasing 'hot particle' densities). Plutonium appears to be more mobile than radiocaesium, possibly due to the movement of soluble Pu-humic complexes formed within the surface organic soil horizons.

1. INTRODUCTION

The contaminated 30km exclusion zone surrounding the Chernobyl Nuclear Power Plant (ChNPP) presents a wide variety of ecological situations ranging from farmland to semi-natural meadow ecosystems and forests. All these ecosystems were contaminated by fallout from the explosion at the ChNPP on 26th April 1986 and subsequent atmospheric releases which occurred up to 6th May 1986. The degree of contamination at different sites varies widely, according to location, and the type of contamination also varies with some sites having received a large input of irradiated UO_2 fuel fragments, or 'hot particles'. Thus the 30km zone has for the last few years provided a large outdoor 'laboratory' in which it has been possible to measure the migration rates of a number of fission and activation products and actinides. This 'laboratory' is unique because, whereas many other areas of the Earth are contaminated by plutonium, radiocaesium and radiostrontium, the 30km zone is the only location where much of the original radionuclide inventory was associated with irradiated UO_2 fuel fragments.

Information on the environmental behaviour of radionuclides to be gained from the 30km zone is, therefore, primarily relevant to the most severe reactor accidents in which massive core damage and loss of containment occurs. Such events are extremely rare, yet it is important to maximise our opportunity to learn about the likely impact of any similar events in the future. This paper presents information on the soil migration rates of isotopes of caesium and plutonium at three similar forest sites within the 30km zone which received different radionuclide inventories in 1986.

2. MATERIALS & METHODS

2.1 Soil Sampling: With the assistance of the Radioecology Laboratory of Moscow State University soil samples were collected from three sites within the 30km zone (Table 1) in 1993 and 1994. Composite samples of AoL, AoF, AoH Ah and A1 layers were obtained from between 5 and 7 individual sampling locations at each site. These were placed directly into polythene bags for transport to the UK where radiochemical analyses were carried out.

2.2 Analytical Protocols: Radioisotopes of interest in this study were ^{137}Cs and ^{134}Cs (together with other gamma emitting radionuclides), ^{238}Pu , $^{(239+240)}\text{Pu}$, ^{238}U and ^{235}U . The uranium isotopes are of interest as the fuel matrix comprises uranium oxide with associated fission products, including isotopes of caesium and plutonium. The ratio of ^{238}U : ^{235}U is informative in identifying the presence of irradiated reactor fuel fragments (see below). Instrumental neutron activation analysis and delayed neutron counting were used to determine ^{238}U and ^{235}U concentrations, respectively, in soil samples. Full details of these analyses are given elsewhere (Wang, 1996). After oven drying the soil samples were thoroughly homogenised, packed into 75 ml cylindrical containers and placed on a high resolution gamma ray (GeLi) spectrometer. Count rates of ^{137}Cs , ^{134}Cs were converted to absolute activity concentrations using a detector efficiency calibration obtained from a mixed gamma standard (Amersham International, UK) made up in an identical geometry to the unknown samples. After ashing at 450°C for 8 hours the soil samples were dissolved in concentrated nitric acid and radiochemical separation of plutonium achieved using the IAEA's published method (IAEA, 1989). Throughout this process ^{242}Pu was used as a yield tracer and internal standard. Plutonium isotopes were then electroplated onto stainless steel planchettes and detected by alpha spectrometry using a surface barrier detector *in vacuo*. Absolute disintegration rates of ^{238}Pu , $^{(239+240)}\text{Pu}$ were obtained by direct comparison of peak areas with the ^{242}Pu internal standard.

Site	Approximate Distance (km)	Inventory (kBq m ⁻²)		Tree Species	Soil Type
		239+240Pu	137Cs		
Shepeliy	6km WNW	250	47 000	<i>Pinus, Betula, Quercus</i>	podzol on sand
Kopachi	6km SSW	24.5	2 800	<i>Pinus</i> spp.	podzol on sand
Diyatki 1	25 km S	4.5	410	<i>Pinus, Betula</i>	podzol on sand

Table 1: Locations and characteristics of the three study sites (NB, distances and directions are in relation to the ChNPP).

Soil Horizon	Caesium				Plutonium			
	Shepeliy		Kopachi		Shepeliy		Kopachi	
	τ	v	τ	v	τ	v	τ	v
AoL	0.99 ± 0.01	1.01	1.11 ± 0.42	0.90	1.01 ± 0.03	0.99	1.02 ± 0.03	0.98
AoF	1.76 ± 0.04	1.42	3.04 ± 0.70	0.82	3.60 ± 0.07	0.69	3.31 ± 1.08	0.76
AoH	199 ± 16.8	0.01	23.9 ± 3.39	0.06	10.5 ± 1.59	0.14	12.9 ± 8.81	0.12
Ah	1.29 ± 0.01	0.78	6.04 ± 1.14	0.17	3.32 ± 2.08	0.30	2.02 ± 1.63	0.50
A1	6.61 ± 0.06	0.61	16.7 ± 13.2	0.24	28.0 ± 18.7	0.14	78.1 ± 45.1	0.05
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2.3 Data Analysis: Vertical migration rates of radionuclides in soil profiles were evaluated using a compartmental leaching model similar to that described by Bunzl *et al.* (1992; see Figure 1). Unlike a 'physically based' model no supplementary information on the soils (soil moisture, soil water flow velocity, solute K_d) is required; such information is difficult to assemble for soils under natural conditions. Although migration rates derived from compartmental models cannot easily be extrapolated to other soils and situations such models can be powerful tools in assessing migration rates at specific sites.

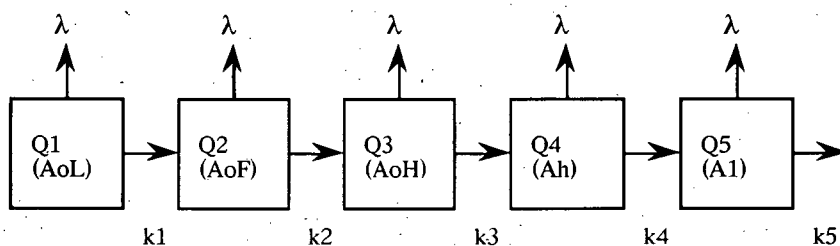


Figure 1: Structure of the five compartment model applied to the data in this study.

The total soil inventory at each site was calculated by summing the individual horizon inventories (Q , kBq m^{-2}), then the vertical distribution of the radionuclide of interest was represented using fractional inventories (Figure 2). The use of fractional inventories means that a $Q(o)$ value of 1.0 can be used as the initial input to the AoL compartment; a series of simultaneous ordinary differential equations describing the movement of radionuclides from one compartment to the next can then be solved either analytically or numerically to give the relative distribution of the soil inventory at any time after deposition. The values of the rate constants k_1 to k_5 were derived by fitting analytical solutions (the Batemann equations) for the five compartments shown to the observed fractional inventory profiles. This fitting technique ignores the fact that at the time of deposition approximately 60 - 80% of the total fallout inventory is captured by the tree canopy overlying the soil surface (Tikhomirov *et al.*, 1993), which is subsequently transferred to the soil surface with a half time of 3 - 6 months. The accuracy of the fitted rate constants was therefore evaluated by numerically solving a six compartment model (SSLM.6 - Shaw & Wang, unpublished) in which initially only 20 - 30% of the total radionuclide inventory contaminated the AoL horizon, the remainder being associated with the overlying tree canopy. The results indicated that the rate of canopy to soil surface transfer was so rapid that after 7 or 8 years migration the soil distributions predicted by SSLM.6 were only negligibly different from the observed values.

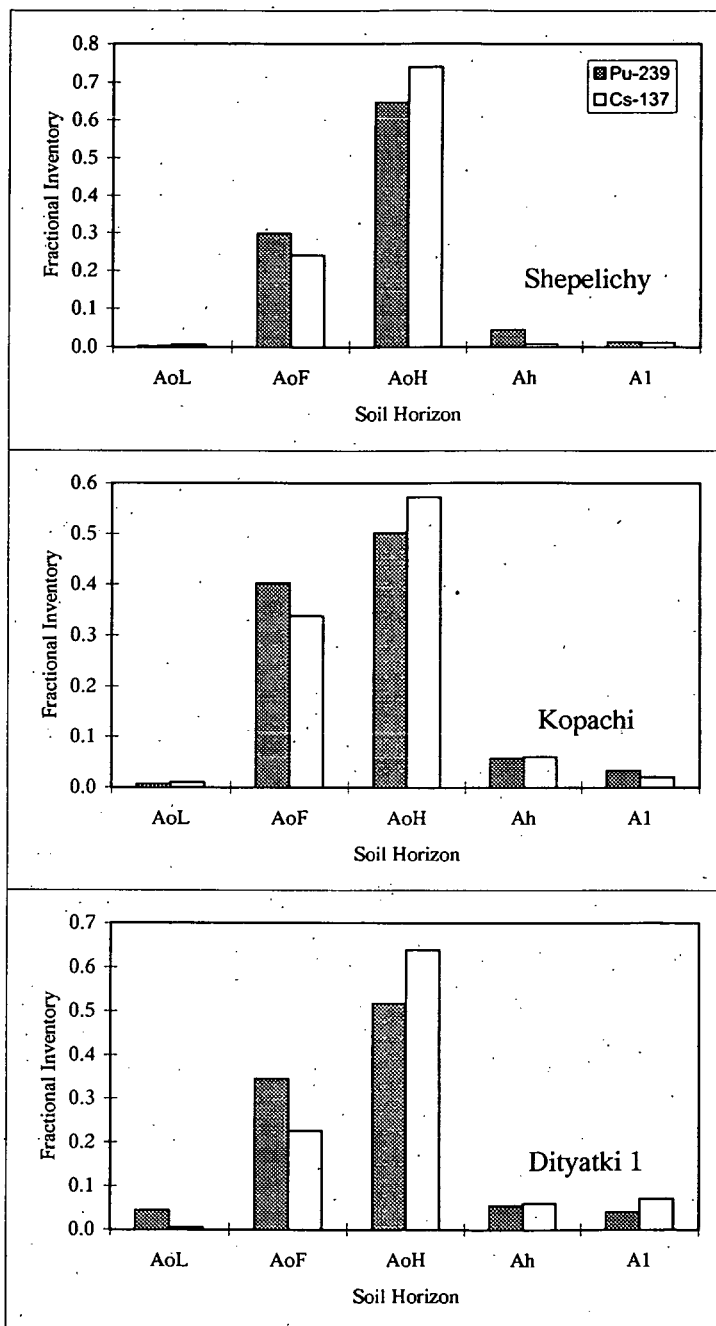


Figure 2: Vertical distributions of fractional inventories of radiocaesium and plutonium at Shepelichy, Kopachi and Dityatki 1 in May 1994.

3. RESULTS & DISCUSSION

Figure 3 shows the variation of the uranium isotopic ratio ($^{238}\text{U}:^{235}\text{U}$) with soil depth for the first five horizons at Shepelichy, Kopachi and Dityatki 1. At the two sites nearest the ChNPP there is clear evidence of ^{235}U enrichment, particularly in the AoF and AoH horizons and this is especially evident at the site with the highest radionuclide inventory (Shepelichy). This indicates the presence of a source of uranium which is non-natural in origin, most likely derived from irradiated reactor fuel in the form of 'hot particles'. Even at the relatively distant Dityatki 1 site there is significant ^{235}U enrichment in the AoF horizon, although the relative occurrence of fuel fragments at this site is evidently much lower than at Shepelichy and Kopachi. The sites chosen for study, therefore, provide a gradient of both total radionuclide inventories and of 'hot particle' populations.

The fractional radionuclide inventory distributions in Figure 2 give a first indication of the relative mobilities of caesium and plutonium at each site. Mobilities of both caesium and plutonium at Kopachi and Dityatki 1 appear broadly similar, with a greater relative migration below the AoH layer than at Shepelichy. This is confirmed by the mean migration half times reported in Table 2 which indicate that for the AoH horizon at Shepelichy both caesium and plutonium had much lower mobilities than at the other two sites. For caesium isotopes there was a systematic decline in AoH residence half time from Shepelichy \rightarrow Kopachi \rightarrow Dityatki 1. It seems likely that this is due to the decreasing importance of 'hot particles' at each of these sites as evidenced by the uranium isotopic ratios in Figure 3. Konoplev & Bulgakov (1995) have reported experimental determinations of leaching half times for 'hot particles' derived from Kopachi which range from 2.2 to 12.5 years, so it is likely that currently a substantial fraction of plutonium and caesium isotopes still remains within intact fuel particle matrices. AoH residence half times of plutonium also showed a similar though less consistent decline from site to site.

Figure 2 indicates that, at Shepelichy, plutonium has penetrated deeper into the soil profile than caesium, and at Shepelichy and Kopachi the AoH residence half times of plutonium are significantly lower than those for caesium. It is therefore clear that while the AoH horizon presents a significant 'barrier' to the migration of both caesium and plutonium it is particularly effective in retarding isotopes of caesium. It seems likely that plutonium isotopes will be complexed by humic molecules within the highly organic AoF and AoH horizons (Agapkina *et al.*, 1995) and this could lead to more rapid leaching of plutonium than caesium over the long term.

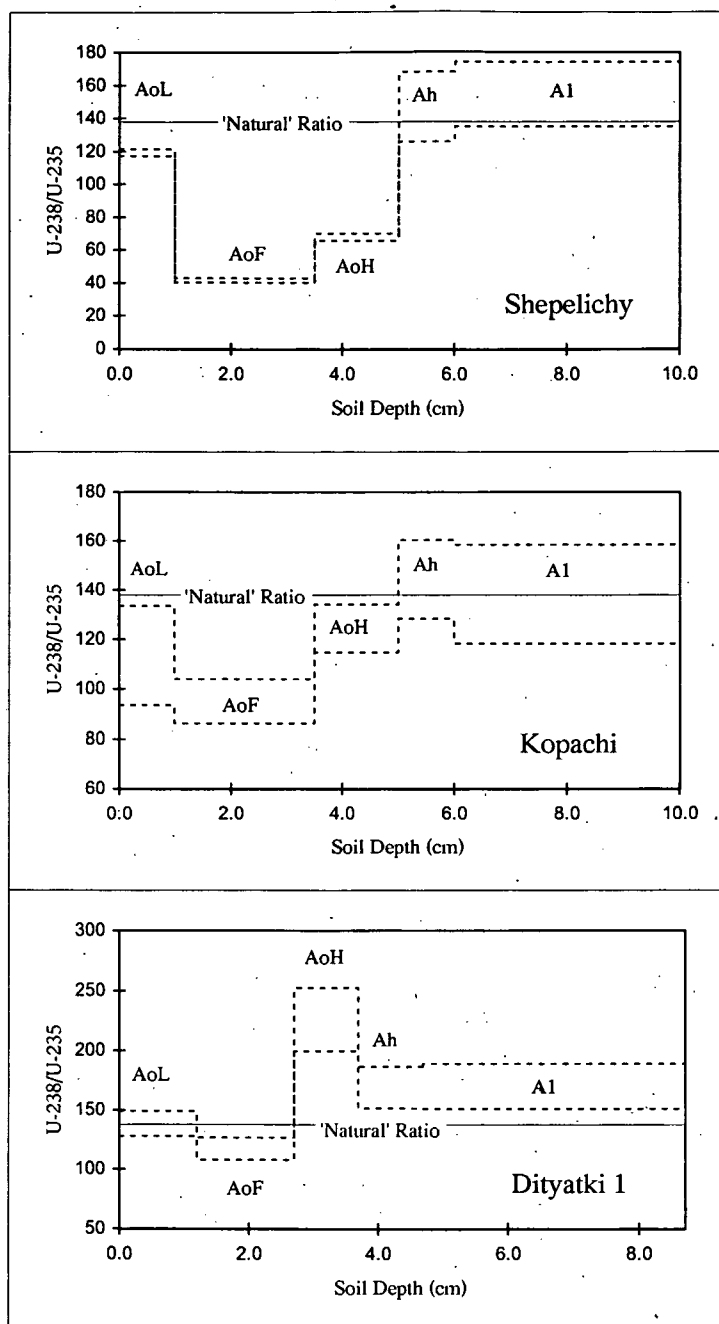


Figure 3: Vertical distributions of uranium isotopic ratios in forest soils at Shepelichy, Kopachi and Dityatki 1 in May 1994 (the dotted lines represent 95% confidence limits of U-238/U-235 ratios).

Our compartmental model evaluations of migration rates do not reveal anything about the mechanisms responsible for the reduced mobility of radiocaesium in AoH horizons in forest soils. Biological recycling processes have been implicated by several workers, although this hypothesis remains to be convincingly demonstrated and appropriate pathways and rates evaluated.

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APPLICATIONS AND LIMITATIONS OF MATHEMATICAL MODELS IN RADIOECOLOGY WITH PARTICULAR EMPHASIS ON RADIOCAESIUM IN SOIL

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ABSTRACT

Mathematical models play an essential rôle in the prediction of the long term fate of radionuclides in ecosystems. This paper reviews some of the modelling approaches adopted in radioecology, with particular emphasis on soil systems. The advantages and limitations of the various levels of approach are discussed. The importance of mechanistically based models is highlighted. Such models can provide insight into the relative importance of the numerous processes which influence bioavailability in ecosystems. They may thus assist in the formulation of efficient research strategies. They may provide a rational basis for input data in large scale 'black-box' models. The need for testing of models, and the associated difficulties are discussed.

1. INTRODUCTION

The presence of radionuclides in soil systems usually poses a long term problem. Society however demands an immediate prediction of the duration of the disruption to normal life: such a demand cannot be addressed by the experimental approach. Contamination also typically occurs on a landscape scale, affecting a wide variety of soils and ecosystems, each of which will behave slightly differently. For both the above reasons, mathematical models have an important role to play in the predictive process (Myttenaere *et al.* 1993; Coughtrey, 1988). Mathematical prediction of the fate of such radionuclides requires an excellent understanding of the various governing processes: predicting the effect of modifications of the system is even more difficult. The need for such models has been highlighted in previous Workshops (e.g. Desmet *et al.* 1990) and an increasing number of models have been developed to fulfil this need. Other models which have been developed in other fields of research could also be applied to radioecology. The aim of this paper is to review modelling approaches which are relevant to radioecology, and in particular the bioavailability of radiocaesium in soil-plant systems. We will not provide an exhaustive review of all published models, nor will we

compare their predictions. Our aim is to draw attention to the usefulness and the limitations of mechanistic mathematical models of bioavailability, and to indicate how models should be designed. The emphasis is placed on models of soil-plant transfer of radiocaesium.

2. REVIEW OF EXISTING MODELLING APPROACHES

The use of mathematical models in all fields has greatly increased in the last twenty or thirty years, largely due to progress in information technology. They are potentially valuable research tools with a wide range of applications. The most basic aim of a model is to assemble and provide a concise mathematical description of a complex system, especially one where the component sub-processes are impossible to test individually. Another application is the extrapolation of an existing system into the future, testing the effect of various changes. At another level it may provide some insight into the understanding of a phenomenon or aid in the design of more sensitive experiments. It is vital to recognise that mechanistic models are simply a set of hypotheses and require rigorous testing against experimental data before they can be considered to have any predictive validity. In radioecology the greatest demand is for models which are reliable predictive tools. However, as we hope to show, mechanistically based models have a very important rôle to play in elucidating the relative importance of various processes and in providing rational input to models not directly based on mechanisms.

Mathematical models are simplified descriptions of systems. Varying degrees of simplification are obtained by making assumptions and the accuracy of a model depends on their validity. The degree of simplification depends not only upon the complexity of the system to be described, but also on the scale investigated. Generally, the larger the scale of the model, the greater the degree of simplification although this may change as computing power continues to increase. For example, at the rhizosphere level, detailed descriptions of root uptake, root size and morphology and adsorption kinetics may be taken into account: while at a landscape level it may be that more processes have to be taken into account but a simpler mathematical description is used for each rhizosphere subprocess. Similarly the nature of the heterogeneity which has to be taken into account depends on the scale of interest. At a rhizosphere level one may wish to investigate the effect of the depletion of the radionuclide or a competing nutrient species, the effect of enrichment in macro-nutrients near the root surface, the weathering of clay minerals by root exudates or the non-uniformity of root distribution. At a landscape level the heterogeneity of deposition, of soil properties or of vegetation cover would be more relevant. Well-designed models rely on smaller-scale models when deciding the degree of simplification that is acceptable.

2.1 The Black-box Approach to Multi-Compartment Models

For very large complex systems, such as ecosystems, there is little alternative to a 'black box' approach where the transfer between various components (e.g. soil to soil solution to plant roots to plant shoots to grazing animals, soil ingestion, animal faeces, leaf fall, stem flow) is described using very simple rate, or transfer, coefficients (Coughtrey, 1988). Relatively few of such coefficients are required but this black box representation takes no account of spatial heterogeneity or time dependent changes in rate constants. It typically has no theoretical basis. The rate constants may be derived from studies in simple closed systems, such as described below, or may be obtained by fitting the model to experimentally observed data. Such models are extremely useful because they are simple to develop and to run but have rather limited predictive power. They may provide reliable extrapolations with time if the system studied remains unchanged but cannot be extrapolated to different environments if changing conditions influence the processes which determined the rate constants. Examples of such changes would be climatic conditions which alter the rooting pattern of plants, nutritional status, seasonal changes in vegetation, changes in soil adsorption capacity with time or because of soil amendments. If the effects of such changes on the appropriate rate constant can be estimated, then a useful sensitivity analysis can be obtained. However this is usually not possible, particularly if the rate constants have been obtained by fitting. If there is little rational basis to the estimation of the input parameters then extrapolation from one to another system is unlikely to be accurate.

A modification of this approach, which is conceptually similar, but is more difficult to model, is to divide the soil into a small number of related layers, in which the rate constants for each process may differ (Crout *et al.* 1990).

2.2 Mechanistic Models of Simple Systems

For very simple systems a limited number of processes are considered and so models may be used to obtain rate coefficients and to support or disprove hypotheses about mechanisms. Such simple systems include the uptake of a radionuclide by plant roots from culture solution or the kinetics of adsorption onto soil or clay in a homogeneous paste or suspension. An example of the former is the description of the root uptake of radiocaesium from solution which is often considered to follow Michaelis-Menten kinetics, possibly with inhibition by competing ions, particularly potassium (Shaw and Bell, 1991). The latter process is usually described by a series of first order reactions (Absalom *et al.*, 1996), but a slight variant is to consider some of the adsorption to follow a non-linear adsorption isotherm, such as the Freundlich isotherm (Comans and Hockley, 1992).

The purpose of such models is to obtain a mathematical description of experimental results obtained in controlled systems. The data expressed thus is suitable for incorporation in

a more complex model. It is also easier to compare them with the results of other studies. In general such models are routinely used and the mathematical techniques are simple and well-known. In some cases these treatments elucidate the mechanisms controlling the processes studied, as for example the inhibition of caesium uptake by potassium in solution.

2.3 Mechanistic Approach to Complex Systems

However systems are usually too complex, even in controlled laboratory experiments, to be described by such simple models. Dynamic mechanistic models can be used at an intermediate level to describe fairly complex systems in detail, with each process having the best possible mathematical description as determined in simple systems. Nevertheless this approach rapidly becomes extremely complicated and various approximations are required. One approximation is that roots are of uniform size, with identical absorption capacities and that they are uniformly distributed in the soil. Another similar assumption is that the soil is a uniform porous medium (i.e. that the diffusion coefficient and the dispersion coefficient are constants) and that the initial distribution of the radionuclide is uniform. One such approximation was that used by Kirk and Staunton, (1989). They argued that because the mobility of strongly adsorbed species, such as radiocaesium in mineral soils, is very low, root uptake is therefore rapidly limited by diffusion and it was therefore reasonable to assume that there would be an instantaneous steady state concentration gradient perpendicular to the root. This simplification gave a simple relation between average concentration and concentration at the root interface which determined the uptake rate. However in poorly buffered soils this assumption may not be valid.

In many respects the problems faced in radioecology are regulated by the same or similar processes as in other related fields such as soil science, plant physiology, crop growth, animal nutrition and ecology. Models developed in other fields may therefore be useful in the study of radionuclides, and may sometimes require very little modification. For example the model of Kirk and Staunton (1989) was adapted from models used in soil science (Nye *et al.*, 1975; Claassen and Barber, 1976; de Willigen and van Noordwijk, 1987). Some of the differences between the models used in various fields arise from the relative importance of various processes and the size and complexity of the system to be. For example the soil chemistry of caesium is very similar to that of potassium. Models which describe the plant availability of soil potassium, of which there are several, should be well adapted to studies of caesium in the soil-plant system. There are however important differences. Firstly the depth distributions of radiocaesium and potassium are different. Radiocaesium is generally highly concentrated in the surface layer of soil because of surface contamination and relatively low movement, whereas potassium is more homogeneously distributed because it is naturally present in soil, even if fertilizers are surface applied. A second difference is in the account

taken of non-instantaneous equilibration of cations between adsorbed and solution phase. All models of caesium availability consider the slow exchange of a fraction of the adsorbed cation. Although potassium undergoes a similar reaction this is not routinely included in models of potassium bioavailability.

In contrast models designed to describe water uptake, or the uptake of an ion such as nitrate which is thought to be largely determined by water absorption, are poorly adapted to the uptake of sparingly soluble radionuclides. Such models emphasize the importance of transpiration and of root architecture.

We have recently introduced a further degree of complication into such models by considering the turnover of caesium in both roots and shoots subsequent to root absorption. We recognised that it is unreasonable to assume that root position is constant over the period of many years. Therefore even at a constant average rooting density, roots do not continue to exploit the same, increasingly depleted, cylinder of soil. Root turnover may contribute to depth redistribution of caesium. Shoot turnover returns a fraction of absorbed caesium to the soil surface. This may be the result of rotting vegetation, or animal grazing and return *via* urine and faeces.

These mechanistic models are designed for use at a level intermediate between the ecosystem and controlled systems in glasshouses or growth cabinets. Their major role in radioecology is the determination of the relative importance of various processes (sensitivity analysis) particularly as many processes are interdependent. Sensitivity analyses may provide a rational basis for the estimation of the input variables to black box models, and to other modelling approaches, such as Geographic Information Systems, which have not as yet been fully exploited in radioecology. They may also be useful in designing remediation strategies for contaminated areas.

3. MODEL INPUT AND OUTPUT PARAMETERS AND MODEL TESTING

3.1 Measurement of Input Parameters: State variables and other boundary conditions

State variables characterize and quantify all properties that describe the current state of the system and have to be known at the onset of simulation (de Wit and Goudriaan, 1978). Examples of such variables are (i) the initial concentration and distribution, with depth or between adsorbed and solution phases, of the isotope and (ii) the root radius and density distribution. Other boundary conditions are the continuing addition of radionuclide to the system, or its removal.

The initial concentration and distribution of a radionuclide in a real system are difficult to measure. Measurements are highly variable, partly because of sampling error and partly because surface contamination and redistribution are highly variable. The root density and

architecture are also difficult to obtain with precision because of sampling difficulties - some field and lysimeter data are available, but only for a limited number of species. In general removal of radionuclides only accounts for a small proportion of the amount present, and so this parameter has little effect on simulation, although it may of course be the desired output variable. However, particularly in forest ecosystems, deposition may not be quasi-instantaneous. Unfortunately accurate data on the rate and extent of throughfall, leaf-fall and stemflow are difficult to obtain, and so input to models must be treated with some caution. Simulation over a range of state variables can be used to determine the range of possible outcomes.

3.2 Measurement of Input Parameters: Rate variables

Rate variables quantify the rate of change of the state variables (de Wit & Goudriaan, 1978). Examples of rate variables are (i) the rate constants of the exchange reactions of the radioisotope between the adsorbed and the solution phases; (ii) the variables which describe the root uptake at the root-solution interface, (iii) the diffusion and dispersion coefficients, or their constituent components, (iv) root growth rate or turnover rate, (v) rate of transfer between roots and shoots and (vi) grazing rate.

Usually these variables are obtained from experimental studies carried out under controlled conditions, such as adsorption measurements in soil suspension or soil pastes or root absorption from culture solution. It is assumed that the difference in conditions does not have a marked effect on these rate variables. Again this assumption must be treated with caution. In the case of adsorption on soil, kinetic parameters are obtained over periods of hours, days and more rarely months. Such studies cannot detect reactions with very slow rate constants, yr^{-1} or lower. If the long term fate of a radionuclide is of interest, then reactions with half lives of the order of hours, days and months will appear to be instantaneous. Similarly for the kinetics of root absorption, it is well known that root development is very different in nutrient solution and in soil. Furthermore absorption properties depend on species, age, and nutrient status. To what extent can measurements made on young plants, usually of species of commercial interest, well supplied with important nutrients, be applied to field conditions? This caveat is particularly relevant to simulations of semi-natural environments where the nutrient supply is poor and plant species are less likely to have been studied by plant physiologists and where fundamentally different mechanisms may apply.

3.3 Choice of Output Variables

The choice of output data depends on the intended use of the model. In the case of simple models, designed to give a simple mathematical description to experimental data it is the experimentally measured parameter. In more complex models the output variable may

correspond to a measureable parameter (which allows the model to be tested by comparison of simulated to real data). For example, the soil-plant transfer factor, the ratio of concentration in the shoot+leaf of the plant to that in the surrounding soil. Alternatively it may be an input parameter, or a rate variable to be incorporated into a more simplified model of a larger, more complex system. For example, the soil-to-plant rate constant in a compartment model. In general the output variable can be measured, so that the model can be verified. However this is not always the case - it is interesting to simulate the changing depth distribution of a radionuclide, although this is difficult to monitor precisely with time.

3.4 Testing of models

In order to test a mathematical model reliable data must be available. This is trivial, albeit time-consuming, for models of simple two-component systems. The poor prediction of the bioavailability of radiocaesium in the organic upland soils by some radioecological models based on the black box approach was made obvious by the Chernobyl accident. However such opportunities for model testing are (happily) not readily available, and when they are, time-dependent data is scanty. Good agreement between data and simulation is not sufficient evidence of a good model; it may be coincidence. Furthermore if simulation does not agree with observation, it is not easy to be certain whether the model is fundamentally at fault (i.e. the simplifying assumptions, the mathematical description of the processes or the selection of the processes taken into account are erroneous) or whether the input variables are wrong. The more complex a model, the greater is the number of input parameters required, with a corresponding increase in the possibility of error. A useful pragmatic approach is to conduct a sensitivity analysis to determine the parameters upon which outcome most depends and then try to obtain data where only this parameter changes or concentrate experimental effort on accurate measurement of this parameter. For example, root uptake is highly sensitive to rooting density and depth distribution. One would therefore expect soil-plant transfer coefficients to be highly correlated to root architecture. Unfortunately such data is extremely difficult to obtain. A more successful example of this approach is to compare soil-plant transfer coefficients with soil adsorption capacity, on condition that a sufficient range of soils is considered (Van Voris *et al.*, 1990).

4. CONCLUSIONS

Mathematical models are useful tools in all areas of plant and animal science. In some areas, such as radionuclide contamination and climate change, where decisions are required before all the experimental evidence can be assembled, they are essential. In such areas, it is vital that the models provide as accurate predictions of future events as possible. Of the models discussed, the mechanistic approach seems to offer the best approach to this requirement.

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SORPTION/DESORPTION PROCESSES OF CESIUM AND STRONTIUM IN SOIL: IS THE K_d -CONCEPT ADEQUATE ?

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Abstract

Interaction with the soil matrix is one of the key processes for the migration of radionuclides in the terrestrial environment. Usually it is modeled by a simple distribution coefficient K_d of trace concentrations between solid and liquid phases. Laboratory experiments using a modified diffusion technique have been performed to study the interaction of cesium with a Eutric Cambisol soil pellet. Of the sorption models tested, assuming first-order kinetics incorporating a Freundlich isotherm resulted in the best fit to the experimental data. The K_d model completely failed to describe the experimental data. Desorption showed a marked hysteresis.

1 Introduction

As it is well known, transport of kationic radionuclides in near-surface soils is a slow process with migration rates in the order of 1 cm y^{-1} or even lower (Coughtrey and Thorne 1983). These data indicate that the mobility in soils is controlled by sorption of the radionuclides to the soil matrix. As Ehlken and Kirchner (1994) recently showed, redistribution of cesium within the rooting zone of grass plants significantly influences soil-to-plant transfer. Hence sorption (and desorption) processes in soil control radionuclide uptake by plants and have to be modeled adequately in any long-term dose assessment.

The sorption of a radionuclide in soil is usually expressed in terms of a distribution coefficient K_d defined by

$$S = K_d C \quad (1)$$

where S , C are the nuclide concentrations sorbed and in solution, respectively. Eqn. (1) only holds if (i) the concentration sorbed is linearly related to the concentration in solution, if (ii) sorption is a quasi-instantaneous process, and if (iii) it is completely reversible. There is, however, experimental evidence that eqn. (1) may be inadequate: In various experiments, sorption of cesium to different soil or rock types was reported to follow a Freundlich isotherm with exponents in the range $0.5 \leq N \leq 0.95$ (Torstenfelt et al. 1982, Bradbury and Stephen (1985), Higgo et al. 1988, Smith 1990). Observations of kinetically controlled sorption processes of cesium with sorption half-lives of days to weeks have been reported also (Nyfeller et al. 1984, Horyna and Dlouhý 1988, Shenber and Johanson 1992, Comans and Hockley 1992).

The objective of this study was to test the performance of various sorption/desorption models versus data of a laboratory experiment investigating the sorption of cesium on a mineral soil.

2 Laboratory Techniques for Studying Sorption Processes

A variety of laboratory techniques for studying sorption processes have been proposed. For strongly sorbing trace substances most widely used is the batch technique (e.g. Relyea et al. 1980). This simple methodology, however, poses a number of methodological problems (Bell and Bates 1988, Lieser and Ament 1993, Beneš et al. 1994) and can not be expected to give reliable results (Relyea and Serne 1979, Bradbury and Stephen 1985). In our opinion, the diffusion technique (Shackelford 1991) offers an attractive alternative, because it enables to study sorption processes under experimental conditions held close to those in the environment.

3 Experimental

The through-diffusion technique used for our experiments is illustrated in Fig. 1. Tracer nuclides diffuse from the reservoir cell across the soil pellet which is fixed mechanically by two membranes into the measurement cell. The experiment is performed in three phases. During phase (1) the fluid in the measurement cell is frequently replaced by synthetic soil solution. In a second phase, tracer concentrations in the measurement cell are allowed to accumulate in order to establish high loadings throughout the soil pellet. Phase (3) is initiated by removing the tracer from the reservoir so that transport of cesium out of the pellet is governed by desorption processes. During this phase, solutions in both cells are periodically replaced. Tracer concentrations accumulated in the exchanged solutions are always measured γ -spectrometrically.

In the following, results are presented of an experiment studying the interaction of ^{137}Cs with soil taken from the A_h horizon of a Eutric Cambisol (Kirchner and Baumgartner 1992). The initial concentration of cesium in the reservoir was 2.5×10^{-7} M.

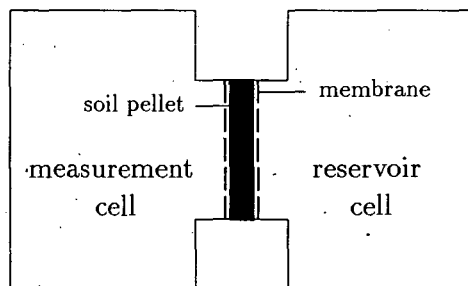


Figure 1 : Schematic of the diffusion cell used

Migration of the tracer nuclides through the soil pellet is described by a diffusion-sorption equation which can be solved analytically if sorption is modeled by a linear isotherm (Kirchner et al. 1993). In order to gain experimental flexibility and to test the performance of various sorption models, we developed a numerical solution based on the Crank-Nicolson method of finite differences and incorporated it into the nonlinear regression code PAR of the BMDP statistical code package (Dixon 1990).

4 Results and Discussion

Concentrations of ^{137}Cs detected in the solutions which were withdrawn from the measurement cell are shown in Fig. 2. The six sorption models listed in Table 1 were fitted to these data. They include various isotherms (linear, Freundlich, and Langmuir) and for each of the isotherms assume sorption to be an instantaneous and kinetic process, respectively. Parameters fitted are (see Table 1) K_d for the linear isotherm, K_{Fr} , N for the Freundlich isotherm, K_{La1} , K_{La2} for the Langmuir isotherm, the kinetic constant r and, additionally, the intrinsic diffusion coefficient. The decision which of the sorption models tested results in the best agreement with the experimental data was based on the Akaike information criterion AIC : if measurement errors are additive and normally distributed with mean zero, it is given as (Cobelli and Ruggeri 1982)

$$AIC = N \ln R_{ss} + 2p,$$

where N is the number of data points, R_{ss} denotes the residual sum of squares of the fit, and p is the number of parameters fitted. The model which results in the lowest AIC value is selected thereby taking into account both the goodness of fit and the principle of parsimony.

As Table 1 shows, to allow for kinetic processes drastically improves the fits for all three isotherms tested. The best fit results if concentration dependence is assumed to conform to a Freundlich isotherm. For this sorption model the parameter estimates are given in Table 2.

For comparison with the experimental data, Fig. 2 includes the results from fitting two of the sorption models tested. Evidently the linear equilibrium (K_d -) model gives a very poor fit, whereas the kinetic model including a Freundlich isotherm adequately matches the experimental data. The remaining deviations may point to an additional slow sorption process (Comans et al. 1991).

Cumulative concentrations of ^{137}Cs diffused out of the pellet into the reservoir cell after start of phase (3) of the experiment are shown in Fig. 3. The experiment is still in progress. Predictions calculated with the parameter values given in Table 2 systematically overestimate the concentrations actually measured (see Fig. 3). This hysteresis effect may be caused by irreversible sorption of part of the cesium. This will be analysed after the experiment will have been completed and the soil pellet dismantled.

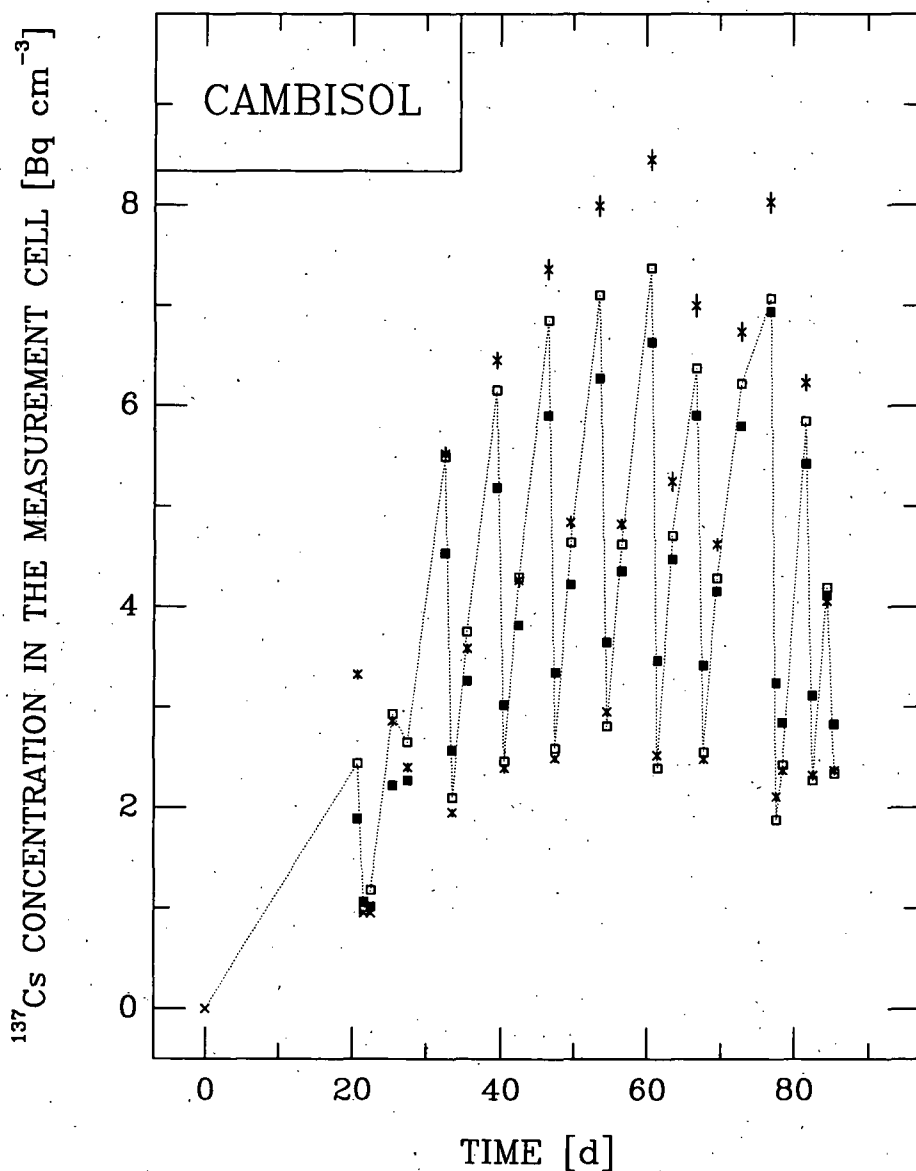


Figure 2: Experimental concentrations (\times) of ^{137}Cs in the measurement cell compared to fits of sorption models no. 1 (\blacksquare) and no. 5 (\square) of Table 1; the dotted lines are for guiding the eyes only

Table 1: Sorption models fitted to the experimental data and computed values of the AIC model selection criterion

No.	Sorption model	R_{ss}^a	AIC ^b
1	$S = K_d C$	10608	310
2	$S = K_{Fr} C^N$	9411	308
3	$S = \frac{K_{La1} C}{1 + K_{La2} C}$	10608	312
4	$\frac{\partial}{\partial t} S = r \{K_d C - S\}$	4337	282
5	$\frac{\partial}{\partial t} S = r \{K_{Fr} C^N - S\}$	1959	258
6	$\frac{\partial}{\partial t} S = r \left\{ \frac{K_{La1} C}{1 + K_{La2} C} - S \right\}$	2173	262

^a residual sum of squares of the fits^b value of the Akaike information criterion

Table 2: Numerical values of the parameters resulting from fitting sorption model no. 5 of Table 1 to the experimental data; uncertainties given denote one asymptotic standard deviation

Parameter	Numerical value
D [10 ⁻⁶ cm ² s ⁻¹]	9.30 ± 0.41
K_{Fr} [Bq ^{1-N} cm ^{3N} g ⁻¹]	9842 ± 2492
N	0.531 ± 0.077
r [10 ⁻⁶ s ⁻¹]	2.02 ± 0.31

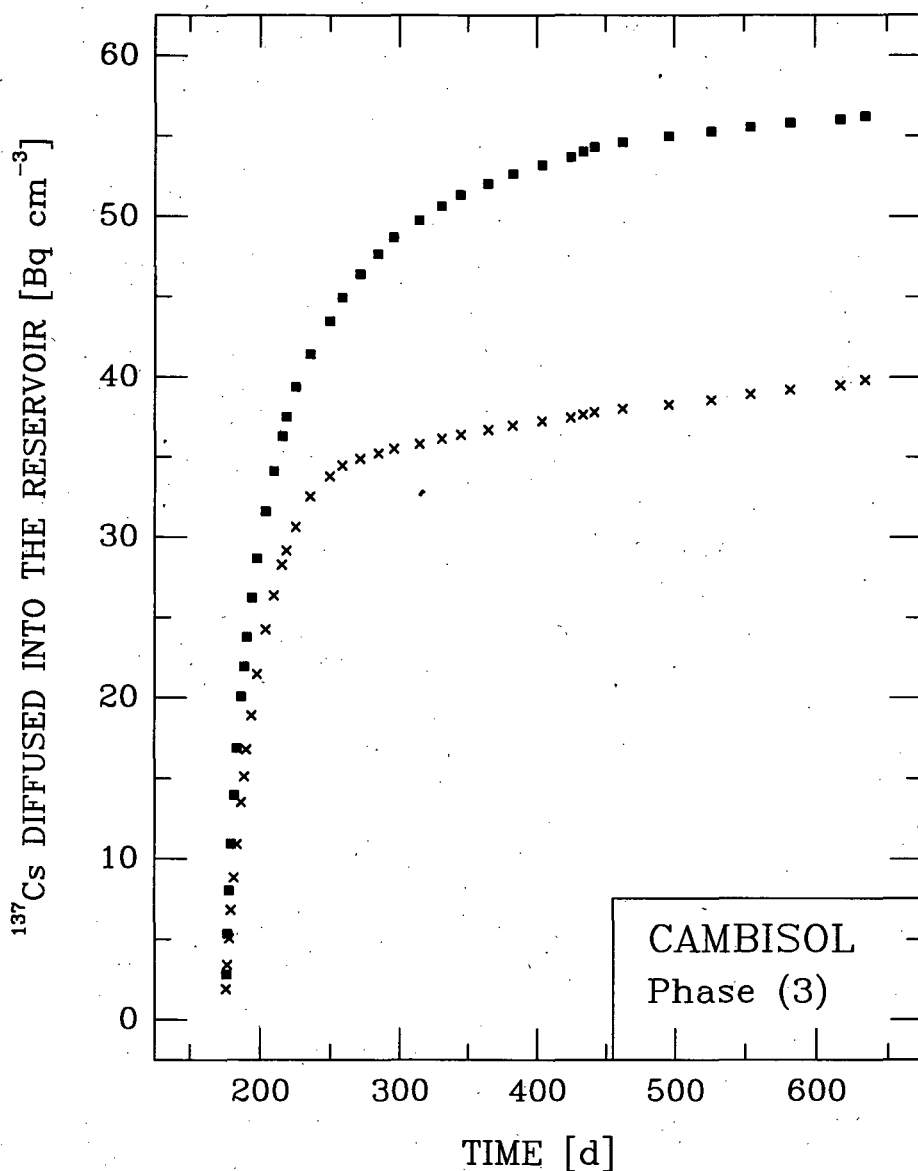


Figure 3: Cumulative concentrations of ^{137}Cs diffused into the reservoir during the desorption phase: \times experiment, \blacksquare predictions of the sorption model with parameter values taken from Table 2

5 Conclusions

Diffusion experiments offer the opportunity to study both sorption and desorption of trace substances in soil and rock under conditions kept close to those in nature. Experimental data for a Eutric Cambisol soil confirm earlier observations that the commonly used equilibrium sorption (K_d -) model may be unsuitable to describe the interaction of cesium with mineral soils.

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LABORATORY STUDY OF THE CAESIUM MIGRATION IN A PODSOLIC SANDY SOIL AS A FUNCTION OF THE IONIC COMPOSITION OF THE SOIL SOLUTION.

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Abstract

On the base of experimental studies realised in laboratory under well controlled conditions, the vertical movement of the caesium in a podsolic sandy soil was investigated using chromatographic columns. The downward movement of the activity was expressed as a percentage of the front of the percolated solution and the result was correlated to the ionic composition of the soil solution (K^+ , Ca^{++} and Mg^{++}). Under our experimental conditions (soil fully saturated with water, inhibition of the microflora, effect of the ammonium not taken into account, Potassium Absorption Ratio kept to a constant value, ...) a simple relationship was found. For a more quantitative approach, this equation must be adapted in order to take into account all these parameters, and especially the influence of the water status of the soil.

1. Introduction

In case of a major nuclear accident (e.g. Chernobyl - 1986), fission and activation products are released to the environment. Considering the committed collective effective dose for the entire population of the different contaminated countries, the two most damageable radioisotopes are the ^{137}Cs and the ^{90}Sr (long physical half-lives, high transfer to the foodchain and big amounts released) (Askbrant *et al.*, 1995). Knowing that one of the largest uncertainties on the predictions is often generated by an insufficient understanding of the fundamental bio-physico-chemical processes which take place in the soil compartment, the study of the behaviour of these radionuclides in the soil is of significant importance. The aim of this work is to try to qualify and to quantify the parameters of the vertical movement of the radiocaesium in a

podsollic sandy soil under well controlled conditions (chromatographic columns) as a function of the main cation composition of the soil solution.

2. Materials and methods

2.1. Chromatographic columns

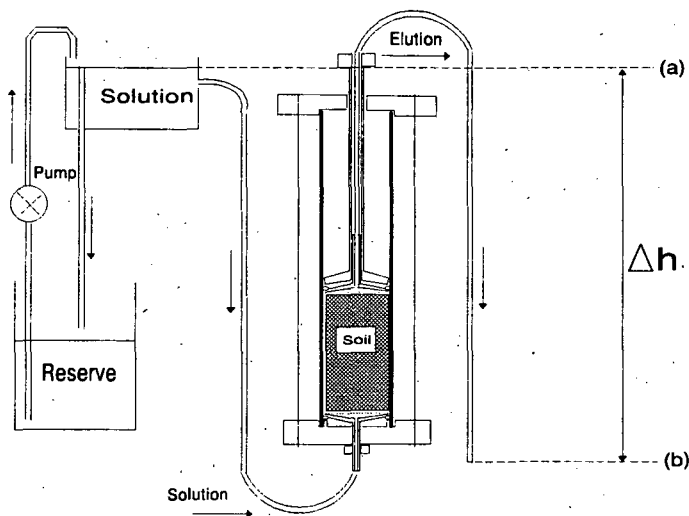


Fig. 1 : Experimental system controlling the solution flux and chromatographic column designed at CEN/SCK.

Home made chromatographic columns represented in fig.1 were filled up with 10 cm of soil. The internal diameter of the Plexiglas tube containing the soil is 4.6 cm and the height of the soil column is adjustable. The tightness of the system is ensure by two compressible o-rings. Successive filters (glass-wool 0.5 cm and paper 0.2 mm) avoid the leaching of soil particles out of the tube. The solution flux is controlled by an adjustable variation of the height (and thus of pressure) between the level of the eluant in the feeding reservoir (a) and that of the exhaust tube (b) from which eluat is collected for analyses. The level in the feeding reservoir is kept constant with a pump and the overflow is recycled.

2.2. Soil sampling and solution used.

The upper 5 cm layer of a podsol soil was collected on a pasture in the region of Mol (Belgium). The soil was air dried, sieved at 2 mm and homogenised. The main physico-chemical characteristics were determined and the composition at equilibrium of the soil solution was estimated :

Density	1300 kg/m ³
Porosity	48 % (v/v)
Field capacity	31 % (w/w)
Cationic Exchange Capacity	15.8 µeq/g
K ⁺ concentration in soil solution at field capacity	1.34 mM/l
Ca ⁺⁺ concentration in soil solution at field capacity	0.87 mM/l
Mg ⁺⁺ concentration in soil solution at field capacity	0.34 mM/l
PAR (at field capacity) = $\frac{[K^+]}{\sqrt{[Ca^{++}] + [Mg^{++}]}}$	1.22
Exchangeable K ⁺	2.0 µeq/g
Exchangeable Ca ⁺⁺	10.1 µeq/g
Exchangeable Mg ⁺⁺	2.0 µeq/g
Exchangeable Potassium Ratio = $\frac{[K^+_{exch.}]}{[Ca^{++}_{exch.}] + [Mg^{++}_{exch.}]}$	0.16
Solid-liquid distribution coefficient for Cs (K _d)	220 ml/g

Table 1 : Main soil and soil solution characteristics of the podsol soil used in this study.

A diluted solution of the main cations in chloride form (KCl, CaCl₂ and MgCl₂) in concentrations close to those measured in the soil solution (respectively 1.0, 0.75 and 0.25 mM → a total concentration of 3 meq/l) was prepared and used for the percolation. The activity of the soil microflora was inhibited by adding 0.01% of Formaldehyde to the solution. In order to quantify the caesium migration phenomena as a function of the total amount of potassium, calcium and magnesium, two other percolation solution's were prepared. For both, the Potassium Absorption Ratio (PAR) was kept constant to his initial value of 1.0 but the concentrations of the main cations were changed. The total ionic concentration was increased from its original value of 3 meq/l to 10 and even to the unrealistic (in natural condition) value of 55 meq/l.

A total of 220 g of dry soil (corresponding to a column high of 10 cm) was introduced in each column. The void volume of the columns was determined by injection of tritiated water

(breakthrough curve) and amounted to 48 ± 1 %. Four columns were used for each scenario's. The soil was equilibrated by passing at least 70 void volumes (V_0) of the eluant before the contamination was applied. Thereafter, 0.2 ml of $^{137}\text{CsCl}$ was introduced at the bottom of each column and the elution was carried out. The experiment was realised under fully saturated conditions (solution coming from the bottom of the column) and the totality of the eluted solution was collected at regular time intervals and measured for his radioactive content. The solution flow rate was equal to one void volume per day ($0.2 \text{ ml h}^{-1} \text{ cm}^{-2}$). The elution from the different columns was stopped successively after the passage of a pre-defined number of column void volumes (different for each column). The activity profiles were obtained by turning the columns upside down, draining them out under gravity for a week and then extruding the soil from the Plexiglas into successive slices of 0.5 cm. For the first scenario, the procedure was repeated in order to ensure the reproducibility of the phenomena. The detector used for the determination of the gamma activities was a Packard Minaxiy Auto-gamma 5000 Series with an efficiency of 13.6 % for the ^{137}Cs . The counting time was set to 10 minutes in order to ensure a statistical error lower then 5 %.

3. Results

The radioactivity profiles obtained respectively after elution of 94, 106, 158 and 219 void volume equivalents for the first scenario are reported in figure 2.

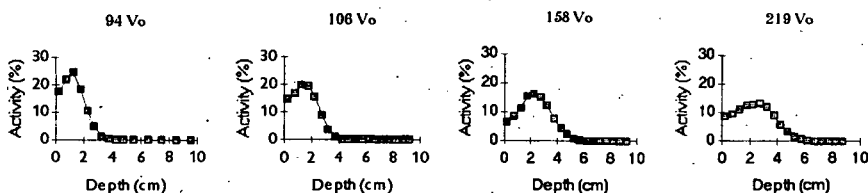


Fig.2 : Profiles of the radiocaesium activities in the soil columns after elution of 94, 106, 158 and 219 void volumes (lowest ions concentration scenario).

The activities of the soil layers are expressed as a percentage of the total activity of the soil column. The depth of penetration of the peak varied from less than 1.4 cm for an elution of 94 void volumes of solution to 2.9 cm for 219 volumes. If we plot these points (+ other points) on

a graph (fig.3) representing the position of the peak as a function of the total eluted solution (in void volumes), one can observe that the phenomena is linear (regression coeff. $r^2 = 0.973$).

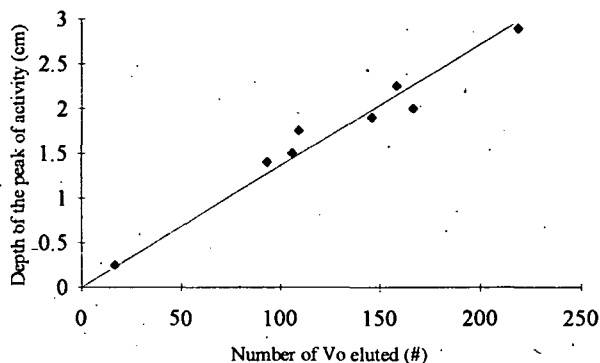


Fig. 3 : Depth of penetration of the maximum of activity expressed as a function of the number of eluted volume for the lower ion concentration scenario.

As it was expected, the three scenario's give three different migration velocities observed by a higher penetration depth of the activity with the increasing total ionic concentration for a same number of elution volumes.

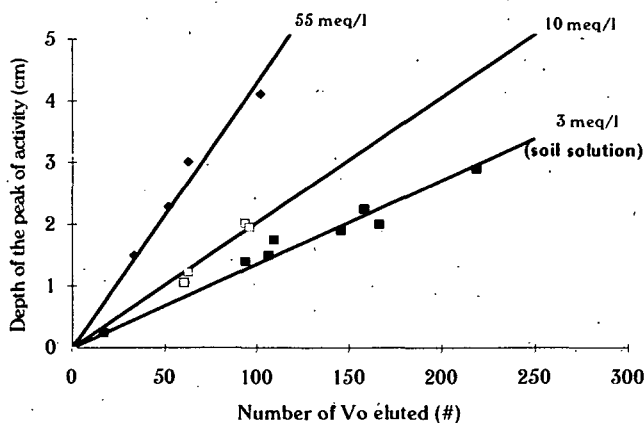


Fig. 4 : The depth of the maximum of activity observed in the soil profile expressed as a function of the total amount of Vo solution having been passed through the soil for the 3 scenario's.

The migration velocities of the caesium, estimated by the slopes of the lines (fig.4), can be related to that of the eluant. The caesium relative 'velocity' (V_r) amount to $0.135 \pm 0.008 \%$ of that of the eluant for the lowest ionic strength scenario (equivalent to the soil solution) and shows increasing relative rates for the two other solutions with a higher ionic concentrations, respectively 0.203 ± 0.014 and $0.429 \pm 0.034 \%$.

4. Discussion

The observed relative migration 'velocity' V_r of the radioisotope is a function of the ratio of the sum of the concentrations (expressed in mM) of potassium, calcium and magnesium divided by the total amount of potassium:

$$V_r \propto f\left(\frac{[K^+] + [Ca^{++}] + [Mg^{++}]}{[K^+]}\right) \quad (1)$$

For the simplicity of the following discussion, let us rename the ratio of the concentrations by the letter a , so that equation (1) can be rewritten as :

$$V_r \propto f(a) \quad (2)$$

Of course V_r is also dependent of other parameters as the PAR value, the ammonium status of the soil or the fact that the experience was carried out under fully saturated conditions. Nevertheless, semi-quantitative information can be extracted from our experience if we consider the ratio of the relative 'velocities' of two different scenario's conducted in the same experimental conditions. Doing that, the dependence of the other parameters is cancelled and the following relationship must than be verified :

$$\frac{V_i}{V_j} = \frac{\left(\frac{[K^+] + [Ca^{++}] + [Mg^{++}]}{[K^+]}\right)_i}{\left(\frac{[K^+] + [Ca^{++}] + [Mg^{++}]}{[K^+]}\right)_j} = \frac{a_i}{a_j} \quad (3)$$

where i and j represent the values obtained for two different scenario's. The relation (3) can be re-expressed in the following form :

$$\frac{V_i}{a_i} = \frac{V_j}{a_j} = \text{Constant} \quad (4)$$

V_n and V_r are the experimental results while the different concentrations are well defined from the beginning of the experience. The knowledge of only one value of V_r induced thus automatically the knowledge of any other values if we stay in the same experimental protocol.

In this study, the observed V_r ratio of 1.504 ($=0.203/0.135$) obtained between the two first scenario's was in very good agreement with the 1.5 expected value of the ratio of a (3 divided by 2), while the accuracy of the correspondence for the second and the third scenario's with the experimental result was slightly less good (2.113 to compare to 2.0). Between the first and the third scenario's, the 'predicted' ratio must be a factor of 3.0 to compare with the obtained value of 3.178. The 6 % error observed can easily be explained by the sum of the experimental errors due to : the soil core slicing, the detection of the activity, the linear regression, the truncation errors... The values obtained for the constants in the three scenario's were respectively 0.068, 0.068 and 0.072 giving an average value for a equal to 0.069 ± 0.002 . For these experimental conditions, the equation (4) gives :

$$V_r = 0.069 \cdot a = 0.069 \cdot \frac{[K^+] + [Ca^{++}] + [Mg^{++}]}{[K^+]} \quad (5)$$

Using the PAR concept, we can rewrite the expression (5) as :

$$V_r = 0.069 \cdot \left(1 + \frac{[K^+]}{PAR^2}\right) \quad (6)$$

Knowing that the PAR value was kept to 1.0 in this work, equation (6) gives :

$$V_r = 0.069 \cdot (1 + [K^+]) \quad (7)$$

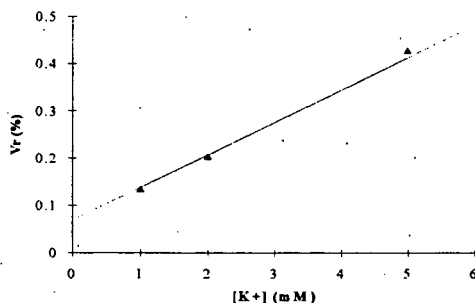


Fig. 5 : Relative 'velocity' V_r as a function of the potassium ion concentration.

The points on Fig.5 represents the observed relative 'velocities' for the three scenario's while the line is the results of the equation 7. Care must be taken with this equation when extrapolation to the $[K^+] \rightarrow 0$ is made. A potassium concentration equal to zero is not allowed (because it is then impossible to obtain a PAR value equal to 1.0) and must be rejected. Furthermore, on the base of our experiment, not data are available to ensure a linearity for the higher potassium concentrations.

We know today that the caesium sorption and retention (as well as the potassium ones) onto the soil solid matrix are governed by different kind of fixation sites (*Cremers et al., 1988*). Some of them are very specific for the considered elements, some are less specific while others are not specific at all but they can be filled by cations as Ca^{++} or Mg^{++} . Furthermore, the bivalent cations which are more hydrated (bigger in size), can play an role by reducing the access of the monovalent ions to the specific and very specific sites. It is thus logical to postulate a relationship (equation 1) between the respective concentrations of K^+ , Ca^{++} and Mg^{++} in the soil solution and the observed migration. For a full quantitative approach, the equation (1) must be adapted in order to take into account the variation of the Potassium Adsorption Ratio, the fact that the experience was carried out under saturated conditions and the concentration of the NH_4^+ , which can induce collapses of some clay mineral (and thus a trapping of the caesium ions (*Jacobs, 1963*)). Including those corrections, the final equation will probably be able to explain partially the well known "apparent migration velocity" parameter used by most of the modelers in the different versions of the convective-diffusional models.

5. Conclusions

In the experimental conditions described above (a soddy posolic soil well homogenised, suppression of the biological components, work under saturated conditions and Potassium Adsorption Ratio kept to a constant value, ...), it is possible to make a semi-quantitative analyse of the vertical migration of radiocaesium down to the soil profile. The movement is linear with respect to the volume of eluted solution and is directly correlated with the main cations concentrations. Furthermore, if the vertical movement is experimentally quantified for one scenario, it is possible to forecast the migration for other ionic composition of the soil solution on the base of a very simple equation. The migration is clearly controlled by the

potassium ion concentration but the contribution of the bivalent cations can not be neglected. In order to be more realist and relevant for the radionuclide migration modelling purpose, the proposed equation must still be adapted for a variation of the :

- PAR value.
- experimental conditions (effect of the dry-wet cycling), and
- retention properties of the soil as a function of the NH_4^+ concentration.

Acknowledgement

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KINETICS OF RADIONUCLIDE LEACHING FROM FUEL PARTICLES IN THE SOIL AROUND THE CHERNOBYL NUCLEAR POWER PLANT

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Abstract. Empiric models of radionuclide leaching from fuel particles of the Chernobyl origin are presented. Modelling is based on experimental data for Sr-90 chemical forms in soils at various distances and directions from the Chernobyl nuclear power plant. Method of indirect assessment of Sr-90 portion incorporated in fuel particles is suggested. Dependence of leaching rate on directions and distances as well as on soil properties are analyzed.

1. Introduction.

During the explosion and the fire at the fourth unit of the Chernobyl Nuclear Power Plant in April-May 1986 a great amount of dispersed nuclear fuel (fuel hot particles), structural materials, and substances dumped into the reactor and formed in it (condensation hot particles) was released into the atmosphere. The fuel particles were of dense or loose structure and consisted of uranium oxides. Their sizes ranged from hundreds of microns to fractions of a micron. The radionuclide composition of the fuel particles was similar to the fuel make-up in the damaged unit with some depletion of volatile nuclides (134 , ^{137}Cs , ^{106}Ru etc.). Fuel particles account for more than 90% of the total amount of hot particles. The condensation particles were generally characterized by smaller size and regular form. They can include either a wide spectrum of radionuclides or 1 to 2 radionuclides (for example $^{134,137}\text{Cs}$; ^{106}Ru ; $^{144}\text{Ce} + ^{95}\text{Zr}$; $^{144}\text{Ce} + ^{106}\text{Ru}$ etc.). Release of these fuel and condensation particles into the environment was the main distinguishing feature of the accidental contamination following the Chernobyl accident.

Condensation particles formed after the Chernobyl accident are similar to global fallout resulting from previous atmospheric nuclear tests. Therefore, the behaviour of these particles

in the environment can be predicted with fair reliability. At the same time, the behaviour of fuel particles deposited mainly in 60-km zone around the Chernobyl nuclear power plant is not known, and therefore presents a serious scientific problem *Konoplev et al., (1992)*.

2. Calculation of the fraction of Sr-90 in particles from Sr-90 occurrence forms in the soil.

Studies show that the fuel particles of the Chernobyl origin do not dissolve in neutral solutions and as a result in the first years after the accident the fraction of the exchangeable form of radionuclides (determined by extraction with 1 M ammonium acetate) was much lower in the soils from the immediate vicinity of the Chernobyl NPP as compared to the soils to which radionuclides entered in the soluble state. This difference in the exchangeable form fraction can become a basis for quantification of the fraction of radionuclides occurring in the soil as part of fuel particles. In this respect the most suitable radionuclide for studies is Sr-90. This radionuclide has a long half life which permits changes in its speciations occurring in the soil to be traced during a certain time period. It takes radiostrontium to reach an equilibrium several days in soil. Thus, speciations of Sr released from fuel particles can be considered equilibrium. What is more, the equilibrium fraction of exchangeable radiostrontium is close to 100% in most soils and is weakly dependent on soil properties.

The fraction of Sr-90 occurring as part of fuel particles was calculated for several points on the Chernobyl contaminated area with the following equation:

$$F_t = 1 - \frac{EX_t}{EX_{\infty}} \quad (1)$$

where F_t is fraction of Sr-90 occurring as part of fuel particles at time moment t ; EX_t and EX_{∞} are fractions of exchangeable Sr-90 at time moment t and at equilibrium, respectively.

There are several methods to determine the radionuclide fraction occurring in the soil in exchangeable form. These methods differ by composition of extracting solution, extraction time and ratio of the volume of extracting agent to the soil weight. Data used for calculation of F_t can be obtained by any of these methods provided that EX_t and EX_{∞} are determined with the same method. The data used in this paper were obtained by the method of extraction of 1M solution of ammonium acetate at phases ratio of 1:8 and extraction time of 1 day.

The quantity EX_{∞} can be found in three different ways. The most accurate of all is to apply a tracer radionuclide, for example Sr-85, to the soil under study. Another option is to determine EX_{∞} in the lower soil layers in which fuel particles do not occur.

In the initial time period when the value of EX_t is far from equilibrium, it may be

sufficient to use expert judgment of EX_{∞} . For example, for soddy-podzolic soils, prevailing in the contaminated area and for other acidic soils with low organic content the value of EX_{∞} varies from 70 to 90% according to *Pavlotskaya (1974)*. Thus, using $EX_{\infty} = 0.8$ as an expert judgment more results in a mistake in calculating F not more than 10% if F_i is more than 0.5.

3. Verification of suitability of different mathematical models to describe radionuclide leaching from fuel particles.

Modelling processes leading to release of radionuclides from fuel particles is not easy because of non-uniformity in size, forms and chemical nature of these particles. Therefore, it seems reasonable to use integral parameters describing leaching rate of radionuclides in different areas of the contaminated zone. There are two such terms being used in the literature: radionuclide leaching rate $\frac{dF}{dt}$ (day⁻¹), where F is fraction of radionuclide incorporated in fuel particles, and uranium oxide matrix dissolution rate $\frac{dP}{dt}$ (g/day), where P is particle weight. In this case, the reduction with time of fraction of radionuclides in particles is described by the following equations:

$$\frac{dF_i}{dt} = -k_i F_i \quad (2)$$

and respectively

$$F_i = F_0 * \text{EXP}(-k_i t) \quad (3)$$

where F_i and F_0 are fractions of radionuclides incorporated in fuel particles at moment t and in depositions, respectively; k_i is first order rate constant of radionuclide leaching from fuel particles, t is time after the accident.

If the particles have approximately the same size and resistance to dissolving, the leaching rate constant will grow with time due to an increase in the ratio of surface area of particles to their weight. In this case, it is better to use as a parameter the dissolution rate normalized to surface area of particles v (g/cm²day):

$$v = -\frac{1}{S} * \frac{dP_i}{dt} \quad (4)$$

where P_i is particle weight at time moment t ; S is surface area of particle.

Assuming that the radionuclides are uniformly distributed over the particle volume and taking

into account that $\frac{dP_i}{dt} = \rho S \frac{dR_i}{dt}$ and $\left(\frac{F_i}{F_0}\right)^{\frac{1}{3}} = \frac{R_i}{R_0}$, where R_i and R_0 are current and initial.

$$\left(\frac{F_t}{F_0}\right)^{\frac{1}{3}} = 1 - \left(\frac{\nu}{\rho R_0}\right) t \quad (5)$$

On a semilogarithmic scale, time dependence of Sr fraction in fuel particles should be linear, given leaching goes at a rate described by Eq. 3

$$\ln F_t = \ln F_0 - k_1 t \quad (6)$$

If Eq. 5 is valid, time dependence of $(F_t/F_0)^{1/3}$ should be linear.

Considering significant non-uniformity in depositions of fuel particles in the zone near the Chernobyl NPP, the agreement between the experimental and theoretical dependencies may be considered as satisfactory for both models. An appreciable deviation from the linear dependence is observed at one site only. Eq. 5 gives a better agreement of the calculations and experimental data. This is also indicated by the linear regression equations derived for Eq. 5 and 6 (see Table 1). Correlation coefficients are higher for Eq. 5 except two cases when they were the same.

Table 1. Linear regression equations for Eq.5 and 6.

Soil	Linear regression equations	
	Equation 5	Equation 6
Acid meadow alluvial Site 1	$(F_t/F_0)^{1/3} = (1.01 \pm 0.04) - (1.3 \pm 0.3) \cdot 10^{-4} t$, R=0.95	$\ln F_t = (1.3 \pm 0.2) \cdot 10^{-2} - (4.6 \pm 1.2) \cdot 10^{-4} t$, R=0.93
Site 2	$(F_t/F_0)^{1/3} = (1.01 \pm 0.02) - (2.4 \pm 0.1) \cdot 10^{-4} t$, R=0.996	$\ln F_t = (0.06 \pm 0.15) - (9.7 \pm 1.2) \cdot 10^{-4} t$, R=0.98
Site 3	$(F_t/F_0)^{1/3} = (1.11 \pm 0.13) - (3.9 \pm 1.1) \cdot 10^{-4} t$, R=0.93	$\ln F_t = (0.7 \pm 1.0) - (2.2 \pm 0.8) \cdot 10^{-3} t$, R=0.88
Acid sod alluvial	$(F_t/F_0)^{1/3} = (1.00 \pm 0.06) - (1.3 \pm 0.5) \cdot 10^{-4} t$, R=0.89	$\ln F_t = -(0.06 \pm 0.01) - (4.4 \pm 0.2) \cdot 10^{-4} t$, R=0.89
Cultivated sod podsollic	$(F_t/F_0)^{1/3} = (1.00 \pm 0.01) - (5.1 \pm 0.7) \cdot 10^{-5} t$, R=0.98	$\ln F_t = -(2.8 \pm 2.7) \cdot 10^{-2} - (1.5 \pm 0.2) \cdot 10^{-4} t$, R=0.98

The validity of Eq.5 is also supported by good agreement in dissolution rate constants calculated by the change of Sr speciations in the soil and determined in the laboratory experiment in which the fuel particles collected in the "near zone" of the Chernobyl NPP were exposed to solutions simulating water media. Values of ν determined in laboratory

ranged from $5.7 \cdot 10^{-6}$ to $1.6 \cdot 10^{-4}$ g/(cm² day) and by using kinetics of Sr-90 speciations in the field, changed within $(0.1-1) \cdot 10^{-5}$ g/(cm² day) for particles of 10 µm and $(0.1-1) \cdot 10^{-4}$ g/(cm² d) for particles of 100 µm.

4. Relationship between leaching rate and direction towards and distance to the Chernobyl NPP.

Rate of leaching of radionuclides from particles depends on properties of the medium they occur in and characteristics of the particles themselves. For example, in the laboratory experiment the fuel particles collected in the "near" zone of the Chernobyl NPP were exposed to solutions simulating natural water media to determine dissolution rate and it was found to vary within two orders of magnitude - from $5.7 \cdot 10^{-6}$ to $1.6 \cdot 10^{-4}$ g /cm² day; Bogatov et al., (1990).

At the initial stage of the accident the radioactive materials released from the reactor were transported north and north-west and later-west, south, east and north-east. As the condition of the reactor did change at that time it may be expected that characteristics of deposited particles are different in different directions. Besides, the average size and density of particles is obviously decreasing at a distance from the source. Therefore, the effect of properties of particles on leaching rate of radionuclides can be analyzed by comparing rate constants determined in different points of the contaminated area. By the classification proposed in Bogatov et al. (1990) the region contaminated after the Chernobyl accident can be divided into three sectors: a west sector (north-west, west and south west), a south sector (south, south-east and east) and a north sector (north and north-east).

The data show that west and south-west of the Chernobyl NPP the leaching rate constants vary from $3 \cdot 10^{-5}$ day⁻¹ to $1.1 \cdot 10^{-4}$ day⁻¹, the average being $6.6 \cdot 10^{-5}$ day⁻¹ and north-west of the NPP they ranged from $1.1 \cdot 10^{-4}$ day⁻¹ to $7.9 \cdot 10^{-4}$ day⁻¹ with the average of $3.1 \cdot 10^{-4}$ day⁻¹. In these directions, no relation is noticed between distance to the station and leaching rate constant, while in the south and south-east directions the trend for increase in leaching rate constant with increase in distance to the Chernobyl NPP is obvious. At distances up to 5 km, the average leaching rate constant is $(2.1 \pm 1.4) \cdot 10^{-4}$ day⁻¹ and at 5 km and more - $(8.9 \pm 3.7) \cdot 10^{-4}$ day⁻¹. Dissolution rate constants show a similar pattern.

Since the soil type in the 30 km zone is more or less the same, it may be assumed that particles of higher resistance to leaching of radionuclides were deposited north, north-west, west and south-west of the Chernobyl NPP and in the vicinity of the NPP, south and south-east of it where leaching rate constants varied from 10^{-5} to 10^{-4} day⁻¹. As one moves south of the NPP, rate constants grow and become 10^{-3} day⁻¹ at a distance of 20 km. The relationships of rate of Sr-90 leaching from particles are different in the west and south sectors, probably, because in the west sector within the "near zone" where the gamma background level was

more than 2 mR/h on the 15th day after the accident, the Sr-90 to Zr-95 enrichment coefficient of deposited particles changed insignificantly with distance: from 0.4 to 0.6, while in the south sector this coefficient increased from 0.4 in the immediate vicinity of the plant where the gamma radiation level is more than 10 mR/h to 3.2 in the area with the gamma radiation level from 2 to 10 mR/h. All this suggests that characteristics of fuel particles changed with distance more drastically in the south direction than in the west.

Outside the 30 km zone, leaching rate constants are expected to be higher because of small size of particles. For example, in the soils of the Bryansk region (Russia) the first determination of speciations was carried out in 1987. The Sr-90 speciations were then found to be practically at equilibrium and, hence, the leaching rate constant in this region is at least $5 \cdot 10^{-3} \text{ day}^{-1}$. A similar conclusion may be made for the regions of Belarus lying at a distance 200-250 km from the ChNPP where Sr-90 speciations in 1987 were close to equilibrium too.

So, depending on characteristics of fuel particles the rate of leaching of radionuclides from these particles in natural conditions may differ by several orders of magnitude.

5. Relationship between constants of radionuclide leaching from fuel particles and soil properties.

In the previous section it was shown that rate of radionuclide leaching from fuel particles is essentially dependent on characteristics of particles themselves. Therefore, the effect of soil properties on rate of particles break-down can be studied on experimental plots on which different soil types occur as close to each other as possible thus minimizing differences in nature of hot particles causing contamination in a particular point.

This requirement is best met by experimental grounds near Benevka, 10 km away from the Chernobyl NPP and Kopachi, 5 km from the Chernobyl NPP. The largest distance between the sampling points on these grounds was about 0.5 km and they differed in both soil type and soil water content since they were located on the slope at varying distance from Pripyat.

For the soils of the Benevka grounds some soil properties have been determined. Specifically in alluvial soils, the humus content is 1-7% for soddy soil and 5-13% for meadow soil, the value of pH is 5-6 in water suspension for soddy soil and 4-5 for meadow soil, the capacity of cation exchange - 3-12 meq/100 g for soddy soil and to 33 meq/100 g for meadow soil. In soddy-podzolic soil the humus content was 4.0; pH was 5.8 and the capacity of cation exchange - 9.4 meq/100 g.

Comparison of rate constants determined in different points is not indicative of any well-defined dependence of leaching rate constants on soil type, soil pH, humus content and capacity of cation exchange. This may be caused by the difficulty to identify the role of soil properties against the effect of nature of particles and non-uniformity of their surface

distribution on leaching rate. Another reason for it may be a weak dependence of leaching rate on properties of the medium they occur in. This may be the case, for example, when the release of radionuclides from particles is determined primarily by processes in particles themselves such as those associated with radioactive decay of isotopes in particles. In any case possible effects of the medium properties on destruction rate of particles can, probably, be studied only in laboratory experiments.

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FACTORS AFFECTING CAESIUM FIXATION IN UPLAND ORGANIC SOILS

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Abstract

A new sequential sorption method is presented which determines the total number of potential Cs-fixing sites in acid organic soils, characteristic of those contaminated in upland areas of the UK following the Chernobyl accident. The total number of potential Cs-fixing sites in the upland soils exceeded those present in lowland mineral soils studied elsewhere. However, although a soil may have the potential to fix Cs ions, evidence is presented that a minimum concentration of Cs⁺ (0.60 to 0.75 mM) is required before Cs fixation occurs, and that this is unlikely to be reached in any contaminated soil. The observed Cs⁺ fixation in lowland mineral soils and its absence from acidic upland soils is explained by the action of K⁺ ions, which can also induce interlayer collapse. Although Cs⁺ ions are unlikely to be fixed in acid organic soils, they can be strongly sorbed on any unoccupied Cs-specific sorption sites, located in the narrow parts of illitic wedge zones. Cs⁺ ions sorbed on such sites are not susceptible to exchange by other ions, but can be desorbed if the concentration of Cs⁺ in solution is decreased. Thus, radioactive Cs in such soils will remain available for plant uptake, unless interlayer collapse can be induced.

1. Introduction

Radioactive caesium (Cs), deposited on upland areas of the UK following the Chernobyl accident in 1986, is continuing to cycle through the soil-plant-animal system. Sheep grazing some of these areas are still accumulating sufficient radiocaesium to prompt restrictions on their movement and sale (MAFF, 1995). It was thought initially that, due to the large organic matter content of the contaminated soils, Cs fixation was prevented by the lack of sufficient quantities of Cs-fixing clay minerals (Livens & Loveland, 1988). However illite, a Cs-fixing clay mineral, has been found in the samples analysed (Hird *et al.*, 1995). In this paper

determination of the Cs fixing potentials of some acid organic soils is reported and factors which affect the realisation of these potentials are considered. Reasons for the continued cycling of radioactive Cs are discussed in light of the results obtained.

2. Materials and Methods

Samples were collected from three sites in upland areas of Cumbria (Table 1). The surface organic-rich horizon and the underlying mineral horizon were sampled at Ennerdale (Harthope series) and Linbeck Gill (Moor Gate series). A surface (0 - 10 cm depth) sample only was taken at Corney Fell (Winter Hill series), where the soil is a deep peat. Samples were not dried, but were sieved (< 4 mm) and stored in a field-moist condition at 3°C until required. Some properties of these soils are given in Tables 1 and 2.

Table 1. Some properties of the soils

Soil series (grid reference)	Soil type	Horizon	pH (water)	Organic matter content (g kg ⁻¹)	CEC (mmol kg ⁻¹)	Exch. K (mmol kg ⁻¹)
Harthope (NX065127)	Ferric stagnopodzol (Histic Epiaquod)	O	4.3	571	166.3	0.8
		Eag	4.3	96	128.1	0.5
Moor Gate (NX154967)	Humic brown podzolic soil (Entic Haplorthod)	Ah	4.5	205	97.7	2.2
		Bh	4.5	131	46.9	1.2
Winter Hill (NX150896)	Raw oligofibrous peat (Typic Borofibrst)	O	4.3	713	221.7	0.4

¹ Avery (1980) and equivalent classification according to US Soil Taxonomy (Soil Survey Staff, 1992)

Table 2. Radioactive Cs and clay contents of the soils

Soil		¹ Caesium content (Bq kg ⁻¹)		Clay content (g kg ⁻¹)		Illite content (g kg ⁻¹)	
		¹³⁷ Cs	¹³⁴ Cs	² standard	³ buffered	standard	buffered
Harthope	O	245.0	2.4	90.6	407.7	61.6	318.0
	Eag	18.3	0.8	317.2	317.2	244.2	276.0
Moor Gate	Ah	156.6	6.8	115.6	139.5	3.5	22.3
	Bh	19.3	1.6	83.0	106.1	1.7	9.7
Winter Hill	O	129.2	4.1	59.0	nd	15.3	nd

¹ Determined October 1992; ² using standard H₂O₂ method of digestion; ³ using buffered H₂O₂ method of digestion (Hird *et al.*, 1994); nd - not determined

2.1 Batch-equilibrium Cs sorption and desorption

Cs sorption was carried out in the presence of silver thiourea (Ag(TU)₄⁺) ions, which are

sorbed on most of the cation exchange sites, but excluded from Cs-specific sites in clay interlayers (Cremers *et al.*, 1988). Added Cs^+ ions are, therefore, sorbed mainly on specific sites. Samples of moist soil (1 g) were first saturated with $\text{Ag}(\text{TU})_4^+$ by washing the soil in two 20 ml aliquots of 0.015 M $\text{Ag}(\text{TU})_4^+$. CsCl solution (30 cm^3), ranging between 0.0 and 4.0 mM, was then introduced to each soil sample in a background solution of 0.015 M $\text{Ag}(\text{TU})_4^+$, to ensure that non-specific sites remained saturated with $\text{Ag}(\text{TU})_4^+$. The suspensions were shaken overnight, centrifuged and the supernatant solutions analysed for Cs using AAS. Desorption was carried out by removing most of the supernatant solution, and determining the volume of CsCl remaining. The solids were resuspended in 30 cm^3 0.01 M KCl , in a background solution of 0.015 M $\text{Ag}(\text{TU})_4^+$. The suspensions were shaken for 16 h and the supernatant solution analysed for Cs using AAS. The amount of Cs^+ desorbed was determined (allowing for the amount of CsCl solution remaining in the moist soil plug). Although three replicates were measured in each case, as the equilibrium solution concentration is dependent on the amount sorbed, it was not possible to take means and estimate errors for sorption and desorption data. Consequently, each result is reported separately.

2.2 Sequential Cs sorption

Moist soil (1 g), saturated with $\text{Ag}(\text{TU})_4^+$, was suspended in 30 cm^3 of 0.007 mM CsCl solution (in a background solution of 0.015 M $\text{Ag}(\text{TU})_4^+$). The suspension was shaken overnight (16 h) and the supernatant solution removed and analysed for Cs using AAS. The procedure was repeated introducing increasing concentrations of CsCl to the same soil sample until no further increase in the amount of Cs^+ sorbed was detected. The total amount of Cs^+ sorbed on the soil was plotted against the concentration of Cs^+ in solution to obtain a sequential sorption isotherm. The experiment was conducted in triplicate.

3. Results and discussion

3.1 Batch-equilibrium method for determination of Cs-specific sites

A typical batch-equilibrium sorption isotherm is shown in Fig. 1a. Cs^+ sorption increased with solution Cs^+ concentration until the concentration reached approximately 1.5 to 2.5 mM. In this range, Cs^+ sorption fell to varying degrees before increasing again. As no sorption maximum was obtained, the total number of Cs-specific sorption sites could not be determined. Despite this, the amount of Cs adsorbed on the Cumbrian soils already greatly

exceeded the number of Cs-specific sorption sites present in four lowland mineral soils (Cremers *et al.*, 1988), which were in the range 0.58 to 2.78 mmol kg⁻¹. Desorption experiments confirmed that the large numbers of specific sites determined for the Cumbrian soils were genuine, and not simply the result of Cs⁺ ions sorbed on cation exchange sites not blocked by silver thiourea (Hird *et al.*, 1995).

It is proposed that, due to the introduction of relatively large concentrations of Cs⁺, the batch-equilibrium method (Cremers *et al.*, 1988) determines not only the number of Cs-specific sorption sites available, but also the number of sites present within expanded clay interlayers which are not initially specific for Cs⁺ ions, but which have the potential to specifically sorb and subsequently fix Cs⁺ ions if collapse is induced. In other words this method determines the Cs-fixing potential of a soil. Decreases in the sorption isotherms, and the absence of sorption maxima are thought to be due to the occurrence of edge interlayer trapping (Hird *et al.*, 1995) where collapse of the outer edge of an interlayer occurs before sites in the still expanded middle region are fully occupied. Reasons for the absence of edge-interlayer trapping and relatively small Cs-fixing potentials found in lowland soils are given elsewhere (Hird *et al.*, 1995)

3.2 Sequential method for determination of total Cs-fixing potentials

Sequential additions of small concentrations of Cs⁺ ions to the same soil sample enables all interlayer sites to be occupied by Cs⁺ ions without inducing edge-interlayer trapping. Fig. 1b shows that, in the presence of Cs⁺ concentrations up to approximately 1 mM, the amount of Cs⁺ sorbed (when introduced sequentially) was similar to that sorbed when Cs⁺ was introduced in batches. Above 1 mM, the amount of Cs sorbed dramatically increased before reaching a sorption maximum. To ensure that the large amounts of Cs sorbed were located on potential Cs-fixing sites, and not simply the result of Cs⁺ ions sorbed on cation exchange sites not blocked by silver thiourea, desorption of the Cs⁺ ions was carried out by shaking in two successive 30 cm³ aliquots of 0.01 M KCl (in a background solution of 0.015 M Ag(TU)₄⁺). Total Cs-fixing potentials of the Cumbrian soil samples studied were taken, therefore, as the amount of Cs⁺ remaining sorbed after desorption by KCl (Hird *et al.* 1995). These varied between 29.6 mmol kg⁻¹ for the Moor Gate Bh soil and 152.5 mmol kg⁻¹ for the Winter Hill soil. It is proposed that sorption maxima, obtained in the sequential sorption isotherms, are a more accurate measure of the total potential of a soil (especially acid organic

soils) to fix Cs^+ than the batch-equilibrium method previously developed (Creemers *et al.*, 1988), which is only applicable for soils not prone to edge-interlayer trapping.

All of the Cumbrian soils studied possessed the potential to fix much more Cs than that deposited following the Chernobyl accident (Table 2). However, as indicated by the continued uptake of radiocaesium by sheep grazing contaminated pasture, conditions necessary for the realisation of this potential are not being satisfied in the field.

3.3 Factors affecting interlayer collapse and Cs fixation

Fig 2 shows the results of a batch-equilibrium sorption and desorption experiment conducted on the Harthope Eag soil using low concentrations of Cs. When extremely small amounts of Cs^+ (up to 0.1 mmol kg^{-1}) were sorbed on this soil, the desorption results indicated that approximately 9% of the total number of Cs^+ ions sorbed were sorbed on non-specific cation exchange sites not blocked by $\text{Ag}(\text{TU})_4^+$. However, when the amount of Cs^+ sorbed on this soil was between 0.1 and 10 mmol kg^{-1} , the proportion susceptible to desorption rose to 80% before falling back to 20%. The same trend was shown in the Moor Gate Ah sample, where 26% of the sorbed Cs^+ ions were initially sorbed on sites not blocked by $\text{Ag}(\text{TU})_4^+$. As the amount of Cs^+ sorbed on the soil increased, the proportion susceptible to desorption rose to 100% before falling back down to around 30% again.

When Cs^+ ions are added to a soil containing illite, they initially occupy specific sorption sites in the narrow parts of the wedge zones and are not readily accessible to exchange by K^+ ions, although they are still potentially desorbable as interlayer collapse has not yet been induced. As the concentration of Cs^+ added is increased and Cs^+ ions occupy sites in the more expanded parts of the wedge zones, a greater proportion become susceptible to exchange by K^+ ions, as shown by an increase in the proportion of Cs^+ desorbed (Fig. 2b). However, there is a point beyond which there is a decrease in the proportion of Cs^+ susceptible to exchange by K^+ (Fig. 2b). We interpret this as being due to the collapse of part of the interlayer, because the amount of Cs^+ sorbed in the wedge zones is sufficiently great to induce it. After this has occurred, Cs^+ ions which once occupied sites in the wide parts of the wedge zones become part of the collapsed micaceous core of the mineral and are no longer susceptible to exchange or desorption, i.e. become fixed. As a result, only Cs^+ ions that are present on non-specific cation exchange sites not blocked by $\text{Ag}(\text{TU})_4^+$ can be desorbed.

For all of the soils, the total amount of Cs^+ sorbed just before interlayer collapse

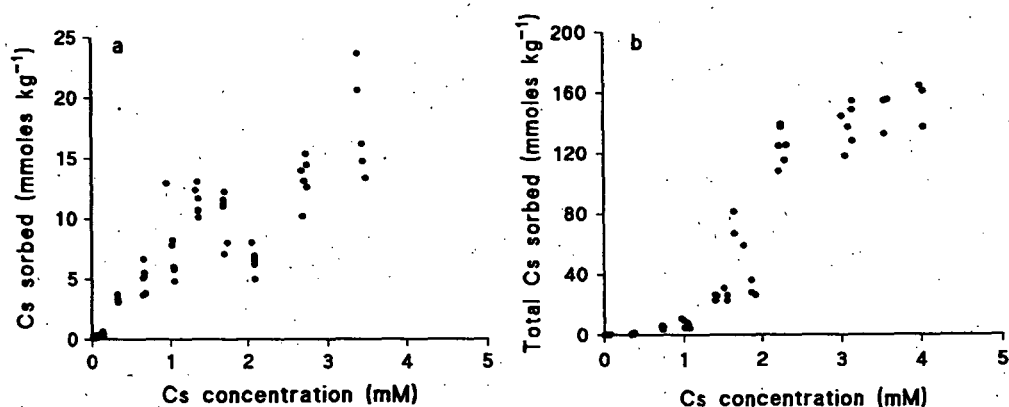


Fig. 1. Cs sorption isotherms in the presence of silver thiourea for the Winter Hill soil: (a) batch-equilibrium method; (b) sequential method.

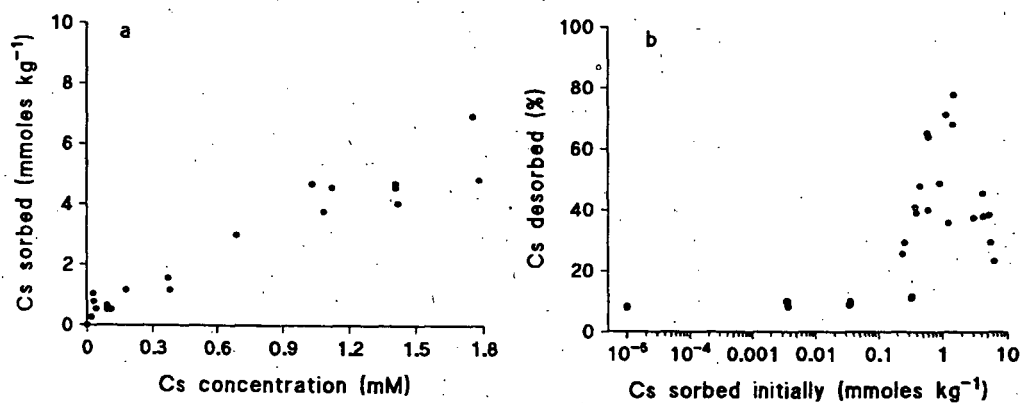


Fig. 2. (a) Batch-equilibrium Cs sorption isotherm in the presence of $\text{Ag}(\text{TU})_4^+$ for the Harthope Eag soil sample and (b) proportion of sorbed Cs^+ ions susceptible to desorption by 0.01 M K^+ .

occurred varied over an order of magnitude. This is because the number of specific sites depends on the number of wedge zones and hence on the amount of illite present. However, irrespective of the soil sample investigated, the concentration of Cs^+ present in solution when interlayer collapse occurred lay within a narrow concentration range, between 0.60 and 0.75 mM. Thus it is the concentration of Cs^+ in the vicinity of the wedge zones that determines whether or not interlayer collapse and subsequent Cs^+ fixation occurs.

It should be noted, however, that for one of the soils investigated (Winter Hill) the vast majority (greater than 90%) were susceptible to desorption by K^+ ions, even when relatively small amounts of Cs^+ were sorbed. This indicated that there were few unoccupied specific sites, despite the presence of illite (Hird *et al.*, 1995). The specific sites present must already have been occupied by Cs^+ ions, which were either deposited previously or present naturally in the soil.

Soil solution concentrations of Cs are likely to be much less than the concentration (0.60 - 0.75 mM) required to bring about Cs fixation. Thus, fixation by this mechanism in both upland and lowland soils is most unlikely. However, field and experimental observations (Coughtrey & Thorne, 1983) clearly indicate that Cs^+ ions are effectively fixed in lowland mineral soils, and in limed and fertilized upland soils containing illite. Fixation, which takes place in these latter soils, but not in the soils of many semi-natural areas, is probably due to differences in the concentration of K^+ in soil solution, which is usually in the range 1 - 10 mM in mineral soils (Edmeades *et al.*, 1985) and of the order of 10^{-3} mM in unimproved upland soils (Ferguson, 1994). K^+ also induces illitic clay interlayer collapse, though not as effectively as Cs^+ (Sawhney, 1972). Such K-induced collapse is reversible on the addition of Ca^{2+} and Mg^{2+} (Sawhney, 1972), and this leads to the 'frayed-edges' of illitic crystallites, which provide the wedge zones for the specific sorption of Cs^+ . Consequently, any additions of Cs^+ will diffuse to these specific sites. Although the concentration of Cs^+ on its own is insufficient to induce collapse, collapse will be induced by K^+ . Because of the presence of Cs^+ in the inner part of the newly collapsed area, this cannot be re-expanded by the presence of Ca^{2+} and Mg^{2+} , resulting in the fixation of Cs^+ .

Although Cs^+ ions are unlikely to be fixed in acid organic soils, they can be strongly sorbed on any unoccupied Cs-specific sorption sites, located in the narrow parts of illitic wedge zones. Cs^+ ions sorbed on such sites are not susceptible to exchange by other ions, but can be desorbed if the concentration of Cs^+ in solution is decreased. Thus, radioactive Cs in

such soils will remain available for plant uptake, unless interlayer collapse can be induced.

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TIME DEPENDENT VERTICAL DISTRIBUTION OF ^{137}Cs IN AN ACID FOREST SOIL

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Abstract

Vertical migration of ^{137}Cs from the Chernobyl fallout was investigated in an undisturbed forest soil (Dystric Cambisol) under spruce. Comparing ^{137}Cs data of different layers (litter, 0-5 cm, 5-10 cm, 10-20 cm) from 1993 with those measured in 1995 reveals a decreasing trend which exceeds the effect of radioactive decay. But 9 years after the deposition event 48% of radiocaesium soil inventory derived from the Chernobyl accident remained in the litter layer of the forest soil. The vertical distribution of ^{137}Cs contamination (corrected for decay to 86-05-01) was determined as 27.8 kBq m⁻² in the litter layer, 22.8 kBq m⁻² and 4.4 kBq m⁻² in the subsequent 5 cm soil increments and 2.4 kBq m⁻² in 10-20 cm depth of mineral soil. Residence half-times were evaluated by application of a compartment model. The values increased significantly in each of the investigated layers between 1993 and 1995, in litter (5-0 cm) from 5.3 a to 7.3 a, in the Ah₁ layer (0-5 cm) from 9.6 to 12.6 a, in the Ah₂ layer (5-10 cm) from 1.6 to 2.5 a and in the A/B layer (10-20 cm) from 0.7 to 1.0 a. The results support the theory, that the binding of radiocaesium to different soil components is a rather slow process which retards the vertical migration of radiocaesium in forest soils with growing efficiency. The high mobility of caesium in the deeper soil layers partly could be interpreted as a result of cycling processes, where root uptake leads to an upward migration of cations into the living biomass of vegetation.

1. Introduction

Due to the high complexity and productivity of coenoses, forests are very sensitive to pollutants. In such natural ecosystems nutrient cycling leads to a long persistence of ^{137}Cs within the different compartments and the foodchain (Myttenaere et al. 1992). Soil conditions of forests tend to be extreme e.g. in respect to low pH-values or high organic matter contents. Such factors as well as acidification and the lack of fertilizer amendment or other cultivation actions are thought to favour vertical mobility and plant availability of radionuclides (Frissel et al. 1990). Since 1988 a monitoring program including soil, vegetation and game animals is carried out in the Weinsberger forest, a large spruce stand, which was significantly affected by the Chernobyl plume (UBA 1995). In 1993 an extended sampling and investigation program was carried out (gamma-measurements, physico-chemical soil analyses). In 1995 soil samples from the same spots were taken to reveal the time-dependence of ^{137}Cs vertical migration in forest soils.

2. Material and Methods

2.1. Site description

The study area, Weinsberger forest, is situated in Lower Austria (48°23' N, 15°03' E). Acidic brown earth soils (Dystric Cambisols), partly podsollic with mor humus types, are developed on granite. The organic matter content is very high even in greater depths. High exchangeable aluminium contents and the vertical pattern of pH-values indicate acidification.

Table 1: Physico-chemical soil characteristics of forest soils under spruce in Weinsberger forest (mean values)

	litter (5-0 cm)	Ah ₁ (0-5 cm)	Ah ₂ (5-10 cm)	A/B(10-20 cm)
bulk density (g cm ⁻³)	0.2	0.5	0.5	0.6
sand: silt: clay (%)	not analysed			
% humus	66	30	18	11
pH-value (CaCl ₂)	3.2	3.0	3.3	3.6
CEC (mval 100g ⁻¹ soil)	not analysed	33.1	24.3	19.2
base saturation (%)	not analysed	10.5	5.2	3.1
fulvic-/humic acids ratio	0.4	0.9	2.3	3.2

2.2. Sampling and measurements

Vertical distribution of $^{134/137}\text{Cs}$ was investigated in six pooled soil profiles, each consisting of 10 single cores. Besides the litter layer (which was taken according to its morphology) the profiles were divided into 3 strata with fixed sizes: 0-5 cm (Ah_1 - horizon); 5-10 cm (Ah_2 - horizon); 10-20 cm (A/B - horizon). The litter layer was collected with a frame (25 x 25 cm), the three mineral layers with an auger (10 cm diameter). Gamma-measurements ($^{134/137}\text{Cs}$) were carried out with air dried material in 1-l Marinelli beakers using Ge-Li and HPGe detectors (25-35 % efficiency). Measurement times were extended until the statistical uncertainty was below $\pm 10\%$.

^{134}Cs data can be used for the distinction between ^{137}Cs from the Chernobyl fallout and older ^{137}Cs contaminations from the global fallout (nuclear weapon tests) of the 50ies and 60ies, as ^{134}Cs is a product of neutron activation and was not developed during the atomic explosions of weapon tests. For this purpose ^{134}Cs data were corrected for decay (ref. date: 86-05-01). Taking into account the sum coincidence of ^{134}Cs γ -lines, the measured values were multiplied with a factor of 1.9 (BOSSEW et al. 1995) instead of 1.76, the isotopic ratio typical for the Chernobyl fallout in Austria ($^{137}\text{Cs} : ^{134}\text{Cs} = 1.76$, MÜCK 1988). The difference between this calculated values and the measured ^{137}Cs data (decay corrected) is attributed to depositions prior the Chernobyl contamination.

2.3. Calculations

^{137}Cs - soil inventory was determined only to a depth of 20 cm of mineral soil. In 1993 less than 3 % of the inventory were located in strata deeper than this range (Strebl et al. 1995). Residence half-times were evaluated using a compartment model (for details see Strebl et al. 1995). Model assumptions included only one single pulse deposition taking place at time $t = 0$ and immediate homogenous mixing of each layer; later inputs (litter fall, tree wash out) as well as losses (e.g. root uptake, bioturbation, leaching) were thought to influence the residence time of caesium within the investigated layers; these and other (e.g. binding by soil components) ecological processes together with physical decay were described by so-called effective loss constants ($\lambda_{\text{eff}} = \lambda_{\text{ecol}} + \lambda_{\text{phys}}$). Calculations were carried out separately for each layer and each of the pooled profiles. The yielded loss constants (λ) are transformed to resi-

dence half-times (τ) by calculating $\tau = \frac{\ln 2}{\lambda}$. Division of the thickness (cm) of the investigated layer through τ (a) can be interpreted as a velocity (migration rate in cm a^{-1}).

3. Results and discussion

The ^{137}Cs activities (kBq m^{-2} in dry matter, corrected for decay to 86-05-01) in soil samples taken in 1993 and 1995 (after 1.75 years) are shown in Figure 1. The vertical distribution is very similar with highest ^{137}Cs contents in the upper 10 cm of the soils. Nine years after the reactor accident 48 % of the soil inventory are still found in the litter layer. Due to migration and ecological loss processes (e.g. root uptake, leaching) values are always lower in the samples from the second sampling date. According to an applied t-test, the differences are not significant, but there is an evident decreasing trend.

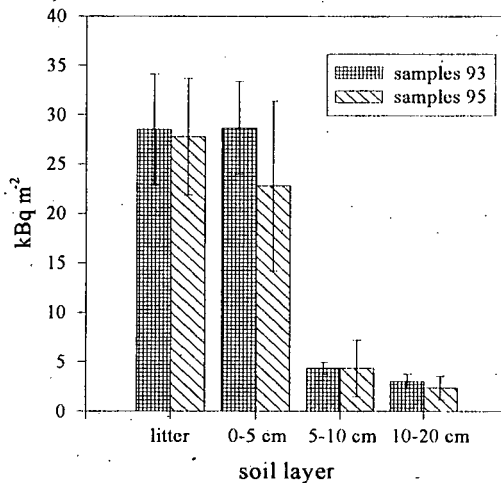


Figure 1: Vertical distribution of ^{137}Cs (kBq m^{-2} in dry matter, corr. for decay to 1986-05-01) from Chernobyl fallout in forest soil profiles from two sampling dates (each bar: $n = 6$)

In Figure 2 ^{137}Cs contents from Chernobyl fallout are compared to those from global fallout. The values are fitted by an exponential regression. Both curves yielded describe the depth-dependent decrease of activities with sufficient accuracy ($R^2 = 0.81$ for global fallout;

$R^2 = 0.92$ for Chernobyl fallout). ^{137}Cs from global fallout already penetrated deeper into the soil, this is expressed by the less steep slope of the regression line. But even this „old“ contamination shows highest values in the litter layer, which supports the finding of other authors (Schimmack et Bunzl 1994) that infiltration of radiocaesium into forest soil is a rather slow process.

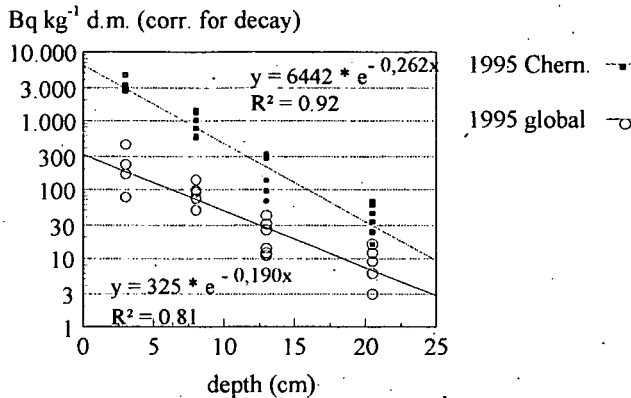


Figure 2: Exponential regression of ^{137}Cs content (Bq kg⁻¹ in dry matter) from Chernobyl and global fallout in different layers of a forest soil (data basis 1995, $n = 6$)

Residence half-times and migration rates were calculated separately for both years of observation, the results are listed in Table 2. The migration rates decrease significantly ($\alpha = 0.01$) in each of the investigated layers from 1993 to 1995. This result is in agreement with findings of other studies (Chamard et al. 1993). Trapping of Cs⁺ cations in the inter-layer binding sites of clay minerals is a nearly irreversible mechanism, but it occurs only with low probability. Formation of clay - humic substance aggregates can protect enclosed organic molecules as well as cations from degradation and leaching, respectively (Gerzabek 1994). On the basis of data from 1995 ^{137}Cs shows an average migration velocity of 0.7 cm a⁻¹ in the litter layer. The transition zone to the mineral layer (0-5 cm) seems to be an effective trap for radiocaesium, in this layer the migration rate decreases to 0.4 cm a⁻¹ corresponding to a effective residence half-time of 12.6 years (see Figure 3). In the subsequent layers radiocaesium mobility increa-

ses distinctly with migration rates of 2.1 cm a^{-1} (5-10 cm) and 11.0 cm a^{-1} (10-20 cm), resulting in low residence half-times.

Table 2: Effective residence half-times (τ_{eff}) and migration rates in different layers of a forest soil calculated with a compartment model. Comparison of data from samples 1993 and 1995 (1.75 years later) (mean values \pm standard deviation; $n = 6$)

	litter (5-0 cm)	Ah ₁ (0-5 cm)	Ah ₂ (5-10 cm)	A/B(10-20 cm)
1993				
τ_{eff} (a)	5.3 ± 0.8	9.6 ± 1.3	1.6 ± 0.3	0.7 ± 0.1
migration rate (cm a^{-1})	1.0 ± 0.1	0.5 ± 0.1	3.1 ± 0.6	14.5 ± 2.4
1995				
τ_{eff} (a)	7.3 ± 2.3	12.6 ± 3.4	2.5 ± 0.6	1.0 ± 0.3
migration rate (cm a^{-1})	0.7 ± 0.1	0.4 ± 0.1	2.1 ± 0.4	11.0 ± 2.3

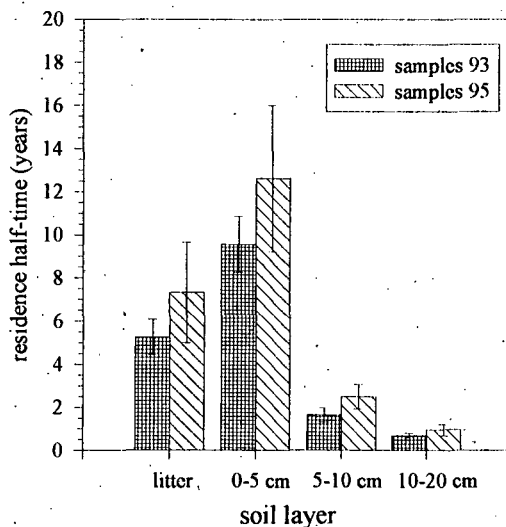


Fig. 3: Effective residence half-times of ^{137}Cs (Chernobyl) in different layers of a forest soil. Comparison of results derived from samples 1993 and 1995 ($n = 6$ each, mean values with standard deviation)

Bunzl and Schimmack (1994) found the same behaviour of ^{137}Cs in German forest soils. The rapid increase of caesium mobility in the deeper layers is rather unexpected, as the vertical changes of most of the physico-chemical properties (see Table 1) are only moderate. The most significant differences are a decrease of humus content by nearly 50% and the relative dominance of mobile fulvic acids in soil extracts ($\text{Na}_4\text{P}_2\text{O}_7 \cdot 10 \text{ H}_2\text{O}$) of humic substances from Ah₂ and A/B horizon (see Table 1). Such conditions can lead to higher plant availability and root uptake of radiocaesium, an upward transport into living biomass would enhance the migration rate, even when losses due to leaching are low.

The estimated development of ^{137}Cs vertical distribution in forest soils for 20 years after deposition is shown in Figure 4. According to the model estimates and taking into account radioactive decay, even 20 years after a fallout event 45% of the initial ^{137}Cs deposition are expected to be found in the first 10 cm of the soil profile. In forests the Chernobyl fallout contaminated the soil surface not in a single pulse (model assumption), considerable amounts of caesium were deposited on tree canopies and reached the forest floor continuously during the first years after the accident through leaf/needle litter fall and crown wash-off. Therefore, migration rates calculated for the first years after the accident may be underestimated considerably, for medium term prognosis this error becomes less important.

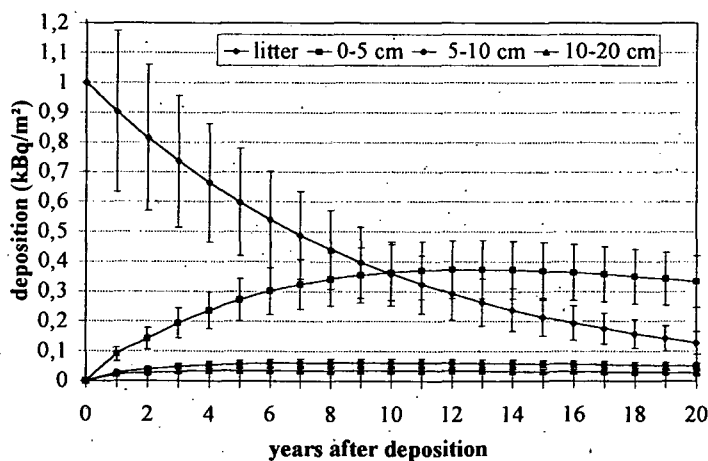


Fig. 4: Time dependent vertical distribution of ^{137}Cs in a forest soil calculated with a compartment model (data basis 1995; $n = 6$ in each layer)

4. Conclusions

Compartment models give only rough estimates of radionuclides' complex behaviour in soils. Many of the processes involved in ^{137}Cs vertical migration are not easy to be quantified, other mechanisms, like interactions with organic matter and living biomass are not yet known in full detail. In this case black-box models are appropriate tools for trend descriptions.

Living and decaying organic matter seems to be an effective storage system, the cycling of ^{137}Cs like nutrients keeps radiocaesium in the organic matter rich surface layers of forest soils and retards its vertical migration significantly. The fixation of ^{137}Cs to soil components is time dependent and seems to increase with time.

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THE USE OF MINERAL AND ORGANIC ADSORBENTS AS COUNTERMEASURES IN CONTAMINATED SOILS: A SOIL CHEMICAL APPROACH

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Abstract

A simple laboratory method is presented to estimate the effect of the addition of organic or mineral adsorbents to ^{134,137}Cs and ⁹⁰Sr contaminated soils. It is based on batch experiments to measure the decrease of the radionuclide solution levels upon addition of the adsorbent, after presaturation with the in-situ soil solution. The method is tested for a number of potential adsorbents (natural and synthetic zeolites, bentonite and organic adsorbents). The results can be readily interpreted on the basis of the (soil) chemical characteristics which govern the solid/liquid distribution of ^{134,137}Cs and ⁹⁰Sr in soil and adsorbent. Results of plant growth experiments on amended soils aimed to check the accuracy of predictions are also presented. Some theoretical and practical aspects of adsorbent applications are discussed.

1. Introduction

After the Chernobyl accident and the widespread contamination of large areas in the Ukraine, Belarus and Russia, a variety of countermeasures have been taken to reduce food contamination, thus lowering the radiation dose to the population (Prister et al., 1992). In the recent past, the emphasis in countermeasure strategy has been shifted and directed at the soil chemical level. In particular, attempts are currently made to reduce the level of radionuclides (mainly ^{134,137}Cs and ⁹⁰Sr) in the soil solution by applying adsorbents characterized by high radionuclide (RN) adsorption properties, thus lowering the levels confronted by the plant roots and resulting in a reduced uptake. The traditional method to test the efficiency of such amendments by measuring the year-to-year variation of the soil-to-plant transfer is costly and time-consuming. In view of the observation that soil-to-plant transfer is mainly function of soil type (Sandalls, 1990), a methodology has been developed to predict the amendment effectiveness for a given scenario at the soil chemical level.

2. Theoretical aspects

In the recent past, the (reversible) solid/liquid distribution coefficient K_d (dm³/kg) of radiocesium and radiostrontium could be reasonably well predicted on the basis of the following equations (Sweeck et al., 1990; Valcke, 1993; Wauters et al., 1995):

$$K_d^{Cs} = \frac{[FES] \cdot K_c^{Cs/K}}{N_K + K_c^{NH_4/K} \cdot N_{NH_4}} = \frac{[K_d^{Cs} \cdot N_K]}{N_K + K_c^{NH_4/K} \cdot N_{NH_4}} \quad (1)$$

$$K_d^{Sr} = K_c^{Sr/Ca} \cdot K_d^{Ca} = \frac{K_c^{Sr/Ca} \cdot CEC \cdot Z_{Ca}}{N_{Ca}} \quad (2)$$

$[K_d^{Cs} \cdot N_K]$ (equiv./kg) refers to the *specific radiocesium adsorption potential*, defined as the product of the capacity of illitic Frayed Edge Sites [FES] (equiv./kg) and the selectivity coefficient $K_c^{Cs/K}$ of Cs towards the competitive cation K. It can be readily obtained by measuring the trace radiocesium distribution coefficient K_d^{Cs} (dm³/kg) in K saturated samples at sufficient high K concentrations. $K_c^{NH_4/K}$ refers to the NH_4 -to-K selectivity coefficient on the FES and varies between 4 to 7 for most soils. $K_c^{Sr/Ca}$, CEC and Z_{Ca} refer to the Sr-to-Ca selectivity coefficient, the Cation Exchange Capacity (equiv./kg) and the Ca saturation on the exchange complex, respectively. Similarly as in eqn (1), $K_c^{Sr/Ca} \cdot CEC \cdot Z_{Ca}$ can be defined as the *radiostrontium adsorption potential* $[K_d^{Sr} \cdot N_{Ca}]$ (equiv./kg). $K_c^{Sr/Ca}$ values generally vary in the range of 1-2 (Bruggenwert and Kamphorst, 1976). Therefore, the product $K_c^{Sr/Ca} \cdot Z_{Ca}$ approaches unity for most soils, as a result of which the CEC value can be taken as a first approximation of the radiostrontium adsorption potential. N_K , N_{NH_4} and N_{Ca} refer to the respective cation solution concentrations (equiv./dm³).

Recently, the solid/liquid distribution behaviour of both radionuclides in a number of Cs and Sr selective zeolites has been quantitatively investigated (Valcke et al., 1995a and b). Similarly as for soils, the zeolites studied could be characterized in terms of *radiocesium and radiostrontium adsorption potentials*, represented by $[K_d^{Cs} \cdot N_K]$ and $[K_d^{Sr} \cdot N_{Ca}]$. These parameters can readily be obtained by measuring the trace radionuclide distribution coefficient in K (Cs) or Ca (Sr) saturated zeolites at sufficient high K or Ca concentrations.

These quantitative insights allow predictions of $K_d^{Cs,Sr}$ values which agree generally within a factor of 2 with observed values. In addition, they are useful to estimate the effect of the addition of an adsorbent to contaminated soils, by comparing radionuclide adsorption potentials before and after adsorbent addition. The procedure presented in this paper aims to verify these predictions by some rapid and simple laboratory tests.

Before embarking on the experimental results, it is of interest to discuss some theoretical and practical aspects involved with the large scale application of mineral or organic adsorbents. Provided the adsorbent is sorption competitive with the soil, mixing of it with the soil may result in two effects. Firstly, RN levels in the soil solution will be decreased. Secondly, a fraction of the reversibly retained RN associated with the soil will be shifted via the soil solution towards the adsorbent. The extent of the effect will depend on the ratio of the "weighed" adsorption potentials of soil and adsorbent, and on the fraction of RN remaining available in the soil. It can be expected to be minimal - particularly in the case of Cs - if several years after the contamination availabilities are already quite low, depending on the fixation potential of the soil.

An important limitation inherent on the use of adsorbents is that of the doses to be applied. In order to obtain a reduction of RN solution levels by a factor of 2, it is required that the RN adsorption potential of the adsorbent to be added exceeds that of the soil by at least a factor of 100 (working at a 1% dose). If adsorbents should be applied shortly after contamination of the soil, when only a thin layer of soil (<5 cm) is contaminated, then a dose of 1%, corresponding to some 5 tons/ha, would be effective. However, such intervention is likely to become ineffective when the amended soil is ploughed and the upper 5 cm layer is mixed with the 20-30 cm ploughing layer, since the adsorption of Cs and Sr on most adsorbents is completely reversible. If adsorbents are to be applied several years after the contamination, when the radionuclides are distributed in the 20-30 cm ploughing layer, then a moderate dose of 1% would amount to some 25-40 tons/ha (depending on the density of the soil layer). However, the most important problem in this case regards the difficulty of mixing the adsorbent with the soil layer in order to obtain a maximum contact with the contaminated soil. Therefore, the maximum effect of such amendment may be expected after several ploughing treatments.

An amendment may also result in a change of the ionic composition of the soil. Depending on the "weighed" CEC values of soil and adsorbent, the ionic composition of the soil (solution) may change considerably. This may have positive or negative consequences for nutrient supply and physico-chemical properties.

3. Materials and methods

3.1. Materials

Eight soils originating from different places in Belgium, Ukraine, Russia and Belarus were studied. Table 1 gives a summary of results on CEC (Chhabra et al., 1975), pH (as measured in 0.01 equiv./dm³ KCl at solid/liquid ratios of 1/2.5), specific radiocesium adsorption potentials and % Cs fixation (as determined using the infinite bath procedure; Wauters et al.,

1994). The following adsorbents were studied: natural clinoptilolite (Hector, California) and mordenite (Karpats, Ukraine), synthetic Na^+ -mordenite (Eko Nobel, Sweden), synthetic zeolite (Ca^{2+}) 5A (Union Carbide), two organic soils (Bragin, Belarus; Donegal, Ireland), organic sapropel (Belarus) and bentonite clay. They are all characterized by (extremely) high adsorption potentials for Cs ($[K_d^{\text{Cs}} \cdot N_K]$) or Sr (CEC for soils and clays; $[K_d^{\text{Sr}} \cdot N_{\text{Ca}}]$ for zeolites). For zeolites, $[K_d^{\text{Cs}} \cdot N_K]$ values are dependent on the PAR value of the soil solution (Valcke et al., 1995a). Zeolites show extremely high adsorption potentials for Cs or Sr, but, unfortunately, the adsorption of these radionuclides is completely reversible.

Table 1: Results of the (soil) chemical characterization of the soils and adsorbents studied.

soil or amendment (origin)	(soil) type	CEC or $[K_d^{\text{Sr}} \cdot N_{\text{Ca}}]$ (equiv./kg)	pH	$[K_d^{\text{Cs}} \cdot N_K]$ (equiv./kg)	% fix.
Mol-1 (BEL)	podzol	0.017	4.5	0.21	15
Mol-2 (BEL)	podzol	0.017	4.5	0.15	15
Wyn-A (BEL)	forest soil, A hor.	0.144	3.2	0.36	67
Poleskoe (UKR)	loam sandy podzol	0.036	6.6	0.46	35
Chistogalovka (UKR)	sandy podzol	0.069	7.7	0.29	25
Vetka (BELA)	sandy podzol	0.024	6.1	0.71	12
Novozibkov (RUS)	sandy podzol	0.050	7.0	0.47	15
Bragin (BELA)	peat (70 % O.M.)	1.030	6.0	0.52	38
Komsomoletz (RUS)	peat (84 % O.M.)	1.140	6.3	0.26	21
Sapropel (BELA)	organic (73% O.M.)	0.891	5.5	0.154	-
Donegal (IRE)	peat (97% O.M.)	0.527	4.0	0.002	3
Bentonite	clay	1.206	-	-	-
Clinoptilolite (Hector, Calif., USA)	natural zeolite	20.317 (20 days)	> 7	35-55 (PAR)	0
natural Mordenite (Karpats, UKR)	natural zeolite	4.973 (20 days)	> 7	60-125 (PAR)	0
Na-Mordenite (Eko Nobel, Sweden)	synthetic zeolite	-	> 7	60-125 (PAR)	0
5A (Union Carbide)	synthetic zeolite	21.334 (1 day) 27.586 (20 days)	> 7	-	0

3.2. Laboratory tests

The protocol to test the effect of the amendment was as follows. Known amounts of soil samples with or without the adsorbents (at the corresponding doses) were weighted in dialysis membranes and dialytically equilibrated with a solution representative for the soil's 'in-situ' solution (end-over-end shaking). After presaturation, dialysis tubings were transferred to polyethylene vials containing the respective solution, labeled with known quantities of ^{137}Cs or ^{85}Sr . Systems were equilibrated for 24 hours or longer (end-over-end shaking), after which time ^{137}Cs or ^{85}Sr solution activities were monitored.

3.3. Plant growth experiments

Growth experiments were conducted on the pure Mol-2 podzol (control) or amended with 1% clinoptilolite (Cs), 1% Na⁺-mordenite (Cs), 1% 5A (Sr), 2% sapropel (Sr) and 2% Bragin soil (Sr). Soils were brought to field capacity by thoroughly mixing with the appropriate amount of ¹³⁷Cs or ⁸⁵Sr labeled 1/2 diluted Steiner nutrient solution and allowed to age for 14 days (3 wetting/drying cycles). Then, the appropriate amount of adsorbent was mixed with the contaminated soil and systems were again allowed to age for 14 days. 1 L pots were filled with 1.14 kg of the different amended soils and three days old seedlings of spinach (*Spinacia oleracea* L., cv. Subito) were planted. Plants were grown in a walk-in growth chamber (Weiss 18'SP/+5JU-PA) with a 8hrs/16hrs day/night cycle (18°/15°C). Soil moisture content was adjusted with alternating deionized water or 1/2 diluted nutrient solution additions. Plants were harvested after 4 weeks of growing and analyzed for their ¹³⁷Cs or ⁸⁵Sr content.

4. Results and discussion

4.1. Laboratory experiments

Results of the laboratory experiments are presented in Table 2 (Cs) and Table 3 (Sr), in terms of $K_d^{Cs,Sr}$ values with (K_d^{+ads}) and without (K_d^{-ads}) adsorbent, the observed ratio of K_d values, and the predicted ratio based on a linear combination of RN adsorption potentials and mass of soil and adsorbent. In Table 3, soil solution pH values, which are important regarding CEC of soils, are given to demonstrate the effect of adsorbent addition on the soil's CEC.

It is seen that addition of adsorbents characterized by high RN adsorption potentials to soils with low RN adsorption potentials effectively reduce the RN soil solution levels (increase of K_d). In the case of radiocesium, a 1% dose of both natural and synthetic mordenite results in an effect of a factor 2 to 5. Clinoptilolite is less effective, but as it is available at much larger quantities and lower prices than mordenite, the application of higher doses (e.g. 2%) of this material may be economically more feasible than of mordenite. Furthermore, it is seen that observed effects agree reasonably well with predicted effects, although in some cases predictions exceed the observations. This is most probably due to the Cs fixation, the effect of which is integrated in the measured K_d^{Cs} values but not in the predictions (eqn (1) describes only the *reversible* Cs adsorption). This is nicely demonstrated for the Wyn-A soil: although [K_d^{Cs,N_K}] for Wyn-A is but 50% higher than the value for Mol-1 (Table 1), the measured distribution coefficient (K_d^{+ads}) is 3 times higher for Wyn-A, which is due to its higher Cs fixation capacity (67% for Wyn-A versus 15% for Mol-1). The distribution behaviour of radiostrontium is clearly governed by the CEC, which is demonstrated by the very good agreement between observed and predicted ratio for bentonite and organic soils. Although the

Table 2: Results of laboratory tests on the effect of adsorbent amendments (radiocesium)

soil	treatment	K_d^{ads} (dm ³ /kg)	K_d^{+ads} (dm ³ /kg)	observed ratio	predicted ratio	PAR
Mol-1	Na ⁺ -Mor 1%	294	1557	5.29	6.95	1.23
Mol-1	Na ⁺ -Mor 2%	294	2751	9.36	12.90	1.23
Wyn-A	Na ⁺ -Mor 1%	897	2024	2.26	4.47	1.23
Wyn-A	Na ⁺ -Mor 2%	897	3353	3.74	7.94	1.23
Mol-1	Clinopt. 1%	311	689	2.22	3.39	1.23
Mol-1	nat.MOR 1%	291	1315	4.52	6.19	1.00
Poleskoe	nat.MOR 1%	807	2569	3.18	2.52	0.24
Chistogalovka	nat.MOR 1%	362	1435	3.96	3.76	0.37
Vetka	nat.MOR 1%	896	1832	2.04	2.41	1.13
Novozibkov	nat.MOR 1%	562	1383	2.46	2.70	0.36
Bragin	nat.MOR 1%	638	1751	2.74	2.25	0.21
Komsomoletz	nat.MOR 1%	151	498	3.30	4.46	0.51

Table 3: Results of laboratory tests on the effect of adsorbent amendments (radiostrontium);

PAR = 0.5; [K] and [Ca] = 1.057 and 8.943 mequiv./dm³

soil	treatment	K_d^{ads} (dm ³ /kg)	K_d^{+ads} (dm ³ /kg)	observed ratio	predicted ratio	pH
Vetka	5A 1%	3.02	31.1	10.30	9.92	7.05
Vetka	5A 2%	3.02	51.3	16.99	18.85	7.27
Vetka	5A 4%	3.02	85.0	28.16	36.71	7.53
Vetka	bentonite 1%	3.02	4.17	1.38	1.50	6.07
Vetka	bentonite 2%	3.02	5.13	1.70	2.01	6.22
Vetka	bentonite 4%	3.02	7.39	2.45	3.02	6.26
Vetka	Bragin 1%	3.02	4.43	1.47	1.43	5.91
Vetka	Bragin 2%	3.02	5.71	1.90	1.86	6.19
Vetka	Bragin 4%	3.02	8.30	2.76	2.72	6.60
Vetka	Donegal 1%	3.02	3.69	1.22	1.22	5.64
Vetka	Donegal 2%	3.02	4.33	1.44	1.44	5.29
Vetka	Donegal 4%	3.02	4.95	1.64	1.88	5.06

effect of organic soils is rather small, these adsorbents may improve e.g. the water holding capacity of the amended soil, which in turn may reduce resuspension effects. Attention should be given to the pH effect on K_d^{Sr} : since the CEC in this poor sandy soil stems mainly from pH dependent organic matter ion exchange sites, a pH increase drastically increases CEC and K_d^{Sr} , and *vice versa*. Although Donegal and Bragin virtually have a similar CEC value (at a given pH), the "actual" CEC of the acidic Donegal soil is but half the value of the neutral Bragin soil. Increasing doses of Donegal soil result in decreasing pH values and hence lower effects. The opposite effect is observed for the "neutral" Bragin soil. Special attention should be paid to the potency of synthetic zeolite 5A: a 1% dose of 5A results in a tenfold increase of K_d^{Sr} (1 day equilibration), but also in a drastic pH increase. Therefore, the enormous increase of the K_d^{Sr} by 5A addition is partly due to the increase of the soil's CEC by this pH increase.

4.2. Plant growth experiment

Results of transfer factors TF, defined as the ratio of Bq/g plant and Bq/g soil, are given in Table 4, together with pH values in the soil solution. It is seen that for this podzol soil, characterized by a low radiocesium ($[K_d^{Cs} N_K]$) and radiostrontium (CEC) adsorption potential (Table 1), addition of organic or mineral adsorbents characterized by higher adsorption potentials considerably reduces the transfer of these radionuclides. Furthermore, it is seen that observed effects coincide very well with predictions for organic substrates (Sr). For zeolites, the agreement is not so good. For mordenite and clinoptilolite, this is very likely due to the drastical changes of the ionic composition (details not shown). For 5A, this disagreement can partly be explained by the enormous increase of the soil's CEC value.

Table 4: Results of the plant growth experiment (standard deviations in parentheses)

treatment	TF (g soil/g plant)	ratio of TF	predicted effect	pH	CEC (equiv./kg)
radiocesium					
control	0.750 (0.093)	-	-	4.67	0.018
Na ⁺ -Mor 1%	0.164 (0.042)	4.57	9.33	5.11	0.019
Clinopt. 1%	0.251 (0.040)	2.99	4.33	6.19	0.022
radiostrontium					
control	35.100 (6.421)	-	-	4.74	0.018
5A 1%	1.407 (0.104)	24.95	16.33	7.68	0.065
Bragin 2%	12.748 (1.455)	2.75	2.14	5.27	0.019
Sapropel 2%	17.039 (2.135)	2.06	1.99	4.86	0.018

5. Conclusions

The use of Cs and/or Sr specific adsorbents as countermeasures will be effective if the radionuclide adsorption potential of the adsorbent exceeds that of the contaminated soil by a factor of 100. In practice, only soils characterized by low Cs or Sr adsorption potentials are to be considered. Based on a soil chemical characterization of soil and adsorbent, (a) the solid/liquid distribution behaviour of Cs and Sr in soils and adsorbents can fairly well be predicted and (b) the effect of addition of an adsorbent on the Cs or Sr solution levels can be estimated satisfactorily. Although the soil-to-plant transfer is influenced by both soil chemical and plant physiological properties, the observed reduction of transfer factors coincides fairly well with our predictions based on a soil chemical characterization. Therefore, the presented laboratory method may be a first step to assess whether the addition of an adsorbent will be effective and, if so, to what dose it must be applied to obtain the desired effect.

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ABILITY OF SPECIMEN VERMICULITIC MINERALS TO FIX RADIOCAESIUM: EFFECT OF THE CHEMICAL ENVIRONMENT

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Abstract

Because of its high ability to fix dried cations like cesium, specimen (commercial) vermiculite minerals were sounded as potential opportune material for soil remediation techniques. The initial presence of mica layers in Transvaal vermiculite (3% K₂O = 30% mica) promote Cs sorption on existing edge sites. In the range of the K-Ca synthetic solution tested, low K level is needed to induce more lattice collapse but also to initiate more competitive effects; as a consequence, the resulting Cs fixation remains relatively constant when K loading increases. For a Russian mineral characterised by more pronounced vermiculitic properties, much higher selective sorption occurs only after the structure has markedly collapsed. At high K loading, competitive effects are balanced by the number and selectivity of the newly created edge specific fixation sites. However, such an optimal structural environment for Cs fixation is only achieved with K levels which are not encountered in soil solution. Independently of the vermiculite specimens and the K-Ca status, the NH₄ loading of the different systems clearly induces additional Cs entrapment. This fixation behaviour is apparently obtained in the presence of NH₄ level certainly below 10⁻³ M, a concentration which is realistic in rich organic acid soils.

1 Introduction

Especially in semi-natural areas, the persistence of a relatively high radiocaesium bioavailability is often related to the presence in soil surface of raw humus layers (Sandalls et al., 1988; Thiry and Myttenaere, 1993). In this context, Vermiculite clay minerals as a countermeasure for site

remediation, offer high selectivity properties for Cs sorption, which could directly reduce the long-lasting availability of radiocaesium for plant uptake.

In case of underground (specimen) vermiculite application in nature, chemical changes in the mineral associated with re-equilibrium with the soil environment complicate interpretation of the nature and capability of the fixation processes involved. The cationic environment can affect the sorption of microquantities of Cs by clays; moreover, since vermiculite is also an exchanger with phase transition (Reichenbach, 1968), its structure and selectivity properties may change while chemical sorption is occurring.

This possible different reaction of radiocaesium with clay led to the present investigation on Cs retention from different chemical forms of vermiculite in the presence of the major soil competing ions able to influence clay properties. Sorption tests were conducted using 2 types of natural vermiculite with a range of K-Ca saturation to determine the effect of chemical changes in exchange sites and of resulting interlayer spacings. In each scenario, the potentials of Cs fixation are determined by submitting the clay samples to 2 types of desorption, in the presence of mixed K^+ - Ca^{++} or NH_4^+ as a counter-ion solution.

2 Material and methods

A South African (Transvaal) and a Russian vermiculite were obtained from Vermiculite et Perlite SIBLI S.A.. Original mineral flakes were ground and the 100-200 μm (5-10 mesh) fraction separated by wet sieving. The clay fraction was purified by the Na-resin method (Rouiller et al., 1972) and washed with distilled water. Two vermiculite were selected because of interest in their different structure properties. The Transvaal specimen is dominated by trioctahedral vermiculite and mica-vermiculite mixed-layer clays. The Russian specimen consists of trioctahedral vermiculite and minor amounts of chlorite.

100 mg of clay were dispersed in dialysis membrane with 5 ml of distilled water. With a batch technique, samples were saturated by K^+ / Ca^{++} ions in various K-Ca scenarios (2 replicates). Five mixed KCl- $CaCl_2$ $10^{-3}N$ equilibrium solutions were used with respective equivalent fractions of 0.0, 0.04, 0.16, 0.51 and 1. The clay-solution ratio was 100 mg/100 ml. After equilibration, the solutions were labelled with trace levels of ^{137}Cs ($10^{-10}N$) and the system gently shaken for 1 week. Sorption rate is regularly monitored by sampling of the solution. Radiocaesium desorption is based upon the generation of "infinite bath" boundary conditions (modified method from Wauters et al., 1992). The desorption (2 weeks) was carried out by introducing in the system a strong Cs^+ sorbent which is renewed regularly to obtain a cumulative desorption yields. Two desorption agents were compared. In one case, use is

made of a K-Ca saturated acid resin (Amberlite IR120, 5 g), previously equilibrated with the same respective K-Ca (10^{-3} N) equilibrium solutions. In the other case, radiocaesium was desorbed by using the Giese Granulate (Ammonium-copper-hexacyanoferrate, 5 g) as an infinite Cs^+ sorbent, equilibrated with a NH_4Cl 10^{-3} N solution as the exchange liquid reagent.

3 Results and discussion

3.1 Radiocaesium sorption

Figure 1 illustrates the variation in K_d with increasing potassium saturation. The Transvaal vermiculite showed higher K_d values in Ca-rich scenarios. In the low K loading region ($Z_K = 0$ to 0.2), K_d values first increased and then decreased with $Z_K > 0.04$. The increase is related to a K-collapse inducing effect (Jacobs, 1963).

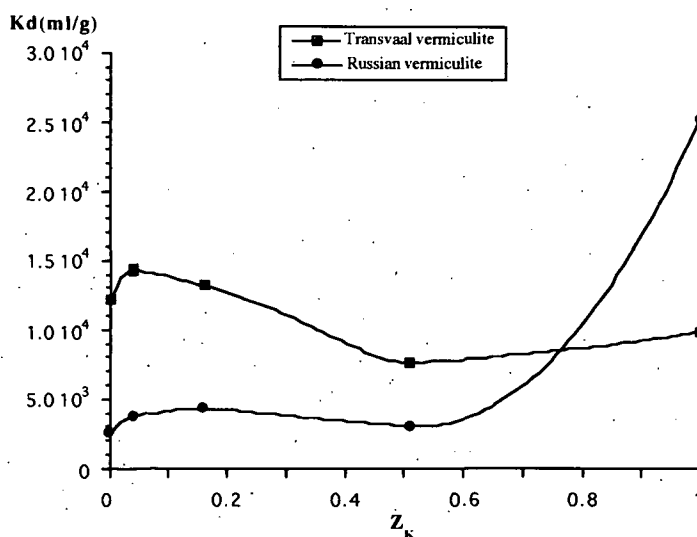


Figure 1: Distribution coefficients from various K-Ca scenarios (mean of 2 replicates).

With higher K loading, K_d values decreased in the 0.2-0.5 K equivalent fraction range; K^+ - Cs^+ competition occurs on similar interlayer sorption sites.

At K saturation, the selected specimens exhibit a strikingly different behaviour: Cs sorption markedly increases in the Russian sample and remains fairly constant in the Transvaal

specimen. Tamura and Jacobs (1960) similarly found that vermiculite expanding lattice collapses after K saturation, giving rise to higher Cs sorption. The K_d for Cs increases rapidly probably because of an increase in edge fixation sites made available by a clear K-collapse inducing effect.

It is generally accepted that micas and vermiculitic clay minerals sorb more Cs when saturated with Ca and inversely with K, due to respective effectiveness of the competing cation. In the sorption of micro-quantities of Cs by vermiculite saturated with mono-ionic solutions of KCl or CaCl_2 , the K cation having size, charge and hydration similar to that of Cs^+ was shown to be more effective than Ca in competing for Cs exchange sites (Shawney, 1964). However, in a more complex medium such as mixed K-Ca solution, the normal ion-exchange adsorption and competition processes seem insufficient to explain the selective Cs sorption by different vermiculite types. Depending on the initial "steric" conditions, the saturating cation differently influences the amount of selective sites for Cs^+ sorption on the minerals, by either decreasing or increasing it. In the Transvaal specimen, mixed-layering of vermiculite with mica generates frayed edge sites with a high selectivity for radiocaesium (Cremers et al., 1988). Increasing K loading mainly results in increasing ion exchange competition between K^+ and Cs^+ ions. For the Russian mineral, a high K loading is necessary to promote interlayer collapse inducing a higher Cs^+ sorption.

3.2 Radiocaesium fixation

In the current investigation, desorption of radiocaesium in various cationic environments was first performed using an acid exchange resin, previously equilibrated with the respective K-Ca solutions. The resulting Cs net retention is herein computed as the Cs relative quantity which was not desorbed in our experimental conditions. The desorption yields are illustrated in figures 2 and 3, using the K/Ca Amberlite resin and the $\text{NH}_4\text{-GG}$ as respective desorption agents.

The figures shows two main features:

- (1) for the K equivalent fraction range $0 < Z_K < 0.51$, the Transvaal vermiculite invariably exhibits higher Cs^+ net retention than the Russian specimen while the "reverse situation" is observed for the K-homoionic scenario ($Z_K = 1$),
- (2) the radiocaesium fixation is distinctly higher when using $\text{NH}_4\text{-GG}$ as the desorbent, whatever the tested material and the ionic scenario.

Using a K/Ca Amberlite resin as desorbent leads to low radiocaesium fixation values in the range $0 < Z_K < 0.51$: the values vary from 28 to 17% and 12 to 5% for the Transvaal and Russian minerals respectively. Under such ionic scenarios, the occurrence of frayed edge sites on the Transvaal specimen may explain a higher Cs fixation. Nevertheless increasing K loading ($Z_K = 1$) does not enhance the net retention of radiocaesium. For the Transvaal mineral, K loading therefore has little influence on the radiocaesium net fixation. This observation is supported by other results (Klobe and Gast, 1970) which showed that neither residual K, nor K added before Cs, affected Cs fixation, measured by isotopic exchange on a K-extracted hydrobiotite-vermiculite clay ($0.2-62 \mu\text{m}$). For a K-treated hydrobiotite, Jacobs (1963) and Tamura (1964) interpreted this behaviour as a collapse of the lattice during K pretreatment and as the cause of more pronounced competitive effect at the edge of the interlayers. This last effect would be particularly predominant if, as suggested by Rhoades and Coleman (1967), the Transvaal vermiculite failed to contract completely, even though the analytical data showed nearly complete saturation with K.

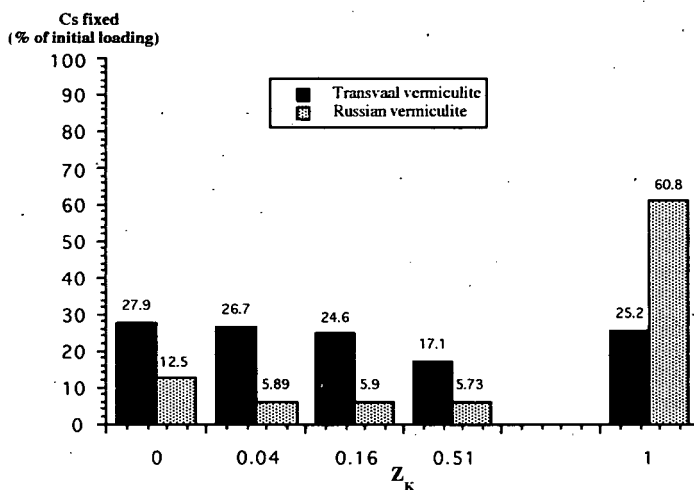


Figure 2: Radiocaesium net fixation in various K-Ca scenarios (extraction with K-Ca acid resin).

Similar observations emerge from the net retention values computed for the Russian specimen (fig.2) except for the $Z_K = 1$ scenario. The Cs fixation is indeed much higher for the K^+ -saturated Russian vermiculite. This is due to a K-collapse inducing effect giving rise to frayed

edge sites. At that scenario $Z_K = 1$, the differing behaviour of the tested materials is probably due to different layer charge properties.

Using the NH_4-GG as desorbent leads to much higher Cs net retention values.

It is well known that ammonium, like other alkali metals can compete with Cs^+ for specific sorption sites. In comparison to K^+ , the strong effectiveness of NH_4^+ in competing for Cs^+ frayed edge sites has been shown by Sweeck et al. (1990) for various reference micaceous clay minerals and Belgian agricultural soils. Their results indicate that NH_4^+ is 4 to 7 times more selective than K^+ for Cs^+ specific adsorption sites. Although the ammonium is generally considered as competitive with other cations for interlayer adsorption sites, collapse-inducing effect of the vermiculite and consecutive ions entrapment can promote the unavailability of radiocaesium (Jacobs, 1963) in addition to other different cations (Barshad, 1954). In Figure 3, it is shown that desorption is strongly reduced, independently of the chemical scenario and of the mineral type.

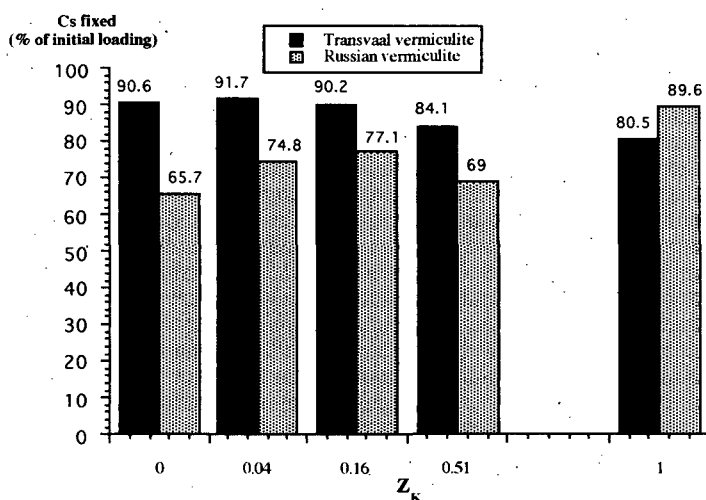


Figure 3: Radiocaesium net fixation in various K-Ca scenarios (extraction with NH_4 -Giese Granulate).

The entrapment effect is such that for all samples, more than 85% of the sorbed Cs remain fixed. Observation of the desorption curves has shown that well defined plateau were obtained after first extraction (24 hours). These data are in agreement with the proposed edge collapse and consequent interlayer entrapment that hinder Cs extraction. A similar effect have been

described for K (Klobe and Gast, 1970) when a Cs contaminated vermiculite were immersed in a solution containing a K amount > 20% of the clay exchange capacity. In the same context, Grütter et al. (1986) observed that caesium concentrations in the exchange solutions as low as 10^{-7} M can initiate the collapse of the edge adjacent to the surface of chlorite grains and increase the Cs fixation.

4. Conclusions

Radiocaesium sorption and desorption yields on the tested vermiculitic specimens depend upon the ionic scenario and the type of mineral. The presence of K^+ ions has two main effects: (1) a collapse inducing effect, increasing Cs^+ fixation, (2) a cationic competition for specific interlayer sites, decreasing Cs^+ fixation. NH_4^+ ions have a strong entrapment effect: the trapped radiocaesium is scarcely desorbed, whatever the K-Ca status of the clay.

At low K saturation scenarios, as occurs in realistic soil conditions, the tested Transvaal mineral behaves as a powerful sink for radiocaesium. The specimen consists of a micaceous vermiculite: low K^+ and NH_4^+ loadings strongly promote net radiocaesium retention. This observation is of major importance: in acid forest soils, K^+ and NH_4^+ are major cations involved in ion exchange process. Indeed, rich organic surface layers are generally characterized by a low mineralization rate, allowing high levels of $N-NH_4^+$ in the soil solution. In another hand, the K produced by decaying organic matter is not affected by organic complex formation and consequently, it remains also highly soluble in surface horizons.

It can be concluded that interstratified mica-vermiculite minerals should be preferred as vermiculite minerals countermeasure to reduce radiocaesium bioavailability in soil. In these conditions, two effects are combined: high charge vermiculitic interlayers promote the entrapment process while the specific mica frayed edge sites fix radiocaesium.

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TRANSFER OF RADIOCAESIUM IN SENSITIVE AGRICULTURAL ENVIRONMENTS 1986 - 1994 AFTER THE CHERNOBYL FALLOUT IN SWEDEN

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ABSTRACT

In 1986, 25 sites in the county of Gävleborg and 17 sites in the county of Jämtland in Sweden, were investigated regarding ^{137}Cs transfer to grass on different types of pasture and to cereal grain. The aim was to study the impact of site and soil characteristics ones a long-term perspective from 1986-1994. Grass and grain samples were radioassayed and chemically analysed. The transfer of ^{137}Cs was higher to grass on permanent pasture land than on temporary grassland. It was much higher to grass than to cereal grain. For both crop products, however, there was a considerable annual reduction. In the fallout year, 1986, the contamination level depended on the interception capacity of the stubble and grass sward and on dilution by growth, especially on grassland. To some degree, the transfer was also influenced by soil fertility and by the level of K-fertilization. In the following years, the transfer to grass was reduced by a factor of 2 to 100. Apparently, the ploughing down and mixing of the contaminated surface soil layer with a larger soil volume was effective in reducing the transfer to crops. However, this measure was less effective on organic soils than on mineral soils. Measures to reduce the long-term transfer to crops are discussed in relation to a new concept for description of the development with time of the transfer, "Tar".

INTRODUCTION

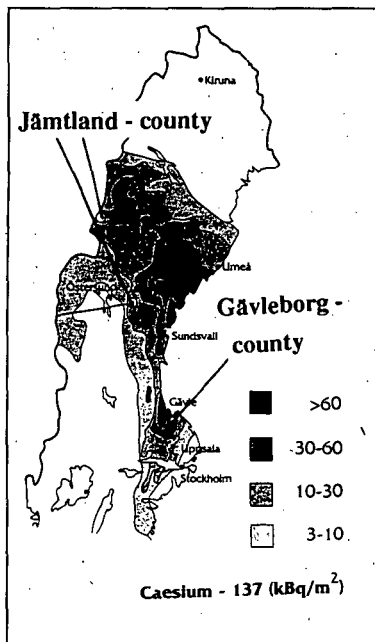
The agricultural environment is complex and varies due to climate and soil conditions, to farming practices, and the type of animal husbandry. It is susceptible to for atmospheric pollution, including radioactive fallout. Part of the land is used as pastures and for cultivation of grass crops for the feeding of animals, other arable lands used for grain production. Farming

intensity varies regionally in Sweden and usually less intensive where the land is used for pasture compared with hay production.

After the Chernobyl accident in April 1986, investigations began into the transfer of radiocaesium to grass and cereal crops in the most heavily contaminated counties of Sweden. The aim was to investigate the short- and long-term situation after the fallout. The districts selected were parts of the Gävleborg, Västernorrland, Jämtland and Uppland counties. These counties were reported to suffer from high contamination levels in the grass crops during the early part of the growing season (cf. Eriksson & Rosén, 1991).

The studies were to be continued for a sequence of years and cover the period of equilibration for radiocaesium on grassland as well as the beginning of an expected later lag period. Results from Gävleborg county, where the transfer to grass was compared with the transfer to cereals, and from Jämtland county, where the transfer to grass in a mountain area is compared with that in a river valley area, are to be published in Rosén, (1995) and Rosén et al., (1995) respectively. A summary is given at this symposium. The investigation was financial supported by the Swedish Radiation Protection Institute and the Swedish University of Agricultural Sciences.

MATERIALS AND METHODS



Study areas and sampling procedures

In 1986, 15 farms with 25 sites in Gävleborg county and 17 grass sites in Jämtland county were selected for study, Figure 1. On most farms in Gävleborg county, one site was located on cultivated grassland with hay production and the other on ploughed land with cereals or oil crops, often located on adjacent fields. In Jämtland county, 9 sites were temporary grassland and 8 sites permanent pasture in a mountain area and river valley area respectively. The investigations were carried out in the growing seasons of 1986-1994.

Figure 1. Deposition of fallout ^{137}Cs from Chernobyl, based on inflight measurements performed from May to October 1986 in Sweden (SGAB, 1986) and the study areas.

On each site, an area of 100 m² was randomly selected for sampling of soil and crop. The sampling of the crop was performed on 4 microplots of 0.25 m² at the normal harvest time every year. In Gävleborg county, grass was sampled twice and cereals once per year, except in 1986, when cereals were sampled twice. In Jämtland county the grass was harvested only once per year. The grass was cut about 3-5 cm above the ground. The cereals were cut at a height of about 5 cm above the ground and threshed. The four grass or grain subsamples were pooled into a bulk sample, which thus contained the yield from an area of 1 m². The soil was sampled by taking three or four soil cores, 57 mm in diameter and to a depth of 5 cm or 10 cm, on each microplot and pooled into a bulk sample for each site in 1986. Soil characteristics are given in Table 1.

Table 1. Soil characteristics of the surface layer (Gävleborg county 0-5 cm and 0-10 cm in Jämtland county) at the sampled sites. Soil type, organic matter %, pH, easily soluble potassium (K_{AL}) mg per 100 g soil and ¹³⁷Cs deposited, kBq / m²

Farm & Site	Soil type	Org. matter	pH H ₂ O	K _{AL}	Dep. ¹³⁷ Cs	Farm & Site	Soil type	Org. matter	pH H ₂ O	K _{AL}	Dep. ¹³⁷ Cs
X-A1	l. sand	13	5.0	7.1	203	Z-1	l. sand	8.1	5.7	30.0	37
X-B3	peat	37	6.7	8.7	163	Z-2	l. sand	6.7	5.9	17.2	37
X-C5	peat	50	5.9	19.3	53	Z-3	h. sand	9.9	6.0	16.0	34
X-D7	peat	45	5.5	27.0	72	Z-4	sand	5.4	5.7	8.0	40
X-E9	peat	53	5.6	37.2	97	Z-5	s. loam	8.2	5.7	9.6	27
X-F11	peat	56	6.3	27.4	131	Z-6	s. loam	10.4	5.7	17.2	21
X-G13	peat	51	6.7	8.8	202	Z-7	s. loam	10.5	5.8	16.8	27
X-H15	sand	5	6.5	6.1	190	Z-8	sand	7.7	5.5	10.8	21
X-I17	peat	57	6.3	25.3	95	Z-9	s. loam	-	-	-	37
X-J19	peat	31	5.5	15.9	89	Z-10	s. loam	11.0	5.3	34.8	34
X-K20	S. cl. l	11	5.8	20.6	157	Z-11	sand	14.5	5.9	20.4	43
X-L21	S. loam	16	6.0	9.4	209	Z-12	s. loam	9.3	5.9	18.0	44
X-M22	S. cl. l	11	6.2	5.0	179	Z-13	l. sand	10.8	4.6	9.7	17
X-M23	S. loam	12	5.3	6.9	149	Z-14	l. sand	6.0	6.4	7.2	34
X-N24	peat	58	5.5	22.9	83	Z-15	silt loam	9.7	5.9	15.1	38
X-O25	peat	41	5.8	29.4	20	Z-16	peat	28.7	5.5	31.6	33
						Z-17	loam	5.0	5.4	9.2	23

S. cl. l. = Sandy clay loam

Analysis

The grass and grain samples were artificially dried at 70-90 °C for 1-3 days, weighed and ground, passing through a 2 mm sieve, and then stored in 1 litre Marinelli beakers or 330 ml

plastic containers. The pooled soil core samples were air-dried for one week, weighed, ground and passed through a 2 mm sieve. Aliquots of these samples were used for radiometric analyses, the soil samples also for chemical analyses (Egnér et al., 1960), for determination of soil type by the hydrometer method (Day, 1965), and for estimation of the deposition level of radiocaesium. The latter was recalculated on the basis of volume and surface of the soil cores.

Radiocaesium activity concentrations of ^{137}Cs of the soil and vegetation samples were determined using high purely germanium detector systems, housed in a low-background laboratory. The measurement errors were in the range of 1-5 % for ^{137}Cs . All activity concentrations in soil and crop samples refer to dry weight (d.w.) basis. The soil activity were recalculated to the date of the Chernobyl fallout in 1986 and the vegetation to sampling day.

Transfer factor

Comparison of radionuclide transfers to feed and food items between studies of different environments cannot easily be made without referencing the nuclide activity concentration to the deposition level on the ground surface. Accordingly, the ratio obtained is a coefficient or transfer factor which, should be independent of the deposition level, but vary according to environmental and / or experimental conditions (Eriksson, 1977; Haak, 1983).

In the formula below, this transfer factor, TF_g , is used to describe the transfer of ^{137}Cs to grass and cereal grain. TF stands for transfer factor and index g for ground. TF_g then denotes the ^{137}Cs activity concentration in the plant, Bq per kg dry weight, as related to the total ground deposition, Bq per m^2 (IAEA, 1987).

$$\text{TF}_g = \frac{\text{Activity concentration in plant (Bq / kg d. w.)}}{\text{Activity deposited on ground (Bq / m}^2\text{)}} \quad (1)$$

Unit: $[\text{m}^2 (\text{kg d.w.})^{-1}]$

A reduction with time in the nuclide transfer to crop products, independent of decay, is observed in most environments. The reduction is sometimes called ageing, and is attributed to decreased plant availability of the nuclide in the soil. An ecological half-time, T_{ec} , $= \ln 2/A$, can be calculated, based on the assumption that the reduction in transfer, like radioactive decay, is exponential (cf. Eq. 2). As found by Eriksson (1994), an exponential reduction of the nuclide transfer does not apply on contaminated pasture sites, at least not during the first years after the fallout. Instead, a function or equation of the following type is recommended (cf. Eq. 3).

$$Y_t = Y_0 \cdot \exp(A \cdot t) \quad (2)$$

$$f(t) = \exp(A \cdot t^N + B) \quad (3)$$

In Eq. 3, the coefficients A, N and B are estimated by a least square fit of the function to the annual TFG-values observed during a sequence of years. From this equation, the "half-time for the annual reduction rate", denoted *Tar* can be calculated. Generally *Tar* starts from a low value and increases with time. *Tar* is thus inversely related to the annual decrease of TFG. An increase in the *Tar* indicates that the annual reduction of the nuclide transfer decreases with years.

RESULT AND DISCUSSION

Fallout conditions

The deposition range of ^{137}Cs in the sampling area in Gävleborg county was 20-209 kBq per m^2 of ^{137}Cs and in Jämtland county 17-44 kBq per m^2 of ^{137}Cs . The mean depositions for Gävleborg and Jämtland were 114 and 32 kBq per m^2 of ^{137}Cs , respectively (Table 1).

Transfer of ^{137}Cs to the grass and grain crops

The data presented in Figures 2 and 3 give a description of the development with time in the average ^{137}Cs transfer to sampled crops from each site in Gävleborg and Jämtland counties. Figure 2 for Gävleborg county is based on 12 sites sampled for temporary cultivated hay crops, on 4 sites sampled for pasture grass, and on 9 sites in grain-grass rotation with grain in the first two years after fallout. Figure 3 for Jämtland county is based on 9 sites sampled for temporary grass crops and 8 sites sampled for permanent pasture grass.

It should be noted that TFG-values, $\text{m}^2 \text{ kg}^{-1} \times 10^{-3}$, obtained from a survey like this, as well as others after Chernobyl, are not equal in precision to those obtained from experimental setups, where the deposition level is known exactly. The TFG-values obtained reflect the influence of various field conditions. In this case, the variability in conditions on pasture sites and temporary grassland, also influenced by ploughing and K-fertilization, have contributed to the significant variation observed. The TFG-values for grass of the first cut was lower than those of the second. This may depend on lower straw content from root uptake and higher mineral content in the grass of the second cut.

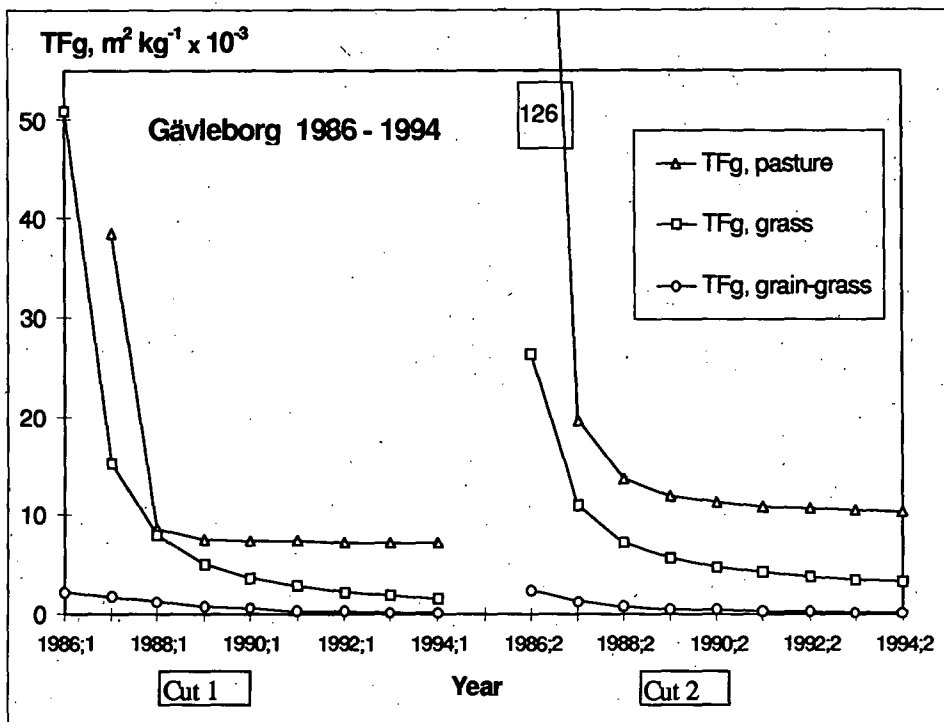


Figure 2. Change in radiocaesium transfer to temporary grass crops, pasture grass and to grain crops during the period 1986-1994. The averages for the sites in Gävleborg county have been used for estimation of the curves by equation 3. The lowest curves refer to grain and grass of crop rotations, with grain as initial crops in 1986 and 1987.

The main impression of Figure 2 is the different and persistent levels in ^{137}Cs transfer to the grass crops from temporary and pasture grassland. The latter was more sensitive to fallout and showed a higher transfer and a slower reduction rate throughout the whole period.

The transfer to the grain-grass rotations was considerably lower than that to the temporary grass crops throughout the period and stresses the importance of ploughing the contaminated land and the use of a crop rotation to reduce the transfer of radiocaesium to the animal feed.

The main feature in Jämtland county was the high transfer to grass in 1986, especially on the pasture sites. It was often reduced to less than half the value in 1987 on sites where this comparison was possible (Figure 3). Also, the radiocaesium transfer to grass was much higher during the first years on pasture sites than on temporary grass sites. After a period of years this difference seemed to diminish. The *Tar*-values were inversely related to the values of the

functions for the average TFg-values. Consequently the *Tar*-values for temporary grass and pastures described different developments with time.

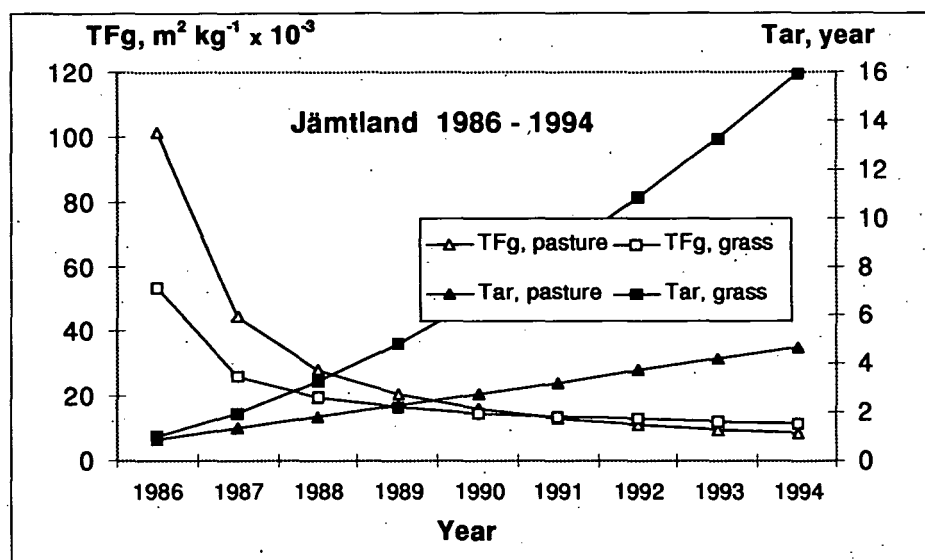


Figure 3. Change in radiocaesium transfer to grass crops during the period 1986 - 1994 in Jämtland county. The annual averages for the group of sites with temporary grass fields and that of the sites with pastures have been used for estimation of curves by the least square method according to equation 3. The *Tar*-values are calculated from the function for TFg and shows the half-time of change in radiocaesium transfer to the crop from one year to next year (cf. Eriksson, 1994).

The first values for 1986-1987 were about the same but in the following years the curves indicated an increasing difference between sites representing these types of grassland. During the observation period of 9 years the higher reduction rate for TFg in the pasture group than for the temporary grass caused lower *Tar*- values or shorter "half-times between years" for the former than for the latter.

If no countermeasure was employed, the level of ^{137}Cs transfer generally was higher on unploughed than on ploughed and K-fertilized land. K-fertilization alone was effective to decrease the ^{137}Cs transfer. In line with earlier Swedish field studies (Lönsjö et al., 1990; Rosén, 1991), repeated K-fertilization was effective in retaining a reduction in the level of ^{137}Cs transfer to grass. If no K-fertilization took place for some years, ^{137}Cs transfer usually

potential value on these poor soils. They were necessary measures, at least during the first decade after the Chernobyl fallout.

As shown in Table 1, many of the soils in the two counties were sandy with low contents of available nutrients. The content of organic matter ranged from 5-58 % and 5.4-29 % in the Gävleborg and Jämtland respectively. The plant cover was a varying mixture of domestic grasses and natural herbs. As a consequence of these varying background conditions, the radiocaesium transfer to grass varied considerably.

Sensitive grasslands like the pastures investigated here often show high interception of fallout as well as high transfer to the grass produced and a slow reduction rate with time of the transfer (cf. also Rosén et al., 1995, a and b). K-fertilized grasslands showed lower transfers to grass already in the first year, 1986.

CONCLUSIONS

In two Swedish counties discussed here, grass production is more important than cereal production. For the farmers, the development with time of ^{137}Cs transfer, and measures to decrease it are important, both with respect to short and long-term perspectives. As regards the development with time, ^{137}Cs transfer was much higher in the year of fallout than later. As observed in the Gävleborg county ^{137}Cs transfer was significantly lower to cereal grain than to grass. In Jämtland county it was lower than to grass on permanent pasture.

^{137}Cs transfer varied with site due to soil fertility. It decreased with increased growth and K-fertilization, and by ploughing down the contaminated sward on sites where this measure was possible. While soil fertility is partly site-specific, i.e. influenced by texture and organic matter, K-fertilization increases growth and thereby counteracts the ^{137}Cs transfer to crop products, indirectly by growth dilution and directly by potassium dilution.

Ploughing decreases ^{137}Cs transfer in both arable and grass land. This stresses the importance of early ploughing after fallout and also grass production in rotation. Liberal annual application of K-fertilizer for some years during the lag period of increasing T_{ar} is recommended in agricultural systems sensitive to ^{137}Cs fallout.

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SOIL-TO-PLANT TRANSFER OF CS AND SR IN AUSTRIA AFTER THE CHERNOBYL ACCIDENT

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Abstract

In field studies after the Chernobyl fallout in Austria distinct differences in soil-to-plant transfer of ^{137}Cs and ^{90}Sr between crops were observed. However, within single plant species transfer values varied over one to three orders of magnitude. The main influencing factors are the soil properties, the soil adhesion phenomenon and probably the unhomogeneous vertical distribution of the radionuclides after plowing. On the basis of the presented results we should be aware of the uncertainties of the simple soil-to-plant transfer model, which partly mask the influence of soil parameters on root uptake and translocation.

1 Introduction

The soil is the most important compartment of the ecosystem with respect to immobilisation of radionuclides. However, in long term the soil is a source for the uptake of radionuclides into plants and thus plays a major role in modeling the ingestion dose response to large scale contaminations. The radioactive fallout in Austria after the Chernobyl accident in 1986 made studies on the behaviour of ^{137}Cs and ^{90}Sr in the soil-plant system feasible. The median values of ^{137}Cs soil contamination of Austrian federal states ranged from 4.2 kBq/m² (Vienna) to 44.1 kBq/m² (Upper Austria; Gerzabek et al. 1994). ^{90}Sr soil contamination is much lower and was measured between 0.40 and 2.0 kBq/m² (Gerzabek et al. 1991). However, the contribution of the Chernobyl fallout to the total amount of ^{90}Sr is only one quarter (Meisel et al. 1992). Therefore, in the case of strontium ^{90}Sr from the weapons fallout still has to be considered the main source for plant uptake.

2 Materials and Methods

In 1987 and 1988 soil and plant samples were taken at more than 100 sites in Lower Austria, Upper Austria, Burgenland and Styria. Transfer factors ($[\text{Bq/kg plant f.w.}]/[\text{Bq/kg soil d.w.}]$) for ^{137}Cs , ^{90}Sr and stable Sr were obtained and correlated to soil parameters. The ^{137}Cs measurements were conducted on a high purity Ge-detector with 30 % relative efficiency in a well-shielded configuration in a low-level laboratory. ^{90}Sr was determined by low-level beta measurements of its daughter nuclide ^{90}Y after separation by radiochemical precipitation according to Hermann and Erdelen (1959). Stable strontium was determined by means of ICP-AES after aqua regia extraction of the soil samples and extraction of plant ashes with 6 M HCl. Soil mass loading on plant surfaces was derived from Sc measurements using the ^{46}Sc neutron activation method (Li et al. 1994). Measures for the ^{137}Cs -extractability from soil texture fractions were obtained using the method described elsewhere (Gerzabek et al. 1992).

3 Results and Discussion

Distinct differences in ^{137}Cs transfer values (TF) occurred between the investigated crops. Zucchini and cauliflower showed TF of only 0.0004. Cereal straw exhibited the highest values (Table 1). ^{137}Cs -TF into cereal grains were 2.3, 3.4, 4.9 and 5.0 times lower compared to straw for barley, rye, wheat and maize, respectively. Within single plant species TF varied over one to three orders of magnitude. Soil parameters like pH, clay and humus content showed minor

Table 1: Transfer factors for ^{137}Cs , ^{90}Sr and stable Sr for selected crops on a plant fresh weight - soil dry weight bases

crop		^{137}Cs	median ^{90}Sr	stable Sr
maize	straw	0.0116	0.494	0.208
	grain	0.0023	0.010	0.003
wheat	straw	0.0294	0.455	0.243
	grain	0.0060	0.049	0.026
rye	straw	0.0236	0.369	0.284
	grain	0.0069	0.097	0.027
barley	straw	0.0265	0.897	0.265
	grain	0.0116	0.095	0.038
potato	shoot	0.0100	0.625	0.359
	tuber	0.0024	0.029	0.015

correlations with TF. On the other hand, TF into straw of cereals were significantly correlated with ^{137}Cs -concentrations in the soil (Table 2). This effect can be explained by an influence of soil adhesion or an intercorrelation of soil parameters and contamination levels. Due to differences between wet and dry deposition areas, resuspension of ^{137}Cs -contaminated soil particles was higher in low contaminated regions as compared to high deposition areas (Garland and Pattenden 1989). Thus, the contribution of the soil adhesion effect to plant contamination should be higher in low contaminated regions. Recent results of soil adhesion measurements (Li et al.

Table 2: Correlation of ^{137}Cs soil-to-plant transfer factors with ^{137}Cs -concentration in soil

crop	equation	correlation coefficient
barley straw	$y = 0.325 \cdot x - 0.696$	-0.86
wheat straw	$y = 0.040 \cdot x - 0.486$	-0.85
maize straw	$y = 0.022 \cdot x - 0.525$	-0.79

y : transfer factor x : Bq ^{137}Cs /kg soil

Table 3: Soil adhesion on plant shoots from field studies after the Chernobyl fallout in Austria obtained by ^{46}Sc neutron activation method and its impact on radiocaesium plant contamination (Li et al. 1994).

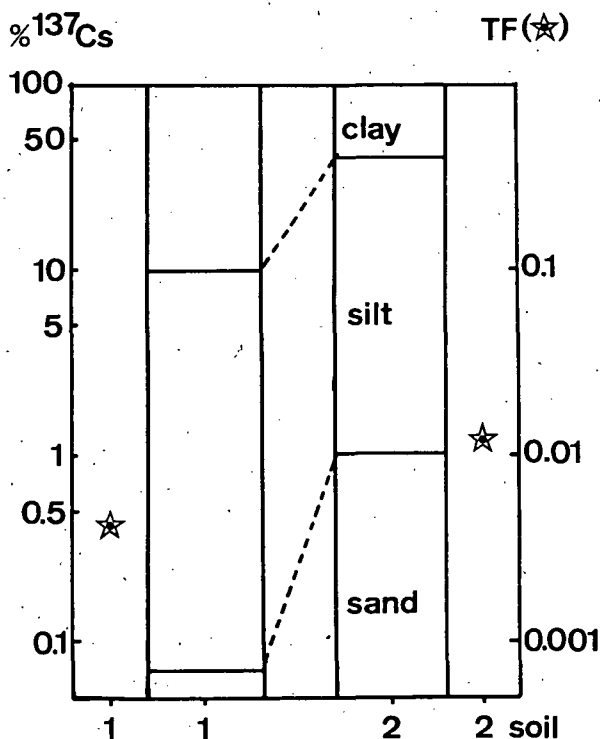
Sample	mg soil/g dry plant a)	Bq ^{137}Cs b)/kg dry plant (% of total)
barley straw	6.01	0.141 (18.34)
barley straw	1.33	0.063 (05.53)
barley straw	2.31	0.087 (03.41)
barley straw	4.23	1.328 (23.18)
wheat straw	1.16	0.023 (02.61)
wheat straw	1.53	0.223 (14.48)

a) Detection limit 0.05 mg soil/g dry plant;

b) Radioactivity derived from soil adhesion.

1994) show that e.g. for barley and wheat straw ^{137}Cs derived from outer contamination can contribute in a range from 2.6 to 23.2 % to the total ^{137}Cs content (Table 3). However, this effect seems not to be sufficient to explain the phenomenon exclusively. Low transfer values were obtained in Upper Austria, where Eutric Cambisols are dominating. High transfer values were determined on the Chernozems of Lower Austria. Despite the fact that chemical and physical properties of these soils are quite similar, differences could be shown in a particle size

Figure 1: ^{137}Cs in the texture fractions of an Eutric Cambisol (1) and a Calcic Chernozem (2) and median values of soil-to-plant transfer factors (TF).

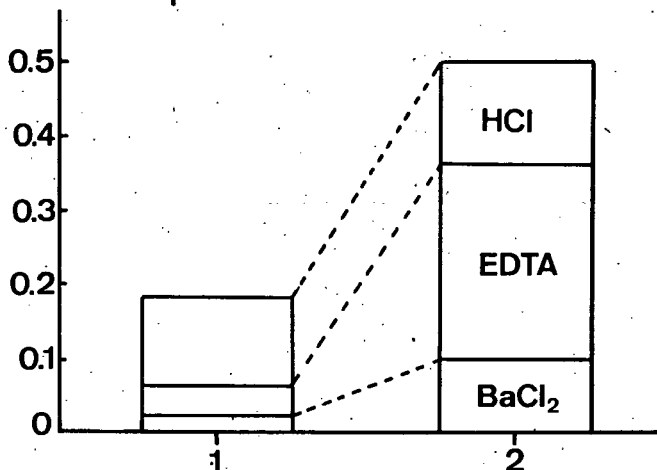


fractionation experiment (Gerzabek et al. 1992). ^{137}Cs in silt fraction was four times higher in the Calcic Chernozem than in the Eutric Cambisol (Figure 1). Extractability of radiocaesium

from the organic matter of the silt fraction was considerably higher for the Calcic Chernozem as compared to the Eutric Cambisol (Figure 2), which corresponds to the investigated transfer factors. Obviously, the total soil organic matter content does not reflect the Cs-availability of soils with exception of very high values of more than 15 % (Frissel et al. 1990). Further uncertainties in calculating plant contamination by the transfer model can be attributed to a lack of homogeneous ^{137}Cs vertical distribution in the soil after the first and second plowing, which may lead to under- or overpredictions using transfer factors based on a well defined contaminated soil layer (Meisel et al. 1991).

Figure 2: Extractable radiocaesium in the silt fraction of an Eutric Cambisol (1) and a Calcic Chernozem (2) in Austria.

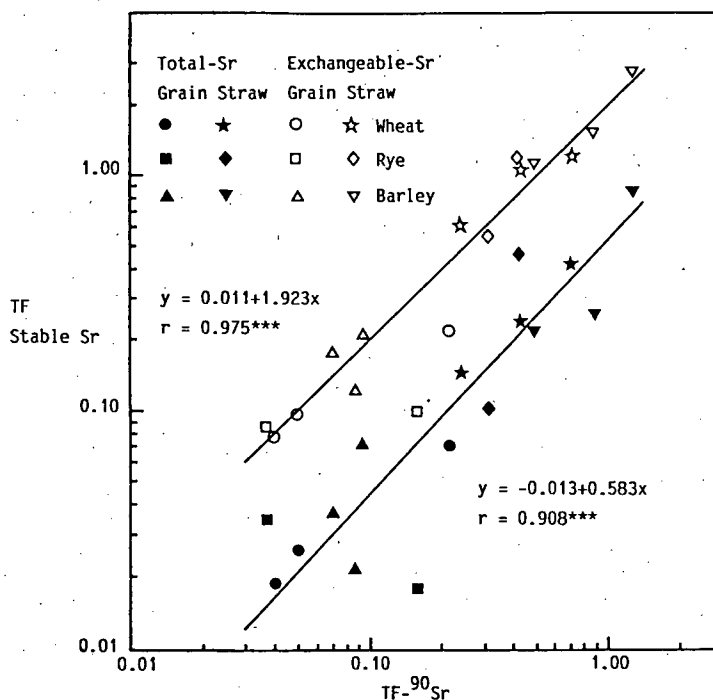
% of bulk sample



^{90}Sr -TF for cereal grains were approximately 3 to 14 times higher than the respective values for ^{137}Cs obtained for the same samples (Table 1). Maize straw exhibited a 50 times higher TF compared to the grains, which is the ten fold discrimination compared to ^{137}Cs . This effect is due to the clearly lower translocation rate of strontium. Translocation occurs to plant parts just in development. Thus, vegetative plant parts are supplied by the transpiration flow with ^{90}Sr during the whole growing period, grains only during a significant shorter time span.

In all cases soil-to-plant transfer of stable strontium was distinctly lower compared to the radioactive isotope. The TF for total stable strontium amounted on average only up to 60% of the respective values for ^{90}Sr (Figure 3). The respective linear regression was statistically significant ($r = 0.908^{***}$). Thus, natural stable Sr was less plant available than ^{90}Sr from the Chernobyl and atomic bomb fallout. Statistical analysis showed a satisfactory correlation between exchangeable soil calcium and ^{90}Sr -TF. Due to ion competition Sr-uptake diminishes with increasing exchangeable Ca-contents of the soil. Soil adhesion obviously had no significant impact on Sr-TF.

Figure 3: Relation between ^{90}Sr -transfer factors and plant/soil concentration ratios for stable Sr (Gerzabek et al. 1992)



4 Conclusion

We have to be aware of uncertainties of the simple soil-to-plant transfer model, which partly mask the influence of soil parameters on root uptake and translocation. Special attention should be given to the mobile radionuclide fraction, the radionuclide depth distribution in the

soil during three years after fallout and the soil adhesion phenomenon in the case of radionuclides with low plant availability and low TF values.

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SOIL FACTORS INFLUENCING THE LONG-TERM TRANSFER OF ^{90}Sr AND ^{137}Cs TO ARABLE CROPS

Results from experimental studies under Swedish field conditions

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ABSTRACT

The transfer of ^{137}Cs and ^{90}Sr from arable soils to barley and peas has been experimentally studied in microplot experiments during 1961-1980. The experiment included the combination of 12 topsoils and 2 subsoils. Clear differences in the TFg were found between nuclides, crop products, topsoils and subsoils, and, furthermore, there were strong interactions between the parameters studied. The TFg were a factor 5-10 higher for ^{90}Sr than for ^{137}Cs and for both nuclides higher for grain of peas than for grain of barley. A high availability of Ca and K in the subsoil lowered the transfer of ^{90}Sr and ^{137}Cs by a factor of up to 2 as compared with unfavourable conditions in this respect. The TFg for ^{137}Cs were found to decrease with increasing contents of clay and potassium in the topsoils but increased with increasing content of organic matter. For ^{90}Sr the TFg decreased with increasing pH, CEC and Ca-saturation of the topsoils. The long-term trend in the TFg differed between the nuclides and was strongly influenced by the feeding of Ca and K in the subsoil.

INTRODUCTION

The testing of nuclear weapons in the 1950s and early 1960s initiated extensive studies on the transfer of the long-lived nuclides ^{90}Sr and ^{137}Cs from soils to crops and further to man. At that time the detection techniques, in combination with the low deposition levels, limited the use of environmental monitoring or conventional field experiments for studies of the influence of soil type and fertilisation practice on the transfer of the nuclides from soil to crops under long-term field conditions. Therefore, in Sweden a new field experimental technique, using microplots and artificial contamination of the soils studied, was introduced

(Fredriksson, 1963).

Results from an investigation using this technique, performed during 1961-1980, will be presented and discussed below with respect to the influence of topsoil and subsoil characteristics on the long-term transfer of the two nuclides to arable crops. Data from parts of the experimental period have been published earlier (Fredriksson et al., 1969; Haak & Lönsjö, 1975). A more detailed final paper is in preparation (Lönsjö & Haak, 1995).

MATERIALS AND METHODS

In this investigation 12 topsoils, representing main agricultural districts of Sweden, were sampled from the plough-layer, homogenised and uniformly labelled with carrierfree solutions of ^{90}Sr or ^{137}Cs . The contaminated soils were placed in steel frame works, enclosing single microplots of 0,25 m², on two different subsoils, one artificial subsoil of sand to 1 m depth and the subsoil *in situ*, a heavy clay. Each soil-nuclide combination was replicated 4 times. In total the experiment comprised 192 plots. Details on the experimental technique, lay out of the experiment etc. were given by Fredriksson (1963).

The topsoils covered a wide range with

Table 1. Characteristics of the topsoils (1-12) and subsoils (I-II)

Soil	pH-H ₂ O	SOM %	Clay %	Ca _{AL} mequiv/100 g	CEC
Topsoils					
1	6.3	1.7	5.1	6.8	6.6
2	5.3	4.0	7.1	3.8	6.9
3	7.5	2.5	10.5	22.0	11.7
4	7.3	2.8	17.5	14.5	15.0
5	6.0	5.4	17.6	9.5	11.8
6	5.6	5.2	29.7	11.5	19.1
7	6.1	5.3	33.3	10.2	15.8
8	6.8	3.3	37.0	12.5	15.5
9	7.0	3.6	40.0	15.0	17.8
10	6.7	2.8	41.1	14.5	17.9
11	5.2	6.6	48.1	10.0	20.1
12	7.4	5.7	50.0	37.5	40.0
Subsoils					
I	6.9	-	4.9	4.6	3.8
II	7.6	-	48.5	16.0	16.2

respect to soil type and physico-chemical properties (Table 1). The ranges were for contents of clay and organic matter 5-50 and 1.7-6.6 per cent, for pH 5.2-7.5, and for CEC and easily exchangeable Ca (Ca_{AL}) 7-40 and 4-38 mequiv. per 100 g of soil, respectively. The subsoils differed, besides in the clay content, also in contents of exchangeable Ca and K (Table 1). More soil data were given by Fredriksson et al. (1969).

The soils were annually fertilised with N, P, K, Mg and S. The crops, barley and peas, were grown to maturity. The harvested material was dried artificially, threshed and ground. The ^{90}Sr -contents were determined by GM-counting, using the briquette technique, and the ^{137}Cs -contents by gamma-spectrometry and a 4x4" NaI-detector. The measurement errors were usually in the range of <1-3 per cent. Chemical analyses on Ca and K were performed using conventional methods. All data refer to dry weight (DW) basis. The activity contents were corrected for decay to the start of the experiment in May 1961 and normalised to the activity deposited at that time, resulting in the transfer factor TFg, i.e.:

$$\text{TFg} = \frac{\text{Bq (kg DW)}^{-1}}{\text{Bq m}^{-2}}$$

with the dimension $\text{m}^2 (\text{kg DW})^{-1}$ (Eriksson 1977, IAEA, 1987). The TFg-data were subjected to analyses of variance and multiple regression according to methods described in the references earlier cited.

RESULTS AND DISCUSSION

Differences due to nuclide and crop type

The soil-plant transfer of radionuclides differs between species and also between cultivars of one and the same species growing under different environmental conditions. Differences among species are caused by a combination of plant characteristics and environmental factors as reviewed by Davis (1963). The annual TFg-values for grain of barley in 1961-80 and for grain of peas in 1961-68, averaged over the 12 topsoils, are shown for the two subsoils in Table 2. As can be seen the TFg for as well ^{90}Sr as ^{137}Cs varied considerably from one year to another. The transfer of ^{90}Sr to barley grain was 5-10 times higher than that of ^{137}Cs , the difference being strongly influenced by the subsoil type. A low or high annual transfer of ^{137}Cs was not necessarily accompanied by a corresponding high or low transfer of ^{90}Sr and *vice versa* (Table 2). This indicates that the relative availability of the two nuclides has varied due to unequal rooting and uptake of Ca and K from topsoil and subsoil in different years. The transfer of ^{137}Cs to grain of peas was about a factor 5 higher than to grain of barley, while for ^{90}Sr the transfer was about the same (Table 2). These differences were due to a much higher content of potassium in grain of peas than in grain of barley, thus

also giving a higher TFG for ^{137}Cs , while the corresponding contents of calcium and hence also the ^{90}Sr -contents were almost the same in the two crop products (compare Fredriksson et al., 1969 and Haak & Lönsjö, 1975).

Effects and time trends due to the subsoil

As can be seen in Table 2 and Fig. 1 the long-term trends in the nuclide transfer were different for as well ^{137}Cs and ^{90}Sr as for the subsoils. The variation in the annual TFG-values was mainly due to differences between single years in the rooting depth and hence in the uptake of Ca and K from the subsoils, resulting in unequal dilutions of ^{90}Sr and ^{137}Cs respectively in the crop products (Haak & Lönsjö, 1975).

For ^{90}Sr the annual TFG-values were throughout the period higher on Subsoil I than on Subsoil II both for barley and peas (Table 2). Also the yields and the Ca-contents were significantly higher on the former subsoil (data not shown; compare Haak & Lönsjö, 1975), which indicates that the growing conditions have been more favourable on the sandy subsoil than on the compact clay subsoil. Despite the lower Ca-contents in the grain on the clay subsoil, a relatively larger proportion of calcium may have been taken up from this subsoil than from the sandy subsoil during the grain formation. This unequal uptake of calcium from the two subsoils

Table 2. Annual Tfg for ^{137}Cs and ^{90}Sr in grain of barley and peas, averaged over the 12 topsoils, on Subsoil I and Subsoil II, $\text{m}^2 \text{kg DW}^{-1}$

Year	^{137}Cs		^{90}Sr	
	I	II	I	II
Barley				
1961	0.030	0.054	0.58	0.46
1962	0.038	0.044	0.27	0.21
1963	0.050	0.065	0.56	0.46
1964	0.028	0.045	0.33	0.28
1965	0.041	0.057	0.42	0.32
1966	0.048	0.074	0.33	0.32
1967	0.035	0.063	0.18	0.10
1968	0.023	0.032	0.34	0.22
64-68	0.036	0.054	0.38	0.30
1969	0.047	0.042	0.48	0.17
1970	0.066	0.053	0.52	0.25
1971	0.047	0.036	0.47	0.22
1972	0.048	0.048	0.55	0.19
69-72	0.052	0.045	0.50	0.21
1974	0.066	0.030	0.54	0.28
1975	0.058	0.034	0.40	0.19
1976	0.045	0.028	0.32	0.15
1977	0.034	0.024	0.54	0.23
1978	0.024	0.019	0.52	0.23
1979	0.028	0.022	0.41	0.21
1980	0.029	0.023	0.44	0.22
74-80	0.041	0.042	0.45	0.22
61-80	0.041	0.042	0.43	0.25
Peas				
1961	0.154	0.176	0.67	0.46
1962	0.192	0.238	0.25	0.27
1963	0.180	0.289	0.37	0.43
1964	0.222	0.269	0.31	0.30
1965	0.206	0.246	0.24	0.26
1966	0.321	0.322	0.33	0.32
1967	0.242	0.229	0.25	0.20
1968	0.176	0.147	0.28	0.20
61-68	0.212	0.240	0.34	0.31

explains the lower ^{90}Sr -contents and $^{90}\text{Sr}/\text{Ca}$ -ratios on the clay subsoil (Haak & Lönsjö, 1975).

As illustrated in Fig. 1:a the ^{90}Sr -transfer to barley on Subsoil I increased irregularly with year during the period, while on Subsoil II a slight and more regular decrease with year was obtained. This difference may partly be due to a slight depletion of major nutrient elements, such as P and K, in some of the topsoils. On the clay subsoil the crops seem to have compensated this depletion by an increased nutrient uptake from this subsoil. The sandy subsoil, relatively poor in nutrients (Table 1), has not been available to the same extent by the crops, which has resulted in eventually decreasing yields and hence in increasing transfer of ^{90}Sr .

Although the averaged TFg for ^{137}Cs in barley during 1961-80 were almost the same on the two subsoils (Table 2), the time trends varied considerably during the course of the period as can be seen in Fig. 2:b. During the first 8 years of the experimental period the more favourable growing conditions on Subsoil I, giving higher yields and higher uptake of potassium, thus resulted in a lower transfer of ^{137}Cs as compared with Subsoil II. Later a negative potassium balance took place in some of the topsoils, which increased the average transfer of ^{137}Cs . This depletion seems to have been compensated for by uptake of potassium from the subsoil, whereby a much better effect was obtained on the clay subsoil than on the sandy subsoil. Therefore, a faster decrease with time and lower TFg-values were obtained on Subsoil II than on Subsoil I. Other effects, such as ageing, may also have contributed to the decreasing transfer of ^{137}Cs at the end of the period.

Effects of topsoils and topsoil/subsoil combination

A wide and consistent variation in the TFg among the 12 topsoils was recorded for both nuclides (Table 3). Thus for the transfer of ^{137}Cs to grain of barley this variation amounted to a factor of about 6 on Subsoil I and to a factor of up to 16 on Subsoil II. For peas a similar variation and relation between the subsoils was found. For ^{90}Sr the variation was somewhat less apparent, a factor of ca. 3.3 for barley, relatively independent of the subsoil, and a factor of 4.8 and 6.5 for peas on Subsoil I and II respectively (Table 3; compare Fredriksson et al., 1969; Haak & Lönsjö, 1975).

As shown by Fredriksson et al. (1969) the soil-plant transfer of ^{137}Cs increased with the content of soil organic matter and decreased with the content of clay (or with the content of slightly soluble potassium, K_{HCl} , which is strongly intercorrelated with the clay content). These relations, based on data for barley during the period 1961-68, are shown in Fig. 2. However, also a low content of easily soluble calcium and hence a low pH of the soil will increase the transfer of ^{137}Cs to the crop. This is illustrated by the data for Topsoil 2 (Table 1), which all favour an extremely high and consistent transfer of ^{137}Cs (and also of ^{90}Sr , see below).

Haak and Lönsjö (1975) showed that the transfer of ^{90}Sr decreased with the soil content of easily soluble calcium (Ca_{AL}) and also by pH and CEC. These relations are shown in Fig. 3. As shown by the curves in Figures 2-3, a certain increment in contents of clay or in CEC causes a large decrease in the transfer of ^{137}Cs or ^{90}Sr at low levels but small ones at increasing levels. This also means that the effect of K-fertilisation or liming in order to reduce the nuclide transfer will be most effective on soils with a low clay content or with a low Ca-saturation degree.

Table 3. Transfer of ^{137}Cs and ^{90}Sr to grain of barley and peas, averaged over the years 1961-68, on Subsoil I and Subsoil II, $\text{m}^2(\text{kg DW})^{-1} \cdot 10^{-3}$

Top-soil no	Barley		Peas		Barley		Peas	
	I	II	I	II	I	II	I	II
	TFg for ^{137}Cs				TFg for ^{90}Sr			
1	0.026	0.041	0.227	0.240	0.56	0.41	0.36	0.38
2	0.077	0.121	0.572	0.772	0.60	0.43	0.82	0.78
3	0.025	0.043	0.124	0.118	0.44	0.29	0.25	0.25
4	0.016	0.024	0.153	0.135	0.26	0.20	0.20	0.19
5	0.077	0.117	0.370	0.435	0.47	0.34	0.33	0.34
6	0.041	0.054	0.236	0.224	0.41	0.36	0.43	0.37
7	0.045	0.070	0.250	0.300	0.31	0.29	0.28	0.28
8	0.038	0.051	0.136	0.172	0.33	0.27	0.34	0.24
9	0.041	0.064	0.138	0.161	0.31	0.26	0.32	0.23
10	0.016	0.015	0.074	0.068	0.24	0.22	0.22	0.20
11	0.022	0.035	0.170	0.171	0.40	0.34	0.36	0.31
12	0.014	0.018	0.089	0.079	0.18	0.13	0.17	0.12
Mean	0.036	0.054	0.212	0.240	0.38	0.30	0.34	0.31

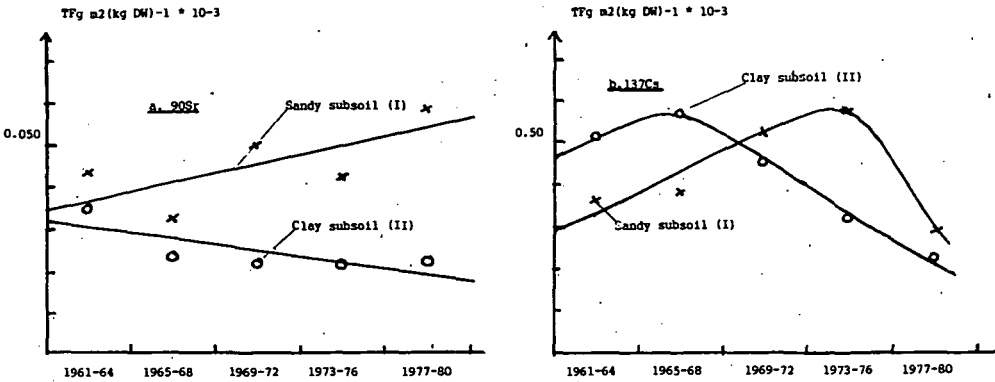


Figure 1:a-b. Time trends in the transfer of ^{90}Sr (a) and ^{137}Cs (b) to grain of barley during 1961-1980. The annual TFg-values in Table 2 are averaged over each 4-year period.

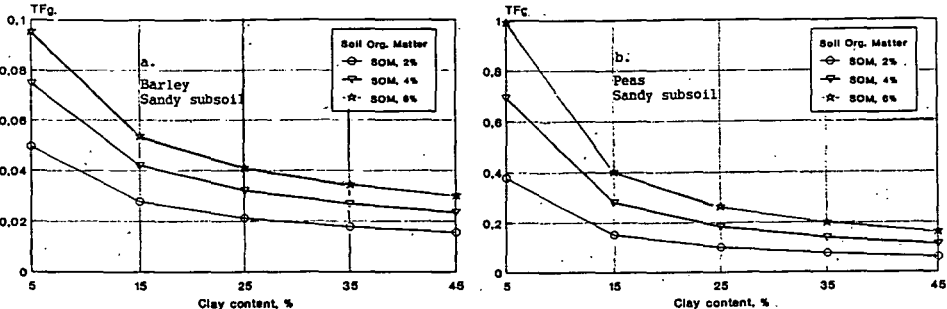


Figure 2:a-b. Expected transfer of ^{137}Cs to barley and peas, related to the contents of clay and organic matter in the topsoils. Calculations based on data in Tables 1 and 3 (compare text and Fredriksson et al., 1969).

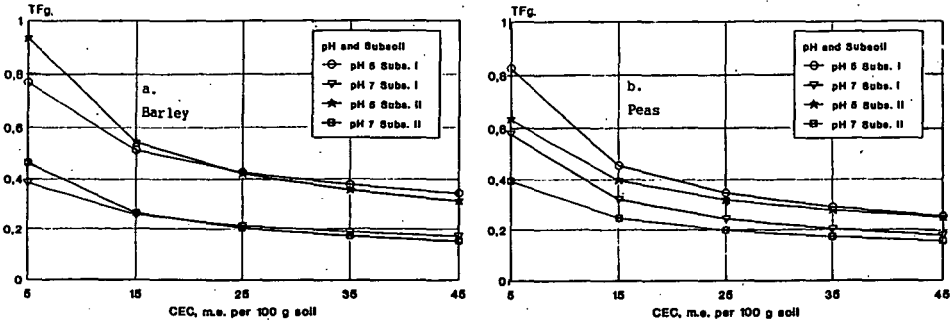


Figure 3:a-b. Expected transfer of ^{90}Sr to barley and peas, related to CEC and pH in the topsoils. Calculations based on data in Tables 1 and 3 (compare text and Haak & Lönsjö, 1975).

CONCLUSIONS

The following may be concluded from the experimental data reported:

- On arable soils, uniformly contaminated in the plough-layer, the TFG-values for ^{90}Sr will usually be a factor of 5 to 10 higher than for ^{137}Cs depending on the crop cultivated and on the root availability and nutrient supply in the subsoil.
- The transfer of ^{137}Cs and ^{90}Sr will strongly be influenced by uptake of K and Ca respectively from the subsoil. As shown in the experiments the higher uptake of these elements from the subsoil, the lower nuclide contents are found in the crop products. The two nuclides do not necessarily show the same relation in TFG on different topsoil/subsoil combinations.
- Increasing content of clay in the topsoils will usually decrease the transfer of both ^{137}Cs and ^{90}Sr , while increasing content of organic matter will increase the ^{137}Cs -transfer and decrease the ^{90}Sr -transfer. With increasing pH and Ca-saturation the transfer of both nuclides will decrease, the effect usually being more clear for ^{90}Sr than for ^{137}Cs .
- The long-term trends in the nuclide transfer to the crops will be influenced by the nutrient status in both topsoil and subsoil. The maintenance of a positive potassium balance in the topsoils will result in a sustainable low ^{137}Cs -transfer with the years. It is also of importance that acid soils are limed in order to prevent an increased transfer of ^{90}Sr . Such measures will be most effective when a negative influence of the subsoil can be expected.

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LONG-TERM TRANSFER OF ^{137}Cs AND ^{90}Sr TO GRASS ON CONTRASTING TYPES OF SWEDISH PASTURES

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ABSTRACT

In 1961 two contrasting types of Swedish pastures were artificially contaminated with ^{90}Sr and ^{137}Cs . One site, Lövsta, was a natural pasture on a sandy soil. The other, Risslinge, was a grazing ley on a clay soil. On both sites field experiments had been running for 10 years. Unique possibilities, to study the long term transfer to grass under equilibrated field conditions, were thus at hand. Results of the two experiments for the period 1961-1981 are treated in this article.

In years of fallout direct deposition was the main transfer to grass. It was larger on the permanent natural pasture than on the grazing ley. For some years after a radioactive inventory, or during a lag period, the nuclides are retained in the surface mat zone of pastures and more available for uptake.

The type of pasture has large impact on the behaviour of the radionuclides. The lag period is longer for ^{137}Cs than for ^{90}Sr . TF_g decreases from year to year due to downward migration, which is always effective at surplus precipitation between growth periods. Although the migration of ^{90}Sr is larger than of ^{137}Cs the transfer to grass is much higher for ^{90}Sr than of ^{137}Cs of all types of pasture on mineral soils.

Liming and fertilization has a large impact on nuclide transfer to grass on permanent pastures established on coarse soils. Use of acidifying N fertilizers, AS, may give a $^{90}\text{Sr}/\text{Ca}$ -ratio twice that of CaN. Fertilization with AS increases the leaching of ^{90}Sr and thereby decreases the $^{90}\text{Sr}/\text{Ca}$ -ratios in the long run. On coarse soils the ^{137}Cs transfer to grass is lower on P- and K- fertilized pasture than on unfertilized pasture.

INTRODUCTION

Direct deposition on growing crops is the main way of radionuclide transfer in a fallout situation. A considerable fraction of ^{137}Cs and ^{90}Sr can be retained by the crops. Rainfall may effectively reduce this contamination but the nuclide transfer to crops will be much higher in the year of fallout than later (Rosén, et al., 1995).

Grass crops are sensitive for transfer during some years after the radioactive inventory. The main reason is the nuclide contamination of the surface mat zone, which consists of the stubble and grass sward created during the pasture or ley establishment and during regrowth of grass year by year. As a consequence radionuclides are then easier transferred to grass than to arable crops.

Results from the first six years (1961-1966) of the two experiments to be presented here, have been treated in detail in (Haak et al. 1973). The aim of this article is to summarize these experiments and their extension up to 20 years, i.e. between 1961-1981. We also discuss countermeasures to decrease the nuclide transfer to grass.

MATERIALS AND METHODS

In Sweden two types of grassland are used for grazing. One type is permanent natural pasture with low yield potential due to meagre soil and a mixture of wild grass and other plant species. The other type is grazing ley on arable fertile soils, with high yield potential due to domestic grass species. Liming and fertilization are also less frequent on natural pastures than on grazing leys.

To cover this range of soil and growth conditions, two sites were selected in 1961, where microplots were artificially contaminated with ^{90}Sr and ^{137}Cs . One site, Lövsta, was a natural pasture on a sandy soil. The other, Risslinge, was a grazing ley on a clay soil. On both sites field experiments had been running for 10 years. The fertilizers were superphosphate (SSP), muriate of potash (MUK), ammonium sulphate (AS), calcium nitrate (CaN) and ammonium nitrate (AN).

Grass harvesting and N-fertilization, 31 kg/ha N, took place four times per year in

Table 1. Design of experiments

Treatm. Fertilization, amounts per hectare

<u>Lövsta, permanent pasture</u>	
A.	Unfertilized
B.	300 kg/ha SSP + 31 kg/ha N as AS
C.	300 kg/ha SSP + 150 kg/ha MUK + 31 kg/ha N as CaN
D.	31 kg/ha N as AN
E.	300 kg/ha SSP + 150 kg/ha MUK
a. & c.	Unlimed
b.	4 tons/ha CaCO_3
d.	4 tons/ha $\text{Ca}(\text{OH})_2$
<u>Risslinge, grazing ley</u>	
A.	Unfertilized with P and K
B.	150 kg/ha SSP
C.	150 kg/ha SSP + 75 kg/ha MUK
D.	300 kg/ha SSP + 150 kg/ha MUK

1950-60, 23 times in total during 1961-66, and later two times per year. From 1961 onwards one third of each PK-plot at Risslinge received N-fertilizers as CaN and two thirds as AN, in both cases to each harvest.

For transfer of nuclides the observed activity contents in herbage, Bq per kg DW, has been normalized to transfer factors, as initially defined by Eriksson (1977).

$$TF_s = \frac{Bq(kg\ DW)^{-1}}{Bq\ m^{-2}} = m^2(kg\ DW)^{-1} \quad (1)$$

The TF_s -values thus express the nuclide transferred per kg dry weight of grass from a deposition of 1 Bq per m^2 ground. TF_s is a suitable transfer unit for comparison between sites and treatments.

Reduction with time in nuclide transfer to crops, independent of decay, is observed in most terrestrial environments due to reduced plant availability. An ecological half-time, T_{ec} , has often been calculated from the general exponential equation

$$Y_t = Y_0 * \exp(A*t) \quad (2)$$

As identified in these experiments and later in field surveys of ^{137}Cs after the Chernobyl accident (Rosén et al, 1996), annual reduction in nuclide transfer is not constant on grassland during a lag period after fallout. Instead an equation of the type

$$f(t) = \exp(A*t^N + B) \quad (3)$$

can be used (Eriksson, 1994), where A, N and B are constants and t years, as obtained by least square fit of observed annual mean TF_s values. From Eq. 3 the annual reduction rate in nuclide transfer T_{ar} , can be calculated. The T_{ar} concept depends on the real annual reduction in nuclide transfer.

RESULTS AND DISCUSSION

Effect of nutrient regimes on soil fertility

In 1967 soil profiles were taken at both sites to quantify the effect of liming and repeated fertilization on soil properties. The most striking feature was the high contents of soil organic matter and easily soluble nutrients in the surface zone. This reflects the typical situation on grassland sites, which in this case has impacts on nuclide uptake and transfer to grass. The reason that the contents of organic matter and nutrients were higher in the surface mat zone at Lövsta than at Risslinge, relates back to the time of establishment of pasture - Lövsta centuries, Risslinge 10 years ago, but also to differences in plant species.

Nuclide transfer to grass

Table 2 and Table 3 give an overview of the TF_g -values obtained for the transfer of ^{137}Cs and ^{90}Sr to grass, presented as annual means and ranges for single cuts taken per year. As can be seen, the transfer of ^{137}Cs was much less than that of ^{90}Sr . This is the general case on mineral soils.

Effect of year

There was a larger reduction in transfer of ^{137}Cs than of ^{90}Sr in 1961-1963. The reduction in ^{137}Cs was larger on the Lövsta natural pasture than on the Risslinge grazing ley. After recontamination with ^{137}Cs in 1964 the difference between the two sites was repeated.

The differences between nuclides and sites are due to the much thicker stubble and grass sward at Lövsta than at Risslinge.

The transfer of ^{137}Cs and ^{90}Sr to grass continued to decline, as displayed in Figure 1-4, also during the long-term period and relatively more at Lövsta than at Risslinge. This decline is to be attributed to increased contact between nuclide and mineral soil surfaces, the contact being more effective at Risslinge than at Lövsta.

The nuclide transfer did not always decrease with year, which indicates unequal rooting depth relative to nuclide distribution in the soil profile. The long-term reduction is shown in Figure 1, as evaluated by Eq. 3. The annual reduction in transfer decreased from year to year, as also is shown by the increase of the inverse relation, Tar .

Effect of site

The differences in transfer between the nuclides and sites were confirmed in the years 1972-1981. The average transfer of ^{137}Cs was six times higher and of ^{90}Sr three times

Table 2. Mean TF_g of ^{137}Cs and ^{90}Sr in grass on Lövsta, $m^2(\text{kg DW})^{-1} \times 10^3$. Means and ranges per year 1961-1981

Year	^{137}Cs		^{90}Sr	
	Mean	Range	Mean	Range
1961	22.2	17.4-29.6	73.0	54.7-89.4
1962	6.85	3.52-10.4	50.4	35.3-76.1
1963	3.48	1.52-4.89	33.0	17.0-51.6
1964 ^a	31.1	14.6-39.4	24.8	11.5-37.2
1965	15.3	11.5-18.9	21.9	7.4-35.4
1966	7.16	1.55-10.1	18.7	8.1-30.2
1967	2.48	1.19-3.79	18.4	6.7-31.4
1968	1.86	1.02-2.86	18.3	8.5-30.9
1969	1.10	0.60-2.29	13.7	6.2-23.7
1970	0.68	0.32-1.72	15.3	6.4-28.2
1971	0.83	0.30-1.94	14.6	5.3-27.2
1972	0.96	0.58-2.20	10.9	3.1-17.7
1973	0.70	0.21-1.65	11.8	4.0-18.4
1974	0.74	0.46-1.58	12.8	4.0-15.5
1975	0.72	0.38-3.48	13.7	2.8-17.9
1976	0.78	0.19-2.72	10.8	3.0-16.9
1977	0.67	0.15-4.37	10.3	2.5-14.4
1978	0.82	0.23-2.15	12.2	2.6-21.5
1979	0.82	0.16-1.64	12.0	1.8-23.2
1980	0.53	0.07-0.76	12.6	3.3-27.2
1981	0.44	0.24-0.90	10.7	2.6-24.0
Mean 72-81	0.72	0.27-2.14	11.8	3.0-19.7

^a Recontaminated with ^{137}Cs in 1964

higher at Lövsta than at Risslinge. The transfer of ^{90}Sr was 16 and 33 times higher than that of ^{137}Cs at Lövsta and Risslinge respectively. This shows the higher fixation of ^{137}Cs to heavy clay soil and the higher dilution of ^{90}Sr with calcium at Risslinge than at the coarse sandy soil at Lövsta.

Effects of fertilization

The TF_g -values in Table 2 and Table 3 show the range of the effects of liming and fertilization at Lövsta and of fertilization at Risslinge. As expected the effects were much larger on the low buffered sandy soil at Lövsta than on the well-buffered clay soil at Risslinge. Treatment effects are presented as means for the years 1967-1971 and 1972-1981 in Table 4. As seen the reduction in transfer between the two periods was higher for ^{137}Cs than for ^{90}Sr and varied with treatment. The means for sites decreased with 50 and 30 % respectively. The reduction was about the same for Tfg of $^{90}\text{Sr}/\text{Ca}$ as for $^{90}\text{Sr}/\text{DW}$.

The reduction in nuclide transfer with year varied with fertilization at Lövsta and was not the same for ^{90}Sr as for ^{137}Cs . For ^{137}Cs the order was

$\text{E} < \text{A} < \text{B} < \text{C} < \text{D}$ in 1967-1971 and

$\text{E} < \text{C} < \text{B} < \text{A} < \text{D}$ in 1972-1981.

It was lowest for P- and K-fertilized pasture (E), and highest for pasture only receiving AN (D). For ^{90}Sr the order was $\text{B} < \text{C} < \text{D} < \text{E} < \text{A}$ in 1967-1971 and $\text{B} < \text{C} < \text{E} < \text{D} < \text{A}$ in 1972-1981. For $^{90}\text{Sr}/\text{Ca}$ the order was $\text{C} < \text{E} < \text{B} < \text{D} < \text{A}$ and $\text{C} < \text{B} < \text{E} < \text{D} < \text{A}$ respectively. For both Tfg it was highest for unfertilized pasture (A).

In treatments limed with CaCO_3 (b) and with $\text{Ca}(\text{OH})_2$ (d), the Tfg of ^{90}Sr and $^{90}\text{Sr}/\text{Ca}$ decreased during both periods, as can be seen by comparison with unlimed pasture, a and c respectively. As seen liming did not have the same consistent influence on the Tfg of ^{137}Cs probably due to a fertility gradient across the site.

Table 3. Mean TF_g of ^{137}Cs and ^{90}Sr in grass on Risslinge, $\text{m}^2(\text{kg DW})^{-1} \times 10^3$. Means and ranges per year 1961-1981

Year	^{137}Cs		^{90}Sr	
	Mean	Range	Mean	Range
1961	9.25	7.39-11.2	23.2	31.3-26.1
1962	0.71	0.54-1.06	15.8	15.1-16.7
1963	0.41	0.33-0.64	11.8	11.4-12.2
1964 ^a	7.93	6.10-11.3	9.9	9.4-10.5
1965	1.37	0.94-2.22	9.7	9.2-10.3
1966	0.80	0.58-1.10	8.7	8.3-8.9
1967	0.35	0.27-0.52	6.6	6.3-6.9
1968	0.24	0.22-0.30	6.1	5.5-6.9
1969	0.10	0.08-0.14	5.1	5.0-5.3
1970	0.11	0.08-0.14	5.5	5.4-5.7
1971	0.18	0.16-0.22	5.7	5.2-5.9
1972	0.18	0.14-0.20	5.4	5.2-5.6
1973	0.10	0.07-0.13	3.8	3.6-3.9
1974	0.22	0.17-0.29	4.5	4.2-4.9
1975	0.08	0.04-0.11	4.2	4.0-4.4
1976	0.06	0.03-0.07	2.8	2.7-3.0
1977	0.06	0.03-0.08	3.6	3.5-3.8
1978	0.13	0.06-0.20	3.5	3.2-3.6
1979	0.10	0.05-0.14	3.5	3.3-3.6
1980	0.12	0.06-0.17	4.8	4.2-5.1
1981	0.10	0.05-0.15	3.6	3.3-4.3
Mean 72-81	0.12	0.07-0.15	4.0	3.7-4.2

^a Recontaminated with ^{137}Cs in 1964

The low transfer of ^{90}Sr and ^{137}Cs at Risslinge was accompanied by small but consistent effects of fertilization. Thus fertilization with P increased (B) and fertilization with K in addition to P (C-D) decreased the nuclide transfer. Fertilization with CaN for ^{90}Sr and for ^{137}Cs (a and b) gave lower T_f than fertilization with AS (c).

Migration of nuclides

In 1967 migration or leaching of the two nuclides was determined in soil profiles at both sites. It was repeated in 1981 at Risslinge. The migration is shown by median depths in Tables 5 and 6 on the next page. Except for treatment B at Lövsta, where acidification of the soil by AS increased migration and more of ^{90}Sr than of ^{137}Cs , no large effect of fertilization was observed (Table 1). The lower median depth of ^{137}Cs in the sandy soil at Lövsta than at the clay soil at Risslinge was unexpected. The much thicker stubble and grass sward at Lövsta than at Risslinge may in some way have interacted.

CONCLUSIONS

Results at the two experimental sites and of the fertilizer and liming treatments employed, can be considered to cover a considerable range of pastures on mineral soils. This makes it possible to estimate the potential transfer of ^{90}Sr and ^{137}Cs in a long-term situation, which fallout on pasture land may give rise to.

In years of fallout direct deposition is the main transfer to grass. Later, plant base absorption is important for transfer on all types of pastures. It is larger on permanent natural pastures than on grazing leys. For some years after a radioactive inventory, or

Table 4. Effect of nutrient regime on transfer of ^{137}Cs and ^{90}Sr , $\text{m}^2(\text{kgDW})^{-1} \times 10^{-3}$ and $^{90}\text{Sr}/\text{Ca}$, $\text{m}^2(\text{g Ca})^{-1} \times 10^3$ at Lövsta and Risslinge 1967-1971 and 1972-1981

Treat- ment	^{137}Cs		^{90}Sr		$^{90}\text{Sr}/\text{Ca}$	
	67-71	72-81	67-71	72-81	67-71	72-81
<u>Lövsta natural pasture on sandy soil</u>						
A	1.09	0.60	27.5	20.4	3.07	2.02
B	1.26	0.65	7.2	3.0	2.32	0.84
C	1.47	0.44	10.6	8.0	1.23	0.71
D	2.52	1.63	15.7	14.7	2.35	1.68
E	0.73	0.28	20.0	12.7	2.11	1.23
a	1.73	0.82	23.8	15.3	3.03	1.64
b	1.58	0.79	13.5	10.3	1.79	1.13
c	0.94	0.61	15.9	11.2	2.15	1.32
d	1.32	0.66	11.2	10.5	1.58	1.06
Mean	1.39	0.72	16.1	11.8	2.19	1.32
<u>Risslinge, grazing ley on clay soil</u>						
A	0.20	0.14	9.6	5.7	0.61	0.42
B	0.26	0.15	5.8	4.2	0.66	0.44
C	0.17	0.08	6.0	4.0	0.61	0.43
D	0.15	0.07	5.7	3.8	0.57	0.40
a	0.16	0.10	5.5	3.8	0.61	0.40
b			5.5	3.9	0.55	0.40
c	0.24	0.12	6.4	4.2	0.69	0.47
Mean	0.20	0.11	5.8	4.0	0.61	0.42

during a lag period, the nuclides are retained in the surface mat zone and more available for uptake than later, when soil-plant transfer increases relatively with year.

The type of pasture has large impact on the behaviour of the radionuclides. The migration of especially ^{137}Cs can be less and the transfer definitely larger and of longer duration on permanent pastures than on grazing leys. The lag period is longer for ^{137}Cs than for ^{90}Sr . TF_s decreases from year to year due to downward migration,

which is always effective at surplus precipitation between growth periods. Although the migration of ^{90}Sr is larger than of ^{137}Cs the transfer to grass is much higher for ^{90}Sr than for ^{137}Cs on all types of pasture on mineral soils.

Some years after fallout the soil properties largely determine the nuclide transfer to grass. The $^{90}\text{Sr}/\text{Ca}$ -ratio can thus be four times higher on acid soils than on neutral soils. The ^{137}Cs content in grass can be ten times higher on coarse soils than on clay soils.

Fertilization has a large impact on nuclide transfer to grass on permanent pastures established on coarse soils. Use of acidifying N fertilizers, AS, may give a $^{90}\text{Sr}/\text{Ca}$ -ratio twice that of CaN. Fertilization with AS increases the migration of ^{90}Sr and thereby decreases the $^{90}\text{Sr}/\text{Ca}$ -ratios in the long run. On coarse soils the ^{137}Cs transfer to grass is lower on P- and K- fertilized pasture than on unfertilized pasture.

Regarding N fertilizers, the influence is less consistent. While AS usually increases the ^{137}Cs transfer, AN and CaN may increase or decrease it. On clay soils rich in Ca, the influence of type of fertilizer on ^{90}Sr - and ^{137}Cs -transfer is small compared to that on coarse soils low in Ca. For both nuclides, however, the transfer decreased with K-fertilization and increased with P-fertilization.

Table 5. Median depth of activity in the soil profile at Lövsta in 1967, cm

Nuclide	NPK-treatment					Lime treatment				Mean
	A	B	C	D	E	a	b	c	d	
^{137}Cs	2.0	2.6	1.7	1.7	1.7	1.8	1.8	2.1	1.9	1.9
^{90}Sr	3.8	10.3	4.6	4.0	5.2	4.1	5.0	5.7	5.4	5.6

Table 6. Median depth of activity in the soil profile at Risslinge in 1967 and 1981, cm

Nuclide	Year	PK-treatment				Lime treatment			Mean
		B	C	D	E	a	b	c	
^{137}Cs	1967	3.0	2.6	3.2	3.1	3.3	3.8	2.6	3.0
	1981	5.0	5.4	5.3	5.7	5.5	5.6	5.0	5.3
^{90}Sr	1967	5.0	5.1	5.2	5.7	5.3	5.2	5.2	5.2
	1981	8.7	8.4	9.3	10.2	9.2	9.0	9.1	9.1

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SEASONAL CHANGES IN THE DISTRIBUTION OF ^{137}Cs , ^{133}Cs AND K IN BENT-GRASS (*AGROSTIS CAPILLARIS*)

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Abstract

Experimental swards of *Agrostis capillaris* were established in pots with peaty soil artificially contaminated with ^{137}Cs . Through regular clipping a sward height of 6 cm was maintained and plant material and soil were sampled between July and December. During July to August ^{137}Cs concentrations in leaves of all age classes increased with time and were highest in the youngest leaves. After late August only ^{137}Cs concentrations in very mature, semi-dead and dead leaves continued to increase and concentrations in very mature and semi-dead leaves exceeded those in younger leaves. The seasonal changes in concentrations of ^{137}Cs and ^{133}Cs were very similar. However, the seasonal pattern for K differed markedly, showing a steady decline in tissue concentrations from July to December. During senescence the relative loss of K was greater than that of ^{137}Cs and ^{133}Cs . In the soil, concentrations of exchangeable ^{137}Cs and K decreased by a factor of 2-3 between July and December. The ratio of $^{137}\text{Cs}/\text{K}$ in plant tissues increased while the ratio in soil extracts decreased. Relative to exchangeable concentrations in the soil, plant uptake of K was 1-3 orders of magnitude greater than that of ^{137}Cs .

1. Introduction

Semi-natural ecosystems with soils rich in organic matter and poor in clay minerals are particularly sensitive to inputs of radiocaesium (Livens and Loveland 1988) due to high plant availability compared to mineral soils (Sandalls and Bennett 1992). The affected soils are also

typically poor in plant nutrients such as potassium. Many authors suggest that in the soil-plant system radiocaesium behaves similarly to its physiological analogue potassium (e.g. Bünzl and Kracke 1989). However, results on hill pasture in the UK (C.A. Salt, unpub.) suggest that seasonal changes in plant uptake can differ markedly between radiocaesium and K with important implications for the radiocaesium intake by grazing animals. These differences may arise when radiocaesium has not reached equilibrium with naturally occurring stable caesium (Salbu et al. 1994). But it is equally possible that genuine differences exist between Cs and K in terms of seasonal changes in plant uptake or supply from the soil solution.

In previous field experiments, levels of radiocaesium in plants fluctuated strongly during the growing season (Salt and Mayes 1991; Salt and Mayes 1993) possibly due to changes in soil moisture. The current experiment was designed to supply plants with near constant moisture in order to be able to study seasonal effects without the effects of soil moisture.

The impact of plant senescence on radiocaesium and K is studied by following uptake patterns for leaves of different age classes from summer to winter. *Agrostis capillaris* was chosen since it is a common species of unimproved acidic grasslands and preferentially grazed by sheep. Analysis of stable caesium, mostly lacking from radioecological studies, is carried out to trace any dissimilarities between artificially introduced and naturally present Cs isotopes.

2. Materials and Methods

Topsoil from a peaty podzol at Devoke Water, Cumbria with an organic matter content of 50% was artificially contaminated with ^{137}Cs in September 1993 and used for growing *Agrostis capillaris* in a pot experiment. The pots were kept under a perspex roof and the soil was maintained at 80% field capacity by regular addition of rainwater. During July-December 1994 pots were consecutively harvested on 8 dates by removing 5 pots per date and separating the biomass into immature, mature, very mature, semi-dead and dead leaves. Fresh soil samples were extracted with 0.5M BaCl_2 . Plant samples were analysed for ^{137}Cs , ^{133}Cs and K, soil extracts only for ^{137}Cs and K. Results refer to oven-dry plant tissues and air-dried soil. Analytical methods were gamma ray spectrometry using a 3-inch NaI crystal detector for ^{137}Cs ; HNO_3 digestion followed by ICP-MS for ^{133}Cs ; flame AAS for K.

3. Results

Mean ^{137}Cs concentrations in *A. capillaris* tissues were affected by both sampling date and age of plant tissue. Figure 1 shows a consistent rise in ^{137}Cs concentration in very mature and semi-dead leaves until December, whilst in immature and mature leaves concentrations began to level off in early September. Until this time the youngest tissues were the most contaminated, thereafter the very mature and senescing tissues showed higher concentrations. In comparison concentrations in dead leaves remained low over the experimental period.

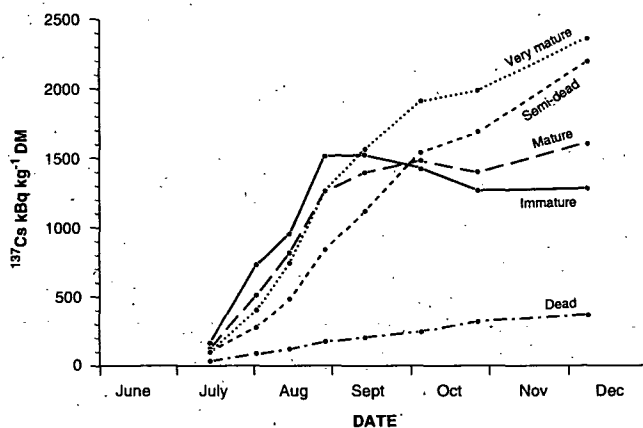


Figure 1: Time trends of mean ^{137}Cs in dry matter (kBq kg^{-1} ; $n=5$) of immature, mature, very mature, semi-dead and dead leaves of *Agrostis capillaris*.

The temporal changes in ^{137}Cs and stable ^{133}Cs concentrations in the different leaf categories were very similar. Figure 2 illustrates this for mature leaves and also shows that time trends for K were very different. While concentrations of Cs isotopes were rising, those of K were declining. This applied to both live and dead tissues. Linear correlations between ^{137}Cs and ^{133}Cs gave R^2 values of 95-98 % ($P<0.001$, $n=40$). During senescence the mean relative loss of K from plant tissues was estimated at 94-98%, while losses for ^{137}Cs and ^{133}Cs were typically 78-87%. Losses of tissue water as this ranged from 32-58%.

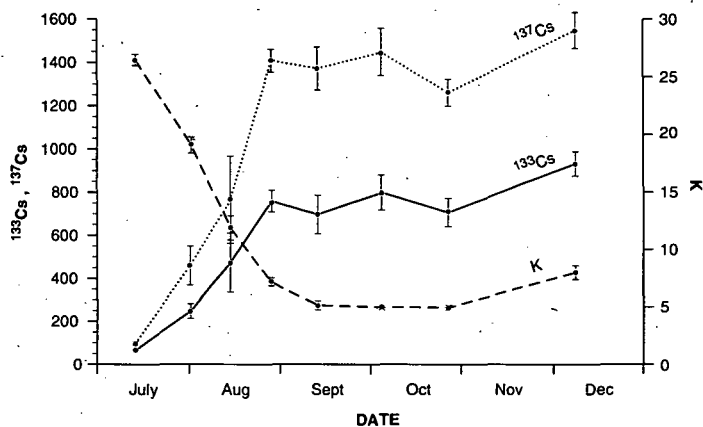


Figure 2. Time trends of mean ^{133}Cs ($\mu\text{g kg}^{-1}$), ^{137}Cs (kBq kg^{-1}) and K (g kg^{-1}) in dry matter of mature leaves with standard errors.

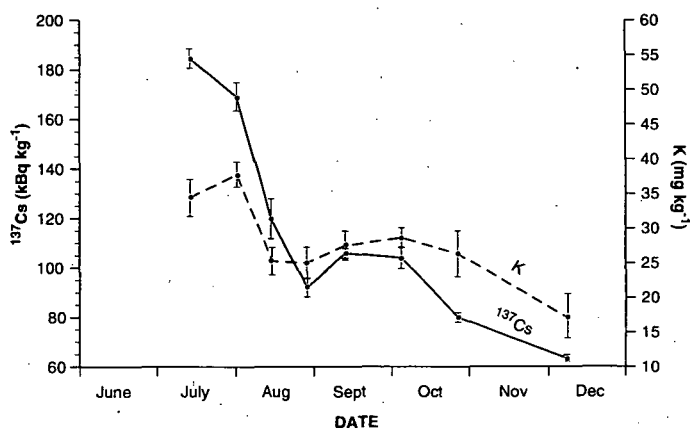


Figure 3. Changes in exchangeable mean ^{137}Cs (kBq kg^{-1}) and K (mg kg^{-1}) in air-dried soil during the period of vegetation sampling with standard errors.

Total concentrations of ^{137}Cs in soil and soil moisture did not change over the experimental period, however, seasonal changes in the exchangeable concentration of ^{137}Cs and K were measured (stable Cs was not analysed). Figure 3 shows a decline of these isotopes with time,

though correlation between $\log^{137}\text{Cs}$ and the square root of K was not very strong ($R^2=40\%$, $P<0.001$, $n=40$). The percentage of total ^{137}Cs extracted from the soil followed the same trend and fell from 32% in July to 12% in December. Concentrations of exchangeable ^{137}Cs and K were not correlated with concentrations in plant tissues. In fact the ratio of $^{137}\text{Cs}/K$ increased in plants but decreased in soil extracts. Table 1 shows the relative magnitude of soil/plant transfer of K versus ^{137}Cs for green and dead tissues. Compared to exchangeable concentrations in the soil, the K uptake was 1-3 orders of magnitude greater than that of ^{137}Cs .

Table 1. Relative soil/plant transfer of $K/^{137}\text{Cs}$, mean values, $n=5$.

Date	Immature leaves	Mature leaves	Dead leaves
14 July	1534	1609	338
1 August	223	232	94
15 August	168	145	83
29 August	30	16	14
12 September	35	14	15
3 October	29	11	8
24 November	32	13	10
5 December	47	23	10

The findings reported here still require more rigorous statistical analysis to identify statistically significant results. The following data interpretation should therefore be regarded as preliminary.

4. Discussion

Seasonal changes in ^{137}Cs and ^{133}Cs concentration in plant tissues followed the same pattern, indicating that the artificially applied isotope was behaving as its natural counterpart. The initial rapid increase in Cs concentration in immature and mature leaves levelled off in August coinciding with a decrease in net production and an increase in the proportion of dead matter in the swards. This resulted in a slower increase in concentration in unseparated vegetation. It appears that peak uptake of Cs in summer led to a delayed increase in tissue levels over a

considerable time and there was little change in contamination of total biomass towards winter. This behaviour of *A. capillaris* contrasts with field studies of ryegrass/fescue/clover pasture where contamination declined each autumn for 3 years (Salt and Mayes 1991; C. A. Salt unpub.).

Sombre et al. (1994) reported a similar seasonal pattern for both ^{133}Cs and K in spruce needles, with a gradual decline towards winter followed by an abrupt increase in spring. In this study potassium displayed a very different seasonal trend, decreasing while ^{133}Cs was increasing. Bunzl and Kracke (1989) reported very similar seasonal patterns for ^{137}Cs and K concentrations in *Trichophorum caespitosum* and *Molinia caerulea*, both declining from spring to winter. However, other research shows that K and Cs do not necessarily behave in the same way (Wolterbeek and De Bruin 1986; Wytenbach et al., in press). Plants seem more able to regulate tissue concentrations of K than Cs and root experiments show that K^+ -selective ion channels are much less selective towards Cs (Gassmann and Schroeder 1994).

Peak concentrations of ^{137}Cs were found in very mature leaves from October onwards showing that youngest tissues are not necessarily the most contaminated, as reported in many studies (e.g. Handley and Babcock 1970; Salt and Mayes 1993). Relative to the dry matter content of plant tissues, concentrations of both Cs isotopes and K were low in dead compared to live tissue. These differences are likely to be, at least partly, due to differences in tissue moisture levels (Leigh and Johnston 1983).

Losses of K due to senescence were greater than for ^{137}Cs and ^{133}Cs . This agrees with studies by Wolterbeek and De Bruin (1986) and Clint et al. (1992) which demonstrate lower mobility of Cs in plants compared to K and greater loss of K from litter. Witkamp and Frank (1969) demonstrated greater loss of Cs and K compared to mass during litter decomposition, resulting in a net decrease in concentration of both elements in the dry matter. The same trend was apparent in this study in dead leaves prior to detachment.

Results not presented here show that percentage loss of water during senescence may only explain up to 50% of the element loss. They also show that most of the element loss occurs while dead leaves were still attached to the parent plant.

Temporal changes in exchangeable ^{137}Cs in the soil bore no relationship to temporal changes observed in plants, while for K there was general agreement of trends in soil and plant. Similar

results were reported by Nishita et al. (1960). Measurements of exchangeable fractions highlighted that K uptake was 1-3 orders of magnitude greater than uptake of ^{137}Cs relative to the amounts potentially available in the soil. Since trends in exchangeable ^{137}Cs and K in soil were similar, but trends within the plant differed greatly, it may be concluded that plant specific factors played a key role. The discrimination against Cs at the soil solution-root interface (Gassmann and Schroeder 1994) may be stronger during periods of highest growth when the plant requires K. Neither levels of K in soil or plants were particularly low, indicating a sufficient supply for the plant.

5. Conclusions

The effect of plant tissue age on Cs and K concentrations was time-dependent. Within the plant, stable and radioactive Cs behaved similarly, but both differed from K. The relative supply of Cs and K from the soil as well as plant specific factors seem to influence the temporal changes of Cs in the plant. It appears that the selective uptake of K diminished with a decline in plant growth. The results suggest that stable Cs can be used as an indicator of the long term behaviour of radiocaesium in the environment. The possibility of high ^{137}Cs levels in plants in winter has important implications for the radiocaesium intake of grazing animals and needs to be supported by evidence from the field.

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EFFECT OF AFCF ON THE SOIL-PLANT TRANSFER OF ^{134}Cs

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Abstract

AFCF [Ammonium-ferric-hexacyano-ferrate(II)] was proven to be an effective countermeasure against radiocaesium uptake in domestic animals. In this paper we evaluate the possibility of using AFCF as a countermeasure for radiocaesium soil-plant transfer. On a sandy soil, AFCF application rates of 10 and 100 g AFCF/m² reduced the radiocaesium transfer to ryegrass by a factor of 25 and 225, respectively, without affecting plant growth. Even additions of 1 g AFCF/m² resulted in a fourfold reduction of the radiocaesium transfer factor. On a loamy soil, the picture was slightly different. 10 g AFCF/m² resulted only in a twofold reduction of the transfer. The difference in effectiveness of AFCF in reducing the transfer on a loamy soil compared to a sandy soil, was explained by the higher inherent Cs-fixation capacity of the former (presence of micaceous clays). Due to the decreased dry weight production at elevated AFCF application rates and the effective reduction in Cs transfer by mixing the radiocaesium through the profile, mixing can be considered as a more effective countermeasure for radiocaesium uptake on a loamy soil.

1. Introduction

The use of AFCF [(Ammonium-ferric-hexacyano-ferrate(II), $\text{NH}_4\text{Fe(III)Fe(II)(CN)}_6$)] and related compounds as countermeasure against radiocaesium uptake in domestic animals dates back to the late 1960s, the era of the atmospheric bomb testing. Researchers looked for methods to reduce the sorption of radiocaesium in the gastrointestinal tract and enhance its elimination from the body and AFCF came out as being very effective (Nigrović, 1965; Madshus *et al.*, 1966; Bozorgzadéh

and Catsch, 1972). AFCF research regained interest (Giese, 1988) with the Chernobyl accident, where radiocaesium was one of the major fall-out isotopes ranking high as a hazard because of its long half life. The use of AFCF as a countermeasure for radiocaesium uptake in domestic animals, leaves us with at least three major topics to investigate: 1) What is the long-term availability of radiocaesium to the plants when contaminated faeces, containing AFCF is used as manure? 2) Will the AFCF present in the manure affect plant growth? 3) Is AFCF also effective in reducing the soil to plant radiocaesium transfer? This article will deal with the third question.

2. Materials and Methods

^{134}Cs (Amersham International plc, Amersham, UK) and AFCF (Riedel -deHaen, Industrial Giese Salt, containing 60-65% AFCF and 35-40% NH_4Cl) were used throughout the experiments. For the transfer studies in greenhouse an Orthic Podzol (so called sandy soil) was used. In a first experiment on sandy soil, 3500 g of moist (11 %) soil was mixed with 400 ml distilled water and transferred to rectangular shaped darkened containers ($20 \times 15 \times 11 \text{ cm}^3$). 2 g of Italian ryegrass (*Lolium multiflorum*) was spread evenly over the soil and were covered with a 0.4 cm moist soil layer, homogeneously contaminated with 1 MBq ^{134}Cs . The final contamination level per pot was 303.2 Bq/g, the final soil depth 10 cm. 5 different concentrations of AFCF (0, 0.0316, 0.1, 0.316 and 1 g/m^2) were evenly sprayed over the soil surface with three different amounts of water (30, 60 and 90 ml/container). All treatments were set up with 4 replicates. Plants were given water every two days. Ryegrass was harvested every two or three weeks, depending on growth. Dry weight was determined and activity measured by γ -counting (minaxi, auto-gamma 5000 series-gamma counter). The transfer factors are defined as $(\text{Bq/g dry plant material})/(\text{Bq/g dry soil})$. Difference in transfer between treatments were analysed with SAS-statistical software. After every harvest, plants were given $\text{N}^{13}\text{P}^{21}\text{K}^{13}$ -fertilizer (30 ml/ dm^2 of a solution containing 17 g fertilizer/L). On the whole 15 cuts were harvested.

Given the results of this first experiment, a second experiment was similarly set up using higher AFCF-doses (0, 1, 10, 100 and 1000 g/m^2). Since the first experiment showed that the amount of water used to spray the AFCF did not influence ^{134}Cs transfer, only one water level (90 ml) was used in experiment 2. The final contamination level was 312.5 Bq/g. Plants were treated and

data were analysed as in Experiment 1.

For the transfer experiments on loamy soil, 1150 g of moist (20.98 %) soil was transferred to 1 L cylindrical ($d=11$ cm), dark containers. 0.634 g ryegrass was sown and covered with a 0.4 cm ^{134}Cs contaminated layer (final contamination level: 1441 Bq/g, final soil depth: 10 cm). Five concentrations of AFCF were applied on top of the contaminated soil (0, 0.3, 1, 3, 10 g/m²) by spreading 10 ml of an appropriate solution. To simulate the effect of ploughing on transfer, in a sixth treatment, ^{134}Cs was homogeneously mixed through the profile (822 Bq/g) and 1 g AFCF/m² was applied superficially. For the seventh treatment, both ^{134}Cs (1274 Bq/g) and AFCF (1 g/m²) were homogeneously mixed through the profile. All seven treatments were set up with 4 replicates. Plants were treated and data were analysed as in the experiments on sandy soil. 6 cuts were harvested.

3. Results

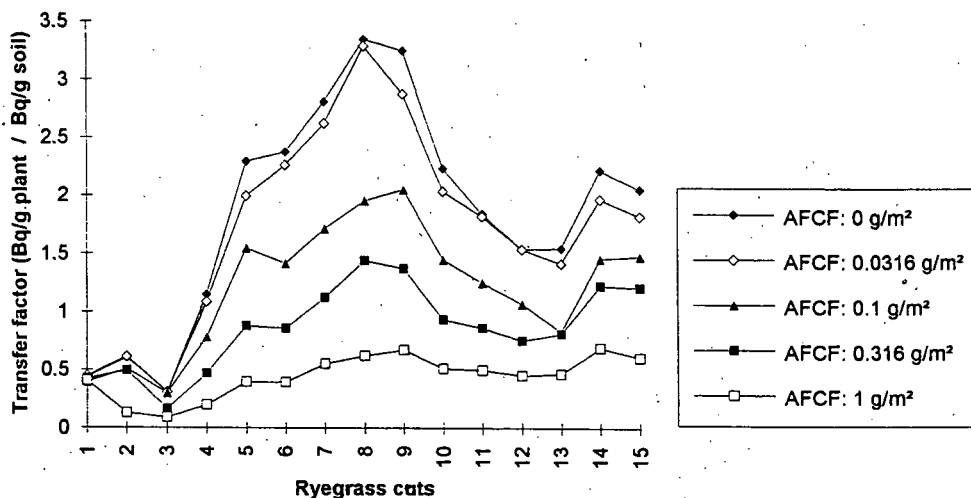
3.1 Experiment 1 on sandy soil

As already mentioned, the amount of water used for the spraying of the AFCF did not influence transfer. The mean values for the transfer factors per treatment and per cut as appearing in Figure 1 are the averages of 9 measurement points (3 water levels * 3 replicates). The transfer value for the control (1.869) is in agreement with literature data (Alexakhin, 1993). Addition of 0.0316 g AFCF/m² did not result in a significant decrease in transfer. Higher levels of AFCF did significantly reduce the soil to plant transfer of ^{134}Cs , by up to a factor of 4 when 1 g AFCF/m² was applied (Table 1). AFCF applications did not affect dry weight production.

Table 1: Effect of AFCF dose on the ^{134}Cs transfer to rye grass grown on sandy soil

AFCF g/m^2	Transfer factors (Exp. 1), Average for 15 cuts	Transfer factors (Exp. 2) Average for 15 cuts
0	$1.869 \pm 0.946^{\text{a},*}$	$1.853 \pm 0.636^{\text{a}}$
0.0316	$1.742 \pm 0.857^{\text{a}}$	
0.1	$1.210 \pm 0.548^{\text{b}}$	
0.316	$0.869 \pm 0.369^{\text{c}}$	
1	$0.446 \pm 0.185^{\text{d}}$	$0.489 \pm 0.183^{\text{b}}$
10		$0.075 \pm 0.038^{\text{c}}$
100		$0.008 \pm 0.008^{\text{d}}$

Transfer factors with a different letter are significantly different at the 5% level: * = stdev

Figure 1: Effect of AFCF-additions on ^{134}Cs -transfer to ryegrass (Sandy soil, Exp. 1)

3.2 Experiment 2 on sandy soil

Since low levels of AFCF additions were effective in reducing transfer and did not affect plant growth, a second experiment was started with elevated AFCF additions. At 1000 g AFCF/m², seeds were unable to emerge through the clogged AFCF layer. The other AFCF concentrations

did not affect growth. As shown in Figure 2 and Table 1, AFCF clearly reduces transfer: by a factor 4 when 1 g AFCF/m² is applied, as was found in the first experiment (results are thus reproducible), and by a factor of 25 and 225, respectively, when 10 and 100 g AFCF/m² were used, resulting in transfer factors of 0.0748 and 0.0083.

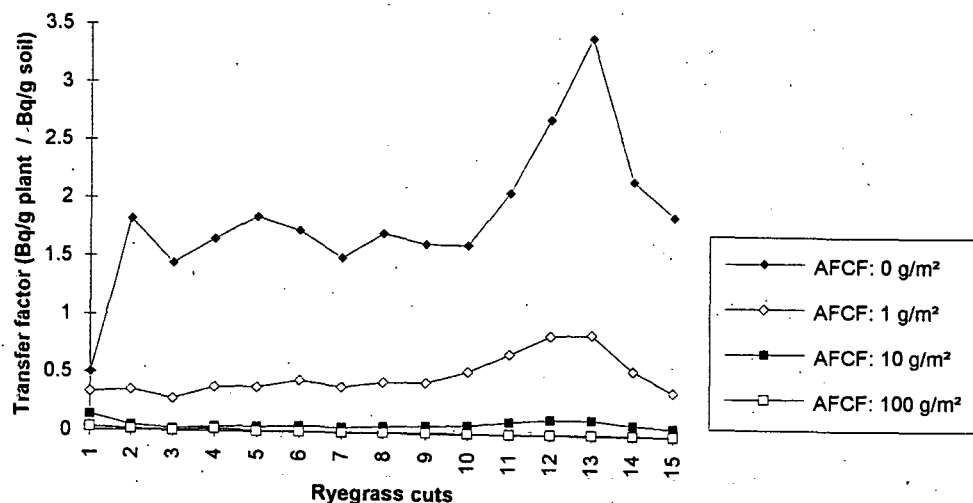


Figure 2: Effect of AFCF-additions on ¹³⁴Cs transfer to ryegrass (Sandy soil, Exp. 2)

3.3 Experiment on loamy soil

When considering the first 5 treatments (¹³⁴Cs applied superficially), the results obtained for the loamy soil (Fig. 3 and Table 2) differ from those obtained for the sandy soil on three major: 1) For the loamy soil, AFCF applications of up to 1 g AFCF/m² do not result in a significant decrease of the transfer factor. 2) 10 g AFCF/m² only reduces the transfer by a factor of 2, compared to the 25-fold reduction on a sandy soil. 3) For the 5 first cuts, 10 g AFCF/m² resulted in a growth reduction of 20 % (results not shown). For the 6th cut, no such negative effect on growth was observed.

For both treatments 3 and 6, 1 g AFCF/m² was applied superficially but ¹³⁴Cs was either applied superficially or mixed through the profile. When comparing both treatments, we can deduce the beneficial effect of ploughing (reduction of transfer factor with factor 2, Table 2). Mixing both ¹³⁴Cs and AFCF (treatment 7) reduces the TF even more (Table 2).

Table 2: Effect of AFCF dose and ^{134}Cs and AFCF application mode on ^{134}Cs transfer to ryegrass on a loamy soil

Treat.	AFCF g/m^2	Mode of ^{134}Cs application	Mode of AFCF application	Transfer factor Average over 6 cuts
1	0	Superficial	Superficial	$0.0759 \pm 0.0336^a, *$
2	0.3	"	"	0.0869 ± 0.0192^a
3	1	"	"	0.0808 ± 0.0348^a
4	3	"	"	0.0705 ± 0.0126^a
5	10	"	"	0.0437 ± 0.0147^b
6	1	Mixed	"	0.0361 ± 0.0166^b
7	1	"	Mixed	0.0229 ± 0.0099^c

Transfer factors with a different letter are significantly different at the 5% level; *=stdev

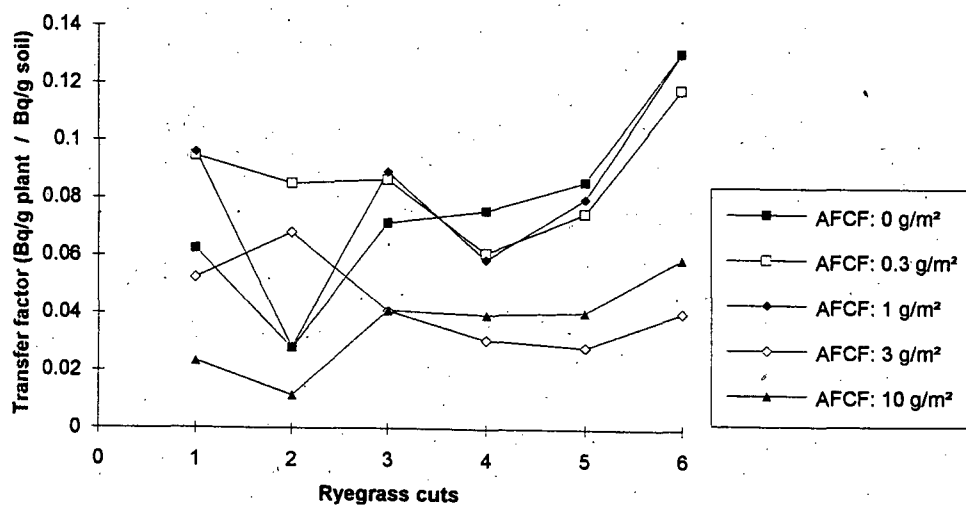


Figure 3: Effect of AFCF -additions on ^{134}Cs transfer to ryegrass (Loamy soil)

4. Conclusions

On a sandy soil, AFCF is undoubtedly effective in reducing ^{134}Cs transfer to ryegrass without affecting its growth. Among some of the countermeasures applied after the Chernobyl accident, ploughing and application of mineral fertilizers yielded at most a 4-fold reduction in transfer and results were often inconsistent (Alexakhin, 1993; Konoplev *et al.*, 1993; Lembrechts, 1993). A 25fold-reduction of the transfer factor was, however, obtained with 10 g AFCF/m² on a sandy soil. Its low cost (± 60 \$/kg: at 1 g/m² the cost is 60 \$/ha), its easy application (it can be sprayed making use of the machinery for applying pesticides) and its non-toxicity for plants (at least under the prevalent experimental conditions) make it a seemingly ideal countermeasure for low-Cs-affinity soils. One should, however, be conscious of the fact that AFCF might enhance the migration of radiocaesium so that it will, eventually, reach the phreatic water table. Meeussen *et al.* (1995) found that Prussian blue, a chemical form similar to AFCF [$\text{Fe}_4(\text{Fe}(\text{CN})_6)_3$] can reach the groundwater. Migration experiments recently initiated in our laboratory also indicated an enhanced ^{134}Cs mobility in sandy soil due to AFCF.

On a loamy soil AFCF showed to be less effective in reducing the transfer. Undoubtedly so because the loamy soil itself already has a high ^{134}Cs fixation capacity, which is related to its micaceous clay content (qualitative XRD-analysis). In control treatments, the ^{134}Cs transfer factor for the loamy soil is 22 times lower than for the sandy soil. Only when 10 g AFCF/m² was applied to the sandy soil, was its transfer comparable to the transfer on the loamy soil without AFCF addition. These findings were confirmed by ^{134}Cs adsorption/desorption studies (data not shown), using dialysis membranes, separately filled with soil and AFCF in the proportion of the quantities used in the pot experiments. For the loamy soil, even without AFCF, no ^{134}Cs was detectable in the solution surrounding the membrane and when an equivalent of 10 g AFCF/m² was present in the second membrane, still more than 50 % of the added ^{134}Cs was present on the soil, pointing to the high sorption capacity of the loam. For the sandy soil on the other hand, even with the equivalent of 10 g AFCF/m² in the second membrane, ^{134}Cs was low but still detectable in the solution surrounding the membrane. More than ± 80 % of the added radiocaesium was recovered from the solution by the AFCF and only ± 15 % by the soil.

The reduction of plant growth with 10 g AFCF/m² might be caused by the possible release of CN after the breakdown of the AFCF molecule. Meeussen (1992) investigated the stability of Prussian blue in different soils, and found that the molecule was less stable at elevated soil clay

contents.

Mixing ^{134}Cs through the profile of the loamy soil (treatments 6&7) resulted in a two fold reduction of the transfer. Similar reductions were found after shallow ploughing (Voyk *et al.*, 1993) This reduction in TF was comparable with the decrease in TF when 10 g AFCF/m² was applied. Given the above and given the growth reduction caused by elevated AFCF concentrations, it is doubtful whether AFCF should be recommended as a countermeasure for ^{134}Cs on a loamy soil. An important conclusion, however, remains in that AFCF distributed on the field together with the faeces of AFCF-treated animals, will not be harmful for AFCF concentration up to at least 3 g m⁻²

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Uptake and Transport of Radioactive Cesium and Strontium into Strawberry Plants and Grapevines after Leaf Contamination.

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Abstract - Radioactive strontium in the form of a carrier-free aqueous solution of $^{85}\text{SrCl}_2$ was taken up by the leaves of strawberry plants and grapevines at different rates. Only a limited amount was transported from the contaminated leaves into other plant organs. However no strontium was found in the fruit. These findings show that bivalent strontium is mobile in plants only to a very minor extent. In contrast, radioactive cesium applied as an aqueous solution of carrier-free $^{134}\text{CsCl}$ was taken up very quickly from the leaf surface, transported from there to other plant parts and released to some extent into the soil via the roots. In our investigations strawberry fruit showed a high cesium content. The cesium content in grape berries rose during growth and decreased again in a late phase of maturation and the nuclide was partly redistributed to other plant parts. Cesium reaching the soil may interact with clay particles resulting in a very limited availability for plants. First results of investigations with grapevines in hydroculture demonstrated a close relationship between potassium supply and cesium release. The foliar uptake of radioactive strontium into plants is minor and represents therefore a negligible risk for the consumer of fruits, berries and nuts. Radioactive cesium nuclides may however reach the human food chain through the leaves of food plants.

Introduction

Radioactivity in fruit, berries, nuts and some processed fruit products was slightly elevated in Switzerland in the summer and autumn of 1986, as a consequence of the Chernobyl disaster of April 26 (Zehnder 1988). This increase in the radioactivity of plant parts which were not even formed at the time of the accident was mainly due to radioactive isotopes of cesium,

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Cs-137 and Cs-134. Cesium is an alkali metal and its interactions with soil particles are similar to those of potassium (Rich 1968). The availability of extraneous cesium to crop plants depends on the soil properties (e.g. soil texture, composition of clay minerals, contents of other cations, pH). In general, this element becomes less available with the increasing clay content of the soil (Andersen 1967). In soils with a high clay content the uptake of cesium into the roots can be considerably lowered and delayed, but not completely eliminated. From the above findings it appears unlikely that the cesium isotopes found shortly after the Chernobyl accident in fruit, berries and nuts grown on loamy soils in Switzerland were taken up from the soil via the roots. These isotopes must therefore have followed another pathway into fruit, berries and nuts, e.g. through the leaves (Bucovac et al. 1965). Consequently, investigations have been carried out on strawberry plants and grapevines over a period of several years in order to determine the foliar uptake of radioactive cesium and strontium - another interesting nuclide in this context - and to trace the transport of these isotopes to other plant organs as well as their release from the plants into the soil (Zehnder et al. 1993, 1995). The accumulation of radioactivity in the berries was of particular interest since this represents a pathway for these undesirable isotopes into the human food chain.

Material and methods

Refrigerator-stored strawberry plants (Variety Elvira) and 2- to 3-year-old grapevines (Variety Riesling x Sylvaner) were planted in large plastic pots. The top surface of each of the pots with grapevines were covered with black plastic to prevent contamination of the soil. The plants were cultivated in a greenhouse using normal procedures. Pesticides were used only if necessary and only before starting the experiments. The floor of the greenhouse was covered with plastic sheeting and the two short sides (front and back) were made of fine plastic netting; for the two long sides polyester sheets were used. The Plexiglass roof was fitted with a rain proof ventilation hole. These precautionary measures ensured protection against loss of radioactivity and allowed air circulation. On the upper surface of the first two fully developed leaves of each strawberry plant and each grapevine 50 μ l of $^{134}\text{CsCl}$ - or $^{85}\text{SrCl}_2$ -solutions were applied with a micro pipette as 20 - 25 small droplets which dried within 2 - 3 hours. We used the short-lived isotopes cesium-134 (half-life 2 years) and strontium-85 (half-life 65 days) instead of the long-lived isotopes (Cs-137, Sr-90) for safety reasons.

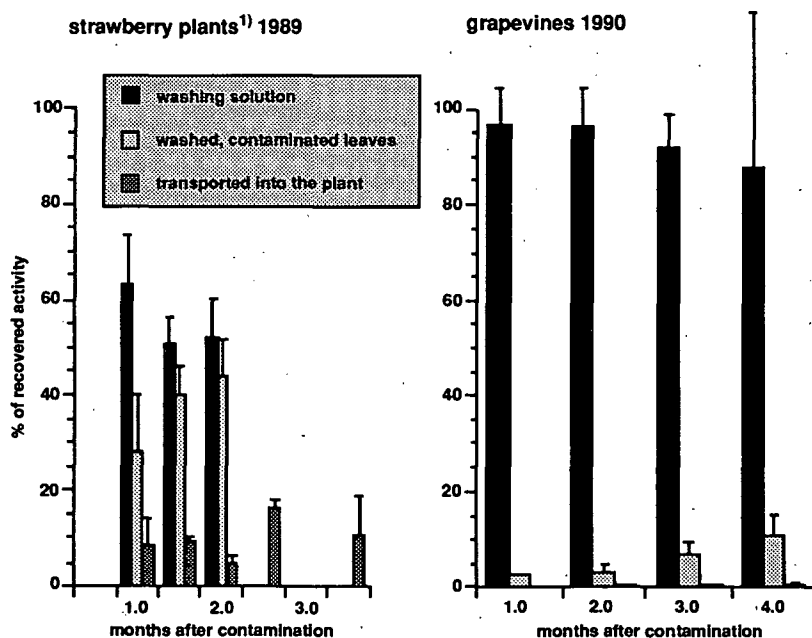
Samples were taken periodically. Per sampling date three strawberry plants and two grapevines were harvested. The two contaminated leaves of each plant were first removed and washed in 50 ml of 0.1 N HCl for five minutes with constant shaking. The washed, contaminated leaves were dried and the washing solution was poured into standard 50 ml polyethylene bottles. The plants were separated into their different organs, chopped and air

dried. Small samples (contaminated leaves, grape stalks) were placed in flat plastic dishes with a diameter of 5 cm and larger samples were put into 50 ml polyethylene bottles. The roots were separated manually from the soil and treated similarly to the other plant parts. The soil was sieved, well mixed, dried and about one kilogram was put into a 1 litre plastic bottle. The radioactivity of all samples was measured in a P-type coaxial solid state intrinsic germanium detector. Because of the different half-lives of the radioisotopes and the different times of analysis all measured activities were calculated back to the application date to provide a basis for comparison of the measured activities at different times. A detailed description of experimental procedures has been published elsewhere (Zehnder et al. 1993, 1995).

Results and Discussion

In both plant varieties most of the applied ^{85}Sr -chloride could be washed off the contaminated leaves. Up to 48 % was taken up in the leaves of strawberry plants but only 12 % in grapevines. In strawberry plants a third of the incorporated radioactivity was transported

Fig. 1 : Foliar uptake of ^{85}Sr into plants

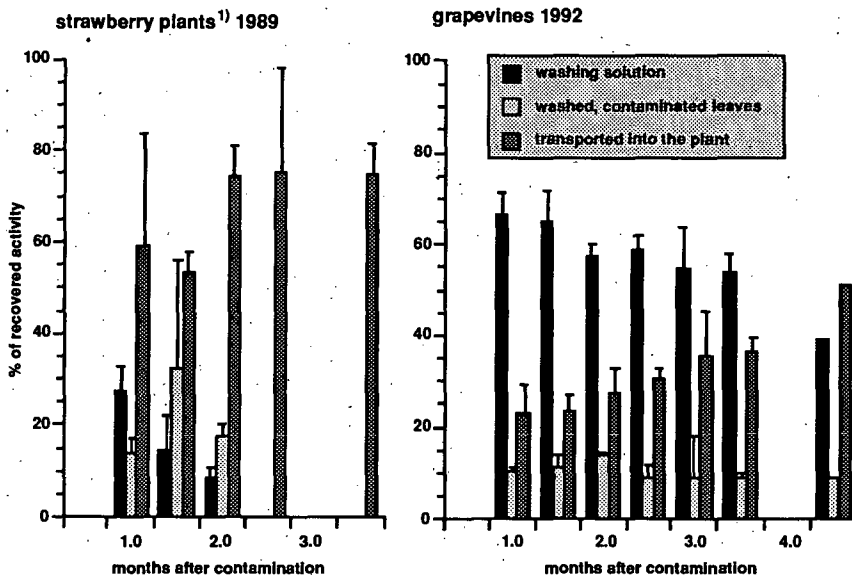


¹⁾ The contaminated strawberry leaves had to be removed after 2 months because of necrotic decay

to other plant parts and the soil, whereas in grapevines the translocated activity was only one percent. No radioactive strontium was detected in the fruit. These results show that strontium taken up by the leaves was relatively immobile in strawberry plants and highly immobile in grapevines (Fig. 1).

In contrast $^{134}\text{Cesium}$ -chloride solution deposited on plant leaves behaved very differently. It was taken up very quickly by both plant varieties and transported from the leaves to other organs and into the soil. Up to 90 % of radioactive cesium was incorporated into the leaves of strawberry plants and 75 % was distributed within the plant. The leaves of grapevines took up 34 - 65 % of the deposited cesium activity and 24 - 55 % was translocated within the plant and into the soil. Fifteen percent of the applied activity was stored in the leaves of the strawberry plants and 6 - 14 % in grapevines. The rest of the cesium-chloride could be washed off the leaves (Fig. 2).

Fig. 2 : Follar uptake of $^{134}\text{cesium}$ into plants



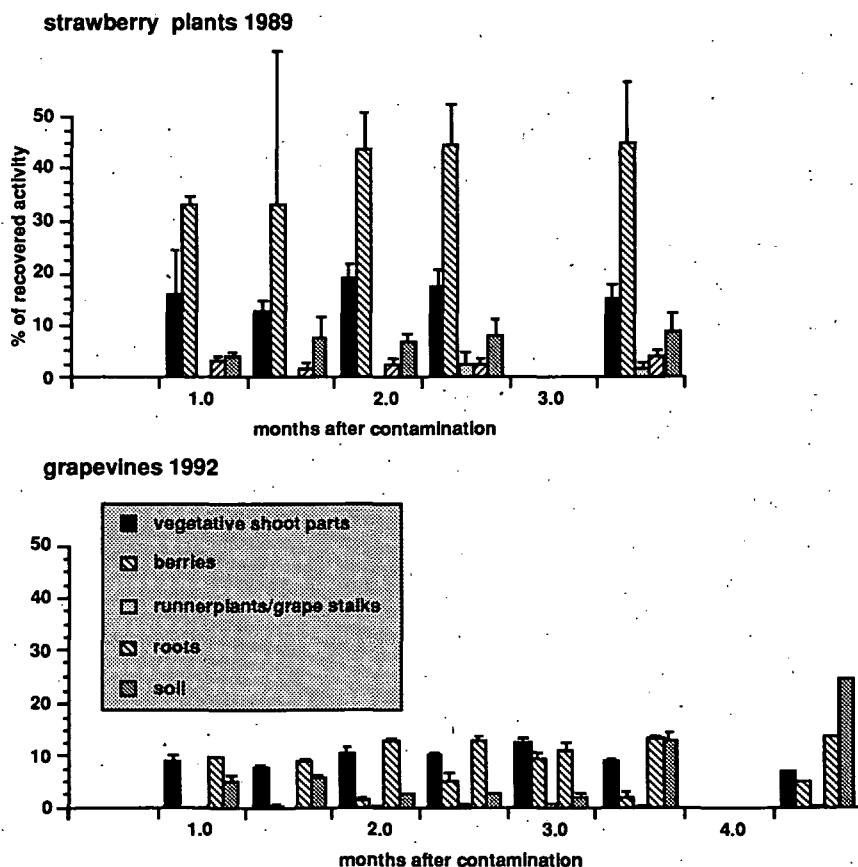
1) The contaminated strawberry leaves had to be removed after 2 months because of necrotic decay

Up to 45 % of the recovered cesium activity was located in strawberry fruits and 7 - 9 % was detected in grape berries. It was found that grape berries acted as a transient sink for cesium.

Cesium accumulated in grape berries during growth and the levels decreased again in a late phase of maturation when the nuclide was partly redistributed to other plant parts (Fig. 3).

Salts may be taken up by the leaves as long as they are in solution (Franke 1984). Once dried, radioactivity can no longer be taken up, except when the salt is dissolved again by mist, fog, water of condensation, guttation liquid, etc. The results of the investigations with grapevines carried out during the hot and dry summer of 1991 showed that the uptake of cesium was lower than during the cooler and wetter summer of 1992. The soil samples from grapevines generally showed a higher cesium activity (up to 25 %) than the samples from strawberry plants (< 10 %). First results of investigations with grapevines in hydroculture show a close

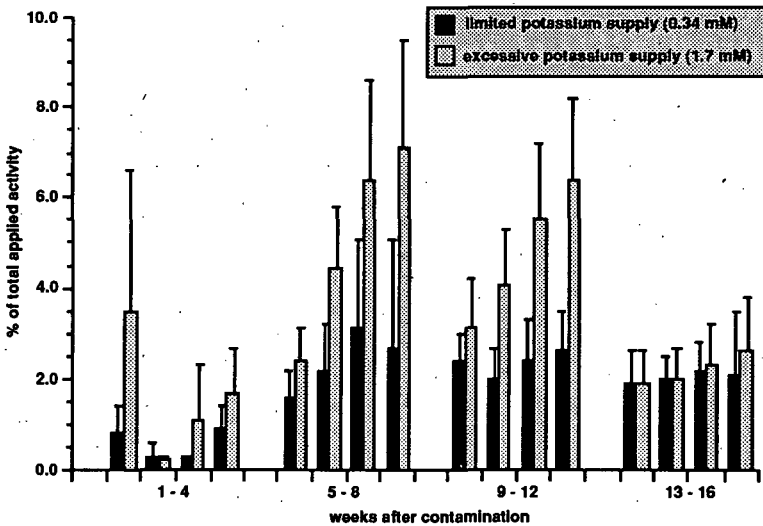
Fig. 3 : Distribution of ^{134}Cs in plants taken up by the leaves



relationship between potassium supply and cesium release. Grapevines with a limited potassium supply release less cesium into the nutrient solution than those provided with an excessive supply (Fig. 4). After having reached the soil, cesium is no longer taken up by the plants, even in a second vegetation period. This observation confirms the finding that radio-cesium, once bound to clay minerals in the soil, is not easily available to plants (Andersen 1967; Rich 1968).

The results of our investigations with strawberry plants and grapevines demonstrate that the foliar uptake of radioactive strontium into food plants represents only a very minor risk to the consumer of fruit, berries and nuts - except in the case of direct external contamination - as these bivalent nuclides are mobile in plants only to a very minor extent. Radioactive cesium nuclides, however, are very mobile in plants and may easily enter the human food chain through the leaves of food plants.

Fig. 4 : Release of $^{134}\text{cesium}$ by grapevines¹⁾ in hydroculture into the nutrition solution²⁾



1) n = 5; 2) the nutrient solution was renewed every 4 weeks

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¹³⁴Cs FOLIAR CONTAMINATION OF VINE: TRANSLOCATION TO GRAPES AND TRANSFER TO WINE

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Abstract.

Grape vines growing in pots placed in open field were contaminated by sprinkling an aqueous solution of carrier free ¹³⁴Cs. Activity was determined in acini and must at the ripening stage. Interception and Translocation Factors (TLF) were quantified. They strictly depend on the state of growth of the crop. Wine was produced by a laboratory process and analysed as to radiocesium. Transfer parameters, F_r and P_e , were calculated to predict radiocesium concentration in wine: it is 0.6 times the activity found in grapes. The leaf-fruit translocation seems to be the dominant process in the contamination of acini when compared to direct deposition.

1. Introduction

Fruit trees play a primary role in the economy of Mediterranean countries and represent an essential part of the diet of populations living in such areas. The radiocontamination of orchards in the case of a serious nuclear accident has to be taken into account by responsible authorities to limit the exposure of populations via ingestion of contaminated food products. The behaviour of radionuclides in fruit trees is not well known. In particular, data about the uptake and translocation of radiocesium are fairly scarce.

After the Chernobyl event research was carried out and data were collected on different kinds of fruit trees in order to understand or improve the knowledge of some pathways of migration and to evaluate some of the parameters determining radiocesium fluxes between different compartments. In particular, it has been established that, in the case of an accident, the foliar translocation is the main pathway for ¹³⁷Cs migration to the fruits (Monte et al., 1990; Anguissola Scotti and Silva, 1992).

In the present research a preliminary study was carried out on vine, perennial plant of high economic value, considering also that Italy is the main producer of wine in the world.

Translocation of radiocesium was studied from leaves to grapes after a wet deposition on aerial part of plants. An attempt was made to separate deposited from translocated activity in acini and to evaluate the transfer of radiocesium to wine.

2. Materials and Methods

Research was performed on three year-old grape vines of "Pinot white" growing in pots of 10 liters capacity and placed in open field. Vines were directly contaminated at the phenological stage of turn of colour by sprinkling an aqueous solution containing 109 kBq of carrier free ^{134}Cs /plant. During treatment the soil surface was covered with a plastic sheet to prevent its contamination. Each treatment was effected on five replicates.

In order to study the translocation of radiocesium from the leaves to the fruits, some vines were sprinkled after covering grapes with a plastic wrapping, removed a few hours later, to prevent the surface contamination of fruits.

The interception fraction was calculated by determining the ^{134}Cs activity of all the leaves picked from each of three plants a few hours after the treatment.

Grapes were picked at the ripening stage, 35 days after contamination. The harvest was of 0.24 ± 0.02 kg/plant. Acini were stripped from the stem and squeezed to obtain must. Acini and must were analyzed to determine the ^{134}Cs activity.

Must was inoculated under controlled conditions with 2.8×10^5 cells of *Saccharomyces cerevisiae* yeast/mL and kept in anaerobiosis at 25°C until constant weight to obtain wine. During the fermentation process sugar changes into alcohol releasing CO_2 with a loss of weight. Wine was filtered and its ^{134}Cs content was determined.

The concentrations of ^{134}Cs were analyzed by γ -spectrometry by means of a HpGe detector with an efficiency of 38% and a resolution FWHM of 1.76 keV at 1.33 MeV of the ^{60}Co . The different matrices were introduced into Marinelli beakers keeping a constant weight for each of them. An efficiency-energy calibration curve was calculated for each geometry.

3. Results and Discussion

As the translocation is highly dependent on the time at which contamination occurs during the growth period of crops (Simmonds, 1985), the contamination of vines was carried out at the phenological stage of turn of colour when the leaf-fruit translocation reaches its highest level.

The interception capacity of a crop depends mostly on LAI (Leaf Area Index). In fruit trees the LAI can change over a rather wide range even inside the same species (0.5 to 4) according to the plantation density and the growing form (Baldini and Bettoli, 1988). The leaf area of one vineyard of 1 ha can range from 10 to 30 thousand square metres. In the present research branches and clusters were not considered for interception. The fraction of activity initially intercepted by vegetation surfaces, expressed as the interception percentage of sprayed activity, was of $36.1 \pm 2.2\%$. The average wet weight of the foliar mass was 109.4 ± 6.9 g. Humidity was 63.6% at 65°C.

Plants were kept under a shed for 48 hours to prevent the wash-off of externally deposited radionuclide by rain and were placed afterwards protected by an antihail net in open field. Foliar absorption is a very rapid process that occurs within a few hours. Levi (1970) suggests an absorption/accumulation period of about 12 hours prior to cellular fixation and translocation. Therefore rain which falls 48 hours after contamination should not strongly affect the activity translocated to fruits.

After deposition a fraction of radiocesium is absorbed by leaves and, being extremely mobile within the plant, is translocated to its different organs. The average activity \pm standard error in acini, must and wine is reported in table 1. 94% of the activity found in the acini (5.7 kBq/kg) is concentrated in the grape juice. Only 6% of the radiocesium remains in peels and grape seeds (pips) after squeezing, but during wine-making 43% of activity is lost. This is a consequence of the process of filtration of wine that removes radiocesium in the sediment.

The mechanisms responsible for the separation of radiocesium can be very varied: precipitation with insoluble potassium tartrate instead of or together with potassium, absorption on organic matter (i.e. bacteria, yeasts), or mechanical deposition because of links with ions and solid particles (van der Waals forces, hydrogen links). There are several different industrial wine-making processes and all entail filtration. The oversight of this stage, carried out also for home-made wine, can lead to an overestimation of the calculated dose to population.

Translocation factors (TLF) have been quantified considering the fraction of activity intercepted by the foliage at the time of deposition and translocated within the acini (table 1). Activity data have been corrected for decay. The field loss of radiocesium was not evaluated.

$$\text{TLF (plant/kg)} = \frac{\text{Activity concentration in acini at harvest (Bq/kg)}}{\text{Activity intercepted by foliage at the time of deposition (Bq/plant)}}$$

TLF were also calculated for must and wine (plant/L or kg). TLF data can be useful to predict the contamination of grapes or wine, if the activity intercepted by the vines is known. They

are $(2.1 \pm 0.2) \cdot 10^{-1}$ for grapes and $(1.2 \pm 0.08) \cdot 10^{-1}$ for wine and agree with data collected by Coughtrey & Thorne (1983).

Determining the activity intercepted by the vines can be very hard in several situations. More frequently, one can rely on the availability of data concerning food, like grapes. The activity of wine can therefore be quantified using transfer parameters relevant to food processing: F_r and P_e (IAEA, 1994). They can be used to predict wine contamination, if the activity of grapes is known.

F_r is defined as the total amount of a radionuclide in processed food divided by the total amount of the radionuclide in the original raw food. It expresses the fraction of radiocesium which is retained in wine after processing of grapes. The total amount of ^{134}Cs in wine divided by the total amount in clusters gives an $F_r = 0.33$: 67% of ^{134}Cs is removed by the conversion of grapes to wine.

P_e is processing efficiency, i.e. the yield, defined as the ratio (kg prepared food)/(kg raw product). In the present research P_e is 0.58.

Radiocesium in wine = $F_r/P_e = 0.6$ times radiocesium in grapes.

In our second year of research, problems occurred at the irrigation system during the first phenological stages of vines. They caused a lesser growth of the plant with a reduction of three times in the foliar mass and four times in the yield, compared with the first year. However the plants were in a good metabolic state. Trials were nevertheless carried out and yield was homogeneous.

The experimental circumstances gave the chance to verify that interception strictly depends on the foliar mass of crop and the translocation factor on the growth. Growth acts both by increasing the hydration of tissues and by increasing the contaminated biomass. Therefore a lesser yield will cause, in the same conditions, a lesser dilution of radioactivity in fruits with a greater concentration per unit of weight and viceversa.

The interception percentage of sprayed activity $-27.7 \pm 1.0\%$ was lower than the previous year, because of a lesser amount of foliar mass, whereas the concentration in acini -31.2 ± 5.5 kBq/kg- and the corresponding TLF -1.50 ± 0.47 - were considerably higher, because of a lesser yield.

A relative comparison between the activity of grapes covered or not covered during treatment indicates that though the total activity of acini is greater in non-covered grapes than in covered grapes, 31.2 ± 5.5 and 23.9 ± 6.7 kBq/kg respectively, the corresponding activities of must do not present a significant difference: 19.8 ± 3.5 and 17.9 ± 5.6 kBq/kg. It seems therefore that the peel-must translocation is negligible compared with the leaf-fruit translocation. The greater concentration of radiocesium in peels and seeds of directly

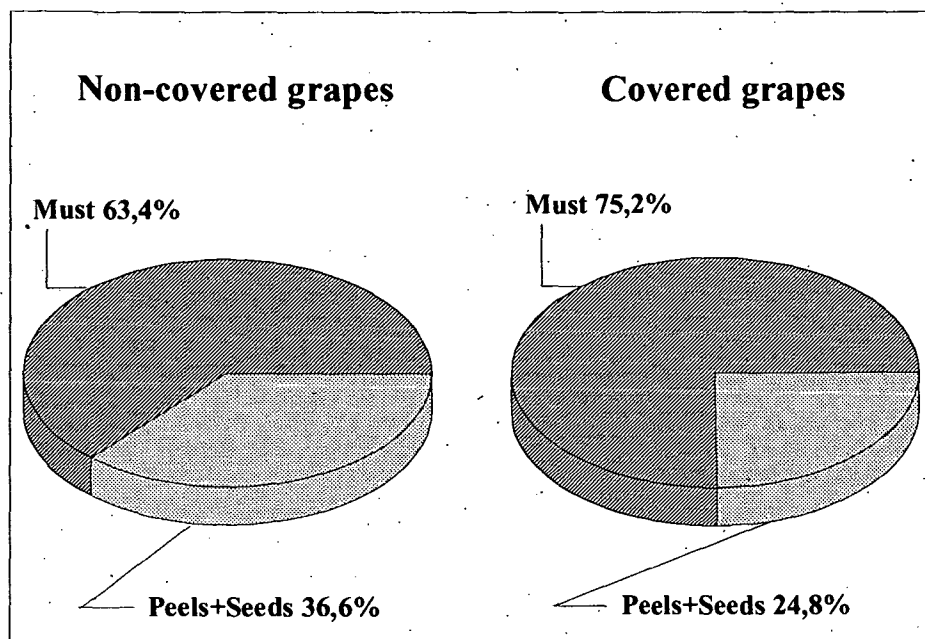


Figure 1: Distribution of radiocesium between must and peels + seeds

Table 1: Activities and translocation factors determined in acini, must and wine

Parameters	Activity kBq/kg	Percentage of acini activity	TLF Plant · kg ⁻¹
Grapes and by-products			
Acini	5.68±0.44	100	(2.1±0.2)·10 ⁻¹
Must	5.35±0.34	94.2	(2.0±0.1)·10 ⁻¹
Wine	3.26±0.20	57.4	(1.2±0.08)·10 ⁻¹

contaminated grapes (37% of acini) (figure 1) can be explained by the rate absorbed in peels without being translocated to other plant parts and not removable by rain, whereas the contamination of peels of covered grapes (25% of acini) is due to the translocation from the leaves to the fruits and, to a lesser extent, to the contact with contaminated leaves and with rain contaminated after the washing of leaves.

4. Conclusions

The analysis of experimental trials on foliar contamination of vines in pots with radiocesium suggests that:

Data are strongly affected by the state of growth of the crop. In particular interception strictly depends on foliar mass and the TLF on harvest.

Transfer parameters relevant to food processing, F_r and P_e , allow one to predict radiocesium concentration in wine: in the present research the foreseen activity in wine is 0.6 times that of grapes.

Non-covered grapes present 23% increase of activity when compared to covered ones, while the activity in must from covered and non-covered grapes does not differ significantly. Leaf-fruit translocation of radiocesium seems therefore to be the dominant process in determining the radioactivity of acini when compared to direct deposition.

Data presented in this paper are the result of a preliminary approach to the understanding of mechanisms involved in the translocation of radionuclides in foliar contaminated vines. They can be used to verify some pathways of migration, but need further investigations to improve a quantitative evaluation of some of the parameters.

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ANALYTICAL MODELS OF THE VERTICAL DISTRIBUTION OF RADIONUCLIDES IN SOIL

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Abstract

Two analytical models for the vertical distribution of radionuclides in soil are presented, one being the traditional diffusion-convection model with linear sorption equilibrium, the second one a non-equilibrium model. The differential equations on which the models are based are established and their solutions are shown. The two models are then fitted to a measured soil profile, both showing reasonable goodness of fit.

1 Introduction

Radionuclides deposited on the soil surface by atmospheric fall-out migrate into deeper soil layers forming a depth distribution. The shape of the profile depends on migration time and physical and chemical properties of both soil and nuclides involved.

In Austria, rather precise empirical data of ^{137}Cs and ^{134}Cs soil profiles are readily available as some areas show high levels of contamination due to Chernobyl fall-out. Maximum values of ^{137}Cs have been found to be around 200 kBq/m^2 , the mean value is 23.4 kBq/m^2 (BOSSEW et al., 1995). However, in low fall-out areas the Chernobyl contribution can easily be distinguished from global fall-out still present in soil (mean value for Austria 2.6 kBq/m^2 ; for all figures the reference date is 1 May 1986).

In most soils caesium migrates rather slowly. Therefore, in undisturbed soil, i.e. assuming only natural redistribution, Chernobyl caesium is still concentrated in the highest few centimeters of the soil column and usually shows a steep concentration decrease with soil depth. In some cases, particularly in acidic alpine soils, no dislocation of the concentration maximum away from the top soil layer can be found in measured profiles even 9 years after initial deposition, thus indicating very strong fixation.

Bomb-test fall-out caesium, on the other hand, with migration times of up to almost 40 years, has usually migrated down comparably deep, now showing concentration maxima typically 5 to 10 cm deep and very flat distributions. However, in the mentioned alpine soils also bomb-caesium maxima may still lie very close to the surface, in fact sometimes indistinguishably close by empirical means.

2 Analytical migration models

As opposed to the compartment type approach where redistribution between discrete soil layers of empiric profiles is studied by numeric means, the analytical method states a physical mechanism which is supposed to govern the migration process. It takes the mathematical form of a set of partial differential equations, the parameters of which are material constants that are typical for soil type and migrating substance, in this case radionuclide.

The traditional approach anticipates that the nuclides are being transported in the liquid phase by diffusion and convection, the migration being influenced or "retarded" by interaction with the solid soil phase, mainly by sorption. The corresponding first-order equations are:

$$J = -DC_L' + vC_L$$

$$\dot{C}_S = aC_L - bC_S$$

where J denotes the activity flux in the liquid phase ($m^2 s^{-1}$), C_L and C_S the concentrations in the liquid and solid phases, respectively, dimension (m^{-3}), D the apparent diffusion constant (m^2/s), v the convection velocity (m/s), and a and b the constants which characterize adsorption (a) to and desorption (b) from the solid phase; linear first-order sorption kinetics assumed.

The first equation is also called Fick's law. Combined with the conservation law:

$$\dot{C}_L = -J' - \frac{1}{W} \dot{C}_S,$$

where W denotes the soil water content (dimension = unity), $C = C_S + W C_L$, and assumption of sorption equilibrium, i.e. $C_S = k C_L$, this leads to the well-known diffusion equation. However, here we shall stick with the first order set of equations,

$$\dot{C}^* = -J' \quad \text{and}$$

$$J = -\alpha C^{*'} + \beta C^*$$

where $\alpha := D/R$, $\beta := v/R$, $R := 1 + k/W$, the so-called retardation factor, and $C^* := R C_L$.

This pair of equations will be the subject of the further investigations of this paper. - We always omit radioactive decay here, as it only adds a $-\lambda C$ type term to the conservation law which reappears in the solution as a $\exp(-\lambda t)$ factor.

The second model proposed here is a physical analogon one, not anticipating an actual soil model. Soil is considered as a "black box", in which there happens to be an activity flux J and a related activity concentration C . The equation governing the flux consists of an "absorption" term, inspired e.g. by photon flux attenuation, $J' = -a J$, and an activation type "desorption" term, $J' = b C$.

The two contributions now combine to a first-order differential equation,

$$J' = -aJ + bC.$$

Together with a simple version of the conservation law, $\dot{C} = -J'$ this yields the dynamics of the system. Like the diffusion model also this one involves two parameters, which, however, are not a priori linked to physical reality. The mathematical analysis shows, that the flux equation, which simulates a sorption situation, plays the role of a transport equation, without anticipating any physical transport mechanism. The model is therefore called the "sorption model" here, in analogy to the diffusion-convection model, where the transport equation is governed by the latter mechanisms.

The idea of this is to see if a model which has a totally different mathematical structure is able, first, to produce a simple analytical solution, and second, of course, to simulate a real, physical situation satisfactorily.

A general drawback of any analytical model is that it anticipates material parameters like D and v to be constants over the entire soil column. This may be true for laboratory soil columns but is certainly not, in general, for naturally occurring multi-layered soils. However, it turns out that, generally, the such idealized analytical models fit well to natural profiles.

3 Solution of the differential equations

Traditionally the diffusion equation is being used in second order form. It can be solved either by single Laplace transformation, see e.g. BUNZL (1978), or transformation into an integral equation and iterative solution, see e.g. KASIMOVSKY (1993). The latter author further

simplifies the equation by omitting diffusion, while, on the other hand, keeping sorption non-equilibrium.

Here, we use double Laplace transform, one for the time and one for space coordinate. The two linear equations are transformed separately and the variable J is only eliminated from the transformed, algebraic pair of equations. The advantage of this method is that initial fall-out $J(0,t)$ remains in the equations, therefore no surface layer needs to be introduced as the quoted authors do in order to satisfy the boundary conditions. In the following considerations C^* has been replaced by \bar{C} for convenience.

Initial and boundary conditions are $C(x,0) = C_0$ and $J(0,t) = J_0$. $\lim_{x \rightarrow \infty} C(x) = 0$ for $x \rightarrow \infty$ follows from the conservation law. Double Laplace transform leads to

$$\bar{\tilde{C}}(u,s) = \frac{u \alpha \bar{C}(0,s)}{\alpha u^2 - \beta u - (s + \lambda)} - \frac{\bar{C}(u,0) + \bar{J}(0,s)}{\alpha u^2 - \beta u - (s + \lambda)}$$

where u and s are the transformed variables corresponding to x and t , respectively. Transforms in respect to x and t are denoted as \wedge and \sim , respectively. Radioactive decay has been included in this formula.

The somewhat lengthy back-transform yields the following solution:

$C(x,t) = C(x,t)_{<J>} + C(x,t)_{<C>}$, with

$$C(x,t)_{<J>} = \frac{1}{\alpha} J(0,t) \# \sqrt{\alpha} e^{\frac{\beta x}{2\alpha}} e^{-\frac{\beta^2 t}{4\alpha}} \left\{ \frac{1}{\sqrt{\pi t}} e^{-\frac{x^2}{4\alpha t}} - \frac{\beta}{2\sqrt{\alpha}} e^{\frac{\beta^2 t}{4\alpha} + \frac{\beta x}{2\alpha}} \operatorname{erfc} \left(\frac{\beta}{2} \sqrt{\frac{t}{\alpha}} + \frac{x}{2\sqrt{\alpha t}} \right) \right\}$$

$$C(x,t)_{<C>} = \int_0^x d\xi C(x-\xi, 0) e^{\frac{\beta \xi}{2\alpha}} \left\{ \frac{e^{-\frac{\beta^2 t}{4\alpha}}}{2\sqrt{\pi \alpha t}} \left(e^{-\frac{(2x-\xi)^2}{4\alpha t}} + e^{-\frac{\xi^2}{4\alpha t}} \right) - \frac{\beta}{2\alpha} e^{\frac{\beta(2x-\xi)}{2\alpha}} \operatorname{erfc} \left(\frac{\beta}{2} \sqrt{\frac{t}{\alpha}} + \frac{2x-\xi}{2\sqrt{\alpha t}} \right) \right\}$$

where $\#$ denotes convolution. Radioactive decay would add another $\exp(-\lambda t)$ factor.

Of particular interest are the initial conditions representing "sudden" fall-out, sudden meaning short compared with the macroscopical change rate of the system. The fall-out is set to happen at $t = 0$; there is no pre-existing contamination, $C(x,0-) = 0$, and $J(0-,t) = J_0 \delta(t)$. The factor J_0 is the total deposition in (m^{-2}). The solution now reduces to a handler

$$C(x,t) = \frac{J_0 \beta}{2\alpha} e^{4\xi} \left(\frac{e^{-z^2}}{\sqrt{\pi \tau}} - \operatorname{erfc}(z) \right), \quad z = \sqrt{\tau} + \frac{\xi}{\sqrt{\tau}}$$

with normalized coordinates $\xi := \beta x / 4\alpha$, $\tau := \beta^2 t / 4\alpha$.

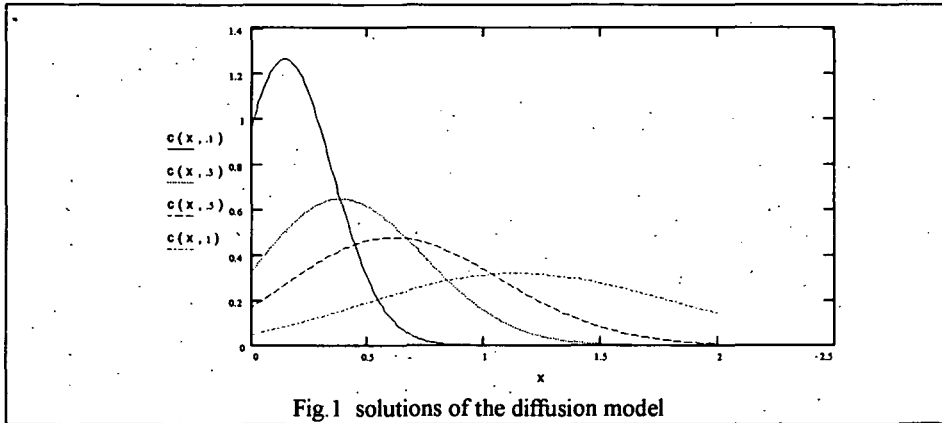


Fig. 1 solutions of the diffusion model

Fig. 1 shows the graphs of the normalized function $c(x, t) := e^{4x}(\dots)$ for four values of t .

The "sorption" model can be solved the same way. The solution is

$$C(x, t) = e^{-bt} C(x, 0-) + e^{-bt-ax} \left\{ \sqrt{\frac{abt}{x}} I_1(2\sqrt{abxt}) C(x, 0-) + a I_0(2\sqrt{abxt}) J(0, t) \right\}$$

where I_0 and I_1 are the modified Bessel functions of 0-th and first order, respectively. With the same initial and boundary conditions as above we get the very simple function

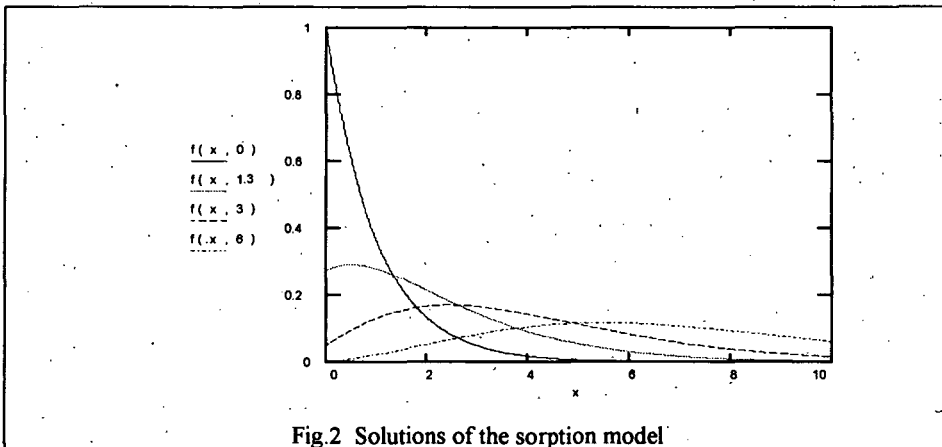


Fig. 2 Solutions of the sorption model

$$C(x, t) = J_0 a e^{-ax-bt} I_0(2\sqrt{abxt})$$

Fig.2 shows the graphs of the normalized function $f(x, t) := e^{-x-t} I_0(2\sqrt{xt})$ for different normalized times:

4 Fitting of solutions to measured soil profiles

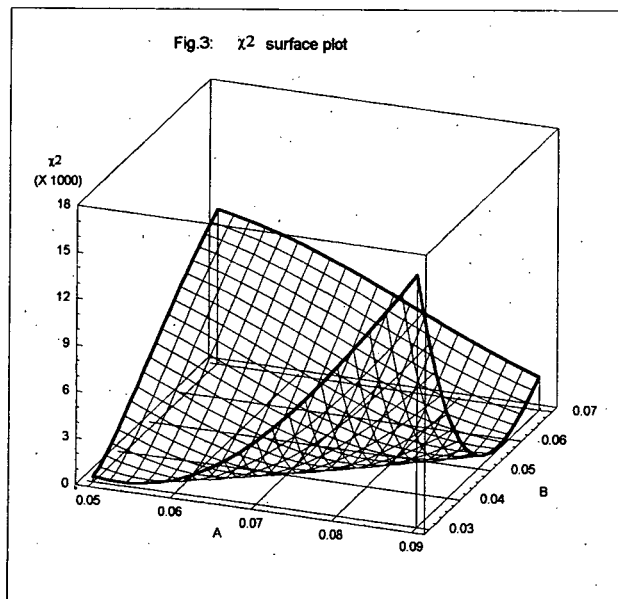
The solutions can be tested by fitting them to empirical profiles. From general regression theory follows that best fit of f to the measured values y_i at the points x_i is achieved if the model parameters $\{a_k\}$ are chosen such that the function

$$\chi^2 := \sum_i \frac{(f(x_i; a_k) - y_i)^2}{\sigma_i^2} \text{ is a minimum; } i \text{ counts the soil layers, } \sigma_i = \text{measuring SD of } y_i.$$

The classical way of doing this is to solve the set of so-called normal equations:

$\partial(\chi^2)/\partial a_k = 0$ ($k=1 \dots n$; in this case, $n = 2$). Since the functions involved are quite uncomfortably non-polynomial ones they have to be solved by numerical means.

Here, the function to be fitted is not simply the concentration $C(x_i, t; a, b)$ but the integrated one, $F_i := \int_{x_{i1}}^{x_{i2}} C(\xi, t; a_k) d\xi$, where x_{i2} and x_{i1} are the upper and lower edges of layer (i),



respectively. Accordingly, the measured points to which F is being fitted, are the fractions of the deposition present in the respective layer, i.e. $y_i = C_{i, \text{measured}} * (x_{i2} - x_{i1})$

Fig.3 shows an example of how the $\chi^2(a,b)$ - function looks like ($A = \beta/4\alpha$, $B = \beta^2/4\alpha$).

The measured values in the example presented here form the empirical ^{137}Cs soil profile in an undisturbed meadow in Salzburg with a measured total deposition of 41.3 kBq/m^2 . The sample was taken in summer 1993, so the migration time of Chernobyl fall-out is 7 years in this profile.

Figs. 4 and 5 show the results of a fitting process. In fig.4, the diffusion model has been fitted to the measured profile which is represented by the step function; in fig.5 the sorption model. Goodness of fit is somewhat better for the diffusion model ($\chi^2 = 13.4$) than for the sorption model ($\chi^2 = 19.3$). As both χ^2 are below the critical value $\chi^2(p=0.05; 11 \text{ d.f.}) = 19.7$ both fits are acceptable from a statistical point of view.

5 Conclusions

Though idealizing natural conditions by ignoring the multi-layeredness of soil both models show reasonable goodness of fit to a natural, undisturbed soil profile. The use of the pair of first-order differential equations rather than of the second-order one facilitates the realization of the sudden fall-out initial condition.

Acknowledgement

This project is being supported by the Austrian Federal Ministry for Science, Research and the Arts.

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Fig.4: measured and calculated profile, diffusion model

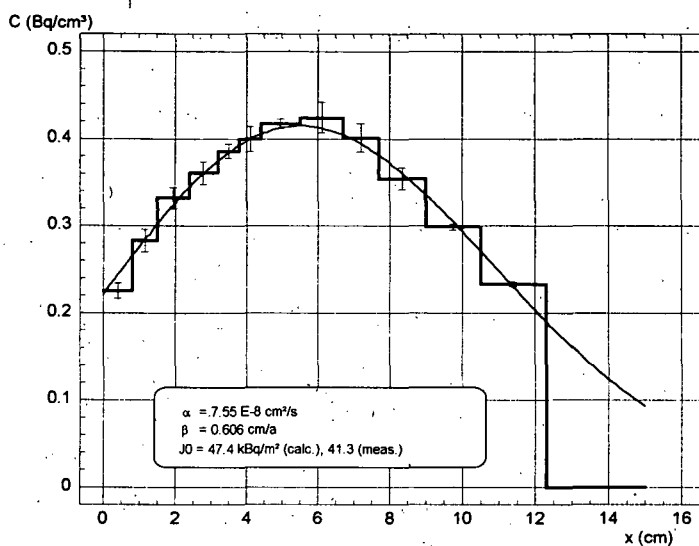
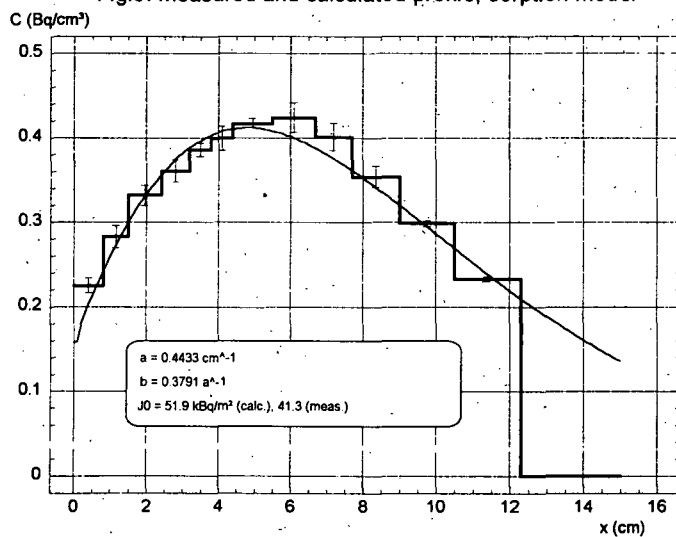


Fig.5: measured and calculated profile, sorption model



SPATIAL VARIABILITY OF FALL-OUT ^{137}Cs

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Abstract

Total process standard deviations of soil deposition measurement originating from spatial variability of fall-out deposition and measuring uncertainties have been determined. The related total coefficient of variation (CV) of ^{137}Cs fall-out in a 1 ha area, measured by soil core sampling, amounts to 21.5%. The measuring process contributes with 12.2% leaving a 17.7% CV purely due to spatial variability of the fall-out, normal distribution of the values assumed.

1 Introduction

In order to assess the statistical significance of soil deposition data measured by soil sampling or in situ gamma spectrometry, information about process standard deviations in general and spatial variability of the fall-out in particular is necessary. Distinguishing between different contributions to the total measuring process standard deviation requires a series of experiments especially tailored to uncover these sources.

In this project, the total process standard deviation, measured as a coefficient of variation and hereafter called TPCV (total process CV), has been determined by multiple fall-out measurement on an undisturbed meadow in Salzburg. The contributions to the TPCV originating from the measuring process itself were determined by separate experiments or estimated from practice.

The data derived from this project are being used in the Austrian soil contamination survey ("Caesium Map", BOSSEW et al. 1995) as input to the calculation of confidence levels of the deposition figures. According to the requirement of the Austrian Caesium Map which defines the contamination at a location as the AM of the deposition density of a 100 x 100 m² area, this area has been chosen as the basic size for this investigation. (In practice such uniform

sampling could not be achieved in the Caesium Map for various technical reasons. However the statistical analysis informs about confidence levels associated with samples which do not fulfil the requirement to have been collected from a 1 ha area.)

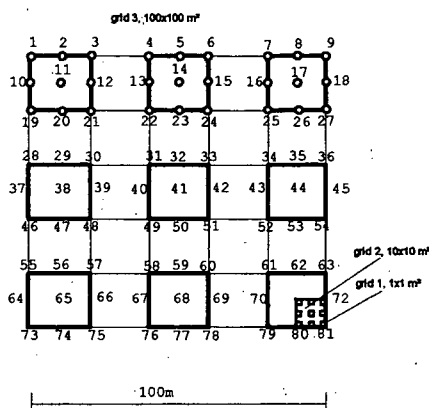
2 Experimental procedure

A meadow in the city of Salzburg which is known to have been undisturbed since the Chernobyl accident in 1986 has been selected for the investigation. The average ^{137}Cs soil contamination is relatively high with 43 kBq/m² (ref.1 May 1986) thus allowing short measuring times. A 100 x 100 m² section of the entire 5 ha area has been sampled extensively according to a grid as follows (Fig.1):

grid 3: the 100 x 100 m² area divided in 25 20 x 20 m² squares, 9 of them (i.e. every second in each row and column) containing 3 x 3 sampling points, i.e. a total of 81 sampling points.

grid 2: starting area 10 x 10 m², subdivision according to the same pattern.

grid 1: starting area 1 x 1 m², subdivision according to the same pattern.



Grid 2 is located in one corner of grid 3, and grid 1 in a corner of grid 2, in turn. As some of the points coincide a total of 235 soil samples was taken. The soil cores were dried, homogenized and measured with HPGe detectors. 24 out of the 235 soil cores were divided in 7 layers (each 2 cm thick) in order to get information about the depth distribution.

Several possible sources of process uncertainties, apart from fall-out deposition variability, were taken into account, among them:

- error in determining the cross section of the soil core;
- cut-off of the profil, i.e. neglecting contamination of deeper layers (systematic error);
- preparing aliquots from the total sample for gamma-spectrometry;
- positioning of the sample on the detector (partly systematic);
- detector calibration;

- photon attenuation in the sample (systematic);
- measuring statistics.

The experimental process cannot be described here in full detail due to space limitations (see LETTNER et al., 1994). The results will be summed up in the following section, however.

Statistical analysis was carried out with the analysis of variance (ANOVA) method.

3 Results and discussion

3.1 Analysis of the soil sample grids

Tab.1 summarizes the statistical parameters related to the three grids. SD(between / within groups) refer to the 9 sub-squares (= groups) of each grid. The principal result is that, according to the F-test, which basically compares the group means, the sub-groups of grid 3 can *not* be anticipated to be representative for the whole area. In other words, samples taken from grid 2 the size of which (100 m²) is of the same order of magnitude as the groups of grid 3, are *not* representative for the deposition of an 10,000 m² area (grid 3). On the other hand, an additional t-test, which compares the AMs of grid 2 and grid 3 shows no significant difference ($p=0.71 \gg 0.05$) from which we conclude that the location of grid 2 within grid 3 has been chosen such as to yield representative (for grid 3) deposition values *by chance*.

The Bartlett-test, finally, indicates homogeneity for the CVs of the subgroups of grid 3, which fits the result that $32.0 = SD(\text{grid 2}) \approx SD(\text{within; grid 3}) = 33.0$. However, from $SD(\text{grid 2}) < SD(\text{grid 3})$ with weak significance (additional F-test: $p = 0.072$) follows that by selecting grid 2 we have found a sub-group with lower SD. - If the CVs would not be homogenous, the selection of one sub-group would not necessarily yield a CV which is representative for an area of this size.

Tab.1. statistical parameters

parameter	grid 1 (1 m ²)	grid 2 (100 m ²)	grid 3 (10,000 m ²)
AM (Bq/cm ²)	171.3	173.3	175.3
SD	38.7	32.0	37.8
CV%	22.6	18.5	21.5
SD(between groups)	44.6	29.7	66.7
SD(within groups)	38.0	32.3	33.0
p(F-test)	0.22	0.56	0.005
p(Bartlett-test)	0.19	0.067	0.31

As grid 2 is concerned in comparison with grid 1, AMs of subgroups of grid 2 are very well homogenous (large $p(F)$). On the other hand, the almost significant Bartlett-value indicates a slight inhomogeneity of the CVs of the sub-groups of grid 2. This fits the observation that $SD(\text{grid 1}) = 38.7$ is quite different from $SD(\text{within groups; grid2}) = 32.3$. Therefore, arbitrary selection of a grid-1-size area may not produce a CV which is representative for this area size. Finally, both SDs between and within groups of grid 1 are relatively high indicating possible very small-scale (10 cm range) inhomogeneity.

It must be emphasized that, so far, all CV values include not only spatial variance but also uncertainties related to sampling and measuring, i.e. are TPVC values.

3.2 Contribution of process uncertainties

Tab.2 overviews the sources of errors related to the sampling and measuring process. Values have been determined or estimated for the methods used by the Austrian Ecology Institute and the University of Salzburg, respectively. All figures are in $CV = SD\%$ and refer to ^{137}Cs measurement.

Tab.2: process related errors

source	Ecol.Inst.	Uni.Salzb.
cross section of soil cores	10	< 2
loss of soil when removing the core (system.)	- 0.2	- 0.2
profile cut-off (systematic)	- 13	- 13
weighing	< 0.2	< 0.2
sample aliquots	3	x
remaining inhomogeneity of the sample	4	n.i.
positioning on the detector	2.7	n.i.
detector efficiency: calibration solution	2.5	2.5
counting error, detector calibration	1	1
calculated efficiency (fitted curve)(system.)	- 2	+ 3
photon attenuation in the sample (system.)	- 1	+ 1
statistical measuring error	3	5
total statistical	12.2	6.8
maximal total systematic	- 17	- 11

x = cannot occur; n.i. = not investigated

In some cases, very disadvantageous assumptions or experimental simulations (like in the case of remaining inhomogeneity and bad positioning) have been made. Therefore we believe that, in general, the figures give upper bounds of the errors.

From a TPCV = 21.5 % (grid 3) and a 12.2.% process contribution follows the net CV due to spatial variability only, equalling 17.7 %.

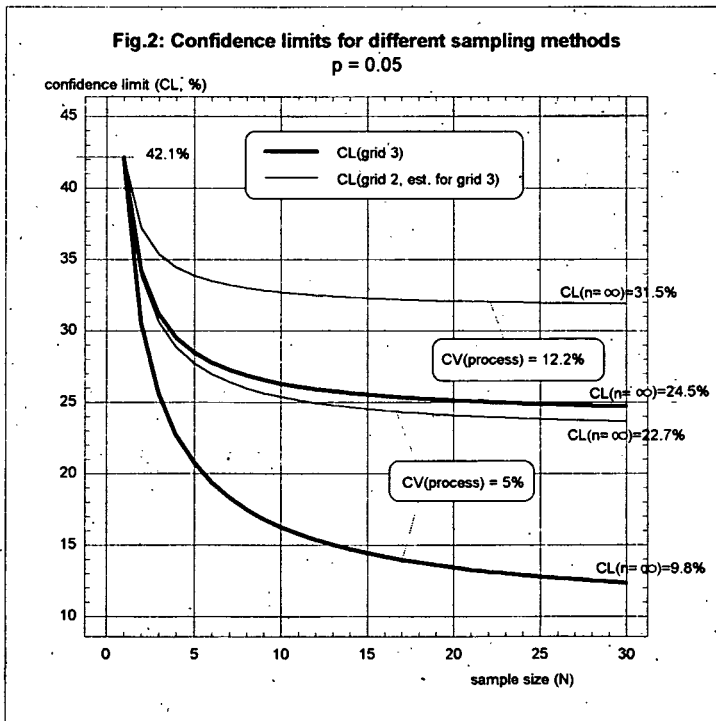
3.3 Confidence levels

From the above findings confidence levels (CL) for measured deposition values can be calculated. The normalized CL (in %) for N random samples out of a *whole* grid 3 size area (1 ha) is

$$CL_{\text{grid3}} = x(p) \sqrt{\frac{CV_{\text{grid3, spatial}}^2}{N} + CV_{\text{process}}^2},$$

$x(p)$ = Gauss distribution value for significance level p . If N samples are randomly taken out of an area of the size of grid 2 (=100 m²), but interpreted as an estimation of the deposition AM of a grid 3 size area, the respective CL is clearly larger:

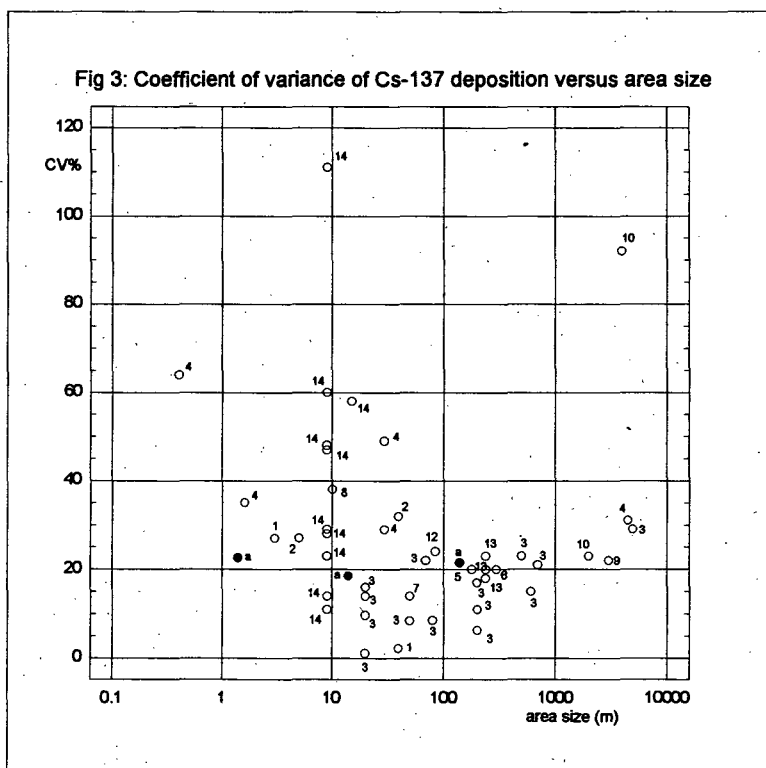
$$CL_{\text{grid2; est. for grid3}} = x(p) \sqrt{CV_{\text{grid3, spat.}}^2 - CV_{\text{within, grid3, spat.}}^2 \left(1 - \frac{1}{N}\right) + CV_{\text{proc.}}^2}$$



$$CV_{\text{spatial}}^2 = TPCV^2 - CV_{\text{process}}^2$$

In our case, $CV_{\text{grid 3, spat.}} = 17.7\%$, $CV_{\text{within, grid 3, spat.}} = 17.5\%$, $CV_{\text{proc.}} = 12.2\%$; $x(p=5\%; 2\text{-sided}) = 1.96$. Normal distribution of the randomly sampled deposition values is assumed here (it is true in our example, as checked by means of χ^2 -tests). Another assumption made here is that $CV_{\text{proc.}}$ is independent of N , which is probably not entirely correct. For example the sample aliquot error probably even increases with N , because the more soil cores, the more total mass out of which the aliquot for measuring must be drawn thus increasing the resulting error. If the CVs are being estimated from an actual sample, $x(p)$ must be replaced by the Student value $t(p; N-1)$.

Fig. 2 shows the CLs as functions of N for two different sampling methods (samples from the whole of grid 3; samples from the sub-area grid 2 as estimation for the AM of the grid 3-size area) and two different assumptions for the process related uncertainty $CV_{\text{proc.}}$. It can be



seen that, the larger the process related contribution to the TPCV, the smaller the difference between the CLs resulting from the two different sampling methods. As the assumption $CV_{proc} = 5\%$ is probably the more realistic one, sampling is much more effective (in terms of minimizing the CL) if done over the whole 1 ha area rather than just over a 100 m² one.

4 Conclusions

The spatial variability of ¹³⁷Cs deposition found in this investigation fits well the values reported in literature, as can be seen from fig.3 which shows coefficients of variation of ¹³⁷Cs deposition versus area size.

The point labels refer to literature as follows:

- a this project
- 1 Austrian Ecology Institute, LETTNER et al. (1994)
- 2 Austrian Federal Environmental Agency, radionuclides in a forest ecosystem, UBA (1995)
- 3 Austrian Federal Environmental Agency, investigations of an agrarian ecosystem, unpublished
- 4 Austrian Ecology Institute, investigation of an area near Gmunden, Upper Austria, LETTNER et al. (1994)
- 5 BACHHUBER et al. (1987) (global fall-out)
- 6 GUSTAFSSON et al. (1987)
- 7 HENRICH et al. (1988)
- 8 NYHAN et al. (1983) (global fall-out)
- 9 PADOVANI et al. (1990)
- 10 SIMOPOULOS (1989)
- 12 McGEE et al. (1995)
- 13 SUTHERLAND & DE JONG (1990)
- 14 HAUGEN (1992)

If the mean deposition density of a 1 ha area has to be determined, the samples should in fact be taken from points spread over the whole of this area, as 100 m² sub-areas may not yield representative figures.

However, it is hard to say how representative the selected area actually is for Austrian meadows and pastures. Further similar investigations on different types of areas and fields would be necessary to answer this question.

Acknowledgement

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CONTAMINATION OF AUSTRIAN SOILS BY CAESIUM-137

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Austria ranks among the countries of the world that are most strongly affected by the Chernobyl fallout. The average contamination in Austria amounts to 23.4 kBq ¹³⁷Cs/m². Higher fallout values (above 200 KBq ¹³⁷Cs/m²) can only be found in the Ukraine, Belarus, Russia and in some parts of Scandinavia.

Of the 70 PBq ¹³⁷Cs that were released from the Chernobyl plant and distributed over the whole world, approximately 2 PBq were deposited on the Austrian territory.

This study was elaborated by the Austrian Federal Environment Agency and the Federal Ministry of Health. It combines all measurements (of ¹³⁷Cs in soil made after the Chernobyl accident) available up to now and presents them graphically. 1,788 measurements could be used for the presentation of the map, 168 of which had been carried out in areas of neighbouring states near the Austrian border.

The data are compiled in a data bank which will serve as evidence in case of radioactive fallout on Austria in the future. The data are stored in the Geographic Information System (GIS) of the Federal Environment Agency and are now part of the Austrian-wide soil information system.

Fallout of radionuclides leads to the contamination of the soil. The fallout activity, however, is not the same as the activity of the soil at the time of the measurement. There are two reasons for this. First, very often measuring took place years after the actual fallout of the radionuclides. In order to compare the measured values, they were calculated for the 1st of

May 1986 taking into account the physical decay of radionuclides according to their half-life. And second, ecologic processes can lead to a dispersion or to an accumulation in soil and thus to a change of the fallout amount at a specific point.

The following are very strongly contaminated sites in Austria with peak values of soil contamination up to more than $100 \text{ kBq}^{137}\text{Cs}/\text{m}^2$:

- A strip leading from the North of Austria to the Hohe Tauern: parts of the areas Waldviertel, Mühlviertel and Hausruckviertel, the area around Linz, the Welser Heide, the Pyhrn Area, the Salzkammergut, and the western Niedere Tauern and Hohe Tauern up to the Zillertaler Alps.
- An area in the South of Austria: Koralpe, Southern Carinthia – this strip stretches into Italy.

Owing to the large number of measuring points a detailed isoline interpolation could be made in some highly contaminated areas.

The following are areas with a very low contamination below $10 \text{ kBq}^{137}\text{Cs}/\text{m}^2$:

- The East Austrian lowland, the Marchfeld, the Weinviertel, the northern Burgenland, and the Tullnerfeld.
- Parts of Carinthia and the southern part of East Tyrol,
- Some of the western parts of the Mühlviertel and the Innviertel,
- Parts of Vorarlberg: the Rhine Valley and the southern part of the Province.

Bodenbelastung durch Cs-137

Fig. 1: Contamination of Austrian soils by caesium-137

Cs-137 bezogen auf 1. Mai 1986

Einheit kBq/m²

< 10

10-15

15-25

25-40

40-60

60-100

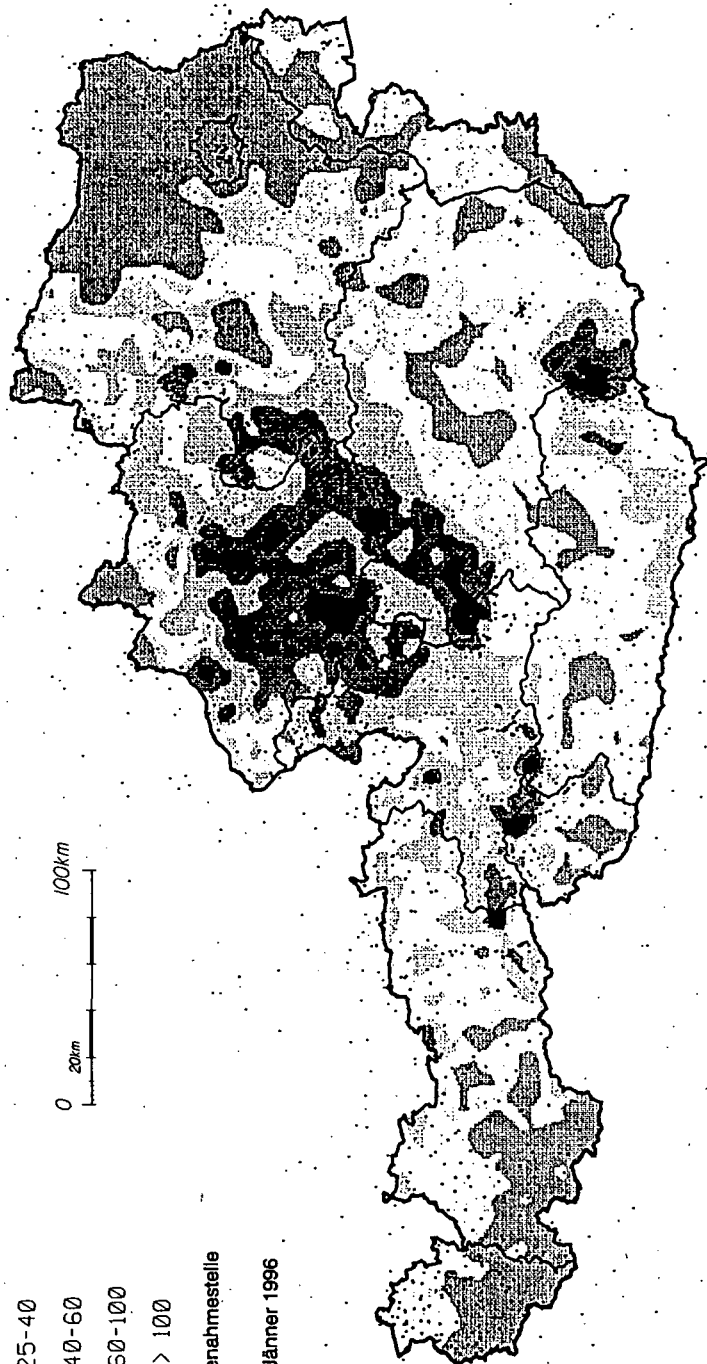
> 100

Probenahmestelle

Stand: Jänner 1996

Umweltbundesamt

0 20km 100km



VARIABILITY OF THE DEPTH-PROFILES OF ^{137}Cs IN SOILS OF THE PROVINCE OF SALZBURG

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ABSTRACT

Radionuclide contamination in soil does not only reveal spatial distribution but is also subject to a great variety of vertical depth-profiles changing with time. Small-scale scattering of the depth profiles due to statistical processes can be distinguished from differences correlating with different soil types. Based only on geographical criteria a classification scheme into a few reduced number of types can be established.

INTRODUCTION

The Chernobyl fallout in 1986 was very unevenly distributed in the environment in general as a consequence of the prevailing meteorological conditions at the sites affected. Different methods for the determination and for the measurement of the surface contamination have been applied. The majority of the surface deposition data originate from analysis of soil samples collected after the fallout. Another source for contamination data are the results of in-situ gamma-spectrometry measurements that became more and more important in the last years. In terms of the statistical interpretation of the data collected the question of representativeness of the results of contamination measurements was recognised to be of great importance but only few data concerning this issue have become available. As a contribution to the important question of spatial variability of the fallout a detailed study of the homogeneity of the surface contamination was carried out in the Province of Salzburg (LETTNER et al, 1994). In the course of this study information about the variability of the depth-profiles of ^{137}Cs was collected and furtheron used for the interpretation of in-situ gamma spectrometric measurements.

MATERIALS and METHODS

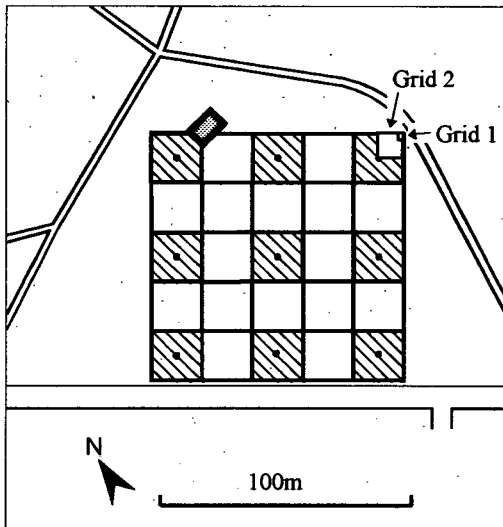


Fig.1: Sketch map for sampling procedure on test-site in the urban area of Salzburg. Dots on hatched squares of grid 3 (100 m * 100 m) mark points for soil sample collection. Analogous procedure for sample collection on grid 1 and grid 2.

For the detailed investigation of the spatial radionuclide distribution (BOSSEW et. al, this issue) and for the depth-distribution of the radionuclides in the soil, soil-core samples were collected on a predetermined test-site in the urban area of Salzburg. On the test site an area of 100 m * 100 m (grid 3) was selected and divided into 25 squares of 20m by 20m each. For the determination of the depth profile the samples from the centre of the hatched squares (Fig.1) were used. The subsquare at the NE corner was divided further into 4 squares of 10 m * 10 m each. The same procedure applied for soil-core sampling at grid 1 was scaled down and repeated on the outermost 10 m*10 m square (grid 2).

Another grid of 1 m * 1 m (grid 1) was defined on the NE corner of grid 2, where the same procedure was repeated again.

The soil cores were sampled with a commercial soil-sampler of 8.1 cm diameter, resp. 50.3 cm² cross-section. For the determination of the depth-profile the cores were cut into slices 2 cm thick, altogether 7 slices summing up to 14 cm core-length. The slices were dried at 105°C, homogenised, measured with a HPGe-Detector and the spectra were evaluated with an internally developed software.

For further investigation of the surface contamination in the Province of Salzburg - covering an area of appr. 7200 km² - in-situ gamma-spectrometry measurements have been performed on about 40 sites and soil samples were collected on each site. Only undisturbed meadows were selected where the depth-profile was determined by cutting the soil-cores into slices, 1 cm thick in the uppermost 2 cm soil core and 2 cm thick below. For the evaluation of the in-situ gammaspectra the depth-profiles were used as a basis for the calculation of the unscattered photon flux.

RESULT and DISCUSSION

The soil depth-distribution in general varies to a great extent both on small-scale and on large-scale. In spite of these variations typical patterns or shapes of depth-distribution curves can be distinguished and attributed to different soil types. The most detailed data are available for the test-site in the urban area of Salzburg, where significant differences between the soil-profiles of grid 1 (1 m²), grid 2 (100 m²) and grid 3 (10.000 m²) can be recognised (Fig.2). In each grid the largest variation of the relative activity tends to be in the uppermost and the lowest layer. The largest scattering is within the depth-profiles of grid 3, that covers the whole area of the

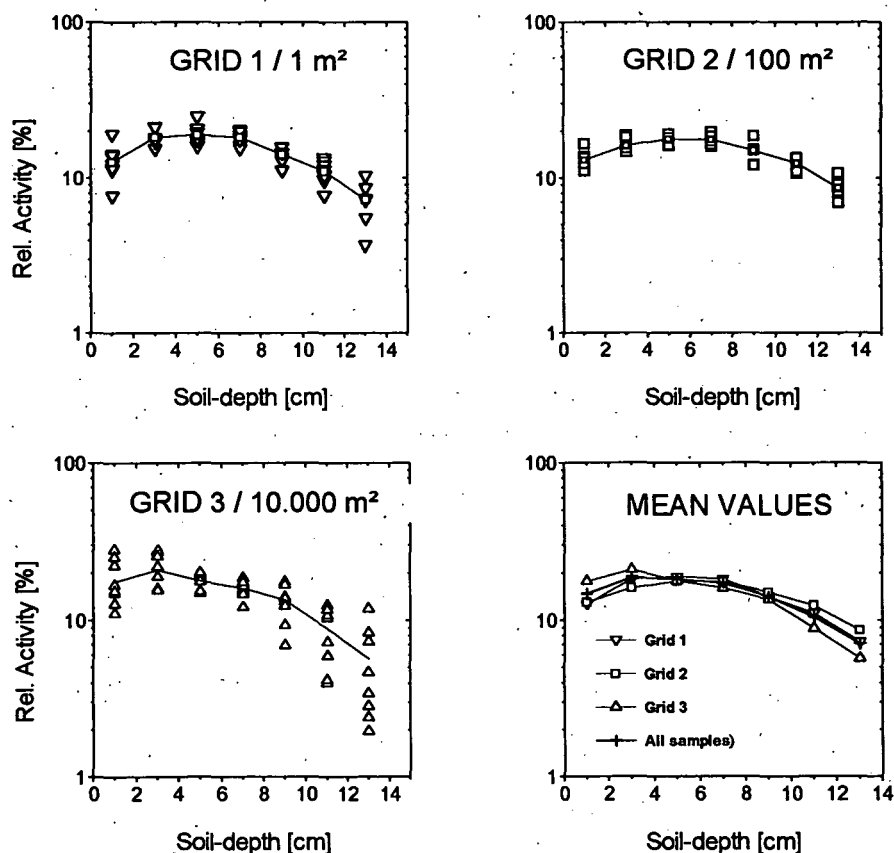


Fig.2: Soil depth-profiles for ¹³⁷Cs in soil cores of grid 1-3 from the test site in the urban area in Salzburg city.

test-site (10.000 m²). If the mean values of the grids are compared it is obvious that the relative activity in the soil layer from 4-6 cm is almost identical in each grid (Table 1). The depth-profile from grid 1 is similar to the average depth-profile. The largest differences are between grid 2 and grid 3, with similar relative activities in the layer from 4-6 cm and increasing differences downwards and upwards as well. In grid 1 and grid 2 the maximum of the relative activity can be found between 4-6 cm, indicating intensive downward-migration, whereas in grid 3 the maximum relative activity is between 2-4 cm. Considering the sampling areas for grid 1, grid 2 and grid 3, the depth-profile for grid 3 is more representative for the whole area than the other depth-profiles. The differences between the soil profiles are significant, accepting an error-probability of 5% for the layer 0-2 cm, 2-4 cm, 10-12 cm and 12-14 cm (Table 1).

The 50% depth is 4.3 cm for grid 3 and 5.1 cm for grid 1 and grid 2. The difference in the position of the 50% depth is significant between grid 2 and grid 3 (Table 2). These findings indicate that a considerable variability in the depth-profiles of the radionuclides in the soil can be expected on a relatively small scale.

Table 1: Mean values (AM) and coefficients of variation (CV%) of the relative activities for ¹³⁷Cs in the soil layers for grid 1, grid 2 and grid 3 from the test-site in the urban area of Salzburg.

Soil layer from - to (cm)	Grid 1 / 1 m ²		Grid 2 / 10 m ²		Grid 3 / 10.000 m ²	
	AM	CV%	AM	CV%	AM	CV%
0-2	12.58	17	12.93	16	17.51	27
2-4	18.11	14	16.01	16	20.91	18
4-6	18.82	22	17.68	13	17.81	25
6-8	18.11	16	17.5	9	15.90	35
8-10	14.1	17	14.95	20	13.41	42
10-12	11.01	24	12.35	14	8.79	54
12-14	7.26	34	8.58	16	5.67	70

Table 2: Position of the ¹³⁷Cs activity 50% depth for soil cores from grid 1 - 3. AM=arithmetic mean, CV% = coefficient of variation, n = number of measurements. Statistical test result: + significant difference with error-probability of ≤ 5%

	Position of the 50% depth [cm]			Statistical test result	
	Grid 1	Grid 2	Grid 3	Grid 2	Grid 3
AM	5.1	5.1	4.3	Grid 1	
SD	0.4	0.4	1.2	Grid 2	+
CV%	7	7	28	Grid 3	
n	8	7	8		

As physico-chemical processes like leaching and downward migration are assumed to be rather constant in one soil-type, similar shapes of depth-profiles without significant differences due to these processes could be expected. It is unclear to which extent biological activities of soil-organisms contribute to the distribution of radionuclides in the soil and to the distortion of depth-profiles. It is obvious that there is an influence of bioturbation which is not negligible and this might be a possible explanation for the differences. These findings could at least be

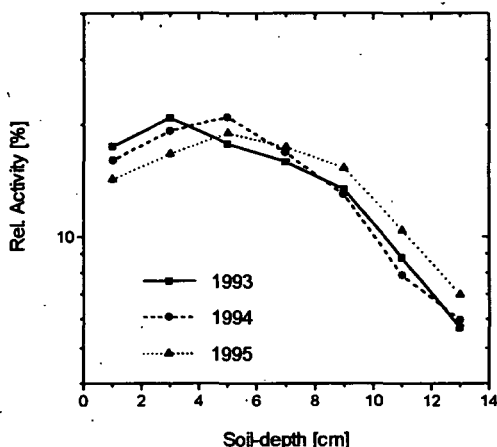


Fig.3: Variation of the depth-profile with time: Mean values of soil samples from grid 3.

qualitatively explained by the activities of moles, producing large mole hills covering between 5 and 10% of the test area of 10.000 m². The depth profiles show only a small variation within the period from 1993 to 1995 by downwards migration resulting in flattening out of the depth profile (Fig.3). The small changes over these years indicate a temporal variation of the migration velocity along the soil profile which was also found by other authors (VELASCO et al, 1993, KONSHIN, 1992a,b).

Further depth-profiles were sampled on agricultural used land in the Province of Salzburg, that can be classified on geographical criteria in soil samples from upland pastures, soil from valley floors and soil from more intensively used pastures in flatland of the forealpine region. The depth profiles show a considerable range of scattering with the only common feature of the depth-distribution to be a function of the soil depth. Only in a small number of profiles the activity seems to be independent of the soil depth. In the majority of the profiles analysed the activity increases downwards, showing the maximum in a soil-depth of 2-6 cm followed by a further decrease which approximates an exponential function. Depth-profiles in soil cores from upland pastures from above 1600 m altitude are significantly different from all other soil profiles (Fig.4). These depth profiles are characterised by the maximum of the specific activity to be concentrated in the uppermost 2 cm of the soil followed by an exponential decrease, halving the specific activity about every 1.5 cm. In some soil-profiles from valley floors the

exponential decrease following the maximum of the specific activity are similar to those from upland pastures, but the maximum is significantly shifted downwards to soil depth of 2-4 cm. In depth-profiles samples from more intensively used agricultural soils the decrease with depth is usually less and more irregular shaped.

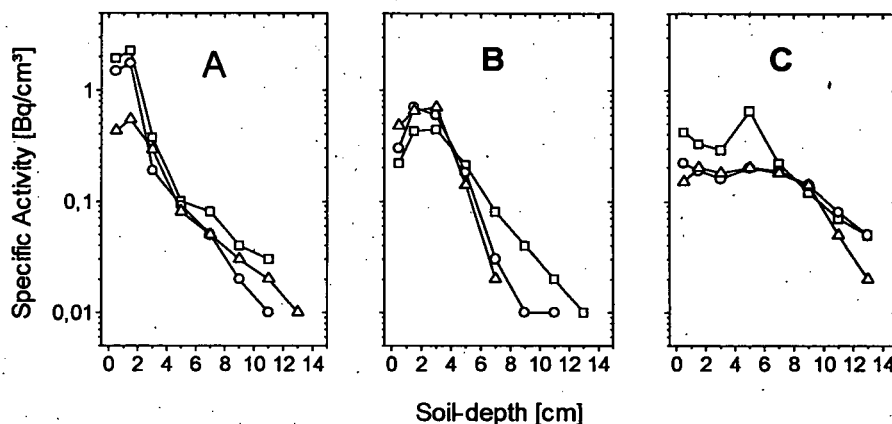


Fig.4: Soil depth-profiles for ^{137}Cs in soil from different locations. A - alpine regions / upland pastures. B: valley floors, C: flatland with more intensive agricultural use.

Summing up these characteristics the different soil profiles can be grouped according to geographical positions correlating with significant characteristic differences in the migration behaviour of ^{137}Cs (Table 3).

Table3: Qualitative overview of the ^{137}Cs migration behaviour in different soil-types based on geographical position

Soil / Situation	Activity maximum	Activity-decrease as a function of depth
Pasture soil	in the uppermost 2 cm soil-layer	large to very large
Soil from valley floors	mostly in the 2-4 cm soil layer	large
Soil from flatland area, or soil from intensively used areas	activity is more evenly distributed in the upper soil layers. Activity maximum tends to be between 4-6 cm	medium

CONCLUSIONS

- The shape of depth-profiles of ^{137}Cs in soil of the same type is considerably large on a small scale (100 m² to 10.000 m²) resulting in statistical differences in the 50% depth. Though these differences are significant the shape of the depth profile in the same soil type is similar on a regional scale.
- The differences between depth-profiles of different soil-types are larger than the variation within the depth-profiles of one soil-type.
- Different soil-types correspond to a certain extent with its geographical occurrence. Due to the geographical occurrence as reduced number of types of depth-profiles can be distinguished: (1) alpine terrain; (2) valley floors and (3) flatland areas.

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LONG-TERM REDUCTION OF ROOT UPTAKE OF CS-ISOTOPES AFTER NUCLEAR FALLOUT

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ABSTRACT

The long-term decrease in activity concentrations of ^{137}Cs in important foodstuffs was investigated in Austria. Using the fallout from the Chernobyl accident, the investigation comprised both annual plants (cereals, potatoes, vegetables) and perennial plants (grass). Large sample collectives were used to avoid artifacts due to single plant characteristics, and local variations influenced by soil properties.

After an initial decrease from first year to second year harvest by an approximate effective half-life of 96 d, the activity concentration of ^{137}Cs in annual plants decreases in the following years by an effective half-life of about 750 - 1170 d, equivalent to a biological half-life of 810 - 1300 d. The comparative value after nuclear weapon's testing was 1600 d (effective half-life).

In grass from first to second year a decrease to about 5 % of the peak value is observed. The long-term decay in the following years follows an effective half-life of about 710 d, equivalent to a biological half-life of 760 d. In semi-natural environments (alpine pastures) effective half-lives of up to 2000 d were observed which are caused by low fixation capacity and low pH of the soil as well as low plant growth and circulation of radiocaesium within living and dead plant biomass.

1 Introduction

The population exposure by ingestion of contaminated foodstuff after a large-scale fallout significantly depends, apart from the initial contamination, on the decrease of the activity concentration in all foodstuffs with time. As this is dominantly determined by the long-term change in the availability of long-lived radionuclides, in particular of ^{137}Cs , the question of radionuclide availability for root-uptake is of great significance for the long-term exposure.

While the short-term decrease in activity concentration in foodstuff is mainly caused by dilution due to plant growth and radioactive decay (Mück 1987, Mück et al. 1994) and thus little influenced by soil properties, the long-term decrease is predominantly controlled by:

- removal of activity from root-layer of soil by transfer into deeper soil layers or other effects
- increased fixation of radionuclides in soil
- transfer into non-edible or non-usable parts of the plant (e.g. roots)
- radioactive decay

It therefore, depends significantly on soil properties. For ^{137}Cs with a half-life of 30 years radioactive decay causes little reduction with time. Therefore, the decrease is dominantly controlled by effects which decrease the radionuclide uptake by roots. These are of main interest with regard to the long-term activity concentration in plants and foodstuff derived thereof.

The above effects are not clearly separable and the contribution by each effect is not easily determined by simple activity measurements, but of main interest is the overall decline of the activity concentration caused by all effects. Therefore, investigations were performed in Austria on this issue at various times after fallout (Mück 1987, Mück 1989, Mück 1994, Mück and Gerzabek 1995, Mück et al. 1990a, Mück et al. 1994, Rabitsch et al. 1994). This paper gives a summary of these investigations and includes most recent data on long-term decay.

2 Method

Special care was given to the proper selection of foodstuff samples for the investigations. On the one hand, fluctuations due to individual animals or single samples of plants were avoided by including a large number of samples with each type of foodstuff investigated (Mück 1995). On the other hand, with large sample sets care had to be taken to obtain comparable, representative sets of samples. With milk, in particular, avoiding individual animals was of great importance as the transfer rates in individual animals may vary significantly for ^{137}Cs which is controlled by the same metabolism in the body as ^{40}K . The latter is known to have significant differences in individual metabolism rates in the animal.

The selection of comparable, representative sets of samples is a particular problem when one considers the time span over which the sets should be comparable. The problem was approached by selecting different areas which were sampled over the whole period of seven years. Each area either covered a whole province or for some foodstuffs smaller areas, i.e. a dairy with a large variety of milk collecting tours. In order to make the sample sets comparable, special care was taken to incorporate the same subsets with each set of samples. The same subsets (same farmers or sites) were surveilled over the whole period of 7 years. The number of samples in each subset ranged between 10 (cows at alpine pastures), about 40 - 60 (cereals, potatoes, vegetables) to more than 2000 (cows in intensive agricultural areas).

In the first year the samples of about 1 to 2 kg were directly measured in a Marinelli beaker on Ge(Li)- and HPGe-detectors of 20 % to 30 % relative efficiency without previous sample preparation. All detectors were shielded by 5 - 10 cm of lead to improve the detection limit. They had been calibrated by standard sources and tested by intercomparison tests showing excellent agreement (STEGER 1987). With measurement times of 100 - 1000 s in the first year after fallout a detection limit of about 10 - 30 Bq/kg was obtained which was sufficient for the elevated activity levels present at that time. In the later phase (2nd to 6th year) the measurement time was increased to about 20000 - 50000 s to achieve a detection limit of 1 - 2 Bq.kg⁻¹.

At the end of surveillance period activity concentrations were so much reduced that all samples had to be specially prepared before measurement. Milk samples were dried by boiling them for several hours. All other samples were dried and then ashed at a temperature of 450°C for 5 hours (at steps of 110 °C, 300 °C and 450 °C, consecutively) to avoid losses of ¹³⁷Cs at higher temperatures. Very long measurements times of the dried or ashed samples of up to 100000 - 200000 s were required to obtain a satisfactory detection limit of 0.02 - 0.09 Bq.kg⁻¹ d.m.

3 Influence of "old" caesium from nuclear weapons' testing

After the nuclear weapon's tests between 1953 and 1964 about 20 times more ¹³⁷Cs was released and deposited worldwide than after the Chernobyl accident (UNSCEAR 1982, UNSCEAR 1989, MÜCK 1991). Since the caesium from this previous fallout had decayed only by a factor of 1.7 since then, its presence might influence the determination of the effective decay constant derived for the Chernobyl fallout. However, average ¹³⁷Cs-deposition rates in Austria after the Chernobyl fallout were about 4 times higher than after the nuclear weapons' tests (MÜCK, 1991). Furthermore, in the years following the fallout the caesium penetrated into deeper soil layers (MÜLLER et al., 1989) and only little was available anymore for plant uptake in 1986. With grass, for instance the amount still to be found in the sod was less than 0.2 % of that of the Chernobyl fallout (MÜCK et al., 1990 b).

A further proof is given by the ratio of ¹³⁴Cs to ¹³⁷Cs determined in the root layer of soil after May 1986. In no soil sample the ratio deviated from the ratio of 1.76 observed directly in the Chernobyl fallout (MÜLLER et al., 1989, STEGER 1987, GERZABEK et al., 1989). Therefore, the old ¹³⁷Cs from weapons' testing has negligible influence on the results of this work.

4 Results

4.1 Annual plants

Since annual plants are grown in a comparable way on tilled soil, significant differences should not be expected between different plants. Nevertheless, the average decrease of the activity concentration was investigated in several annual plants (Mück 1995). The trend in activity concentration in potatoe tubers is given in figure 1. In the first year after contamination a decrease by an effective half-life of 96 d is observed. In the following years the effective half-life amounts to 885 d. The difference in the two values is explained by the fact that the contamination in the first year is mainly due to translocation within the plant (leaves to tubers) while in the following years the activity concentration in the plant is dominantly caused by root uptake. Therefore, the effective half-life observed after the 2nd year is characteristic for soil transfer and thus for the ^{137}Cs -availability with regard to root uptake.

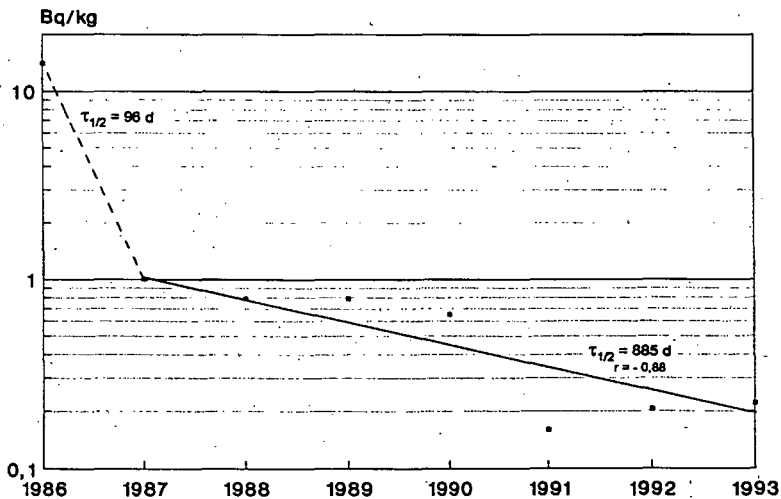
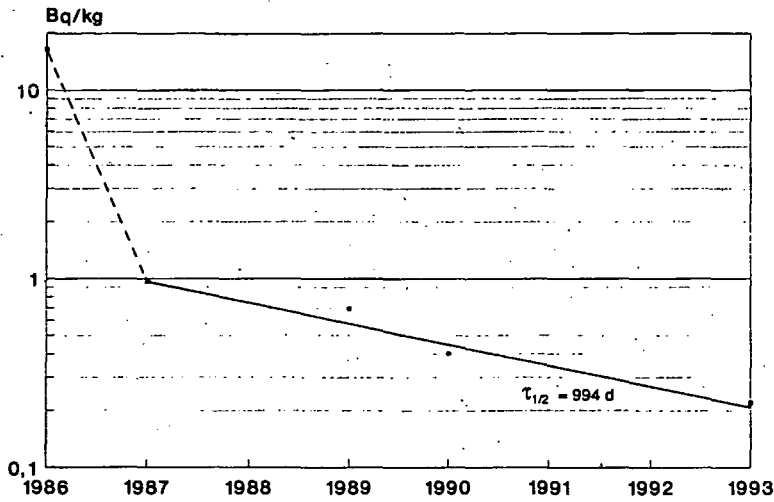
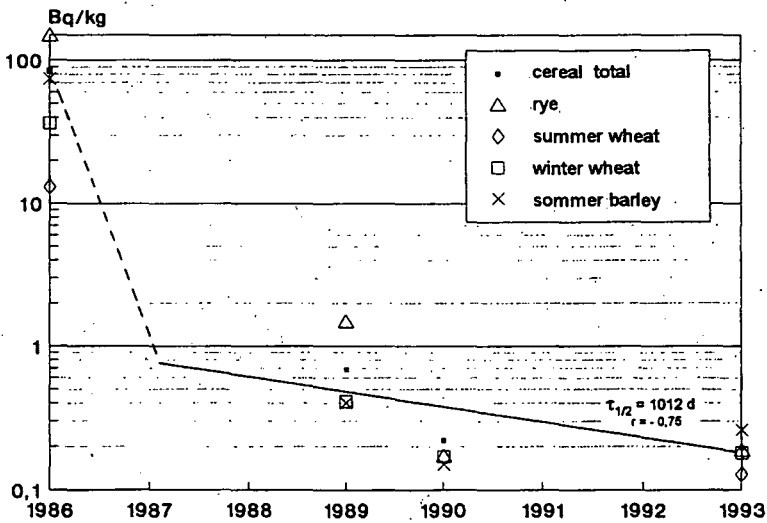


Figure 1 ^{137}Cs -activity concentration in potatoes

Figure 2 shows the comparable trend in a typical vegetable, cabbage. This vegetable was chosen as it is more consumed than most other vegetables. Also the decay characteristics should be comparable to other vegetables. If we compare to potatoes, we find the same typical decline in ^{137}Cs -activity concentration in the first year by about the same average half-life. The long-term effective half-life thereafter is also very similar. The differences are within statistical fluctuations. Thus the long-term decay does not significantly vary between these two plant species.

Figure 2 ^{137}Cs -activity concentration in cabbage

Another important foodstuff is cereal. Its trend in activity concentration is given in figure 3. The steeper decline from first year to second is caused by the more elevated activity levels in the first year due to direct contamination of the small plants. After the second year we observe the same trend as with potatoes and cabbage. The longer effective half-life may be caused by a

Figure 3 ^{137}Cs -activity concentration in cereals

statistical artifact - unfortunately, there are no data on average activity concentrations available for the second and third year. So the decay curve depends only on 3 values (1989, 1990 1993).

Comparing different cereal types we find only differences in activity concentration in the first year which are caused by the different growth stages of the plants at the time of fallout - winter cereals were more advanced in their growth stage than spring varieties. The decrease in activity levels in the 2nd to 7th year follows equal patterns for the different species. This was expected since different cereal species show only minor differences in root uptake.

4.2 Perennial plants

The most important perennial plant with regard to the contribution to the ingestion dose is grass. A significant amount of foodstuff relevant to human consumption is produced by feeding grass: milk, milk products, beef, veal, lamb, mutton and most of venison. Therefore, a major focus of the investigation was on the trend in activity concentration in grass. This was determined not in single grass samples which might show significant individual variations, but by measuring the activity concentration in milk. Milk was taken from dairies whereby a further averaging over individual fluctuations was achieved. Each sample taken from tanks of milk collecting tours averaged over an area of approximately 50000 - 250000 m² (Mück et al. 1994).

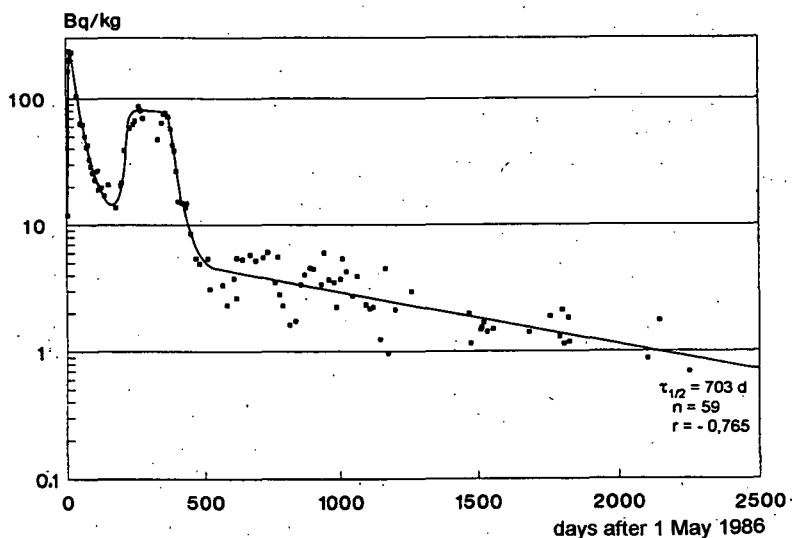


Figure 4 ¹³⁷Cs-activity concentration in milk in Eastern Styria

In figure 4 the average activity concentration of ^{137}Cs in milk in a region in Austria is given. While the variation in concentration in the first year is mainly caused by dilution effects in the initial phase and the re-increase after the 180th day by feeding hay which was produced in the earlier part of the year, the long-term decay after the decline after the 500th day (end of winter hay feeding period) is described by an effective half-life of approximately 700 d. Thus, also in grass with a completely different behaviour with regard to storage of radionuclides an equivalent effective decay is observed as in annual plants.

This effective half-life is, however, only observed in intensively cultivated agricultural areas. In extensive Alpine pastures the effective decay is quite different. Fig. 5 shows the ^{137}Cs -activity concentration in milk of three different Alpine pastures compared to average milk concentrations in the surrounding province. While province average shows about the same half-life as Eastern Styria, the two Alpine pastures show half-lives of 1400 and 2000 d. These significantly higher values are explained by low fixation capacity and low pH of the soil as well as low plant growth and the circulation of ^{137}Cs within living and dead plant biomass as shown by Mück and Gerzabek (1995). The half-life in these extensive pastures shows a whole range of values. This is demonstrated by the third pasture in fig. 6 which shows an effective half-life of 1000 d between the two values. Taking these variations into account, a classification of soils with regard to long-term Cs-transfer was proposed (Mück et al. 1990a, Mück and Gerzabek 1995).

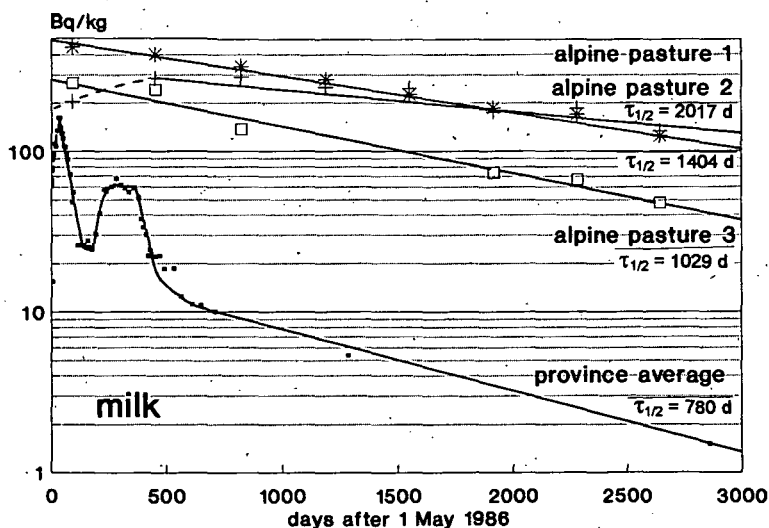


Figure 5 ^{137}Cs -activity concentration in milk in alpine pastures and the surrounding province

5 Conclusions

The long-term decay of the ^{137}Cs -activity concentration in plants after a nuclear fallout follows an effective half-life of about 750 - 1170 d both for annual and perennial plants. This is equivalent to a biological half-life of 810 - 1300 d. Only in semi-natural environments (alpine pastures) effective half-lives of up to 2000 d were observed, but these contribute little to the ingestion dose in Central Europe. After nuclear weapon's testing the comparative value for the effective half-life was 1600 d. This much longer half-life may be only explained by the continuous fallout at a lower level even after the test stop by the major nuclear powers.

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BEHAVIOR OF IODINE-129 IN THE SOIL-PLANT SYSTEM

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Abstract: In order to obtain information on the behavior of long-lived ^{129}I in the environment, investigations were carried out on (1) ^{129}I in soil samples (including a Chernobyl soil sample), (2) sorption of iodine on soil, (3) transfer of iodine from soil to plants, and (4) volatilization of methyl iodide from the soil-plant system. Iodine-129 was found to be accumulated in surface soil. Concentrations of this nuclide were higher in forest soil than in field soil. Sorptions of radioactive I^- and IO_3^- were high in soil samples having high concentrations of total organic C, active-Al and active-Fe. The sorptions were affected by pH, heating treatments of soil and co-existing ions such as SO_3^{2-} and stable iodine (I^- and IO_3^-). Effects of Cl^- and Br^- ions on the I^- and IO_3^- sorption were small. The soil-plant transfer factors obtained for different crops ranged from 0.0003-0.016. Methyl iodide was found to be volatilized from the soil-plant system. A marked emission was observed in rice plants grown on flooded soil. Results obtained in this study may contribute to the environmental assessment of radioiodine in relationship to its release from nuclear facilities as well as in the Chernobyl area.

1. Introduction

Radioiodine is one of the most important radioisotopes released into the environment from nuclear facilities and by nuclear weapons testing. Since iodine is accumulated in the thyroid gland of humans, considerable attention should be paid to the levels and the behavior of radioiodine in the environment. Two radioisotopes of iodine, ^{129}I (half-life: 1.6×10^{17} y) and ^{131}I (half-life: 8 d) are specifically important in terms of radioecology. Since the amount of ^{131}I produced in a reactor is much higher than that of ^{129}I , the former isotope is more important at the time of a reactor accident such as at Chernobyl and Three-Mile Island. However, ^{131}I decays away within several months of contamination. The long radiological half-life of ^{129}I , on the other hand, prevents this nuclide from disappearing, causing it to enter the geochemical and ecological cycles of iodine. Interest in ^{129}I is, therefore, currently increasing, particularly

regarding the operation of nuclear fuel reprocessing plants (Brauer and Strebin 1982, Muramatsu and Ohmomo 1986). In addition to environmental assessment involving nuclear reprocessing plants, ^{129}I analysis has a further significance. After the Chernobyl accident, increased numbers of thyroid cancers were found in children in Byelorussia (Bäverstock et al. 1992). However, it is not yet clear whether these thyroid disorders were caused by the ^{131}I released at that time. To carry out an epidemiological study related to the reactor accident, it is necessary to have data both on the thyroid disorder cases and on the ^{131}I levels of each region after the accident. Because only limited numbers of reliable measurements of ^{131}I were made in the former Soviet Union after the accident, it is no longer possible to detect this nuclide from the reactor because of its short radiological half-life. Since the long-lived ^{129}I should also be produced in fuels in the reactor and released into the environment together with ^{131}I , it might be possible to obtain information on the ^{131}I levels deposited in soil through measurements of ^{129}I .

We have carried out several studies on the levels and behavior of radioactive and stable iodine in the environment from the viewpoint of biogeochemistry (Muramatsu and Ohmomo 1986; Muramatsu et al. 1987, 1989, 1990, 1995; Muramatsu and Yoshida 1995; Yoshida et al. 1992). Our results may provide information not only for the radioiodine assessment of nuclear facilities (e.g. reprocessing plants) but also for the estimation of the levels and behavior of radioiodine in the contaminated areas due to the Chernobyl accident. In this paper, we summarize our recent investigations, together with new data, on (1) determination of ^{129}I in soil samples (including a Chernobyl soil sample), (2) sorption of iodine on soil, (3) transfer of iodine from soil to plants, and (4) volatilization of methyl iodide from the soil-plant system.

2. Materials and Methods

2.1 Determination of ^{129}I in soil samples

Neutron activation analysis (NAA) was applied for the determination of ^{129}I and ^{127}I (Muramatsu and Ohmomo 1986). An outline of the method is as follows. A sample (30-180 g) was mixed with ^{125}I (half-life: 60 d) yield tracer and then placed in a quartz tube and heated at 1000°C under oxygen gas flow. The evaporated iodine was collected in a trap (KOH: 8 g; K_2SO_3 : 2g; H_2O : 180 ml). After this trap solution was acidified with HNO_3 , it was placed in a separatory funnel. 5% NaNO_2 solution (about 1 ml) was added to convert I^- to I_2 , which was extracted into a CCl_4 phase by shaking. Next, iodine was back-extracted into the aqueous phase containing K_2SO_3 as I^- . The volume of the aqueous phase was reduced to about 0.2 ml

by heating and it was transferred into a quartz ampule. The ampule was sealed and irradiated in a research reactor (JRR-4, JAERI) at the flux density of $5.5 \times 10^{13} \text{ n cm}^{-2}\text{s}^{-1}$ for 1-6 hours. The following reactions were considered for quantification of ^{129}I and ^{127}I ; $^{129}\text{I}(\text{n}, \gamma)^{130}\text{I}$ (half-life: 12.3 h) and $^{127}\text{I}(\text{n}, 2\text{n})^{126}\text{I}$ (half-life: 13 d). After cooling the sample, the iodine fraction was purified by solvent extraction using CCl_4 . Finally the iodine was precipitated as PdI_2 and counted on a Ge-detector to quantify ^{130}I peaks (418, 538 and 739 KeV) and ^{126}I peaks (389, 491 and 754 KeV). The detection limits for ^{129}I and ^{127}I in soil by this method were about 0.1 mBq kg^{-1} and 0.1 mg kg^{-1} , respectively. The $^{129}\text{I}/^{127}\text{I}$ ratio of 10^{-9} could be detected.

2.2 Sorption of iodine on soil

Sorption of iodide (I^-) and iodate (IO_3^-) on soils was examined by the batch method. Samples (3 g) were mixed with deionized water (30 ml) in polyethylene bottles (50 ml), and radioiodine tracers ($^{125}\text{I}^-$ and $^{125}\text{IO}_3^-$) were added. The bottles were shaken at 23°C for certain periods (1 to 14 d). Finally the samples were centrifuged and aliquots of the supernatant solution were counted with an NaI scintillation counter. The soil-solution distribution coefficient (K_d) was calculated from the activities remaining in the solution. For examining the effects of co-existing ions, the salts KBr, NaCl, KI, KIO_3 , NaNO_3 , KH_2PO_4 , Na_2SO_3 and Na_2SO_4 were added to make 0.1 M solutions.

2.3 Transfer of iodine from soil to plants

Andosol, which is the most common soil type in Japanese fields, was used. Radiotracer (^{125}I) was thoroughly mixed with the soil (3 kg) in a Wagner pot (3 l). A plant growth chamber (Puffer-Hubbard 38-15 HLE), installed in a hot laboratory, was used for the cultivation. Light intensity of the chamber was about 70000 lux at the top of the plants. During cultivation, temperature and moisture in the chamber were controlled in accordance with the outdoor conditions. After the plants were harvested, they were separated into organ parts (e.g. leaves, grains) and ^{125}I concentrations were determined with an NaI scintillation counter. The soil-plant transfer factor (or concentration ratio) is defined as "the ^{125}I concentration in the plant part (Bq g^{-1} , fresh)" divided by "the ^{125}I concentration in soil (Bq g^{-1} , dry)."

2.4 Volatilization of methyl iodide from the soil-plant system

Radioiodine volatilized from plants was collected with two traps. The first trap contained silver wool for collecting inorganic iodine and the second trap contained activated charcoal

(supplemented with triethyldiamine) for collecting organic iodine. Activities were measured with an NaI scintillation counter. Gas chromatographic mass spectrometry and gas chromatography with an electron capture detector were also used to identify the iodine species.

3. Results and Discussion

3.1 Determination of ^{129}I in soil samples

Results obtained for ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios in soil samples collected from Tokaimura, Ibaraki Prefecture, are summarized in Figure 1. The concentration range of ^{129}I in soil was 0.9 to 180 mBq kg⁻¹. It was found that the levels around Tokaimura, where a nuclear fuel reprocessing plant is located, were higher than at other places in Japan (Muramatsu and Ohmomo 1986). The ^{129}I concentrations in soil samples collected in forests tended to be higher than those in field soils. This indicated that the nuclide released into the atmosphere may be trapped by leaves or needles and transferred to the ground through wash-out by rain and/or falling leaves, thus accumulating in the surface soil. Lower ^{129}I concentrations in field soils than in forest soils might be explained by the lower deposition rate and tilling procedures used in the fields. The $^{129}\text{I}/^{127}\text{I}$ ratios were higher in rice paddy soil than in wheat field soil. This was due to the lower stable iodine concentrations in the former soil. Vertical distributions of ^{129}I in soil collected from Tokaimura were measured. Results indicated that most of the ^{129}I released from the facility during a 20-year operations had been retained in the first 10 cm of the surface soil. These results suggest that most of the ^{129}I derived from the Chernobyl accident might still be accumulated in the soil surface in the contaminated area.

A soil sample contaminated by Chernobyl fallout was obtained from the IAEA in line with our participations in the agency's intercomparison program. The mean value of our three determinations for ^{129}I was 1.6 mBq kg⁻¹ (or 12×10^8 atoms g⁻¹), which agreed well with the IAEA recommended value of ^{129}I (12.2×10^8 atoms g⁻¹ for ^{129}I). The concentration of ^{137}Cs (5180 Bq kg⁻¹) measured in this sample was about two orders of magnitude higher than that observed in common surface soils in Japan. Both the ^{129}I concentration (1.6 mBq kg⁻¹) and the $^{129}\text{I}/^{127}\text{I}$ ratio (1.7×10^{-7}) measured in the Chernobyl sample were higher than the respective values obtained before the Chernobyl accident in areas far from nuclear facilities in Japan, e.g. Nagano: 0.13 mBq kg⁻¹ or 6.5×10^{-9} ; Tokyo: 0.78 Bq kg⁻¹ or 6.0×10^{-9} . This suggested the possibility that ^{129}I levels around the Chernobyl reactor have been enhanced by the release of the nuclide though the accident. Since there is some contribution to the ^{129}I levels in the environment due to nuclear weapons testing, it is necessary to analyze many other

samples to conclude the effectiveness of the ^{129}I measurements for the estimation of the ^{131}I following a reactor accident.

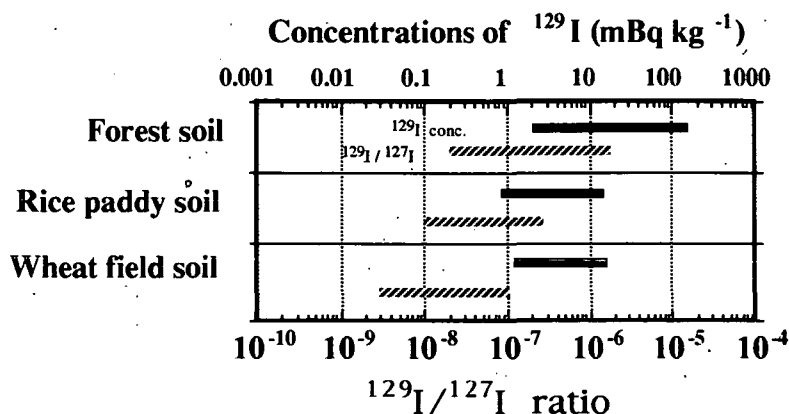


Fig. 1 Ranges of ^{129}I concentrations and the $^{129}\text{I}/^{127}\text{I}$ ratio in soil collected in Tokaimura

3.2 Sorption of iodine on soil

The soil-solution distribution coefficient (K_d) for both I^- and IO_3^- is known to vary very widely, i.e. <0.1 to 8000 ml g^{-1} (Muramatsu et al. 1982; Whitehead 1984; Yoshida et al. 1995). High values were found in soils having high concentrations of total organic carbon, active-Al and active-Fe (Al and Fe extracted by a mixture of oxalic acid and ammonium oxalate). Andosol, one of the most typical Japanese soils derived from deposits of volcanic ash, showed specifically high K_d values. These results indicated that added radioiodine was associated on the surface of sesquioxides of Fe and Al, noncrystalline silicates such as allophane, organic materials and complexes of metals with humus. The K_d s for radioiodine, specifically I^- , were decreased when the samples were air-dried or heated. A marked decrease was found in the soil samples heated to more than 150°C prior to the batch experiment. The sorption of I^- and IO_3^- on soil increased with decreasing pH. Effects of co-existing ions (0.1 M) on the sorption of $^{125}\text{I}^-$ and $^{125}\text{IO}_3^-$ were also examined. Table 1 shows the results obtained for Andosol (shaking time: 1 and 14 d). Compared to the control sample, almost no effects were found for Cl^- and Br^- . The presence of SO_3^{2-} greatly reduced the sorption of both $^{125}\text{I}^-$ and $^{125}\text{IO}_3^-$. Stable I^- and IO_3^- also affected the sorption of $^{125}\text{I}^-$ and $^{125}\text{IO}_3^-$, respectively.

Table 1 Sorption of $^{125}\text{I}^-$ and $^{125}\text{IO}_3^-$ on Andosol in the presence of co-existing ions

Co-existing ions (0.1 M)	$^{125}\text{I}^-$ (%)		$^{125}\text{IO}_3^-$ (%)	
	1 d	14 d	1 d	14 d
Control	96.8	99.2	93.7	94.7
Br^-	97.6	99.2	95.9	97.1
Cl^-	95.9	98.7	93.9	94.5
I^-	0.4	0.4	76.2	24.8
IO_3^-	81.8	79.8	19.1	15.5
NO_3^-	87.2	97.9	88.6	89.2
PO_4^{3-}	59.2	59.7	50.8	54.1
SO_3^{2-}	0	0	0	0
SO_4^{2-}	88.8	92.0	75.5	80.4

3.3 Transfer of iodine from soil to plants

Results obtained for the soil-to-plant transfer factors (or concentration ratio) in the edible parts of crops are summarized in Table 2. The range of the transfer factors was 0.0003-0.016 (Muramatsu et al. 1995). The IAEA recommended value for the transfer factor (concentration ratio) of iodine is 0.02 and it is widely used in environmental assessment for radioiodine. The transfer factor obtained for a common leaf vegetable (komatsuna) in Japan was comparable to the IAEA value. However, the values for tomato, grains and beans were significantly lower than their leaf value. The transfer factor for polished rice (0.0019) was less than 1/1000 of that of the rice plant leaves. Since the transfer factor of iodine for agricultural crops varies widely, we note that only one representative value for the transfer factor is insufficient. Different values of transfer factors should be established for plant groups as categorized by the type of their edible part. In addition to the type of crops, the transfer factors are also expected to be influenced by the sorption characteristics of soils. As mentioned above, we found very high sorption of I^- and IO_3^- on common Japanese field soil (e.g. Andosol). This may explain the relatively low transfer factors observed in our experiments using Andosol.

Table 2 Soil-to-plant transfer factors (TF) of radioiodine for edible parts of agricultural crops

Crops	TF
Spinach	0.0031
Komatsuna	0.016
Tomato	0.0003
Soybeans	0.0029
Rice (polished)	0.0019
IAEA recommended	0.02

3.4 Volatilization of methyl iodide from the soil-plant system

Iodine emission from soil was highly stimulated by the presence of plants (Muramatsu and Yoshida 1995). A marked emission of gaseous iodine from rice plants grown on flooded soil was observed, whereas plants grown on unflooded soil, such as oat plants, showed a considerably lower emission. Seasonal patterns in the iodine emission were observed for rice and oat plants. The emission rate increased with time from planting and the maximum value was observed in the late tillering stages (shortly before heading) of the plants. The chemical species of volatilized iodine was identified as methyl iodide (CH_3I) from gas chromatography.

The following mechanism for the high production of methyl iodide in the soil-rice plant system seems reasonable: Microorganism activities in the soil (particularly the rhizosphere) are increased by flooding and the effects of root exudates and/or root autolysis, so that eventually an anaerobic condition (low Eh) is generated. Under this condition iodine is desorbed from the soil into the soil solution as iodide (I^-). We presume that the iodide dissolved in the soil solution in the rhizosphere is biomethylated, possibly by the effect of enzymes produced by microorganisms or roots. For example, the enzyme methyl halide transferase, as reported by Wuosmaa and Hager (1990), might be a candidate for methylating iodide. The biogenic methyl iodide produced in the soil might be transported through the intercellular gas space and aerenchym system in the plants into the atmosphere, presumably by a mechanism similar to methane emission from rice fields.

Iodine-129 was still detected in the air collected around a reprocessing plant several years after its closing as reported by Brauer and Strebin (1982). This might be explained by the volatilization of the nuclide from the contaminated soil. Volatilization of iodine from the soil-plant system is thought to be important to understanding the behavior of the long-lived ^{129}I in the environment and this pathway should also be considered in establishing a transfer model

for ^{129}I to estimate radiation doses.

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INFLUENCE OF SOIL PROPERTIES AND CLIMATIC CONDITIONS ON ^{137}Cs VERTICAL DISTRIBUTION IN SOME CHILEAN SOILS

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ABSTRACT

Long-term downward distribution of global fallout ^{137}Cs was studied in different soil types of four Chilean climatic zones, in natural and semi-natural environments. The ^{137}Cs soil content showed an exponential decrease with depth; relaxation depth ranged from 0.9 cm in the polar climate environment up to 6.8 cm in the mesothermal warm summer one. In the Antarctic and Westpatagonic ecosystems the long freezing periods hindered the ^{137}Cs downward movement. In temperate and tropical climate zones, the amount of soil coarse pore volume, and annual rainfall rates appear to be the main factors influencing the radionuclide's distribution in the soil profiles.

1 INTRODUCTION

Around 25% of the total global fallout from atmospheric nuclear explosions have been deposited on the Southern Hemisphere (UNSCEAR 1982, UNEP 1984). Schuller¹ et al. (1993) reported for Chilean sites located between latitudes 38 and 41°S (Lake Region), ^{137}Cs soil contents varying between 500 and 6000 Bq m⁻², depending upon mean annual precipitation. No traces of deposit of contaminants released at Chernobyl were detected in soils of this Chilean region with high atmospheric fallout (Schuller² et al. 1993).

In Chilean territory, several climates from polar to tropical occur. Also, soil properties vary strongly, depending on geological and climatic conditions. The objective of this study was to contribute to the evaluation of the effect of soil parameters, under different climatic conditions, upon long-term vertical distribution of ^{137}Cs in soils from natural and semi-natural environments in Chile.

2 MATERIAL AND METHODS

Four natural or semi-natural, sampling areas within different climatic zones were selected: 1) King George Island, located in the South Shetland Islands, Antarctic Territory; 2) Torres del Paine, in Westpatagonia; 3) Lake Region; and 4) Easter Island. Two soils, with different pore texture, and consequently water permeability properties, were studied in each of these areas. The geographical position, climate, and mean annual precipitation (FACH 1991) of the sampling sites are shown in Table 1.

To study ^{137}Cs downward distribution, soil samples were cut with a palette-knife from three of the walls of 0.5 to 1 m² pits, in 1 to 3 cm increments up to 50 cm depth. From the fourth wall, soil samples were taken from each horizon in order to evaluate physical and chemical soil properties: pore size distribution, texture, carbon content, pH (1: 2.5 with CaCl_2 1 M) and cation exchange capacity.

Additionally, a 0.09 m² sample of the moss or lichen cover from the King George Island sites, and 1 to 2 m² of the vegetation cover of the other sites were collected. Plant cover of the Patagonia and Easter Island sites were steppe grass (*Poa spp.* and *Estipa spp.*). The vegetation cover of the Lake Region sites consisted of about 90% grass (*Agrostis capillaris*, *Holcus lanatus*, *Dactylis glomerata*) and 10% forbs (*Leontodon taraxacoides*, *Hypochoeris radicata*, *Trifolium repens*) (Ramírez et al. 1989).

The samples were analyzed for ^{137}Cs using Canberra and Ortec HP Ge detectors of 28% relative efficiency for the 1.33 MeV peak. The overall uncertainty of the activity determination was below 5%. Analytical results were expressed as ^{137}Cs activity per unit mass (Bq kg^{-1}) in each depth increment. This was converted to activity per unit area (Bq m^{-2}) of the

Table 1: Characteristics of the sampling sites

Sampling area	Site code	Coordinates of sampling site	Climate	Mean annual precipitation (mm y^{-1})
King George Island	1A	62.4°S 58.9°W	Polar	800
King George Island	1B	62.4°S 58.9°W	Polar	800
Westpatagonia	2A	51.1°S 72.6°W	Polar isothermic tundra	700
Westpatagonia	2B	51.1°S 72.6°W	Polar isothermic tundra	700
Lake Region	3A	41.0°S 73.1°W	Mesothermal warm summer	1500
Lake Region	3B	40.4°S 73.0°W	Mesothermal warm summer	1300
Easter Island	4A	27.1°S 109.5°W	Tropical	1150
Easter Island	4B	27.1°S 109.5°W	Tropical	1150

depth increment by multiplying it by the increment thickness (m) and by the corresponding bulk density (kg m^{-3}). The areal activity, $A(x)$, below a given depth, x , (referred to here as 'soil content') was obtained by summing the areal activity over all depth increments below x . The total soil content for each site was obtained by summing the areal activity over all depth increments of the soil profile. The estimated uncertainty for the total soil contents measured was around 15%.

3 RESULTS AND DISCUSSION

The measured edaphic parameters are shown in Table 2. The soils of the sites coded A on each sampling area, have a lower coarse pore space than the soils coded B within the same area. Information concerning ^{137}Cs soil content and downward distribution of the sampled locations is also summarized in Table 2.

3.1 ^{137}Cs SOIL CONTENTS

No correlation was observed between the total ^{137}Cs soil content and the mean annual precipitation of the sites analyzed. This because of their different latitudinal positions, type, amount and annual distribution of precipitation.

In King George Island, the Antarctic environment, the total soil content was considerably higher than the total ^{137}Cs fallout reported by Bartarya et al. (1993), which varied from 10 to

Table 2: Soil properties, ^{137}Cs content and vertical distribution in Chilean soils

Site code	Soil characteristics					^{137}Cs total soil content, relaxation and total penetration depth		
	Clay (%)	Carbon content (%)	Coarse pores >10 μm	pH (CaCl ₂)	CEC+ meq/100g	Total soil content++ (Bq m ⁻²)	Relaxation depth (cm)	Penetration depth (cm)
1A	25.9	4.5	22.7	4.8	22.5	420	0.9**	5
1B	34.2	1.1	37.7	4.5	33.8	520	1.0**	5
2A	5.5	6.8	44.1	4.4	17.5	690	1.1	8
2B	13.1	12.5	61.6	4.1	42.5	740	2.6**	14
3A	43.6	13.5	14.0	5.0	39.6	690	3.9**	24
3B	28.3	14.0	20.6	5.2	58.4	770	6.8**	30
4A	78.6	4.1	17.8	4.6	17.5	840	2.2**	14
4B	49.8	9.0	23.1	4.2	32.5	880	3.9**	28

+ CEC = cation exchange capacity

++ referred to January 1, 1995

** significant at 0.01 level

174 Bq m⁻² at different Antarctic stations. The total ¹³⁷Cs soil content can vary considerably from site to site in this ecosystem due to snowdrifts caused by blizzards and to the water and soil particles runoff during the melting periods.

In the Westpatagonic environment, precipitation had a high solid proportion with a more even annual distribution than the Antarctic area. This contributed to a more efficient scavenging of ¹³⁷Cs from the atmosphere and therefore to a higher soil content.

At the sites studied in the Lake Region, with a mean annual precipitation higher than in the Westpatagonic area, total ¹³⁷Cs soil contents were similar to those measured in the latter. The liquid precipitation and its seasonability in the sampled locations of the Lake Region, contribute to making the washout of atmospheric contaminants less efficient. The sites analyzed in this zone have a lower mean annual rainfall than other areas of the same region, where rainfall reaches 4000 mm y⁻¹. Consequently, total ¹³⁷Cs soil contents found by Schuller¹ et al. (1993) in this zone reached much higher values than those obtained in this study.

The amount and homogeneous annual distribution of rainfall can explain the greater total ¹³⁷Cs soil content in the tropical Easter Island environment, compared with the other study sites previously described. In addition, this island is the nearest Chilean territory to the sites of contaminant injection into the atmosphere in the Southern Hemisphere.

The vegetation cover of grass and forbs of the Patagonia, Lake Region and Easter Island ecosystems retained less than 1% of the total ¹³⁷Cs content. However, in the Antarctic environment 20% of the total ¹³⁷Cs content was found in the lichen cover of site 1A, and 24% in moss at site 1B. The long vegetative cycle and the morphology of moss and lichens, together with the long freezing periods which hinder water movement, favour the accumulation of contaminants in these species.

3.2 ¹³⁷Cs DISTRIBUTION IN THE SOIL PROFILES

As shown in Fig. 1, in the natural Antarctic environment the ¹³⁷Cs soil content shows an exponential decrease with depth, according to the equation

$$A(x) = A_0 \exp(-b^{-1}x), \text{ being } A(b) = e^{-1}A_0.$$

The ¹³⁷Cs soil content follows no pattern in the upper 1 to 6 cm depth of the other six soil profiles of semi-natural environments, presumably because of the biological activity and animal

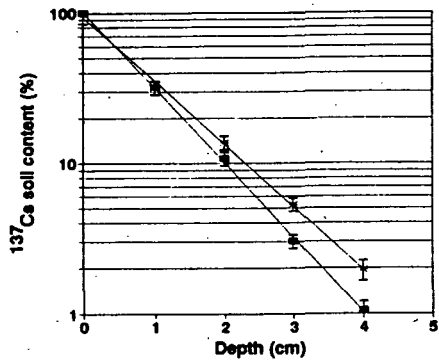


Figure 1: ¹³⁷Cs soil content as a function of depth in sites 1A(=) and 1B(=), King George Island, South Shetland Islands, Antarctic Territory.

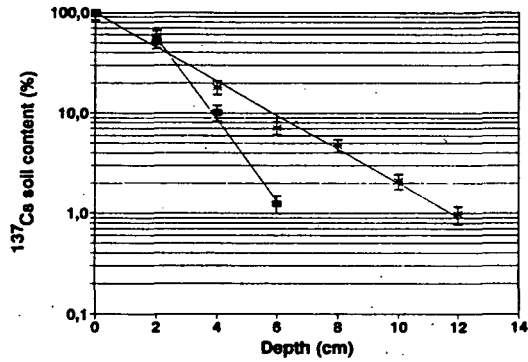


Figure 2: ¹³⁷Cs soil content as a function of depth in sites 2A(=) and 2B(=) in Torres del Paine, Westpatagonia.

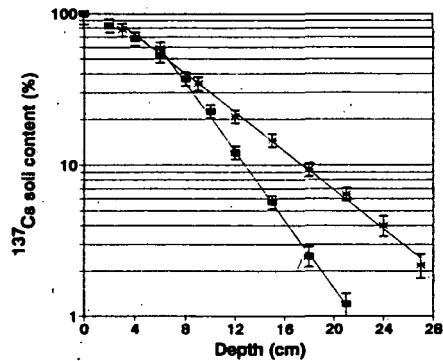


Figure 3: ¹³⁷Cs soil content as a function of depth in sites 3A(=) and 3B(=), Lake Region.

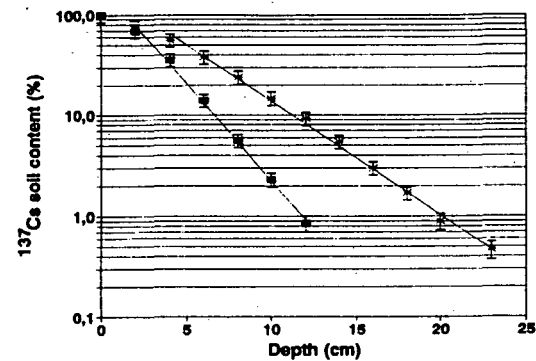


Figure 4: ¹³⁷Cs soil content as a function of depth in sites 4A(=) and 4B(=), Easter Island.

footprints, which influence the migration processes in this layer. Below this depth, ^{137}Cs soil content also decreased exponentially (Figs. 2 to 4).

The penetration depth for each location, i.e. the total depth where ^{137}Cs concentration was above detection limit, is shown in Table 2. The relaxation depth, b , also shown in Table 2, calculated by linear regression analysis for each profile in the depth interval of exponential decrease, was obtained through the slope in the regression line:

$$\ln A(x) = \ln A_0 - b^{-1}x$$

Relaxation depth represents the depth where $A(x)$ will have reduced to e^{-1} of the total soil content within the depth interval showing exponential decrease. It gives an estimation of the long-term ^{137}Cs vertical migration process in the soil profiles. The measured relaxation depths depend on soil and climatic factors.

In the Antarctic ecosystem, the measured relaxation depths indicate a high ^{137}Cs retention in the surface layer and small differences in the vertical translocation of this radionuclide, because the annual freezing periods of the soils are very long. The soil of site 1B with a greater relaxation depth than 1A also has a greater coarse pore volume, which in turn favours leaching. This could indicate a slight influence of this edaphic characteristic on the downward migration of the radionuclide.

In the Westpatagonic area, the difference between the relaxation depth of the profiles in sites 2A and 2B increased. The soil properties could have a higher influence on the radionuclide's vertical translocation since the soil freezing periods are shorter than in the Antarctic environment. Both soils have a coarse texture and consequently a high coarse porosity. In site 2B the coarse porosity has exaggerated high values, which explains the greater relaxation depth in this soil profile.

The selected soils of the Lake Region and Easter Island never freeze. Therefore, the edaphic properties can play an important role in ^{137}Cs downward migration. The dominant clay fraction of the Lake Region soils are allophane and Fe-oxides (Besoain 1985) and in Easter Island hallosyte soils admixed with kaolinite (Besoain et al. 1993).

The greater coarse porosity and lower clay content of soils from sites 3B and 4B in relation to 3A and 4A favoured vertical migration of ^{137}Cs in the B-site soils. In spite of the high clay content and adsorption capacity of all these soils the high amount of annual rainfall contributes to the migration of the radionuclide in the soil profile.

In brief, the magnitude of ^{137}Cs relaxation depth appears to be directly related with the radionuclide's leaching possibility. In the studied soils ^{137}Cs vertical migration depends on climatic characteristics, such as length of freezing periods, as well as annual precipitation rates. Moreover, it appears to be dependent on edaphic factors that favour percolation such as high coarse pore volume and continuity of this system, and to a lesser degree on the clay content and adsorption capacity of these soils.

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ASSESSMENT OF RADIOCAESIUM BEHAVIOUR IN AN AUSTRIAN FOREST ECOSYSTEM

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Abstract

A monitoring program has been carried out since 1988 in Weinsberger Wald, an Austrian forest stand severely affected by the Chernobyl plume. Analyses of forest soils, understorey vegetation and mushrooms showed that ^{137}Cs values decreased significantly between 1988 and 1993. In ferns, mosses and bilberry sprouts the concentrations did decrease to about 35% of the concentrations measured in 1988, however, seven years after the deposition event, the ^{137}Cs contamination of edible forest products and game browse is still considerable. Derived ecological half-times are the longest in mushrooms (5.8 years), followed by values between 3.1 - 3.7 years in mosses, ferns and dwarf shrubs. Ecological half-times of 2.1 years were determined in grasses and the shortest values (1.9 years) were found in herbs (*Oxalis acetosella*). Annual measurements of ^{137}Cs content in soil samples from different layers showed that the ^{137}Cs values decreased in the litter layer, whereas they increased in mineral strata between 1988 and 1993.

1. Introduction

In the course of routine food assessment by Austrian authorities it turned out that the decline of ^{137}Cs contamination of food products from forest ecosystems (game meat, mushrooms) was unexpectedly slow. From 1987 to 1988 ^{137}Cs values even increased in some forested

areas of Austria (Tataruch et al. 1993). As a consequence of this observation an investigation program in two Austrian spruce forests severely affected by the Chernobyl plume was started in 1988 by the Federal Environment Agency. Samples of soils, forest vegetation (mosses, ferns, herbs, dwarf shrubs, trees, mushrooms) and game animals were collected and analysed. Besides contamination assessment this investigation focused on radioecological questions such as the behaviour of ^{137}Cs within the foodchain and its transfer between compartments. The use of vegetation samples for bioindication was tested by analysis of *Vaccinium myrtillus* (bilberry). Since 1988 about 800 samples of soils and vegetation as well as 450 samples of roe deer muscle were analysed by gamma-spectrometry.

2. Material and Methods

2.1. Investigation area

Weinsberger Wald is situated in Lower Austria (48°23'N, 15°03'E; 900 m alt.) precipitation amounts to 910 mm a⁻¹. This large mixed forest due to climatic conditions is naturally dominated by spruce with uniform understorey vegetation consisting of mosses, ferns and forest grasses (*Avenella flexuosa*, *Calamagrostis* sp.). To provide sufficient comparability of data, the investigation site was restricted to an area of 200 ha. Soils are classified as Dystric Cambisols, partly podsollic, the litter layer is degraded to mor humus types. The soil pH - values are rather low (3.2 in litter layer increasing to 3.6 in 10-20 cm of mineral soil). Soil texture fractions in mineral soil were determined as 44 : 29 : 27 (%) sand : silt : clay. Humus contents decrease from 70% in the litter layer to 20-30 % in the Ah-horizon and 10% in the A/B horizon. The soil inventory of ^{137}Cs derived from depositions of Chernobyl was determined as 62 kBq m⁻² (median; 40 - 84 kBq m⁻²).

2.2. Measurements

Measurements were carried out with dried material (soils air dried, plant material oven dry (100°C)). For gamma-spectrometric measurements a multi-channel analyser system with several Ge-(Li) and HPGe - detectors was used (relative counting efficiency 35% and 30%

respectively). Measurement times were extended until the statistical counting uncertainty for radiocaesium was below 10%.

3. Results and Discussion

Results of gamma-spectrometric analyses of ^{137}Cs soil content in different layers of forest soils are shown in Fig. 1. In the litter layer ^{137}Cs values show a decreasing trend over time, while in the underlying layer (0-5 cm of mineral soil) a remarkable increase was observed. Under the transition zone between pure organic and mineral soil the values remain rather low even 9 years after the deposition event. This distribution of ^{137}Cs contents is a result of radiocaesium soil migration. The observed behaviour agrees well with model calculations derived with a compartment model (Strebl et al. 1995).

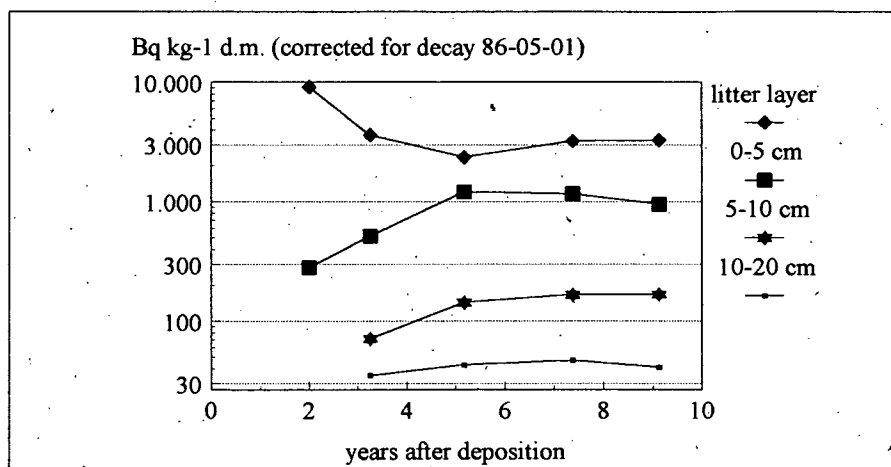


Fig. 1: Time dependence of ^{137}Cs content in soil samples from different layers of a forest soil.

Observed ^{137}Cs concentrations in selected species of forest vegetation are shown in Fig. 2. Between 1988 and 1993 these values decreased significantly in all species. Taking the values of 1988 as a basis (100 %), values of 1993 represent only 33% in ferns, 36 - 39 % in mosses,

36% in bilberry, 19% in grasses, 16% in herbs and 55% in mushrooms. The corresponding results for the organic litter layer show values decreasing to 35% of the 1988 concentrations. Ecological half-times were derived from this comparison of data (see Tab. 1). Rosen et al. (1994) found longer half-lives in Swedish meadow ecosystems (e.g. 4.2 years for *Vaccinium myrtillus*).

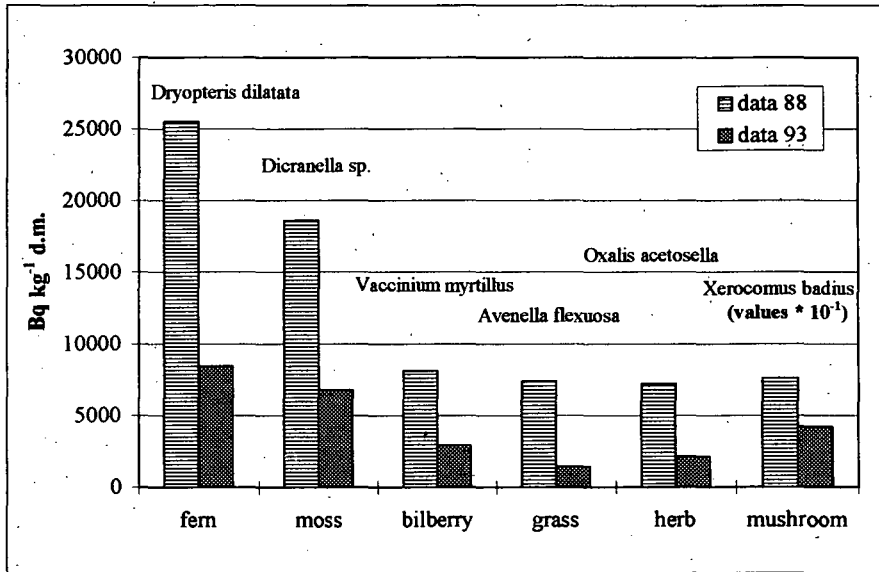


Fig. 2: ¹³⁷Cs content in forest vegetation (Bq kg⁻¹ in dry matter) - comparison of values 1988 - 1993 (decay corrected to 1986 - 05 - 01))

Tab. 1: Ecological half-lives (τ_{ecol}) of ¹³⁷Cs in forest vegetation and the litter layer of Weinsberger forest. (Data basis: decay corrected values from 1988 and 1993)

Species	τ_{ecol}	Species	τ_{ecol}
<i>Dryopteris dilatata</i>	3.1	<i>Avenella flexuosa</i>	2.1
<i>Dicranella sp.</i>	3.4	<i>Oxalis acetosella</i>	1.9
<i>Polytrichum commune</i>	3.7	<i>Xerocomus badius</i>	5.8
<i>Vaccinium myrtillus</i>	3.5	forest litter layer	3.3

Vaccinium myrtillus was investigated to test its suitability as a bioindicator (Kienzl et al. 1992). Time dependent changes of ^{137}Cs concentrations in youngest sprouts are shown in Figure 3.

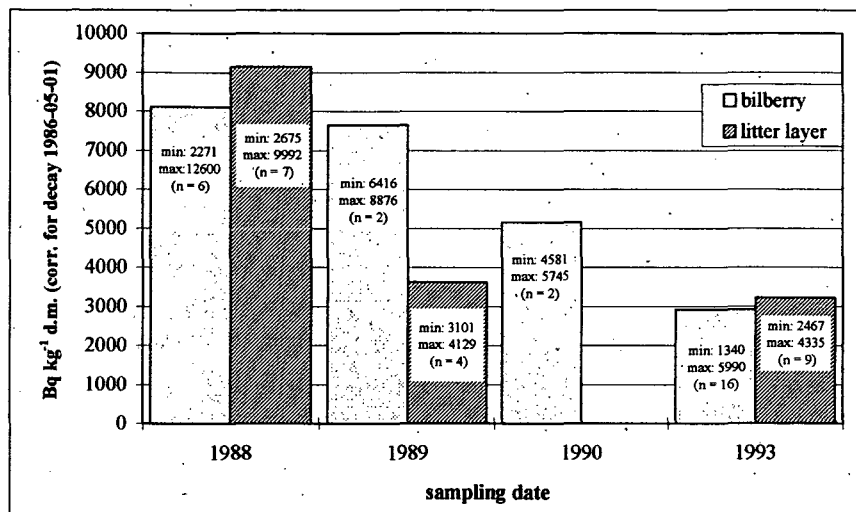


Fig. 3: ^{137}Cs content in samples of bilberry (*Vaccinium myrtillus*) and forest litter from several years after the Chernobyl accident.

Minimum and maximum values (min; max) and number of observations mentioned in the bars.

Fig. 3 indicates that, after 1988, the decrease of radiocaesium content in the litter layer is faster than that in samples of *Vaccinium myrtillus*. The fine root system of forest understorey species is situated in the superficial soil layers (Kutschera and Lichtenberger 1992), highest root densities are found in the first 10 cm of soil. As seen in Fig. 1, the radiocaesium content of the 0-5 cm layer increases because of vertical migration of cations. As this layer also contributes to the nutrient pool of bilberry plants, the concentrations in dwarf shrubs stay high when compared to the litter layer.

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RADIOECOLOGY: DEVELOPMENT AND OBJECTIVES

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Abstract

The development of "radioecology" in the last decade with a short glance to earlier periods is shown. Present activities are discussed. Both assessment and modelling approaches are going to become more developed. Attention is paid to sensitivity analysis and parameter variability. The requirements to measuring techniques are discussed. At present, problems can be solved only by cooperation of measurements and modelling activities. This collaboration is important for not to leave the field to groups, directed by politicians seeking for short term gain, in order to give retrospective justification in a specious manner of decisions not be based on objective analysis.

1. Introduction

Radioecology has developed some independence from radiation protection, but is still to be considered as one of its branches. Radioecology is therefore rather an applied science than an independent pure science. This leads to the requirement that the purpose has to be defined well in advance before starting the investigation. This implies different requirements for parameter quality in different applications

The term "radioecology" is based upon the basic terms: *radio* derived from Marie Curie's initial definition, and *ecology* (Greek: *oikos*, meaning: house, immediate environment of man; coined by Heckel (1834-1919), medical practitioner in Jena. Term later established by Worming), later moved to a political term)

1955 as *Radiation ecology* coined (Ge 55), later changed to *Radioecology* / Co 61/

The present scientific meaning of radioecology: **relation between mankind and environment**, possibly to be interpreted as:

- * understanding the behaviour of radioactive material in the environment (Ts 84)
- * science of the effects of ionizing radiation to living and non living environment (Bo 93)

These definitions are very general. Therefore, further specification is required, which is only possible by defining the purpose of a given program. Purposes of programs might be classified into:

- i) **MONITORING PROGRAMMES:** demonstration of the impact of a given source, demonstration of compliance with limits (assessment of activity in a given compartment (e.g. activity concentration in air, independent from the origin), recently also dose estimates (e.g. Lo 91ff/), where simple models have to be used. Programs can be source related, compartment related, site related.
- ii) **RADIOECOLOGICAL PROGRAMMES:** establishment of basic criteria for the transfer of radioactive materials in environmental compartments, investigations on transfer parameters, development of models for radionuclide transfer and for both retrospective dose estimation and prediction of doses for different conditions (e.g. acute and chronic releases) (e.g. /IA 93,94/)
- iii) **ECOLOGICAL PROGRAMMES:** determination of risk factors from empirically assessed exposure conditions (e.g. health effects from radon: St 93)

The results of programs attributable to the first two groups lead usually, even taking into account the often very large inherent variability of parameters, to reliable and consistent conclusions, provided that they remain on the scientific basis rather than they are biased by political issues. However, since programmes were established for different purposes, some of the results of different programs may not necessarily be consistent or exchangeable. When data are used for another purpose as they were established, misleading conclusions may result.

Ecological programmes lead at present only under particular conditions to proved results. This is mainly because too many parameters are needed for evaluation, but only a few are sufficiently well known. Therefore the conclusions may depend rather on the assumption of parameters than on real parameters. (see e.g. St 93 for the relation between Radon and lung cancer).

2. Development of Radioecology

It might be convenient to show the development in time by a more or less arbitrary separation into five phases and a precursor. The phases are obviously in close relation to the general development in the fields of physics (mainly development of radiation detectors), radiation biology and epidemiology (risk factors), radiation standards (organs of interest). A few examples and keywords to these phases are outlined below.

Precursor phase:

- * epidemiological studies before discovery of radioactivity /Hä 1879/; /Sc 93 reports even on much earlier reports on miners diseases as by Paracelsus (1567)
- * measuring approaches: determination of Ra-A in air and water (Ba 07, Sc 13), cosmic rays (He 29) and others.

phase 1 (until 1960)

- *assessment of activity concentration in environmental materials
- *source: nuclear weapons test
- *used technique: gross beta measurements
- *results: activity concentration gross beta
- *presentation of results: tables, no interpretation, reporting of random figures
- *conceptual development: dose commitment (Lindell, UNSCEAR 1958)
- *selective measurements: mainly based on chemical separation (Sr-90)

These programs have the longest tradition and have to be seen today as documentation

phase 2 : about 1960 - 1970

- *development of research programs , mainly in relation to use of nuclear energy and use of radionuclides
- *recognition of natural background as environmental parameters
- *development of selective measurement techniques
- *only a few radionuclides considered as relevant

phase 3: pre Chernobyl (about 1970- 1986)

- *advent of environmental awareness
- *improvement of measuring techniques (spectroscopy, low level measurements)
- *dose assessment methodology
- *development of prediction models
- *improvement of conceptual background
- *increase of public interest by political influences of "quasicritical" groups with a biased background in contradiction to scientific basic
- *assessment of natural background extended to radon

phase 4 :past Chernobyl and immediately after 1986

- *avalancing increase of the number of "experts" in radioecology, number in the meantime decayed to pre-Chernobyl level

- * interaction between political and scientific issues
- * dose predictions
- * development of scientific basis for decision makers overruled by political influences

phase 5: present development

Among other activities, the following large programs are in progress:

measuring techniques

- i) optimization of measuring techniques towards Low Level measurements
- ii) definition of Lower Limit of Detection (detection limit, decision limit) / ÖN 95, DI 89/

empirical dose assessments

- a) retrospective dose assessments of previous emissions
 - Hanford, Wismut, Maralinga&Emu, Kyshtym, Nevada test site (IA 94a)
 - Semipalatinsk (St 95)
- b) doses assessment after Chernobyl
- c) Radon programmes

model development

- α) improvement of models
- β) model validation
- γ) predictive models
- δ) default sets of environmental parameters
- e) sensitivity analysis

3. Present activities- some examples

- 3.1 Estimation of the doses of Wismut workers/ Ei 93/: annual doses of uranium miners were in the early years (to 1955) in the range of 30-300 WLM, later decreasing. (10 WLM=50 mSv)
- 3.2 Dose reconstruction projects from nuclear fuel cycle : Hanford / Na 93/: routine releases in the Columbia river 1955 bis 1959 were in the order of 10^{15} Bq (gross beta activity). Sellafield /Jo 95/: routine releases of low level liquid wastes in the Irish sea were up to 10^{15} Bq (e.g. Ru-106 in 1975).
- 3.3 Dose reconstruction projects at nuclear weapons test sites e.g.: i)Maralinga and Emu (Australia) /Wi 94, Ha 94/: area was contaminated with Pu-isotopes and still occupied by aborigines. The dose by inhalation is the most important pathway. ii)Semipalatinsk /St95/:

3.4 Continuation of model development and parameter assessment: IAEA Handbook on Parameter values for prediction of Radionuclide transfer in temperate environments (IA 94b)

3.5 Model validation

IAEA: Validation of Model Predictions (VAMP), The International Chernobyl Project

BIOMOVs: BIOSperic MODEL Validation Study: an international study to test models designed to predict the environmental transfer and bioaccumulation of radionuclides and trace substances

IUR: International Union of Radioecology : scientific basis, collaboration with CEC

CEC: support of projects to provide a scientific basis for decision making in accidental cases.

An example of a Model validation exercise

The model is described in / Wi 95/ and /Ts 92/

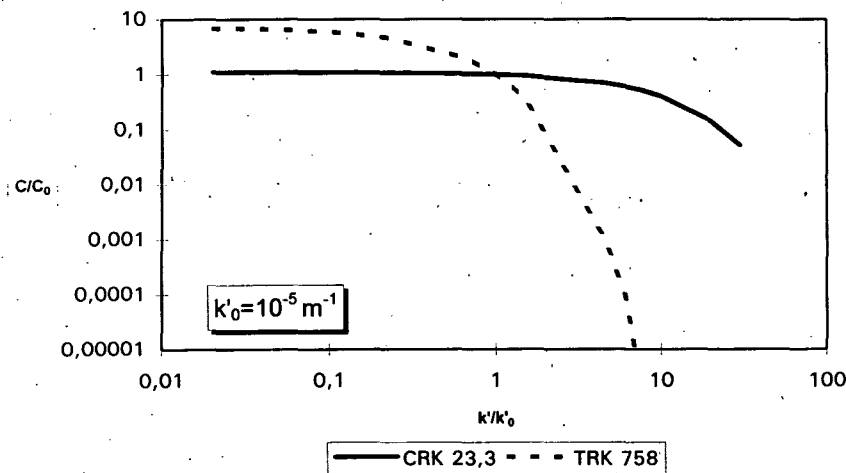


Fig. 1 Sensitivity analysis, Clinch river scenario: Influence of the variation of retention coefficient k' for sediments: predicted activity concentration for two different sites in dependence from the variability of the factor k' /Wi 95/. It can be seen that the predicted activity concentration is at site CRK 23,3 almost independent from k' in given range, but k' is a very sensitive parameter at site TRK 758. This has consequences for the required accuracy of parameters

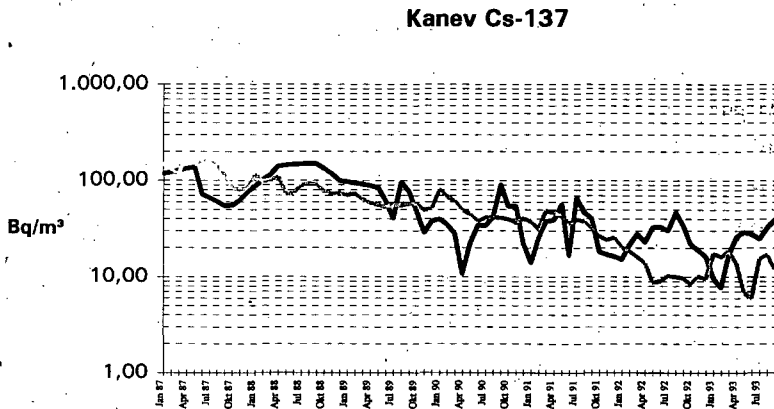


Fig.2: Comparison of measured activity concentration (black) and calculated (grey), Dnjepr river scenario: a simple model gives a reasonable prediction of the activity concentration for Cs-137

4. Conclusions

Substantial effort is put at present into both model development and measuring programs. Previous release sites are investigated not only for retrospective dose assessment, but also for assessment of generalized parameters. Uncertainty assessment, and sensitivity analysis are required to improve data quality and to optimize the assessment procedure. Neither the progressive modelist nor the pragmatic measuring engineer can solve the existing problems alone. Both groups need the interaction with the other to prevent irrationalism. Radioecology developed to a modern environmental science with very sensitive public perception. It is a professional challenge to keep radioecology on a well proved scientific basis for not to leave the field to groups directed by politicians seeking for short term gain in order to give retrospective justification in a specious manner of decisions not be based on objective analysis.

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RADIOCESIUM IN AN AGRICULTURAL ECOSYSTEM RESULTS OF A FIELD STUDY IN THE WALDVIERTEL, AUSTRIA

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Abstract

The transfer of radionuclides in an agricultural ecosystem was investigated at two farms in the Waldviertel, the north-western part of Lower Austria in a field study. Typical for this field study were fluctuations in the daily intake of ¹³⁷Cs which meant that no state of equilibrium was reached.

An average transfer factor from forage into milk of 0.0045 d/l was found. The results correspond with the values from IAEA (1994). Changes in the forage activity affected the milk activity with a certain delay, shown by carrying out a cross correlation, to correspond to one week.

The concentration factors of forage crops to ground contamination depend on the degree of fertilization. The highest concentration factors are found at undisturbed and infrequently fertilized meadows.

1 Introduction

In 1989 a field study at two farms in the Waldviertel (north-western part of Lower Austria) was carried out to investigate the transfer of radiocesium in an agricultural ecosystem. The study was commissioned by the Austrian Federal Environment Agency. At one of the farms the series of measurements was continued in 1990.

The aim of this study was to determine transfer rates under the usual agricultural conditions of production. There were no alterations to normal procedures regarding the feeding of the animals; the main activities were documentation and measurements.

2 Selection and site of the farms

The Waldviertel, situated in Lower Austria, is a largely wooded and upland region. In general, the forage crops are grown on many small and scattered meadows as there are no large and coherent patches of land.

The farms were chosen from a wooded area which had been contaminated relatively high by the Chernobyl fall-out, the Weinsberger Wald in the Waldviertel. Table 1 gives the results of a survey on the contamination of the soil in the Weinsberger Wald, the values indicating a generally homogenous contamination near the farms ($SD < 15\%$), but big differences in the whole region (mean = $37,9 \text{ kBq m}^{-2}$, $SD = 30\%$).

Table 1: Soil contamination in the area of the Weinsberger Wald:

site	number of samples	Cs-137 kBq m^{-2}
Brettles	2	59,0
Stein	1	54,8
Reitzendorf	1	35,4
Weinsberg Wiese	2	66,9
Vord. Waldhäuser	1	42,4
Weichselbaumhof	1	29,4
Ulrichsberg	1	22,9
JH Dorfstatt	1	23,0
Buchegg	1	35,9
Berglucken	1	45,7
Marchstein	1	24,8
Kl. Kamp	1	26,7
Kamplaiten	1	25,5
Leitentümpfl	1	16,2
Dürnberg	1	22,7
Prinzbach	1	27,6
Saggraben - farm B	7	40,1
Elendwiese - meadows of A&B	13	48,4
Hintere Waldhäuser - farm A	6	48,8
Bärnkopf	1	51,2
Spitzwiese	7	48,3

3 Cooperation with the farmers

During the investigation period one of the chosen farms (farm A) was converting to biological farming and the farmer was very cooperative and interested in the measurements carried out at his farm. The other farm (farm B) was organized along conventional farming methods. Here, it was difficult to give the family an understanding of the project and they needed more help for sampling and completing the protocols. The farmers were compensated for their cooperation in the project.

4 Sampling and measurement

4.1 Sampling

After brief instruction the milk was sampled by the farmers themselves. They were given containers for sampling and protocols to complete. The milk was sampled in the form of weekly mixed samples of 1 litre from all milking cows at each farm. The protocol contained information on the average amount of milk given by each cow per week, the amount of fresh grass in relation to hay, silage and additional feed, on the meadow and from which cut the forage comes from. Grass samples from each meadow and each grass cut, samples of hay, silage and additional feed (coarsely ground rye) as well as samples of manure in solid and liquid form were also taken by the farmers.

Soil sampling was carried out by the project assistants; for methods see IAEA (1989). At several meadows botanical analysis was carried out by a member of the Federal Environment Agency.

When continuing the project in 1990, the forage samples were no longer separated according to the meadows they came from but were sampled by the farmer as weekly mixed samples.

4.2 Preparation of samples and measurements

Grass, hay and soil samples were dried at 105°C and homogenized. Gamma spectrometry was carried out with a HP germanium detector (efficiency: 19%) provided by the Canberra company.

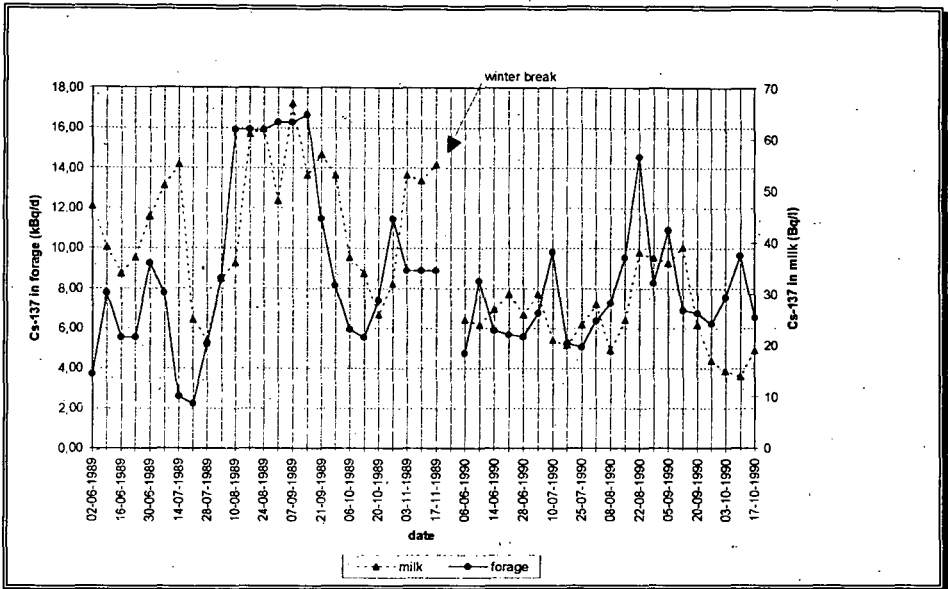
5 Results

5.1 Transfer of radiocesium from forage to milk

The radiocesium contamination of forage varies greatly when only scattered meadows and no large fields are available for the growing of forage crops. This study determined the effect of these variations on the contamination of milk with radiocaesium. Figure 1 shows the results of the series of measurements at the smaller farm (farm A).

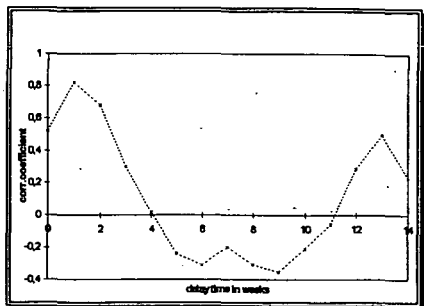
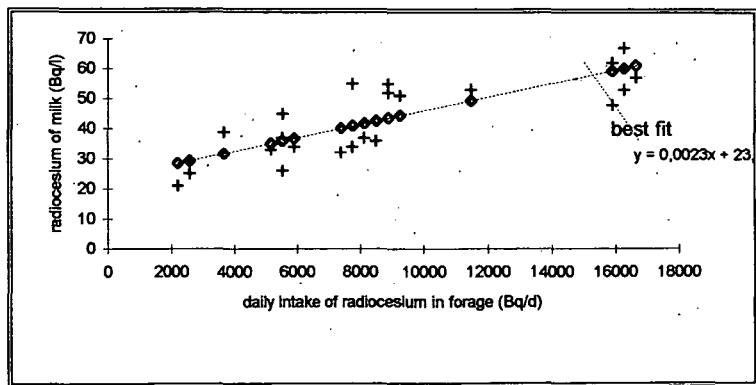
Out of all at both farms observed transfer coefficients the mean value was determined to be $0,0045 \text{ dl}^{-1}$. These result corresponds with the data given in IAEA (1994), which are as follows: expected value: 0,0079; confidence interval: 0,001-0,027. Our data are lower than those given in IAEA (1994) because they represent no state of equilibrium.

Figure 1: Variability over time of ^{137}Cs in forage and milk at farm A



A more detailed analysis of the relationship between forage activity and milk contamination shows, however, that a linear transfer model can only be regarded as a coarse approximation.

The measurements shown in figure 1 reveal that changes in the forage activity show an effect only after a certain delay. This hypothesis was verified via cross correlation and it was found that with a given delay time of one week the correlation between both series of measurements (forage and milk) reaches a maximum. A second maximum is attained after a time of several weeks (the results of farm B show a similar trend). A best fit adapted to the measurement results for both farms shows a significant constant part. These effects can be explained by a two compartment model with a short and a long biological half life, like the model described by [KIRCHNER 1989].

Figure 2: Cross correlation farm A 1989**Figure 3: Best fit : Activity concentration in forage and milk at farm A in 1989 - delay time: 1 week**

5.2. Effect of tillage on the contamination of forage crops

This study aimed at determining the transfer or translocation of ^{137}Cs into forage crops. For this reason we investigated the extent to which tillage and fertilization influence contamination of the plants. For comparison reasons meadows which had been planted after 1986 were chosen, too.

Among other things a comparison between adjacent meadows was made. The chosen meadows differed clearly in terms of vegetation type and intensity of fertilization. No significant difference in the depth distribution of ^{137}Cs from Chernobyl could be observed between the unfertilized and the fertilized meadow. However, a clear difference could be discerned between fertilized and unfertilized meadows regarding the activity of the grass.

Given the same soil contamination, the activity of one kilogram of grass from meadows which had not been fertilized was considerably higher than the activity of the same amount of grass from intensively fertilized meadows. These results were confirmed during the stock-taking of plant varieties at each of the meadows in 1989.

The highest activity concentrations occurred at undisturbed and hardly fertilized meadows; the lowest concentrations were registered at meadows ploughed up after 1986. Significantly lower concentrations, however, were also found for fertilized meadows, even if only biological fertilizer was used.

A Comparison of the concentration factors of forage crops (i.e. ratio of ^{137}Cs in dry plant [Bq/kg]) to ^{137}Cs in soil [Bq/m^2]) at differently treated meadows shows a ratio of 65:20:1 for not fertilized, fertilized and meadows ploughed after 1986, respectively.

5.3 Introduction of radionuclides by means of fertilization

The amount of radionuclides introduced into the environment during fertilization was investigated. The way fertilization was carried out differed greatly at the two farms chosen for this study. At farm A the liquid manure was strongly diluted. At farm B the manure was much more contaminated, when spread on the meadows, however, no specific contamination of the soil could be measured. Nevertheless, the liquid manure of farm B may increase direct contamination (However, our sample was taken at the end of the vegetation period in October).

When comparing the degree of activity and the amount of manure spread it can be concluded that manure brought out onto the meadows does not cause major contamination. Farmer A informed us that the maximum amount of liquid manure which he sprayed onto his meadows is 4 l m^{-2} . This results in a maximum ^{137}Cs concentration of 2500 Bq m^{-2} , which is below 5% of the existing contamination.

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^{137}Cs , ^{90}Sr , K^+ , and Ca^{++} in lichens, mosses, and vascular plants of a mountain area in Styria, Austria

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Abstract

The ^{137}Cs and ^{90}Sr concentrations in plants from a forest (altitude 1000 m) and a pasture (1700 m) in the Styrian Koralpe were found to be still high today, ten years after the Chernobyl accident. Following the reactor accident, the ^{137}Cs activity of lichens rose to over 50 KBq kg^{-1} d.w. from an initial value of approx. 400 Bq . At the Weinebene (1770 m) the biological half-life of ^{137}Cs was 3.1 years for *Pseudevernia furfuracea* and 2.5 years for the terricolous lichen *Cetraria islandica*. The biological half-life of ^{90}Sr in *Pseudevernia* was shorter than that of ^{137}Cs , and was determined to be between 1.2 and 1.6 years. A positive correlation was found to exist between ^{90}Sr and Ca^{++} concentrations among the plants tested, whereas ^{137}Cs showed no correlation to K^+ .

1. Introduction

Aboveground nuclear tests conducted between 1954 and 1965 gave rise to large amounts of ^{137}Cs and ^{90}Sr (Cambray *et al.* 1985). Both radioisotopes have half-lives (T_{phys}) of approx. 30 years and persist especially in alpine and circumpolar environments, because those environments are characterized by a slow biological turnover rate. It has long been known that lichens are able to adsorb more airborne material than flowering plants, and therefore, they are often used as indicators for air contamination. The aim of the present study was to determine the levels of radiocesium and radiostrontium in a seminatural area in Styria, a southeastern province in Austria, and to study the behavior of the long-lived ^{137}Cs and ^{90}Sr radionuclides

over a number of years in lichens, mosses and vascular plants. Special attention was given to the measurement of the biological half-life of ^{137}Cs and ^{90}Sr in lichens, since the time period during which 50% of the radioactivity is lost in lichens has been shown to vary between less than 1 year (Akçay and Kesercioglu, 1990) and 17 years (Lidén and Gustafsson 1967). Since ^{137}Cs is reported to show similarities to K^+ and is thought to replace K^+ in its functions in K^+ deficient areas, whereas Sr^{++} behaves similarly to Ca^{++} (Haunold *et al.* 1987), we analysed the plants with respect to K^+ and Ca^{++} concentrations.

2 Material and Methods

The measurements of ^{137}Cs were made with the help of a NaI crystal detector of a Raytest multichannel gamma analyzer Mod. MCA. The spectrometer was calibrated with a ^{137}Cs standard solution and with the ^{40}K activity present in KCl. The plant samples were cleansed of soil and other debris and dried at 60°C . The time of measurements in a 1-L Marinelli-beaker was between 30 min and 40 hours depending on activity. All values are expressed as Bq kg^{-1} dry weight (d.w.). The atomabsorption spectrophotometry for the analysis of K^+ and Ca^{++} was carried out using a Shimadzu AA660 at the required wave lengths. After drying at 105°C for 48h, 200 mg of each plant sample were decomposed in 2ml HNO_3 and 0.5 ml HCl , using a pressurized microwave decomposition apparatus (Anton Paar, PMD 312). Air-acetylene flame spectrophotometry was carried out on the diluted samples. Most of the plants originated from the Koralpen area with Koralpenblick at 1000m, and Weinebene between 1700 and 1770 m. The area Koralpenblick is a mixed forest composed mainly of *Picea abies*, but *Pinus sylvestris*, *Larix decidua*, *Abies alba*, and *Betula verrucosa* also occur. Weinebene shows separated groups of *Picea abies* situated in a pasture. Between April 28 and May 6, 1986 the precipitation in the Koralpe (980 m) was 65 mm, from May 6 to May 16, 1986 32 mm, and, as a result of the high amount of precipitation, the radioactivity in the Koralpenregion was also higher than e.g. in Graz. The lichen samples collected before the Chernobyl accident (between 1976 and 1985) were taken from the Hafellner Herbarium (Botanical Institute of Graz). ^{90}Sr activity was determined with the Scintillation counter „Quantulus“, (Fa. Wallace, Pharmacia Instruments), according to Schönhofer *et al.* (1982).

3. Results and Discussion

3.1. ^{137}Cs and ^{90}Sr in soil

In June 1992, a soil sample from the Koralpenregion (Koralpenblick 1000 m) showed a 36 fold higher contamination of ^{137}Cs (55 KBq m^{-2}) over ^{90}Sr (1.5 KBq m^{-2}). 80 % of the ^{137}Cs level (in Bq dm^{-3}) and 70 % of the ^{90}Sr could be detected in the first 10 cm of the topsoil. *Pseudevernia furfuracea* measured on 14. 08 1986 had a ^{137}Cs to ^{90}Sr ratio of 149, whereas the ratio for the soil was 36. This means that a large part of the ^{90}Sr present in the soil did not come from the Chernobyl reactor accident but is rather the result of atmospheric nuclear weapons testing

3.2.1. ^{137}Cs - contamination of lichens before and after the reactor accident in Chernobyl

Lichens are able to adsorb more airborne material than flowering plants. The pre-Chernobyl values of ^{137}Cs for flowering plants were $< 1.8 \text{ Bq kg}^{-1} \text{ d.w.}$, whereas *Hypogymnia physodes* from the Koralpe (Glashütten 1270 m, collected on 31.07 1983) showed a ^{137}Cs contamination of $229 \text{ Bq kg}^{-1} \text{ d.w.}$, and *Hypogymnia physodes* (Weinebene, 1650 m, 31.07.1983) had values of 351 Bq kg^{-1} . The ^{137}Cs radioactivity exceeded the natural radioactivity, caused mainly by ^{40}K and ^9Be , by two to three fold. Following the reactor accident in Chernobyl, the ^{137}Cs activity of *Pseudevernia furfuracea* from the Weinebene rose to over 50 KBq kg^{-1} . The man-made contamination of this lichen exceeded the natural one by a factor of 750.

3.2.2. ^{137}Cs gradients on spruce trees for *Pseudevernia furfuracea*

Data from *Pseudevernia* collected in 1991 from dead spruce trunks without branches, showed that the lichens higher up on the trunk took up more radionuclides from rain water than those in the lower sections on the tree (Fig. 1). These ^{137}Cs gradients were already well developed on 2 m high trees. Measurements of lichens from dead spruce trees at different heights above ground conducted in 1993 showed that those gradients become less steep, as the radioactivity from the upper tree lichens leached down and was taken up by the lower lichens in the absence of further supply coming from the air. In some cases the highest radioactivity was not at the top end of the tree, but the somewhat lower inserting lichens had the highest peak of ^{137}Cs .

The same phenomenon was also observed for the litter layer of the soil, which shows a decreasing trend over time, as the ^{137}Cs is slowly washed in slightly deeper layers of the soil.

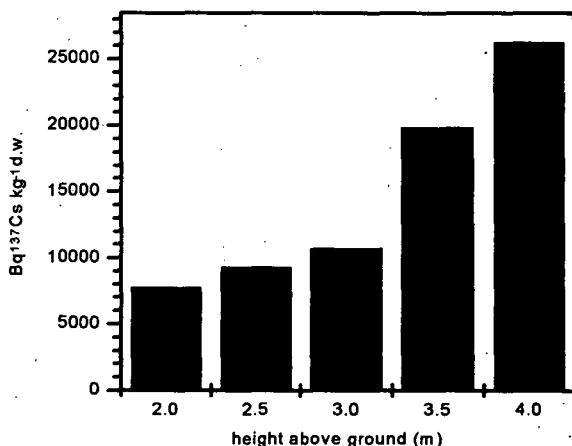


Fig. 1. ^{137}Cs -activity in *Pseudevernia furfuracea* from the Weinebene (1750 m), collected from a dead spruce tree at different height above ground on July 05, 1991.

3.2.3. Biological half-life of ^{137}Cs and ^{90}Sr in lichens

This tree height gradients must be taken into account in the measurement of the biological half-life of the long-lived radionuclides in lichens. Only lichens from branches of the same height of a tree are to be compared.

From our measurements in the Koralpenregion (Weinebene, 1700-1760 m) the biological half-life of ^{137}Cs in *Pseudevernia furfuracea* was 3.1 and in the terricolous lichen *Cetraria islandica* 2.5 years. To determine the T_{bio} of radiocesium in *Cetraria islandica* all samples were taken from an area of 100 m² from the Weinebene (1760 m), to get a similar starting contamination. Measurements carried out on four trees from the Stubalpe on the way from the Gaberl to the Alten Almhaus at approx. 1650 m showed a biological half-life of ^{137}Cs from 2.5 years (Fig. 2). The biological half-life of ^{90}Sr was shorter than that of ^{137}Cs in *Pseudevernia* and ranged between 1.2 to 1.6 years at the Weinebene.

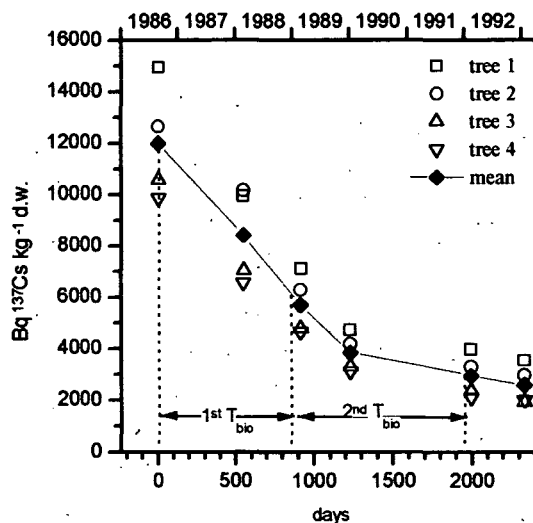


Fig. 2. Decrease of ^{137}Cs -activity in *Pseudevernia furfuracea* collected from lower branches of four spruce trees, Stubalpe 1650 m.

3.3. ^{137}Cs , ^{90}Sr , K^+ , Ca^{++} in plants, and correlation between ^{90}Sr and Ca^{++}

Table 1 shows the values of ^{137}Cs , ^{90}Sr , K^+ and Ca^{++} and the ratio ^{137}Cs to ^{90}Sr in selected plants. As expected lichens showed a high level of radionuclides when compared to flowering plants. The dense canopy of mosses also acts as an efficient filter system in trapping radionuclides and retaining different pollutants for long periods of time (Thomas 1986, Heinrich 1987). But the vascular plants of the seminatural environment from the forest at Koralpenblick and from the pasture of the Weinbene do show unexpected high ^{137}Cs and ^{90}Sr values (Table 1), compared e.g. to crops from Deutschlandsberg from the foot of the Koralpe (*Solanum tuberosum*, tuber, 10.08.1990: 4.5 Bq kg^{-1} ^{137}Cs and 1.1 Bq kg^{-1} ^{90}Sr). The evergreen Ericaceae, e.g. *Loiseleuria procumbens*, *Calluna vulgaris*, and *Vaccinium vitis-idaea* show ^{137}Cs values rather similar to those of the lichens. Most of the dicotyledonous plants and the grasses are eaten by cows. It is not the case for *Veratrum album*. It may be one of the reasons why this plant shows a high radioactivity for both radionuclides. The roots of most of the plants tested do not reach deeply in the soil and therefore the uptake of radionuclides comes

from the upper 10 cm of the topsoil, which contain the mainpart of the radionuclides. The ^{137}Cs to ^{90}Sr ratios of lichens and mosses are high compared to those of vascular plants (Table 1).

Table 1. ^{137}Cs and ^{90}Sr in selected plants, ratio of ^{137}Cs to ^{90}Sr , yield of K^+ and Ca^{2+}

species	location	collection-date	K^+ (mg g^{-1} d.w.)	Ca^{2+} (mg g^{-1} d.w.)	^{90}Sr (Bq kg^{-1} d.w.)	^{137}Cs (Bq kg^{-1} d.w.)	ratio $^{137}\text{Cs} : ^{90}\text{Sr}$
Lichens							
<i>Cetraria islandica</i>	K*	06.07.1993	2.84	0.47	13	1751	134.7
<i>Cetraria islandica</i>	W*	31.05.1994	1.91	0.17	6	4537	756.2
<i>Pseudevernia furfuracea</i>	K	06.07.1993	4.11	0.38	29	7928	273.4
<i>Pseudevernia furfuracea</i>	W	08.08.1987	2.63	1.16	89	38813	434.3
Mosses							
<i>Bazzania trilobata</i>	K	25.06.1988	4.58	2.14	176	20832	118.4
<i>Hylocomium splendens</i>	K	27.10.1989	3.90	5.85	83	12198	147.0
<i>Hypnum cupressiforme</i>	K	27.10.1989	3.76	3.77	63	11155	177.1
<i>Leucobryum glaucum</i>	K	27.10.1989	2.10	2.03	62	19068	307.5
<i>Leucobryum glaucum</i>	K	06.07.1993	3.81	0.76	30	7768	258.9
<i>Sphagnum nemorum</i>	K	20.07.1989	4.28	3.36	92	23449	254.9
<i>Polytrichum norvegicum</i>	W	31.05.1994	3.06	0.05	57	2366	41.5
Vascular herbaceous plants and cow dung							
<i>Anthoxanthum odoratum</i>	W	31.05.1994	10.38	0.52	49	1149	23.4
<i>Calluna vulgaris</i>	K	27.02.1993	3.11	5.78	52	2302	44.3
<i>Calluna vulgaris</i>	W	03.06.1993	4.48	3.69	30	1893	63.1
<i>Calluna vulgaris</i>	W	31.05.1994	3.44	3.15	54	724	13.4
<i>Geum montanum</i>	W	31.05.1994	12.71	4.57	244	2357	9.7
<i>Hieracium silvaticum</i>	K	03.06.1993	26.98	4.20	80	494	6.2
<i>Loiseleuria procumbens</i>	W	31.05.1994	3.05	1.08	99	7459	75.3
<i>Luzula luzuloides</i>	K	01.06.1994	22.86	1.08	44	1117	25.4
<i>Nardus stricta</i>	W	01.06.1994	2.34	0.82	26	597	23.0
<i>Potentilla aurea</i>	W	31.05.1994	13.11	7.51	310	2050	6.6
<i>Vaccinium myrtillus</i>	K	01.06.1994	7.11	5.34	219	1877	8.6
<i>Vaccinium myrtillus</i>	W	31.05.1994	6.12	5.13	71	906	12.8
<i>Vaccinium vitis-idaea</i>	K	01.11.1989	3.87	8.15	269	4819	17.9
<i>Vaccinium vitis-idaea</i>	K	01.06.1994	4.15	3.97	210	1699	8.1
<i>Vaccinium vitis-idaea</i> , leaf	K	30.09.1993	4.09	6.09	116	1801	15.5
<i>Vaccinium vitis-idaea</i> , stem	K	30.09.1993	2.50	4.59	150	1512	10.1
<i>Veratrum album</i>	W	31.05.1994	26.42	3.65	246	3844	15.6
cow dung	W	01.06.1994	2.23	4.41	171	1377	8.1

*) K = Koralpenblick, W = Weinebene

The $^{137}\text{Cs}/^{90}\text{Sr}$ ratio in lichens shortly after the reactor accident was comparable to that found in the air. Often this ratio rose with time in lichens, because of the shorter biological half-life of ^{90}Sr . Whereas lichens showed only a loss of the adsorbed or absorbed ^{90}Sr , vascular plants are quite different from lichens with respect to the strontium. They take it up from the soil much more effectively than ^{137}Cs (Haunold *et al.* 1982, Gerzabek *et al.* 1991). Fig. 3. demonstrates that there is a positive correlation between ^{90}Sr and Ca^{++} concentrations among the plants tested in Table 1. On the other hand ^{137}Cs shows no correlation to K^+ (correlation coefficient $r = -0.252$).

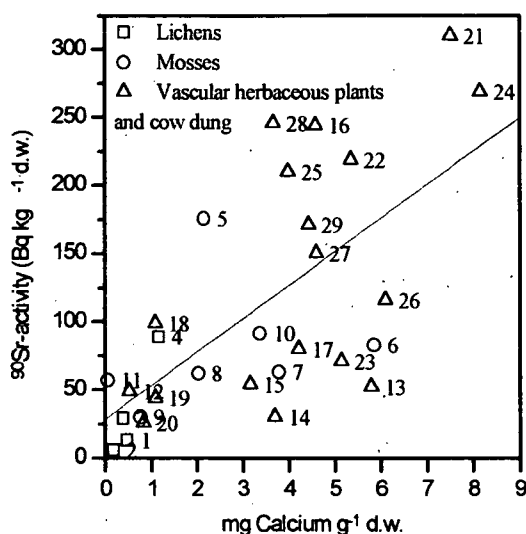


Fig 3. Correlations between ^{90}Sr and Ca^{++} among different plants collected at Koralpenblick (K) and Weinebene (W) between July 1993 and July 1994.

1. *Cetraria islandica*, K. 2. *C. islandica*, W. 3. *Pseudevernia furfuracea*, K. 4. *P. furfuracea*, W. 5. *Bazzania trilobata*, K. 6. *Hylocomium splendens*, K. 7. *Hypnum cupressiform*, K. 8,9. *Leucobryum glaucum*, K. 10. *Sphagnum nemorum*, K. 11. *Polytrichum norvegicum*, W. 12. *Anthoxanthum odoratum*, W. 13. *Calluna vulgaris*, K. 14.,15. *C. vulgaris*, W. 16. *Geum montanum*, W. 17. *Hieracium silvaticum*, K. 18. *Loiseleuria procumbens*, W. 19. *Luzula luzuloides*, K. 20. *Nardus stricta*, W. 21. *Potentilla aurea*, W. 22. *Vaccinium myrtillus*, K. 23. *V. myrtillus*, W. 24,25. *V. vitis-idaea*, K. 26. *V. vitis-idaea*, leaf, K. 27. *V. vitis-idaea*, stem, K. 28. *Veratrum album*, W. 29. Cow dung, W.

Linear regression: $N = 29$, correlation coefficient $r = 0.663$.

4. Acknowledgements

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ACCUMULATION OF RADIOCESIUM AND TRACE ELEMENTS IN MUSHROOMS COLLECTED FROM JAPANESE FORESTS

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Abstract: Concentrations of ^{137}Cs , ^{134}Cs and ^{40}K for 124 mushroom species (284 samples) collected from Japanese forests during 1989 to 1991 were studied. The levels of ^{137}Cs in mushrooms varied very widely ranging from <3 to 16000 Bq/kg (dry wt), while those of ^{40}K were relatively constant. The median concentrations of ^{137}Cs and ^{40}K were 53 and 1180 Bq/kg (dry wt), respectively. The ^{134}Cs discharged through the Chernobyl accident was detected only in 33 mushroom samples. The proportions of ^{137}Cs originating from the Chernobyl accident in these mushrooms were in the range $<4 - 60\%$. The ^{137}Cs concentrations in mycorrhizal fungi tended to be higher than those in saprophytes. The concentrations in many mushrooms reflected the layers in which their mycelia were growing. In addition to the radionuclides, many stable elements including stable Cs in mushroom and soil samples were determined. A good correlation between ^{137}Cs and stable Cs was observed for 33 mushrooms collected from a pine forest on sandy soil near coast in Tokai-mura, Ibaraki, indicating that mushroom available Cs is recycling in the pine forest with the constant $^{137}\text{Cs}/\text{stable Cs}$ ratio. Accumulations from soil to mushrooms were observed for Cd, Rb, Zn, Cu, Cs, K, Mg, Mn, Ni and Tl. Through cultivation experiments in flasks using radiotracers, ^{137}Cs , ^{65}Zn and ^{54}Mn were observed to be accumulated in mushrooms.

1 Introduction

Radiocesium has been discharged into the global environment through nuclear weapons testing and nuclear accidents. After the Chernobyl accident, high concentrations of radiocesium in mushrooms, especially fruit bodies of basidiomycetes, were reported in European forests (e.g. Haselwandter et al., 1988; Baldini et al., 1989; Römmelt et al., 1990). We have also reported relatively high concentrations of radiocesium in mushrooms collected in Japanese forests (Muramatsu et al., 1991; Yoshida and Muramatsu, 1994a, b; Yoshida et al., 1994).

There are many factors controlling the radiocesium concentration in mushrooms, e.g. type of forest (Heinrich, 1992), soil pH (Eckl et al., 1986) and species of mushrooms (Dighton et al., 1991). In addition to these, the authors have considered the relationship between the habitat of the mycelium and the radiocesium concentration in the fruit body (Yoshida and Muramatsu, 1994a; Yoshida et al., 1994).

In order to obtain information on the accumulation of radiocesium in mushrooms due to the contamination by nuclear weapons testing and the Chernobyl accident, we have carried out analyses and radiotracer experiments. In this paper, we summarized our analytical data of ^{137}Cs , ^{134}Cs and ^{40}K for mushrooms (284 samples belonging to 124 species) collected from Japanese forests during 1989 to 1991. In addition to the radionuclides, many stable elements including stable Cs in mushroom and soil samples were determined by using ICP-AES and ICP-MS to understand the accumulation mechanisms. Ability of mushrooms for radionuclide uptake was examined by cultivation experiments in flasks using radiotracers, ^{137}Cs , ^{85}Sr , ^{60}Co , ^{54}Mn and ^{65}Zn .

2 Materials and Methods

2.1 Analysis of ^{137}Cs , ^{134}Cs and ^{40}K

Mushroom samples were collected mainly from forests at different locations in Japan during 1989 to 1991. Soils at different depths were also sampled. Mushroom samples were freeze-dried and pulverized with a cooking blender. Soil samples were air dried and sieved (1 mm). Dried samples (usually 20-40 g for mushroom samples and 50-100 g for soil samples) were placed in plastic bottles (diameter: 50 mm) and concentrations of ^{137}Cs , ^{134}Cs and ^{40}K were determined with a Ge-detector for 40000-80000 seconds. Decay corrections were made to the sampling date. Approximate limits by our counting system for a 30 g dry sample were 3 Bq/kg for ^{137}Cs and ^{134}Cs and 100 Bq/kg for ^{40}K .

2.2 Analysis of stable elements

Mushroom (0.2-0.5g) and soil (0.1g) samples were digested in TeflonTM beakers or TeflonTM PFA pressure decomposition vessels with acids (HNO_3 , HF and HClO_4). A hot plate (at about 150 °C) or microwave digester (CEM, MDS-2000) was used for heating the samples. After digestion, the samples were evaporated to dryness. Then, the residues were dissolved in 1-2% HNO_3 , yielding the sample solutions. Major elements (K, P, Mg, Na, Ca, etc.) were analyzed by ICP-AES. Trace elements (Cs, Sr, Zn, Cu, Cr, Ni, Co, Cd, La, etc.) were measured by ICP-MS.

2.3 Radiotracer Experiments

Three mushroom (*Hebeloma vinosophyllum*, *Flammulina velutipes*, *Coprinus phlyctidosporus*) and one plant (*Medicago sativa*) species were cultivated in the agar media (with yeast and malt extracts for the nutrients) in which ^{137}Cs , ^{85}Sr , ^{60}Co , ^{54}Mn and ^{65}Zn were added as radiotracer. Mushrooms were cultivated in flasks at 22-25 °C for 1-2 months in light condition. The plant was cultivated from seeds at the same condition as mushrooms for 1 week. Some fruit bodies or seedlings taken from a flask were packed in a polyethylene test tube, and the activities of radiotracer were counted with a Ge-detector.

3 Results and Discussion

3.1 Radionuclides in Mushrooms

Table 1 summarized the analytical results for ^{137}Cs , ^{134}Cs and ^{40}K in 124 Japanese mushroom species (284 samples) collected from 1989 to 1991. The concentrations of ^{137}Cs in mushrooms varied very widely (<3 - 16300 Bq/kg, dry wt or <0.4 - 1250 Bq/kg, wet wt), while those of ^{40}K fluctuated within a narrow range. Similar patterns have been reported in Europe (e.g. Dighton and Horrill, 1988; Borio et al., 1991). The median values for the whole mushroom species were 53 Bq/kg (dry wt) for ^{137}Cs and 1180 Bq/kg (dry wt) for ^{40}K as listed in the table. In comparison with the radiocesium concentrations reported in Europe after the Chernobyl accident, our results were generally one or two orders of magnitude lower.

Table 1: Mean, median and ranges of the radionuclides concentration in 124 mushroom species collected in Japan from 1989 to 1991 (Bq/kg, dry wt)

	n*	Nuclides	Mean**	Median	Range
Whole samples	124	^{137}Cs	433	53	<3 - 16300
		^{134}Cs	18	10	<1 - 436
		^{40}K	1150	1180	<39 - 2790
Mycorrhizal fungi	59	^{137}Cs	810	202	11 - 16300
		^{40}K	1310	1290	608 - 2390
Saprophytes	62	^{137}Cs	96	29	<4 - 1520
		^{40}K	1040	1110	<84 - 2790

* n: Number of species

** In the calculation of the mean values, values below the detection limits were regarded as measured values. Therefore, the mean values might be somewhat overestimated.

Among all the species collected from 1989 to 1991, 59 were mycorrhizal fungi and 62 were saprophytes. The ^{137}Cs concentrations in mycorrhizal fungi tended to be higher than those in saprophytes, although the concentration range of each type varied widely. Median concentrations for mycorrhizal fungi and saprophytes were 202 and 29 Bq/kg (dry wt), respectively. Similar findings on the differences in ^{137}Cs concentration between mycorrhizal fungi and saprophytes were reported by Römmelt et al. (1990) and Heinrich (1993).

On the other hand, the relationship between the habitat of the mycelium and the radiocesium concentration in the fruit body was considered (Yoshida and Muramatsu, 1994a; Yoshida et al., 1994). Sixty-three mushroom samples collected from a pine forest on sandy soil in Tokai-mura, Ibaraki were categorized into four different groups according to the habitat of their mycelia, i.e. (1) fallen trees, (2) litter layer, (3) surface soil layer (0-5 cm) and (4) deeper soil layer (>5 cm). The concentrations in mushrooms reflected the layers in which their mycelia were growing, indicating that the habitat of the mycelia would be one of the most important factors controlling the radiocesium concentration in many mushrooms (Figure 1). These results also suggested that the mushroom species inhabiting in the deeper soil layer may have higher radiocesium concentrations in the future, if the nuclide moves downwards in the soil or litter.

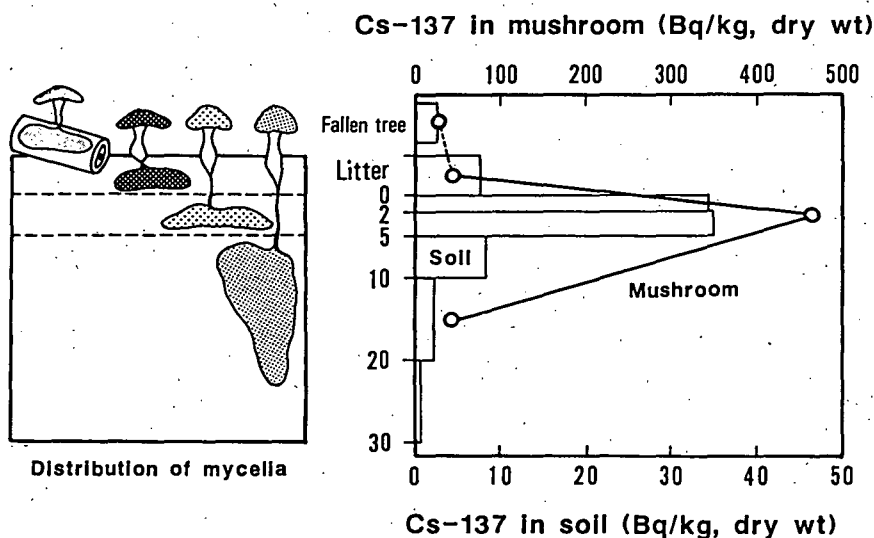


Figure 1: Relationship between the concentrations of ^{137}Cs in mushrooms of each group and respective soil (or litter) layers and fallen tree (Yoshida and Muramatsu, 1994a).

The highest concentration of ^{137}Cs was found in *Hebeloma* sp. (16300 Bq/kg, dry wt), followed by *Tricholoma flavovirens*, *Ramaria formosa*, *Lactarius corrugis*, *Sarcodon aspratus*, *Rhodophyllus* sp., *Cortinarius* spp., *Cortinarius saturninus*, *Agrocybe erebia*, *Rhodophyllus rhodopolius* and *Rhodophyllus crassipes* with average concentrations of >1000 Bq/kg (dry wt). Relatively high concentrations in mushrooms of *Cortinariaceae* such as *Cortinarius* spp. and *Hebeloma* spp. were also reported in Europe (Byrne, 1988; Römmelt et al., 1990), indicating the specific accumulation of ^{137}Cs in this species. Species such as *Kobayasia nipponica*, *Leucopaxillus giganteus* and *Astraeus hygrometricus* contained low amounts of ^{137}Cs . In 33 mushroom samples, ^{134}Cs discharged through the Chernobyl accident was detected. The proportions of ^{137}Cs originating from the Chernobyl accident in these mushrooms, which were calculated from the ^{134}Cs concentrations, were in the range $<4 - 60\%$ (mean value; 20%). The highest proportions were found in *Lactarius hatsudake*. This species was observed to inhabit the surface soil layer where the ^{134}Cs derived from the Chernobyl accident is accumulated. However, the ^{137}Cs in Japanese mushrooms originates mainly from the fallout of nuclear weapons testing, particularly in the 1960s.

3.2 Stable Elements in Mushrooms

Analytical results of 28 major and trace elements in 33 mushroom samples collected from a pine forest on sandy soil near coast in Tokai-mura, Ibaraki, are shown in Table 2 together with the data for ^{137}Cs and ^{40}K . The highest concentration was found for K followed by P, Mg, Na, Ca, Fe, Al, Rb. In comparison with the element composition of plants, the mushroom composition can be characterized by the low Mg, Ca, Sr and Ba concentrations.

A good correlation ($R = 0.991$) between ^{137}Cs and stable Cs was observed for these mushroom samples, although the type of mushrooms and the place of mycelia differed from each other. In this area, the distribution pattern of ^{137}Cs and stable Cs in the soil profile is different, i.e. the highest ^{137}Cs concentration is found in the surface soil layer, whereas the stable Cs concentrations in the soil profile is relatively constant. These findings suggests that available Cs for mushrooms is recycling in the pine forest with the constant ^{137}Cs /stable Cs ratio. The ^{137}Cs /stable Cs ratio estimated was 160 (Bq/mg). The correlation between Rb and Cs was also good ($R = 0.811$). Such stable elements analyses might be useful in predicting the migration of radionuclides also in the areas contaminated with Chernobyl fallout.

In order to estimate the accumulation of each element by mushrooms, tentative transfer factors (TFs) were calculated by using the element concentrations in the surface soils collected in the area. The TF of the element was defined as "median concentration in mushrooms on a dry weight basis" divided by "concentration in the surface soil on a dry weight basis". The highest TF was observed for Cd (TF = 16). The TFs between 1 and 10 were observed for ^{137}Cs , ^{140}K ,

Table 2: Major and trace elements concentration in 33 mushrooms collected from a pine forest in Tokai-mura, Ibaraki

	Mean	Median	Range	Surface soil*	TF**
dry/wet	0.13	0.09	0.05 - 0.79		
	Bq/kg, dry wt				
¹³⁷ Cs	310	127	3 - 3110	35	3.6
⁴⁰ K	873	883	150 - 2130	619	1.4
	mg/kg, dry wt				
K	24100	22800	746 - 56100	26900	0.8
P	5140	4700	742 - 12900		
Mg	978	930	407 - 1680	4190	0.2
Na	867	718	60.0 - 3020	18700	0.04
Ca	393	217	6.95 - 1470	14700	0.02
Fe	298	150	38.0 - 1320	12600	0.01
Al	248	173	32.5 - 870	53200	0.003
Rb	120	92.4	3.05 - 422	56.5	1.6
Zn	82.8	65.5	15.4 - 238	25.5	2.6
Mn	32.1	25.8	3.38 - 125	265	0.1
Cu	27.7	21.4	4.28 - 175	4.40	4.9
V	9.69	0.78	0.05 - 111	27.1	0.03
Cr	3.23	1.13	0.19 - 27.5	12.3	0.09
Sr	2.53	1.42	0.12 - 11.1	160	0.01
Cd	2.17	0.80	0.12 - 11.4	0.05	16
Cs	1.95	0.76	0.03 - 20.1	1.32	0.6
Ni	1.87	0.88	0.29 - 14.8	4.95	0.2
Ba	1.57	0.95	0.21 - 8.49	360	0.003
Pb	1.16	0.79	0.13 - 7.23	13.0	0.06
Ce	0.39	0.19	0.02 - 1.69	18.5	0.01
Co	0.29	0.20	0.05 - 2.34	3.86	0.05
Li	0.22	0.16	0.01 - 1.14	14.0	0.01
La	0.19	0.10	0.01 - 0.94	8.72	0.01
Nd	0.16	0.08	0.01 - 0.78	6.80	0.01
Y	0.12	0.05	0.004 - 1.10	5.42	0.01
Sc	0.11	0.10	0.03 - 0.24	4.24	0.02
Ga	0.09	0.07	0.01 - 0.33	10.4	0.007
Tl	0.07	0.06	0.001 - 0.34	0.36	0.2

* Surface soil was collected in the pine forest (0-5 cm for ¹³⁷Cs and ⁴⁰K) or in a open area beside the forest (0-10 cm for stable elements).

** TF (transfer factor) was defined as "median concentration in mushrooms" divided by "concentration in surface soil".

Rb, Zn and Cu, and those between 0.1 and 1 were observed for Cs, K, Mg, Mn, Ni and Tl. The TFs for the other elements such as Sr, Ca, Ba, Co, La, Pb were less than 0.1. The relatively high TFs for ^{137}Cs and stable Cs indicate the high ability for the accumulation of radiocesium by mushrooms.

Among 33 mushrooms analyzed, the species-specific accumulations were observed in *Tricholoma flavovirens* for Cs, *Amanita pantherina* for V and Cd, *Agaricus* sp. for Co, Tl, Cd and Pb, and *Lycoperdon perlatum* for Cu and Pb.

3.3 Radiotracer Experiment

Results obtained by the radiotracer experiments also indicate that mushrooms have ability to accumulate ^{137}Cs (Ban-nai et al., 1994). The TF (or concentration ratio), which was defined as "activity of radionuclide in mushroom (Bq/g, wet wt)" divided by "activity of radionuclide in medium (Bq/g, wet wt)", had the range from 2.6 to 21 for ^{137}Cs . The highest values was observed in *Hebeloma vinosophyllum*. The value was much higher than that in the plant samples. The findings agreed with our field observations described above, in which *Hebeloma* spp. collected in forests contained large amount of ^{137}Cs . The concentration ratio of ^{54}Mn for the mushrooms was about 10, while the ratio of ^{65}Zn ranged from 15 to 30. No considerable accumulations were observed for ^{85}Sr and ^{60}Co . The results agree with those obtained from the stable element analysis of wild mushrooms described in Table 2.

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THE ROLE OF FUNGI IN THE TRANSFER OF ^{137}Cs IN THE FOREST ECOSYSTEM

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Abstract

Fruitbodies of fungi show much higher ^{137}Cs levels, often more than 10 times higher, than other plants in the forest ecosystems. Roe deer are a major consumers of mushrooms and therefore also show a marked seasonal variation in ^{137}Cs levels. In Sweden, mushroom picking is very common and each person picks about 3 kg fw each year. Assuming that there are two species in the mushroom basket, *Cantarellus cibarius* and *C. tubaeformis* with a TF_g of 0.39 and 0.94 $\text{m}^2 \text{kg}^{-1}$, the total annual transfer of ^{137}Cs from forest ecosystem to man in Sweden due to mushrooms will be very high or 12.8 GBq compared to a transfer of about 6.2 GBq from other pathways, game animals and berries. This 12.8 GBq is an upper limit since we have not assumed any loss during cooking and no counter measure such as reduced mushroom picking in the most contaminated areas in Sweden.

1. Introduction

Fungi are very important in the process of soil-plant nutrient transfer due to the symbiosis between fungi and most of the forest plant species. In forest soil, fungal mycelium is responsible for both the decomposition of litter and for the main uptake of the mineral nutrients. The ^{137}Cs activity concentration in the mycorrhizae of *Suillus variegatus* seems to be rather similar to that found in the fruitbodies and there seems to be an inflow of ^{137}Cs from the surrounding mycelium to the fruitbodies during their formation (Nikolova et al. 1995).

The fruitbodies of fungi show very high ^{137}Cs activity concentrations with transfer parameters (TF_g , $\text{Bq kg}^{-1} / \text{Bq m}^{-2}$) often above 1 (Heinrich 1994, Kammerer et al. 1994). Normal TF_g s for vascular plant species are usually more than 10 times lower, with a few exceptions such as heather (*Calluna vulgaris*).

In Sweden, the consumption of mushrooms picked in the forest is rather common (Hultman 1983). Therefore the potential transfer of ^{137}Cs to man by mushrooms in Sweden is high. Furthermore, many game animals, particularly roe deer, consume quite large amounts of mushrooms (Cederlund et al. 1980). In roe deer rumen it is possible to find 10 to 20 % of the rumen content as mushrooms and the intake of ^{137}Cs may be very high during the mushroom season (Karlén et al 1991). Moose seem to consume much less mushrooms - at least large quantities are not found in the moose rumen (Cederlund et al. 1983, Johanson et al. 1994).

2. Material and methods

2.1. Study area

Samples of plants, mushrooms and game animals were collected from a forest area located in Heby commune about 40 km northwest of Uppsala. The ground deposition of ^{137}Cs in the area was 37 kBq m^{-2} (Nikolova et al. 1995). The dominating trees in the forest are Scots pine (*Pinus sylvestris*) and Norwegian spruce (*Picea abies*). In the field layer, bilberry (*Vaccinium myrtillus*), lingonberry (*Vaccinium vitis-idaea*) and heather (*Calluna vulgaris*) are the dominating species. On clear-cuts, fire weed (*Epilobium angustifolium*) and wavy hairgrass (*Deschampsia flexuosa*) are common. In autumn, fruitbodies of fungi may be very common.

2.2. Sampling of plant material

In July and August 1995, bilberry and lingonberry were sampled in various stands of pine or spruce. The sites were chosen from the forest mapping and attempts were made to choose places representative of the various types of stands. Usually 6 composite samples of the green parts of the plants were collected within a certain stand. The samples were dried at 105°C for at least 24 hours and then milled and transferred to a plastic vial for determination of the ^{137}Cs activity.

2.3. Sampling of mushrooms

In 1992, fruitbodies of various species of fungi were collected from August to October. The fruitbodies were cut into small pieces and transferred to plastic vials for determination of ^{137}Cs activity. The mushroom samples were then dried at 55°C and the ^{137}Cs levels, $\text{Bq kg}^{-1} \text{ dw}$, were calculated.

2.4. Sampling of game animals

Samples of the foreleg muscles of harvested roe deer were prepared directly into plastic vials by the local hunters following an instruction stressing the importance of clean muscle samples without blood or tendons. The samples were checked at the laboratory and if necessary the samples were cleaned from tendons and blood.

2.5. Radiometry

The ^{137}Cs activities were determined using hyperpure Ge detectors at the low background laboratory at the Department of Radioecology, Swedish University of Agricultural Sciences, Uppsala.

3. Results and discussion

3.1. ^{137}Cs in plants

As can be seen in Table 1, there were quite large variations in the ^{137}Cs levels in green parts of bilberry and lingonberry grown in different stands in the forest. The lowest levels were found on clear cuts, 572 Bq kg⁻¹ dw, and the highest levels on bogs and rocky areas, particularly if there was a mixture of rocky and bog sites where levels of more than 5,000 could be found.

Table 1. The ^{137}Cs activity concentrations in green parts of bilberry and lingonberry collected in various forest stands. Site 1; an old pine and spruce stand, site 2; a bog with slowly growing pine, site 3; a clear cut, site 4; an old pine stand on rocks, site 5; an old spruce stand, site 6; a young pine and spruce stand, site 7; an old spruce stand and site 8; a clear cut

	1	2	3	4	5	6	7	8
Bilberry	2260	5251	1740	3189	4582	4004	2260	932
Lingonberry	2901	-	1036	2605	1956	2498	2060	572

The TF_g for bilberry and lingonberry (green parts) varied from 0.015 to 0.14 m² kg⁻¹ depending on the stands. The mean TF_g for bilberry and lingonberry was 0.067 and 0.072 m² kg⁻¹ respectively.

3.2. ^{137}Cs in mushrooms

During 1992, the mean ^{137}Cs in all collected mushrooms was 45 kBq kg⁻¹ dry weight corresponding to a TF_g of 1.2. For *Cantarellus cibarius* the mean value was 14.6 kBq kg⁻¹ and for *Cantarellus tubaeformis* 35.3 kBq kg⁻¹ with TF_g of 0.39 and 0.94, respectively.

3.3. ^{137}Cs intake by game animals

Roe deer is a big consumer of mushrooms. The mean values of the mushroom content in the rumen for three years were 15 % in August, 20 % in September and 30 % in October

(Bergström et al. 1993). Outside this period the intake of mushrooms was much smaller. The total food intake during the period August to October is 0.55 kg dry weight (Drozd and Osiecki 1973). During this period, the most important fodder plants are dwarf-shrubs, mainly bilberry and lingonberry, leaves of deciduous trees and some herbs. The mean ^{137}Cs level of bilberry and lingonberry is 3027 and 1947, respectively, with a mean value for the two species of 2487 Bq kg^{-1} (Table 1). The corresponding value for deciduous trees is $1,500 \text{ Bq kg}$ and for herbs 1000 (Fawaris, 1995). The daily food intake of dwarf-shrubs is 60, 30 and 25 % during August, September and October, respectively. The corresponding values for deciduous trees were 5, 45, and 20 % and for herbs 10 in August and only small levels in September and October. If only these three vascular plant groups are considered, the daily intake of ^{137}Cs without mushrooms would be between 1041 and 1223 Bq per day (corrected up to 100%). The food intakes as mushrooms were 15, 20 and 30 % of the rumen content. Assuming $45,000 \text{ Bq kg}^{-1}$, these correspond to a ^{137}Cs intake due to mushrooms of 3713, 4950 and 7425 Bq during August, September and October respectively. The total ^{137}Cs intake would be 4630, 5691 and 7931, respectively. Compared with the ^{137}Cs intake without mushrooms, there will

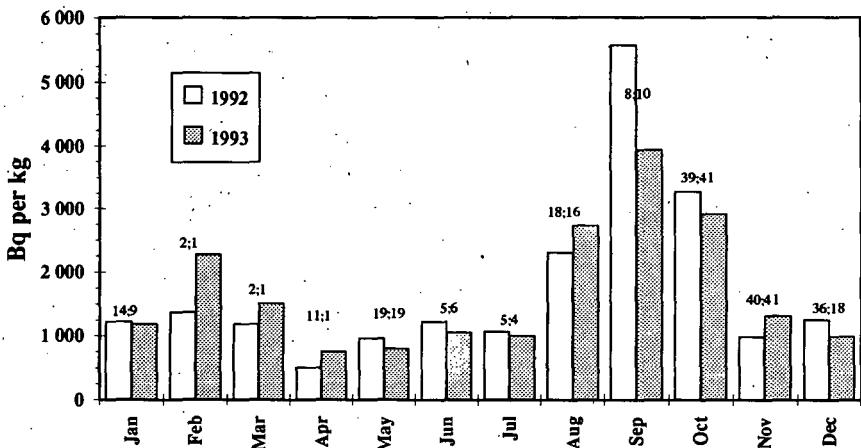


Figure 1. The seasonal variation in ^{137}Cs activity concentration in roe deer during 1992 and 1993. The number of collected samples is shown above the bars.

be an increase between 379 to 706 %. As can be seen in Figure 1, the ^{137}Cs levels increased in roe deer during the mushroom season from a mean value of $1,000 \text{ Bq kg}$ during June and July to about 2,400, 4,800 and 3,200 Bq kg in August, September and October, respectively corresponding to an increase of between 260 to 480 %. In the calculation of daily ^{137}Cs intake

we used the mean levels of all mushrooms collected - $45 \text{ kBq kg}^{-1} \text{ dw}$ - mean levels of all mushrooms collected - 45 kBq kg^{-1} - since we do not know what species roe deer prefer. The calculated increase in ^{137}Cs daily intake is 145, 114 and 231 % higher than the increase in ^{137}Cs levels in roe deer muscles. In August and September, there seems to be rather good agreement between the calculated increase of the daily intake of ^{137}Cs and the observed levels in harvested roe deer. In October the calculated value is much higher, probably due to intake of fungal species with lower levels than 45 kBq kg^{-1} . Therefore, it seems to be more relevant to assume that the mean ^{137}Cs levels in the mushrooms roe deer prefer in October would be about 20 kBq kg^{-1} .

Table 2. The daily intake of dwarf-shrubs, deciduous trees, herbs and mushrooms in per cent of total rumen content and the calculated daily intake of ^{137}Cs of roe deer during the period July to October

	August		September		October	
	%	Bq	%	Bq	%	Bq
Dwarf-shrubs	60	821	30	410	25	341
Deciduous trees	5	41	45	371	20	165
Herbs	10	55				
Sum	75	917	75	741	45	506
	100	1223	100	1041	100	1124
Mushrooms	15	3713	20	4950	30	7425
Total sum		4630		5691		7931
Increase of ^{137}Cs intake due to mushrooms (%)	379		547		706	

3.3. ^{137}Cs transfer to man

In Sweden, the annual consumption of mushrooms has been estimated to be about 3 kg fresh weight or about 0.24 kg dry weight (Hultman 1983). We do not know the distribution of various species among these 3 kg. However, it seems quite clear that *Cantharellus cibarius* and *C. tubaeformis* are the most frequent species in the mushroom basket. We therefore assume

that 1.5 kg fw or 0.12 kg dw of *C. cibarius* and 1.5 kg of *C. tubaeformis* are representative for the Swedish consumption of mushrooms. The TF_g for the two species are 0.39 and 0.94, respectively. Assuming that these transfer parameters are relevant for all parts of Sweden, it is possible to calculate a potential transfer of ^{137}Cs to the Swedish population - 8 million people. The mean ground deposition of ^{137}Cs from Chernobyl in Sweden is $10,000 \text{ Bq m}^{-2}$ (Edvarson 1991) and the mean ^{137}Cs levels in *C. cibarius* would be $3,900 \text{ Bq kg}^{-1}$ dw and 9,400 in *C. tubaeformis*. This will mean that 468 Bq per capita is transferred by *C. cibarius* and 1128 Bq by *C. tubaeformis* or a total of 1596 Bq corresponding to $21 \mu\text{Sv}$. To the whole Swedish population these will mean a transfer of 12.8 GBq, corresponding to a annual collective dose of 166 manSv. It seems reasonable that some radiocaesium is lost during cooking and in the part of Sweden with the highest contamination there seems to be a reduced consumption of mushrooms due to the ^{137}Cs levels. Corresponding values for the annual ^{137}Cs transfer from the forest ecosystems to man by game animals is about 3.5 GBq and by berries about 2.7 GBq or 6.2 GBq together. Obviously, the potential transfer of radiocaesium to man by mushrooms is higher and more important than other pathways although the calculated annual transfer of 12.8 GBq probably is an upper limit.

4. Conclusion

Within the same forest with rather homogenous ground deposition of ^{137}Cs there was a variation from 900 to $5,000 \text{ Bq kg}^{-1}$ in bilberry partly due to various stands.

Fruitbodies of fungi often show 10 times or more ^{137}Cs activity concentrations than vascular plants growing at the same site. Mycorrhizae seems to have rather similar ^{137}Cs levels as the fruitbodies, indicating that a large fraction of the ^{137}Cs activity in soil is within fungal compartment.

Roe deer consume quite large quantities of mushrooms and during mushrooms season there has been a drastic increase of the daily ^{137}Cs intake and a corresponding increase in the ^{137}Cs levels in muscle tissue of roe deer.

People in Sweden also consume mushrooms and the collective potential transfer to man due to mushrooms may be quite large and one of the most important pathways for ^{137}Cs transfer to man.

5. Acknowledgements

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Bioavailability of Cesium Radionuclides in Prealpine Forests and Lakes

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Abstract

In forests of southwest Germany radiocesium from Chernobyl is still accumulated in the organic layers of the soil. This radiocesium is available for the biosphere showing variations of the transfer factor of 3 orders of magnitude due to variations of thickness, degree of decomposition, and pH of the organic soil horizons as exemplified by measurements of the contamination of soil, plants and 5600 roe deer. Bioavailability of radiocesium seems to be mediated by mycorrhiza fungi.

In lakes the persistence of elevated levels of dissolved radiocesium in the water and in the fish population is found - as in forest soil - to be due to the reversible binding of radiocesium to organic matter in sediments and in watersheds and its redissolution as shown for Vorse, a shallow eutrophic lake. A totally different behaviour is observed in Lake Constance, a deep mesotrophic hardwater lake, in which radiocesium was rapidly removed and strongly bound at clay mineral particles in the sediment.

These examples demonstrate the importance of detailed information concerning characteristics of specific compartments in ecosystems like fish, fungi, roe deer, etc. for reasonable decision making in terms of countermeasures to accidental contamination.

1. Contamination of roe deer, mushroom, and fish

The prealpine region of Oberschwaben in southern Germany was among the areas in central Europe which were most severely contaminated with cesium radionuclides from the Chernobyl fallout. The observation of rather high contamination values among individual roe deer from this area resulted in a surveillance program covering all roe deer shot by the state forest authorities in the first 5 years after fallout in that region (Lindner et al. 1994). Since 1987, more than 5600 samples of roe deer shot within an area of about 40 x 40 km² have been analysed with respect to their specific cesium activity.

The results shown in fig. 1 for the State Forestry Ochsenhausen reveal a small scale variation in the regional distribution of roe deer contamination, which was reproduced similarly every year. Contamination levels vary widely between less than 10 Bq/kg and more than 3000 Bq/kg. This can be explained by a variation of the cesium availability for plants in the soil. The total cesium-137 inventory of the soil in the State Forestry Ochsenhausen is about 50 kBq/m². In some districts, the level of roe deer contamination has stayed constant over the years.

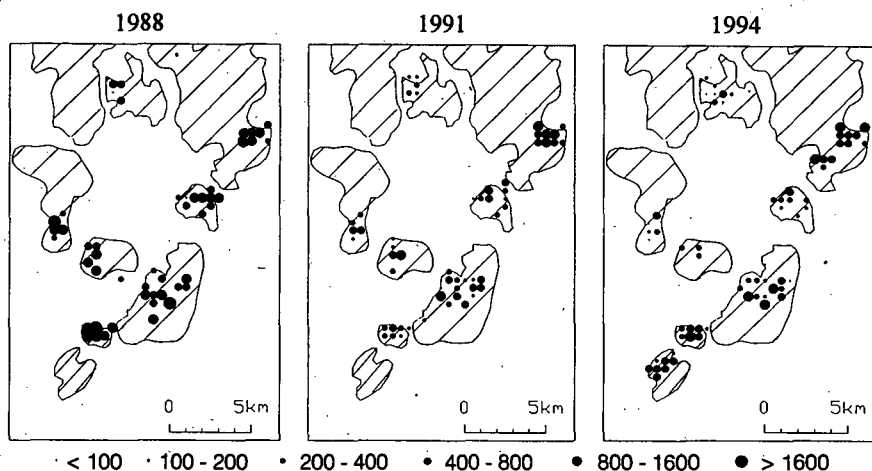


Fig. 1: Regional distribution of contaminated roe deer shot in the years 1988, 1991, 1994 in the State Forestry Ochsenhausen (see map in fig. 4). Forest area is dashed, dots indicate location of shooting and different values of the specific ^{137}Cs activity of roe deer meat in Bq/kg. The numbers of roe deer shot vary between 180 and 280 for the years indicated.

The seasonal structure of roe deer meat contamination (fig. 2a) shows periodic maxima correlated with the mushroom season (fig. 2b). Between 1987 and 1991, an ecological half-time for roe deer meat of about 3 years could be deduced. Moreover in 1993, a drastic increase in the contamination in September was observed due to a strong yield of mushroom which can vary by a factor of 10 between different years (Nikolova et al. 1995). In this way, weather conditions determine the roe deer contamination in fall and so it is difficult to predict the contamination.

In the lakes of the prealpine area in southern Germany, large differences in the bioavailability of cesium radionuclides from the Chernobyl fallout were observed. Two such extreme examples are Lake Constance and Vorsee. Lake Constance is a large and deep mesotrophic hardwater lake, where cesium radionuclides were rapidly removed from the euphotic zone and strongly bound to clay mineral particles in the sediment (Robbins (1992)). Vorsee is a small, shallow eutrophic lake supplied by a swampy watershed, where the persistence of elevated levels of dissolved cesium radionuclides in the water and in the fish population suggests a continuous input both from sediments and from the watershed.

The rapid and efficient removal of dissolved cesium radionuclides from Lake Constance restricted drinking water contaminations to very low levels (below 1 Bq/m^3 ^{137}Cs already in 1991) and the danger of redissolution of cesium radionuclides from sediments is very low (Robbins et al. 1992). The initial accumulation of dissolved cesium radionuclides in the euphotic zone and their early removal explains the time dependence of the observed specific

^{137}Cs activity of herbivorous fish (whitefish, fig. 3c). A rather steep increase in spring 1986 was followed by a rapid and continuous decrease with an ecological half-life of about 4 months. As a consequence, the transfer of ^{137}Cs radionuclides in the trophic chain remained limited and the contamination of carnivorous fish (pike and eel, fig. 3a) never exceeded the initial contamination of herbivorous fish.

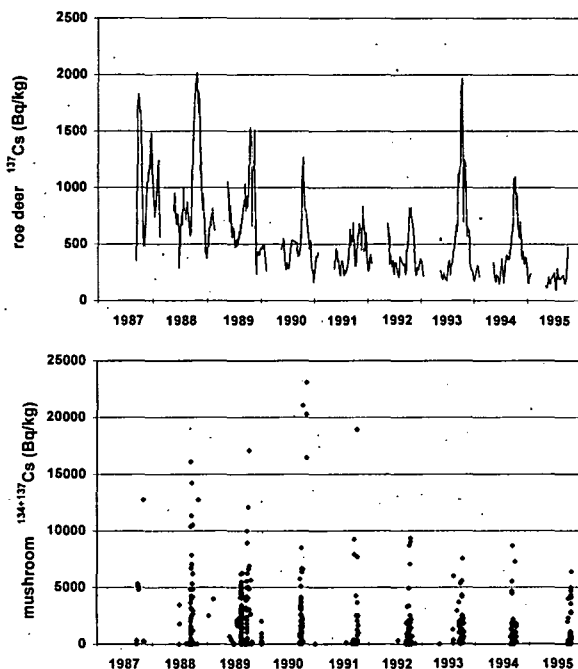


Fig. 2: Seasonal variation of the specific ^{137}Cs activity. a) About 1500 roe deer shot in the State Forestry Ochsenhausen. The full line is the 2 weeks arithmetic average during the hunting seasons. b) About 500 mushroom samples collected in the forests near Ochsenhausen and beyond.

In Vorsee, the specific activity of dissolved ^{137}Cs in the water persisted even in 1995 at a level of about 90 Bq/m^3 . This high contamination level is attributed to a redissolution from sediments due to a decomposition of organic substances, to which organically speciated cesium radionuclides are bound (Kaminski et al (1994)). The implications on fish contaminations become evident from fig. 3b and 3d: The general contamination level of fish is higher than in Lake Constance and the specific ^{137}Cs activity of carnivorous fish is higher than that of herbivorous fish due to food chain accumulation. Additionally, the decrease of contamination is much slower than in Lake Constance.

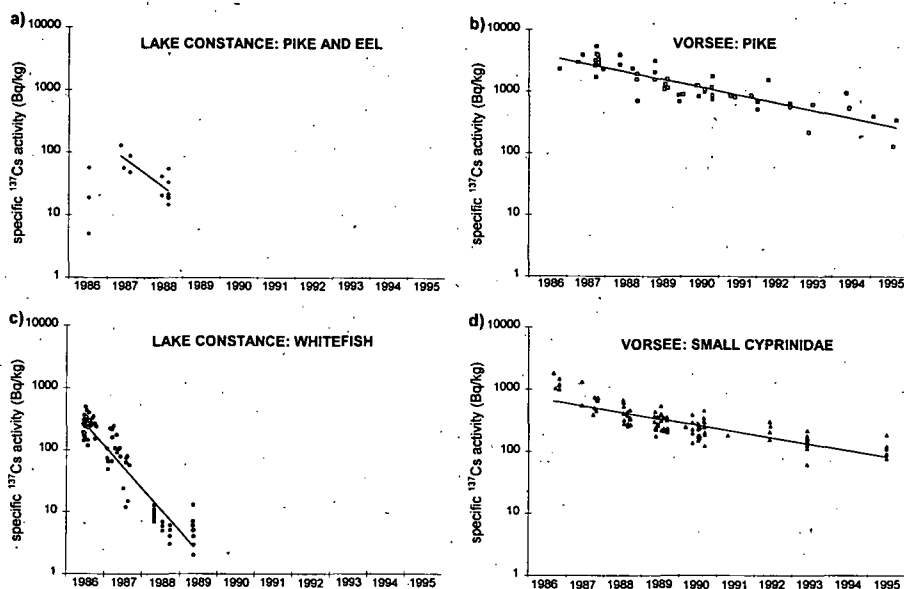


Fig. 3.: Specific ^{137}Cs activity in carnivorous (a,b) and herbivorous (c,d) fish of Lake Constance and Vorsee

2. Transfer factors and classification of soil and sediment

In order to examine the substantial variations of the bioavailability of cesium radionuclides, soil and plant samples were taken at 15 spruce forest sites in southern Baden-Württemberg (fig. 4) in early summer 1994. To measure the plants' uptake of cesium from the soil, the aggregated soil-to-plant transfer factor (T_{ag}) was determined (unit: Bq/kg of dried plant divided by Bq/m² of soil). The T_{ag} was determined for fern (*Dryopteris carthusiana*), bilberry (*Vaccinium myrtillus*), raspberry (*Rubus idaeus*), blackberry (*Rubus fruticosus*), and clover (*Oxalis acetosella*). On all sampling sites, it was highest for fern. Therefore, the T_{ag} of fern, which varied between 0.003 m²/kg and 0.5 m²/kg at the different locations, was chosen to characterize the transfer behaviour of the sampling sites.

Concerning the magnitude of the T_{ag} , a certain order was observed at most sites, i.e. fern showing highest and clover or blackberry lowest values, although soil properties differ considerably at some sites. Therefore, the availability of radiocesium for plants must depend strongly on their physiological properties, e.g. which soil horizon contributes major amounts of nutrients. This is illustrated in fig. 5. It shows a typical vertical root distribution for fern and for clover. Fern with its root system extending down to the A_h horizon has access to a major part of the total cesium inventory, whereas clover roots show a more horizontal structure and extend only in the uppermost 1 to 2 cm of the soil.

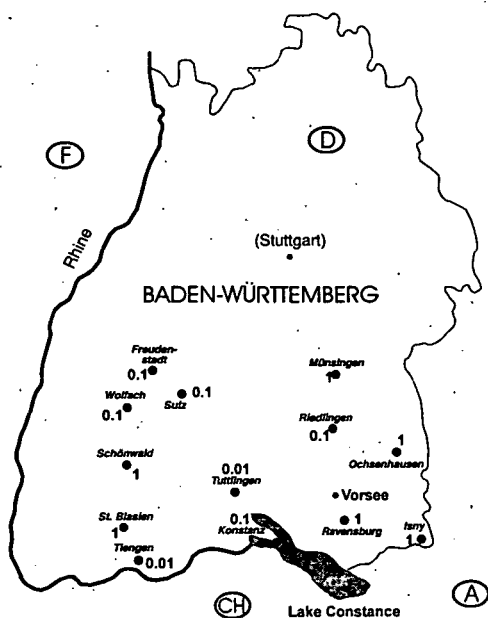


Fig. 4: Sampling locations of soil and plants in spruce stands in southern Baden-Württemberg. The numbers indicate the upper limit of the range of ^{137}Cs aggregated transfer factor T_{ag} for fern in m^2/kg . The lower limit is one order of magnitude smaller.

Highest T_{ag} values are found on sites with thick humus layers (4cm-7cm) showing weak decomposition (raw humus, low pH of 2.3 to 3.5), whereas lowest T_{ag} values are found on sites with thin humus layers (1cm-3cm) showing strong decomposition (O_f mull, high pH of 4 to 6). We conclude that soil-specific characteristics e.g. kind of humus, thickness and pH-value of humus layer, are of dominant influence on the bioavailability of radiocesium.

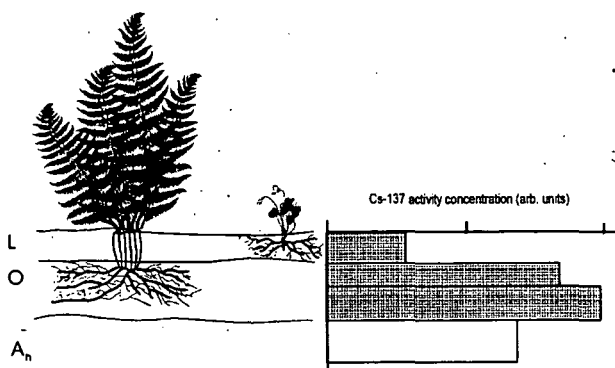


Fig. 5: The root system of fern (*Dryopteris carthusiana*) and clover (*Oxalis acetosella*) and their extension into the different soil horizons compared to the vertical distribution of radiocesium in the soil of a spruce forest. Organic horizons are shaded.

Fig. 6a, b, c show the vertical distribution of cesium radionuclides in the different soil horizons. They illustrate the importance of organic matter for the bioavailability of cesium.

Fig. 6b shows the typical distribution of a spruce stand characterized by a high T_{ag} : A thick organic layer of about 5 cm contains substantial parts of the cesium inventory.

For deciduous forests (fig. 6a) the organic layer is always thin and the T_{ag} is about a factor of 100 lower as compared to high T_{ag} spruce stand locations.

Highest T_{ag} ($>1 \text{ m}^2/\text{kg}$) were found on peaty meadows (fig. 6c), where the organic matter is dominating the whole contaminated profile.

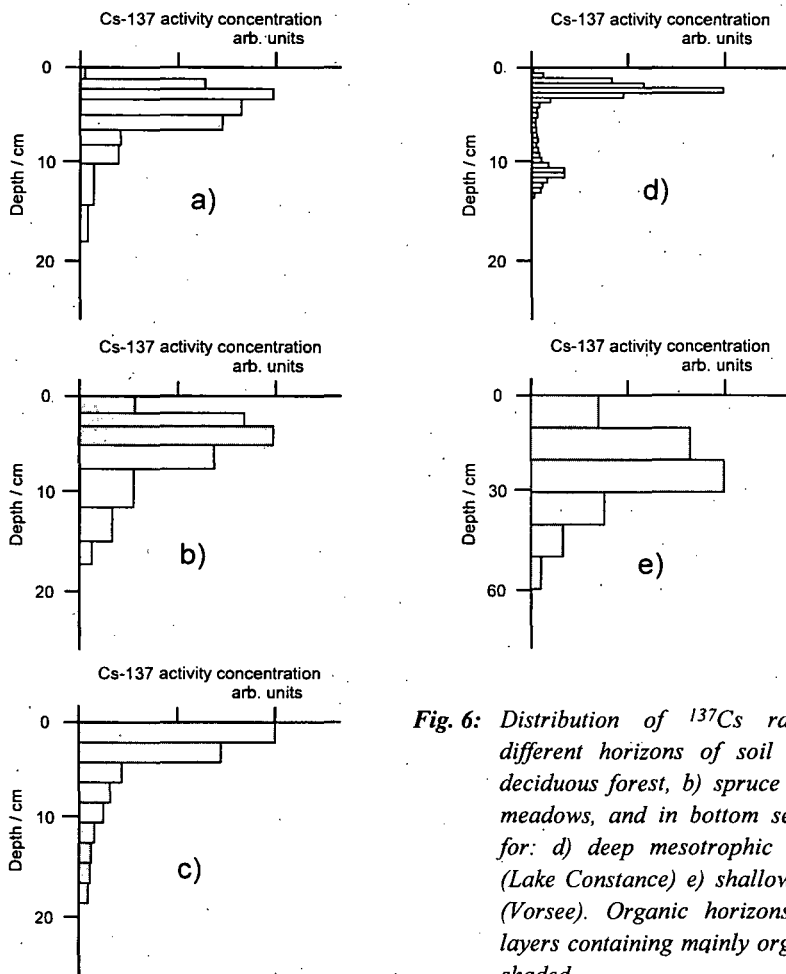


Fig. 6: Distribution of ^{137}Cs radionuclides in different horizons of soil typical for: a) deciduous forest, b) spruce forest, c) peaty meadows, and in bottom sediments typical for: d) deep mesotrophic hardwater lake (Lake Constance) e) shallow eutrophic lake (Vorse). Organic horizons and sediment layers containing mainly organic matter are shaded.

Similar arguments hold for the sediments of lakes. A typical vertical distribution of ^{137}Cs in the sediment of Lake Constance is shown in fig. 6d. The two peaks in the graph result from different events: The lower peak can be related to the fallout due to nuclear weapons testing with a maximum in 1963, the peak in the top layers of the sediment can be related to the year of the Chernobyl accident 1986.

Since cesium radionuclides became fixed in clay minerals at internal binding sites, their solubility from the sediment is very small: Not more than 10% of the Chernobyl-related ^{137}Cs inventory of a representative sediment sample could be extracted in a five-step selective extraction procedure according to Robbins et al. (1992) leaving only clay, quartz and feldspars as solid residues.

The Vorse sediments consist mainly of water (80%) and solid material (20%, mainly organic matter), and the vertical distribution in these sediments (fig. 6e) looks quite different. Due to bio- and/or physical turbation of the sediment the maximum of the activity concentration is found in a depth of about 30 cm. The binding strength of cesium radionuclides to sediments in Vorse is much weaker than in Lake Constance; more than 20% of the cesium inventory could be extracted applying the same procedure. This is attributed to a predominantly organic nature of the binding partners of cesium radionuclides in these sediments and is consistent with the cesium contamination of fish measured in these lakes.

3. Role of mycorrhiza

Mycorrhiza fungi living in symbiosis with plant roots seem to play an important role in the transport of cesium from soil to plants. In our experiments, the length of mycorrhiza hyphae per mass unit of soil was determined under a microscope.

We found higher specific lengths of hyphae by a factor of two at those sites exhibiting considerably higher T_{ag} values of up to one order of magnitude. This result holds for spruce forests (Ochsenhausen and Riedlingen, see map in fig. 4) as well as for deciduous forests in Ticino, Switzerland. Within the same climatic region in Ticino, measurements showed that the specific length of hyphae is about one order of magnitude larger in spruce forests than in deciduous forests. Both results confirm the relationship between density of hyphae and bioavailability of ^{137}Cs .

Olssen et al. (1990) supposed that a major part of ^{137}Cs inventory in the forest soil might be located within the mycelium of fungi. Nikolova et al. (1995) measured the ratio between ^{137}Cs activity concentrations in the mycorrhizae and fruit bodies of fungi and found that up to 50 % of ^{137}Cs soil inventory is located within the mycelium of fungi.

Fig. 7 shows the distribution of the specific ^{137}Cs activity in the different horizons of a spruce forest soil (left), and the density of mycorrhiza hyphae in these horizons (right). Both distributions coincide within the organic horizons, and show a maximum in the A_h layer. This result and that of Nikolova et al. (1995) give strong support to the idea of Olssen et al. (1990) that most radiocesium in the soil is accumulated in the mycelium of fungi.

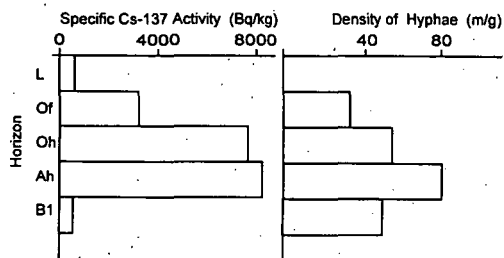


Fig. 7: Distribution of ^{137}Cs radio-nuclides in different horizons of a spruce forest soil (left), and density of mycorrhiza hyphae in these horizons (right).

4. Conclusion

- Radiocesium contamination in forests and lakes can persist during many years due to enhanced bioavailability from organic material in soil or sediment.
- Classification of soils in semi-natural ecosystems in order to predict the bioavailability of radiocesium in an area of about 10,000 km² has been achieved and should be extended to other areas.
- Mycorrhiza mediated transfer of radiocesium from soil to plants is proposed as a field of further investigation to understand the mechanisms underlying the enhanced bioavailability in semi-natural ecosystems.

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MONITORING OF CHERNOBYL FALLOUT ^{137}Cs IN SEMI-NATURAL CONIFEROUS FOREST OF CENTRAL SWEDEN

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ABSTRACT: Monitoring the behaviour of ^{137}Cs in semi-natural coniferous forest ecosystem of central Sweden was investigated between 1990 and 1994. Results demonstrated that soil in the area of study is thin layer characterised with high organic matter fraction and acidic pH. Most of Chernobyl fallout deposited ^{137}Cs retained in the upper 5 cm with venial migration into deeper layers of soil profile. The activity concentration of ^{137}Cs in the dominant plants showed some variation, which presumed to be due to high variations in the different soil parameters and species-specificity of plants in ^{137}Cs uptake. Fractionation of soil bound ^{137}Cs using Sequential Extracted Procedure predicted that easily extractable ^{137}Cs fraction that includes water soluble and NH_4OAc exchangeable comprises ~25% of total soil radiocaesium inventory in the upper 5 cm layer of forest soil. These fractions, represent the soil mobile ^{137}Cs in the forest ecosystem. About 37% of total forest soil ^{137}Cs inventory could be accounted for soil organically bound ^{137}Cs that include oxidizable and acid digestible organic matter. These fractions require a long term bio-degradation process by soil micro-organisms, before becoming available for plant uptake. More over a substantial fraction of ^{137}Cs was firmly bound onto soil compartments of organic and/or mineral nature as a residual (36%). This means that binding processes of ^{137}Cs onto humic forest soil with high fraction of organic matter and acidic reaction is time dependent. The most distinguished soil parameters that might influenced ^{137}Cs transfer to and/or uptake by natural plants in the area of study were soil OM%, and soil pH. Soil biological activity might also be considered, (although it was not determined in this study). The long term ^{137}Cs availability in this type of forest soil must be related to the organically bound ^{137}Cs fractions.

Key words: Forest, ^{137}Cs , Fractionation, transfer, soil parameters, Species specificity

1. INTRODUCTION

The study area is a part of coniferous forest covering about (~2 km²) located in central Sweden about 35 km Northwest of Uppsala (Latitude 60° 8' N and longitude 17° 14' E). Dominant plant litterfall and debris accumulated over solid substratum forming a thin (3-10 cm) humic soil layer supporting growth of vascular plants and fungi. The rocky sites covered with a thin layer of lichens or mosses. Dominant trees: Scots pine (*pinus sylvestris*), Norwegian spruce (*picea abies*) with some intermixture of deciduous trees, mainly birch (*Betula spp*). The understorey vegetation consists mainly of billberry (*Vaccinium myrtillus*), lingonberry (*Vaccinium vitisidaea*), and heather (*Calluna vulgaris*). Herbaceous plants such as, wavy hair grass (*Deschampsia flexuosa*) and fire weed (*Epilobium angustifolium*) are common in clear cuts. The aim of this presentation was to discuss the soil-to-plant transfer of Chernobyl fallout ^{137}Cs

(which will be referred to hereafter as ^{137}Cs) in a coniferous forest ecosystem, under the influence of different forest soil physical and chemical parameters.

2. MATERIAL AND METHODS

2.1 Sampling and treatments

Soil and dominant plants were sampled between July and October, from different locations inside the area of study during the period of investigation (i.e. between 1990-1994). Treatments and preparation of samples for analysis were summarised in (Fawaris, 1995)

2.2 Determination of soil parameters, and Fractionation, of soil bound ^{137}Cs

Soil physical and chemical parameters including soil pH, OM% bulk density, soil CEC, Kd, exchangeable and total cation of K^+ , Ca^{2+} , Mg^{2+} , and P as (P_2O_5) were determined from air-dried soil using conventional standard methods, (for further information see Fawaris & Johanson, 1994; Fawaris, 1995). Fractionation of forest soil bound radiocaesium was performed by use of sequential extraction as described in (Fawaris & Johanson, 1995)

2.3 Data Acquisition and Calculation

Radiocaesium concentration levels (Bq kg^{-1} DM) in soil and plants were determined using high resolution gamma spectroscopy counting system consisting of hyper-pure Ge or NaI (tl) Scintillation detectors, that were available in the Department of Radioecology SLU, Uppsala. Counting time selected to guarantee counting error of less than 5%. Statistical analysis of the data was performed using the Excel Microsoft computer programme and the results are presented as means.

3. RESULTS AND DISCUSSION

3.1 Soil parameters, deposition and distribution of Chernobyl fallout ^{137}Cs

Soil parameters data, which are presented in Table 1 revealed that, the studied forest soil belongs to nutrient deficient soil type when compared to farm land, with acidic pH and high fraction of organic matter. The measured ground deposition of ^{137}Cs ($n = 92$) in 1991-92 was ($27 \pm 8.5 \text{ kBq m}^{-2}$) and was found to agree well with the previously reported ground deposition levels of ^{137}Cs in the area of study (Linde'n & Mellander, 1986; SGAB, 1986). Soil profile studies demonstrated that more than 85% of the total ^{137}Cs inventory confined in the upper 5 cm layer of undisturbed forest soil (Fawaris & Johanson, 1994). The vertical migration of ^{137}Cs into deeper layer is a very slow process and seems to be influenced by OM%, rainfall, soil acidity, the activities of soil living micro-organisms, and may be the fraction of clay minerals although it was very low, (Van-Bergeijk et al., 1992; Thiry & Myttenaere, 1993) In forest ecosystem the litter layer retained a major part of fallout ^{137}Cs (Coughtrey & Thorne, 1993). In some cases, however, up to 90% of ^{137}Cs retained in the upper 3-10 cm of undisturbed soils with a little migration down through soil profile (El-Fawaris & Knaus, 1984; Livens, et al., 1990; Bunzl, et al., 1988; Tobler, et al., 1988).

3.2 Transfer of ^{137}Cs to plant

The ^{137}Cs activity concentration in dominant plants grow within the area of study presented in Table 2. The levels of ^{137}Cs in heather was higher than others all times

Table 1 Summary of coniferous forest soil parameters in the area of study.

Parameter	n	Max	Min	Mean	SD
Soil pH in (H ₂ O)	121	6.1	3.8	4.1	0.40
Soil pH in (KCl)	121	5.0	2.8	3.2	0.20
Soil OM % (LOI)	121	94	10	68	19
Soil Kd (Bq g ⁻¹ / Bq ml ⁻¹)	121	530	6	98	119
Soil Bd (g cm ⁻³)	121	1.15	0.28	0.60	0.20
Soil ¹³⁷ Cs activity (Bq kg ⁻¹ DW)	121	8204	544	3607	1634
Field plants ¹³⁷ Cs (Bq kg ⁻¹ DW)	121	16783	701	5108	4181
Ground deposition ¹³⁷ Cs (Bq m ⁻²)	92	44985	6428	26871	8525
Extracted K ⁺ (mg g ⁻¹)	121	1.68	0.11	0.55	0.27
Extracted Ca ²⁺ (mg g ⁻¹)	121	4.46	0.22	1.57	0.86
Extracted Mg ²⁺ (mg g ⁻¹)	121	0.98	0.10	0.48	0.15
Extracted P as (P ₂ O ₅) (mg g ⁻¹)	121	0.52	0.04	0.17	0.08
CEC meq/100g soil DW	20	201	29	91	41

Table 2 Measured ^{137}Cs activity concentrations in sampled field plants, mean values and (SD) against the elapsed time from same location within the study area between 1986 and 1994

Name of species	Field Plants ^{137}Cs activity in Bq kg^{-1} DM from same site (1986-1994)									
	1986-1989*		1990-1991		1992		1993		1994	
	n	Bq kg^{-1}	n	Bq kg^{-1}	n	Bq kg^{-1}	n	Bq kg^{-1}	n	Bq kg^{-1}
<i>P. sylvestris</i>	8	3500 (523)	4	2303 (713)	5	2178 (613)	12	2205 (933)	17	2163 (1033)
<i>Betula spp.</i>	11	3200 (533)	6	2459 (958)	6	1732 (499)	10	1267 (487)	7	705 (299)
<i>V. myrtillus</i>	7	4662 (273)	11	2932 (743)	9	2373 (608)	12	1899 (553)	8	1489 (567)
<i>V. vitis-idaea</i>	7	7960 (458)	5	4261 (694)	6	2476 (749)	10	1866 (468)	9	987 (413)
<i>D. flexuosa</i>	6	2385 (838)	7	1941 (577)	5	1884 (692)	5	1741 (505)	6	861 (188)
<i>C. vulgaris</i>	9	12269 (909)	15	12580 (899)	6	11960 (951)	19	11336 (940)	21	10318 (967)
<i>Bryophytes</i> §	11	16500 (681)	3	4623 (1124)	6	4653 (864)	7	4560 (751)	11	4573 (604)

* Samples extracted from old data bank at the Deptment of Radioecology.

§ Bryophytes are a mixture of the available species in the area of study.

with ($>11 \text{ k Bq kg}^{-1} \text{ DM}$). In lingonberry and bilberry the levels of ^{137}Cs were slightly above 1.1 and $1.8 \text{ k Bq kg}^{-1} \text{ DM}$ respectively. The ^{137}Cs levels in Scots pine after five years post Chernobyl accident was $2.2 \text{ k Bq kg}^{-1} \text{ DM}$ (Table 3), and seems to reach a steady state between 1991 and 1994. This might reflect that the ^{137}Cs soil-to-plant which transferred through vegetative parts in the early stage of Chernobyl fallout or via root uptake after 5 years is now at least in equilibrium with the loss of ^{137}Cs via litterfall and rain wash of Scots pine canopy. Thus ^{137}Cs will remain for a long time recycling within the coniferous forest ecosystem. Similar conclusion drawn by (Bergman, 1994). In contrast the activity concentration levels of ^{137}Cs in some (moose diet) in central Sweden with vegetation similar to the sampled species where the Chernobyl fallout ^{137}Cs deposition was about 45 kBq m^{-2} showed some variation in the ^{137}Cs levels regardless the slight changes in the levels of ^{137}Cs in moose meet between 1986-1992 (Johanson et al., 1991; Johanson & Bergstrom, 1994). The possible explanation to this is that moose is not foraging a specific site and always migrate from location to another depending upon the season of the year and its feeding habit as well as stand age and density. The ^{137}Cs TCs in forest understory plants growing in different locations within the area of study were within the same order of magnitude (Table 3). Comparing ^{137}Cs transfer parameters of field plants revealed that the variations in ^{137}Cs uptake might be related to species-specificity which was very much dependent upon the type of plant. For example, bilberry ^{137}Cs TCs ranged from 0.04 to 0.24 with a mean of 0.09 ± 0.05 , for heather the range was from 0.39 to 0.64 with a mean of 0.49 ± 0.05 , and for lingonberry the range was from 0.07 to 0.25 with a mean of 0.16 ± 0.06 (Table 2). Heather ^{137}Cs TCs mean value was the highest among all tested plants. An explanation to this might be related to species specific mechanisms on the uptake of ^{137}Cs from a deficient nutrient forest soil. Heterogeneous distribution of ^{137}Cs ground deposition and high ranges of variation in different soil parameters can not be excluded. Variation in ^{137}Cs soil-to-plant transfer always occur even when plant species are grown on the same site. Thus the point that may be raised is that the soil chemistry by itself is not enough to predict the bioavailability and transfer of ^{137}Cs in nutrient deficient forest soil with high OM% and acidic pH. Therefore other parameters must be considered, one of such parameters could be soil biological activity factor (which was not determined in this study).

3.3 Fractionation of forest soil bound ^{137}Cs

Results presented in Table 4 predicted that there are three major fractions of soil ^{137}Cs . The easily extractable ^{137}Cs fraction which should represent ^{137}Cs in water soluble form and NH_4OAc exchangeable forms. This easily extractable fraction is very important for plant root uptake particularly in a short term bases. Similar conclusion were drawn by (Riise, et al. 1990; Oughton et al., 1992). The long term availability of ^{137}Cs , however, must be related to the organically bound ^{137}Cs fractions (the H_2O_2 oxidizable and HNO_3 digestible fractions). However, it should be born in mind that the ^{137}Cs oxidizable fractions might be changed as it is depending upon oxidation reduction processes that controlled by soil micro-organisms activity and their contribution in bio-degradation of soil organic matter which is to a great extent time dependent.

3.4 Influence of soil parameters on soil-to-plants ^{137}Cs transfer

Soil parameters that are expected to contribute to ^{137}Cs uptake and transfer in forest ecosystem were investigated using multiple regression analysis between soil-to-plant

Table 3 Summary of ^{137}Cs transfer parameters (TF & TC) from coniferous forest soil in understorey field plants.

Species	n	Transfer Factor				Transfer Coefficient			
		<i>(Bq kg⁻¹ plantDM/ Bq Kg⁻¹ soil DM)</i>				<i>(Bq kg⁻¹ plantDM/ Bq m⁻²)</i>			
		Max	Min	Mean	SD	Max	Min	Mean	SD
<i>V. myrtillus</i>	48	2.30	0.28	0.85	0.42	0.24	0.04	0.09	0.05
<i>V. vitis-idaea</i>	31	4.46	0.57	1.35	0.78	0.25	0.07	0.16	0.06
<i>Calluna vulgaris</i>	29	8.89	1.80	3.31	1.53	0.64	0.38	0.49	0.05
<i>Mixed bryophytes</i>	13	3.26	1.06	2.06	0.74	0.37	0.16	0.24	0.07

Table 4 Percentage of extracted (F1, F2, F3, F4) and residual (F5) ^{137}Cs fractions from coniferous forest soil to the original soil ^{137}Cs activity concentration in the upper 5 cm due to sequential extraction procedure(SEP).

Sample	Extracted and residual ^{137}Cs fractions				
No.	F1	F2	F3	F4	F5
1	2.3	23.5	35.6	13.3	25.4
2	6.9	22.9	26.3	19.9	24.0
3	9.8	13.4	23.4	32.3	21.1
4	9.3	17.0	14.2	43.7	15.8
5	2.4	16.0	13.3	21.4	47.0
6	5.0	15.3	15.2	25.8	38.6
7	20.0	23.5	18.3	15.7	22.6
8	4.6	12.0	12.9	13.4	57.2
9	9.2	12.9	9.2	16.5	52.2
10	11.5	16.4	12.3	14.4	45.3
11	11.7	16.6	13.4	14.6	43.7
12	9.9	19.3	17.7	19.3	33.7
13	14.2	15.6	15.5	17.0	37.7
14	12.3	11.8	10.3	14.2	51.4
15	13.5	20.6	10.8	28.9	26.1
16	7.2	23.1	12.7	35.6	21.4
17	4.0	14.1	16.1	20.8	45.2
18	2.6	6.8	15.6	19.2	55.7
19	2.0	4.9	11.7	32.4	49.0
20	3.3	7.6	13.8	24.3	51.0
21	11.1	14.1	11.9	20.7	42.2
22	13.3	13.7	16.0	29.8	26.6
Mean	8.46	15.5	15.74	22.42	37.85
SD	4.72	5.13	5.84	8.08	12.66

^{137}Cs TFs of field plants and the different soil parameters excluding soil CEC gave $r^2 = 0.38$, $n = 121$ and $P < 0.001$. The covariation in different soil parameters was surprisingly and some times rather contradictory, as it leads to a buzzing conclusion. For example correlation between K_d and soil OM% was rather good and gave $r^2 = 0.94$, $n = 121$, $P < 0.001$. However, low K_d could result in a rapid migration of deposited ^{137}Cs into mineral layer. In this regard the real situation in the area of study (a coniferous forest ecosystem) was different since a substantial fraction of radiocaesium in forest soils was firmly bound to soil components of organic nature or clay even after treatment with strong oxidising agents regardless the low K_d of forest soil. These reflected that the actual transfer pattern of ^{137}Cs was controlled by other parameters or another mechanism than soil physical and chemical parameters. Additionally the measured ^{137}Cs soil-to-plant TFs in same field plants reflecting a significant difference among the sampled species (Fawaris, 1995). Thus the hypothesis that, not only species-specificity, soil pH and soil OM% were presumed to contribute to ^{137}Cs soil-to-plant transfer and hence fluctuation in the TFs of tested species are still unpredictable.

In boreal coniferous forest, however, most of plants root biomass were quite shallow, which facilitating the uptake and transfer of nutrients as forest floor litter decomposes. Thus soil biological factor and its role in soil organic matter bio-degradation processes must be taken into consideration particularly in forest ecosystem. Moreover, the trees and most associated plants benefit by having mycorrhizae and rapid fine root turnover (Knight, 1991). Bunzl and Kracke (1986) reported that heather (one of ericaceous species) which grow on acid heath, showed higher TFs of ^{137}Cs which could be extended to all plants inhabiting acid bogs.

The role of fungi in the mechanism of radiocaesium translocation and cycling in the forest soil was beyond the scope of this study. However, a large fraction of ^{137}Cs within forest soil is bound within the mycelium of the fungi (Nikolova et al. 1994 personal communication). Taking into consideration at minimum 85% of total ^{137}Cs inventory bound within upper 5 cm of forest soil layer, and about 22% of such amount was readily available for plant uptake from soil solution. Additionally ~36% of total ^{137}Cs inventory in the top 5 cm soil layer which become available for plants root uptake as a result of bio-degradation and decomposition processes by soil microflora and microfauna, one can presume that on the long run between 32-58% of total forest soil ^{137}Cs inventory would be available for plants root uptake. Based upon what has been stated above species-specificity, soil OM%, pH, and soil biological activity were presumed to be behind the dynamics of soil-to-plant ^{137}Cs transfer. This also support the hypothesis that soil-to-plant ^{137}Cs transfer does not depend upon the total ^{137}Cs activity concentration in the soil but upon its availability in soil solution including soil biologically bound ^{137}Cs .

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RADIOCAESIUM LEVELS IN ROE DEER AND WILD BOAR IN TWO LARGE FOREST AREAS IN AUSTRIA

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Abstract

A report is given on the course of radiocaesium contamination in roe deer and wild boar in two large forest areas in Austria. In autumn 1987 and winter 1987/88 radiocaesium levels rose to values higher than those recorded in 1986 in these regions. The reason for this increase was the very specific feeding selection of roe deer in these forest areas resulting in the ingestion of an unusual high amount of blueberries, ferns and mushrooms. An explanation for the changes of wild boar's contamination has not been found yet, but possible reasons are discussed.

1 Introduction

Shortly after the Chernobyl accident in 1986 examinations of radioactive contamination of wildlife were started in Austria (Tataruch et al., 1988). In the beginning the main aspect of these analyses was the radiation protection point of view related to the use of meat from game animals for human consumption. During winter 1986/87 and spring 1987 the levels of ^{137}Cs were low and mostly below the former Austrian legal limit which was defined as 16 nCi/kg ($\sim 592\text{Bq}\cdot\text{kg}^{-1}$) for meat. Surprisingly, in autumn 1987, a significant increase of radiocaesium levels in muscles of roe deer and wild boar was registered in two large forest areas in Austria. In all other regions a slow decrease was noted. These findings were the reason for intensifying research on radioactive contamination of wildlife in the two forest areas. This work is still going on. In this paper a preliminary report will be given.

2. Materials and Methods

Study areas characteristics:

Weinsberger Wald (WW): in the NW-part of Lower Austria, granite, gneiss, brown earth, para brown earth, pseudo podzol, pH 3 - 4, ~ 10.000 ha, elevation of 850 - 1041m, annual precipitation of 900 to 950 mm, unbroken forest with 90% of spruce, beside beech and fir, understory vegetation consisting mainly of blueberries, ferns, wood-sorrel and mosses; high moisture content in soils.

Kobernauber Wald (KW): Upper Austria, ~ 15.000 ha, largest unbroken forest area in Central Europe; soil: tertiary sediments, sands, gravels, semi-podzol, podzol; pH \leq 3.0, elevation 500 - 650m, spruce, fir, beech, heath, understory vegetation mainly blueberries, ferns and mosses.

Radioactive contamination of both regions by direct fallout in 1986 was relatively high (up to 100 kBq \cdot m⁻²; (Bossew et al., 1995)).

Samples were collected during regular hunting activities, immediately after shooting the animal. Usually samples from muscles were taken, additionally kidneys, rumen (stomach) contents and in a few cases liver samples were also taken. The material was kept deep frozen until analyses as described by Tataruch et al. (1988). The numbers of samples collected in the period from 1986 to 1994 were the following:

WW: roe deer: 663 samples of muscle, 36 kidneys and 4 rumen contents; red deer: 85 muscle, 2 kidneys; wild boar: 65 muscle.

KW: roe deer: 77 muscle, 15 liver, 79 kidneys, 57 rumen contents; wild boar: 16 muscle, 3 livers, 14 kidneys, 12 stomach contents.

3. Results and discussion

As already mentioned monitoring of radiocaesium contamination of wildlife continued during 1987. Whereas samples from most areas were contaminated at a low level during summer and autumn, a considerable increase in radiocaesium contamination of muscle of roe deer from WW and KW was found. The highest values for 1987 were 7548 Bq \cdot kg⁻¹ ¹³⁷Cs and 7955 Bq \cdot kg⁻¹ in muscle of roe deer shot in October and November respectively. These values were even higher than the maximum levels recorded in May/June 1986 (4144 Bq \cdot kg⁻¹ and 4551 Bq \cdot kg⁻¹) in these regions. At the same time no elevated activity concentrations were found in red deer. In February

1988, the highest value for wild boar was $17575 \text{ Bq} \cdot \text{kg}^{-1}$ in KW, which was the highest activity concentration of ^{137}Cs ever found in Austrian wildlife. In 1986 wild boars' contamination by radiocaesium had not been very high, so these findings in winter 1987/88 were surprising. Samples from roe deer shot in February 1988 (with special permission) showed lower levels than those in autumn 1987.

Due to these results sampling was intensified, including radiological and botanical analyses of rumen (stomach) contents to find out which components of the animals' feeding might be responsible for the high radiocaesium burden.

3.1. Roe deer (*Capreolus capreolus*)

Figure 1 shows the changes of radiocaesium in muscles of roe deer between the begin of the hunting period in the middle of May until its end in December. Although the individual values varied slightly, the general pattern was the same every year.

At the beginning of the hunting season in the middle of May, the mean ^{137}Cs -levels and the maximum are relatively low, in June, the levels are increasing slightly. From July to September a considerable increase occurs, with the highest median value of the whole season. During autumn high maximum values are found, but the median and mean values decrease.

What are the reasons for these seasonal changes? An explanation is possibly by considering the roe deer's feeding selection from the results of botanical and radiological rumen analyses. Generally, roe deer's diet consists mainly of herbs, leaves, leaf-buds and various sprouts as well as mushrooms, when they are growing (e.g. Onderscheka et al., 1989). If these plants are not available for roe deer in sufficient quality and quantity it has to change to other feeding plants. Botanical analyses of the rumen contents of roe deer from KW showed distinct differences to other roe deer habitats: The proportion of ferns, which is usually low ($< 5\%$) was very high, up to 95% (bracken) as a maximum; blueberry stems and leaves were found more often too in deer from KW; the amount of herbs was lower than in roe deer's diet elsewhere. During the time of their growing period mushrooms were often found in a high percentage. Due to the dense, unbroken forest areas, the understory vegetation in KW is very different from other habitats considered, it is relatively uniform, with only a few species, dominated by blueberries and ferns. Hence, roe deer is forced to feed on these available plants as the forest area is so big that

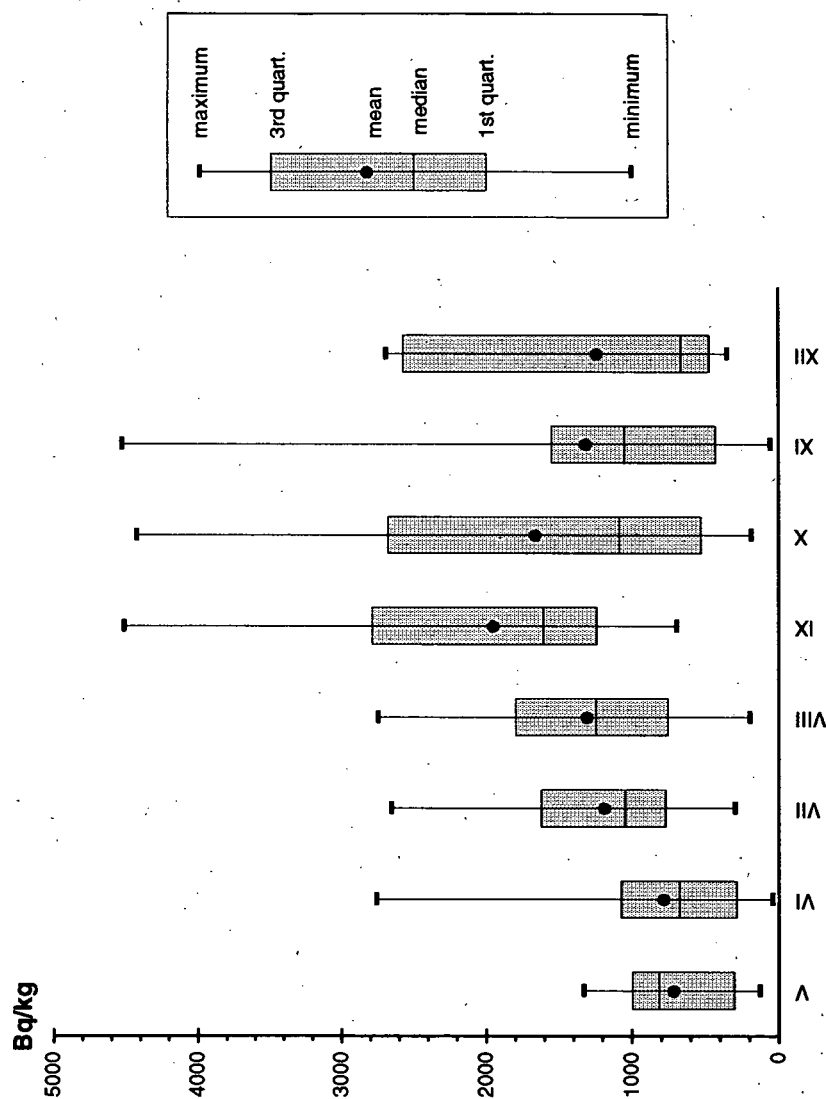


Fig. 1: Cesium-137 levels in roe deer from may to december

changing to other areas is impossible. Some of these species are known to accumulate radiocaesium to a high extent so that their ingestion contributes significantly to the animal's burden. This explains the seasonal changes of radiocaesium levels in roe deer's muscles: in spring ferns turn green again, becoming attractive as feeding plants, mushrooms start to grow etc.

As the regions of the WW and the KW were highly contaminated by direct fallout in 1986 the radiocaesium values in ferns, mushrooms and also blueberries are very high in the two study areas (Fig. 2), reaching much higher levels than in other forests (Tataruch and Schönhofner, 1993). The mobility of Cs is also enhanced by the low pH value of soil, the high organic content and the low concentrations of potassium.

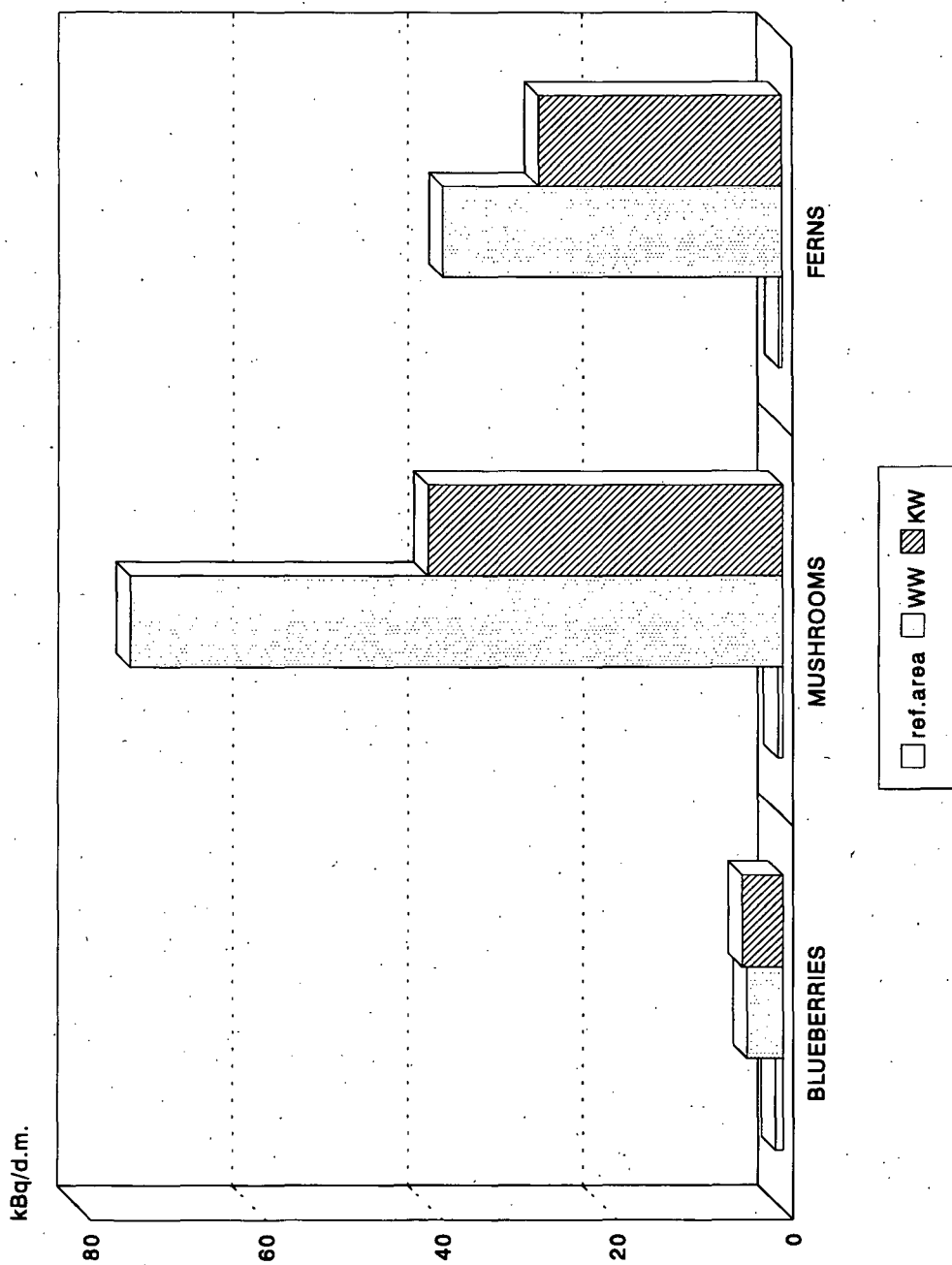
In late autumn, when ferns senesce and mushrooms stop fruiting, radiocaesium activities in roe deer decrease again. Some higher values in December can be explained by higher ingestion of blueberry stems, which during winter are the only attractive and preferred feeding plant for roe deer left in these regions. Blackberry leaves, usually a favourite feedstuff in winter, hardly exist in the study areas.

Seasonal variation in radiocaesium levels discussed above was similar during all years from 1988 to 1994. Only in the summer 1992, when very high temperatures and drought persisted, was there no increase of contamination noted, presumably because there was only a small amount of mushrooms available.

Fig. 3 shows the Cs-137 values obtained for roe deer from the Weinsberger Wald for each of the different years. The highest median was found in 1988 and the highest maximum value in August 1989. In the other study area, the Kobernauser Wald, the maximum value for roe deer was registered in October 1988. According to figure 2 there was a decrease of the contamination level from 1989 to 1992, but thereafter a slight increase took place. The minimum in 1992 is probably due to the extreme dry weather condition during summer.

3.2. Wild boar (*Sus scrofa*)

The changes with time in ^{137}Cs contamination of this species after the Chernobyl accident was even more surprising than that of roe deer. In 1986 radiocaesium levels in wild boar were remarkably lower than those in wild ruminants, even in highly contaminated areas (Tataruch et al., 1988). Surprisingly, Cs activity concentrations in wild boar rose to very high levels in winter



Cs-137 levels in feeding plants of roe deer

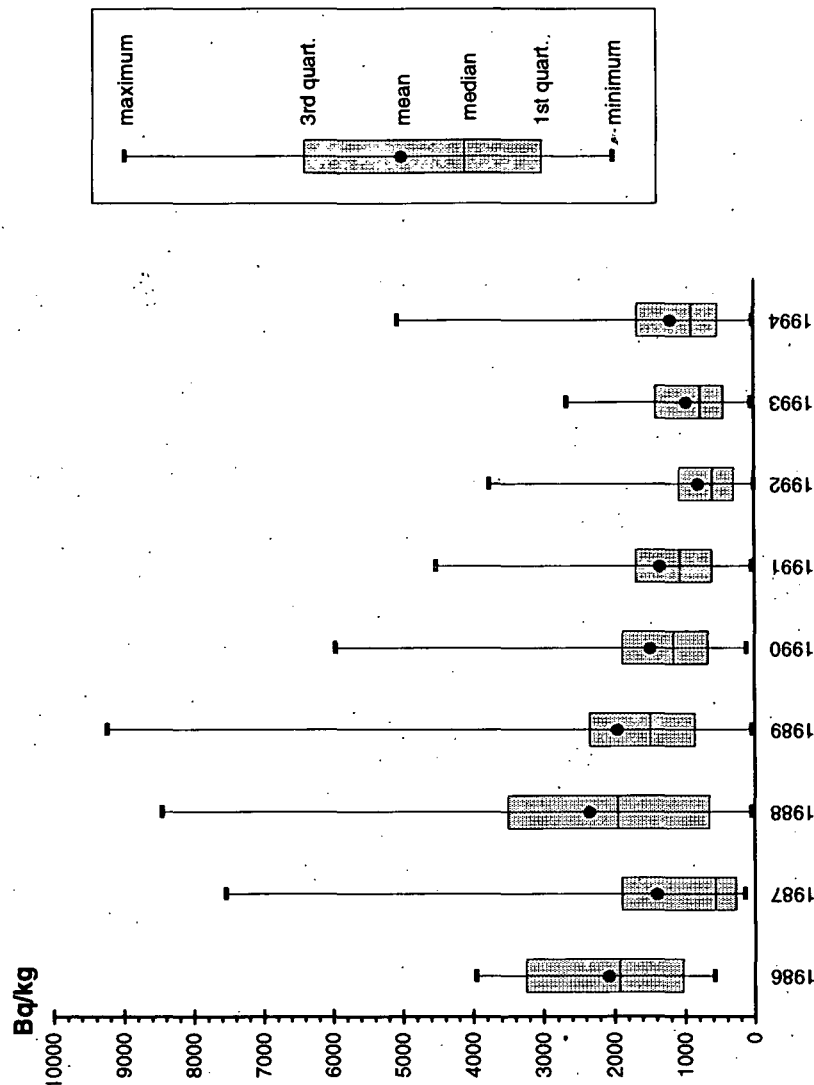


Fig. 3: Cesium-137 levels in roe deer in the period 1986 - 1994

1987/88, with a maximum value of $17583 \text{ Bq} \cdot \text{kg}^{-1} {}^{137}\text{Cs}$ and $4706 \text{ Bq} \cdot \text{kg}^{-1} {}^{134}\text{Cs}$ in February 1988, representing the currently highest level ever found in Austrian wildlife.

Table 1 shows the ${}^{137}\text{Cs}$ activity concentrations in wild boar from the 2 study areas. Unfortunately the number of samples is small, because there are few wild boars in these regions. But it can be seen that the contamination decreased - as for roe deer - from 1988 to 1991 and then started to increase again, showing comparatively high maximum values. The highest activity registered so far in 1995 so far was $8440 \text{ Bq} \cdot \text{kg}^{-1}$. In contrast to roe deer the increase from 1991/92 onward was not limited to the large, unbroken forest areas, but was also noted in several other regions.

Table 1: Cs-137 levels in wild boar from Weinsberger Wald and Kobernauber Wald (in $\text{Bq} \cdot \text{kg}^{-1}$)

	n	x	s	±s	Min.	Max.
1988	44	1105	2302	3307	41	17583
1989	15	1050	1488	1326	205	4899
1990	12	845	1399	1177	278	3916
1991	22	581	657	379	264	1709
1992	22	1007	1637	1801	265	6705
1993	21	1461	1963	1907	107	5988
1994	18	701	1203	1227	73	4252

What is the reason for the special variation of wild boar contamination? Feeding selection of the omnivorous wild boar is obviously different from that of the strictly herbivorous roe deer.

Botanical analyses of stomach contents yielded in addition to grasses, grains and fruits, large amounts of soil together with some worms, larvae, beetles etc. Different species of mushrooms were also found. High contamination of animals can only occur when either the animals' diet is highly contaminated or/and absorption of radionuclides is high. Regarding the stomach contents grass, grains and fruits can be disregarded as potential sources of large amounts of radiocaesium because of their low contamination levels. Ingestion of large amounts of contaminated soil could be one factor causing high contamination of the animals, but in our opinion the high values found

are probably not due to ingestion of highly contaminated soil present in KW and WW, though it might contribute to some extent. In the first year after the Chernobyl accident, the values observed were lower than for ruminants. If soil would be the main source of radiocaesium the animals would probably have also had high values shortly after the Chernobyl accident. This was not the case, so some processes must be responsible which occur over a number of years. We suggest that probably small organisms like worms, larvae, beetles etc. that form a part of wild boars' diet accumulate radiocaesium very strongly and are therefore the source of the high amounts of radiocaesium necessary to cause the extreme contamination of wild boar. These organisms might also be the explanation for the repeated increase of contamination levels in 1991/92.

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1. Der Walter-Kubiena-Preis bezweckt
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3. Es können nur Arbeiten von Studierenden (a) an österreichischen Universitäten, Hochschulen; b) an Höheren Lehranstalten) in unbezahlter Stellung eingereicht werden.
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Heft 44

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Heft 45

1992, 144 Seiten

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Heft 47

1993, 128 Seiten

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BERG, G., C. McCLAUHERT, A.V. de SANTO, M.B. JOHANSSON und G. EKBOHM: Decomposition of litter and soil organic matter - can we distinguish a mechanism for soil organic matter buildup?
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JOERGENSEN, R.G.: The C:N ratio of the soil microbial biomass in soils of deciduous forests.
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MERCKX, R., S. KACHAKA, M. VAN GESTEL and B. ANLAUWE: Decomposition of organic residues in soils: Litter quality and spatial distribution of decomposition products and microbial components.
SMITH, J.U.: Calculating the amount of carbon returned to the soil each year from measurement of soil organic matter.
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CHEN, P. and L. LI: Sulphur deposition distribution and sulphur balance in Sichuan Basin, China.
Kurzfassungen der Vorträge.

Heft 48/49

1994, 442 Seiten

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Abbildungen: Titel unterhalb, numeriert, z.B.: Abbildung 1: Titel der Abbildung

AUTORENINDEX

ANGUISSOLA SCOTTI, I.	163	LETTNER, H.	179, 191
BAN-NAI, T.	251	LEWYCKYJ, N.	51
BOSSEW, P.	171, 179, 187, 191, 237	LINDNER, G.	267
BULGAKOV, A.	61	LIVENS, F. R.	69
BUNZL, K.	11	LÖNSJÖ, H.	119, 129
CARINI, F.	163	MONTRUCCOLI, M.	163
CREMERS, A.	51, 85	MRAZ, G.	237
DARRAH, P. R.	35	MÜCK, K.	199
DE BROUWER, S.	145	MURAMATSU, Y.	207, 251
DELVAUX, B.	93	MURITH, C.	19
DESMET, G.	1	NAGELDINGER, G.	43
DRISSNER, J.	267	NIKOLOVA, I.	259
EIKENBERG, J.	155	OERTLI, J. J.	155
EL-FAWARIS, B. H.	275	REMELE, K.	243
ELLIES, A.	215	RIMMER, D. L.	69
ERIKSSON, Å.	101	ROSÈN, K.	101
FALKNER, T.	187	SALT, C. A.	137
FELLER, U.	155	SCHIMMACK, W.	11
FLÜGEL, V.	267	SCHÖNHOFER, F.	285
GASTBERGER, M.	191	SCHULLER, P.	215
GERZABEK, M.	77, 111, 199, 223	SHAW, G.	27
GURTNER, A.	19	SILVA, S.	163
HAAK, E.	101, 119, 129	STAUNTON, S.	35
HEINRICH, G.	243	STREBL, F.	77, 223
HENRICH, E.	187, 223	TATARUCH, F.	285
HIESEL, E.	237	THIRY, Y.	93
HIRD, A. B.	69	TSCHURLOVITS, M.	229
HUBMER, A. K.	179, 191	VALCKE, E.	85
JAMES, J. W.	137	VANDENHOVE, H.	145
JARVIS, K. E.	137	VANDECASTEELE, C. M.	51, 85, 93, 145
JOHANSON, K. J.	259, 275	VAN HEES, M.	145
KAMINSKI, S.	267	VIDAL, M.	85
KARG, V.	77	WALSER, M.	267
KIENZL, K.	187, 223	WANG, X.	27
KIRCHNER, G.	43	WENISCH, A.	237
KLANSEK, E.	285	YOSHIDA, S.	207, 251
KLEMT, E.	267	ZEHNDER, H.-J.	155
KONOPLEV, A.	61	ZIBOLD, G.	267
KROPP, P.	155		