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Commission of the European Communities

radiation protection

Evaluation of data on the transfer of radionuclides in the food chain

Post-Chernobyl action

Report

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Post-Chernobyl action

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Preface

The Chernobyl accident, which occurred on 26 April 1986, presented major challenges to the European Community with respect to the practical and regulatory aspects of radiation protection, public information, trade, particularly in food, and international politics. The Chernobyl accident was also a major challenge to the international scientific community which had to evaluate rapidly the radiological consequences of the accident and advise on the introduction of any countermeasures. Prior to the accident at Chernobyl, countermeasures to reduce the consequences of radioactive contamination had been conceived largely in the context of relatively small accidental releases and for application over relatively small areas. Less consideration had been given to the practical implications of applying such measures in case of a large source term and a spread over a very large area.

The Radiation Protection Research and Training Programme was influential in a number of important initiatives taken within the Community immediately after the accident. Information was collected by Community scientists and, from it, an assessment made within days of the possible consequences. This showed that the health impact on the population of the European Community was not expected to be significant. About four weeks after the accident, the Programme, together with the US Department of Energy, organised a meeting in Brussels during which the data on dispersion of radioactive material were discussed and evaluated. Several other meetings followed soon after on the transfer of radionuclides in the food chain and possible health effects. These meetings were carried out in close co-operation with the DG XI (Directorate General, Environment, Consumer Protection and Nuclear Safety) within the CEC, and, externally, with international organisations such as the International Atomic Energy Agency (IAEA) and the World Health Organisation (WHO). In addition, the Commission convoked a Committee of high-level independent scientists to assess the scientific evidence from current research in view of recent nuclear incidences, to consider the possible implications for the Basic Standards and emergency reference levels and to advise the Commission on future action in radiological protection including research. (EUR 11449 EN).

Soon after the accident, additional research requirements were identified by the Programme; these were mainly better methods to assess accident consequences and the further improvement of off-site accident management. Several existing contracts were reoriented and new contracts were placed; however, the financial means then available within the Programme were insufficient to fund the additional research identified as necessary. A proposal for a revision of the Programme was, therefore, elaborated in 1986. It comprised 10 specific "post-Chernobyl" research actions. This revision, with an additional budget of 10 MEcu for a period of two years, was adopted by the Council of Ministers on 21 December 1987. With the help of the Management and Coordination Advisory Committee (CGC) "Radiation Protection" a number of institutes was identified to carry out the research in a co-operative manner, and the research began in the spring of 1988.

These post-Chernobyl activities have now been completed. Detailed reports on each of these studies and an additional volume containing the executive summaries of all reports are now available.

- Evaluation of data on the transfer of radionuclides in the food chain,
- Improvement of reliable long-distance atmospheric transport models,
- Radiological aspects of nuclear accident scenarios,
  A. Real-time emergency response systems,
  B. The RADE-AID system,
- Monitoring and surveillance in accident situations,
- Underlying data for derived emergency reference levels,
- Improvement of practical countermeasures against nuclear contamination in the agricultural environment,
- Improvement of practical countermeasures against nuclear contamination in the urban environment,
- Improvement of practical countermeasures: preventive medication,
- Treatment and biological dosimetry of exposed persons,
- Feasibility of studies on health effects due to the reactor accident at Chernobyl.

The research undertaken within the "post-Chernobyl" actions has added considerably to the understanding of the basic underlying mechanisms of the transfer of radionuclides in the environment, of the treatment of accident victims and of how the environmental consequences of accidents may be mitigated. In addition, progress has been made in the setting up environmental surveillance programmes development of predictive and decision-aiding techniques, the implementation of which will lead to significant improvements in off-site accident management. Several new ideas and lines of theoretical and practical research have originated from the post-Chernobyl research and these have already been integrated into the ongoing Community Radiation Protection Research Programme. A further important feature which should not be overlooked, is the close and effective collaboration of many institutes in the research; this has markedly strengthened the ties between Community institutes and scientists. The outcome of all of this work is that the Community and all other countries are now better prepared and co-ordinated should a significant release of radioactivity ever occur again

Further research is continuing within the current Radiation Protection Research and Training Programme 1990-1991 on a number of the "post-Chernobyl" topics; these also form part of the proposal of the specific Programme on "Nuclear Fission Safety" 1992-1993, e.g. real-time emergency management systems, development of countermeasures in the agricultural environment, treatment of radiation accident victims, etc. Moreover, the Community Programme is currently making a significant contribution to an international evaluation, being undertaken by IAEA at the request of the Soviet Government, on the consequences in the USSR of the Chernobyl accident and of the measures being taken to ensure safe living conditions for the affected populations.

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VI CONCLUSIONS

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I. INTRODUCTION (KFA)

During the accident at Chernobyl large amounts of radioactive materials were released into the atmosphere and distributed all over Europe. Depending on the trajectories of the radioctive clouds and the large variation of rainfall during its passage through the countries of the European Community, large local and regional differences in deposition were observed, leading to an enhanced contamination of soil and plants. Consequently, the radionuclides entered the foodchain by several pathways resulting in an internal body exposure by gamma radiation due to ingestion of contaminated foodstuffs.

The radiological consequences of radionuclides discharged from nuclear installations usually are evaluated using doses assessment models. In these models the passage from one compartment into another, e.g. the transfer from soil into plants, from plants into milk or meat is defined by transfer factors, which are influenced by various parameters. From the longterm point of view only the long-lived nuclides like $^{134/137}$Cs are important for longterm radiation risk assessment after the Chernobyl accident.

For routine releases the transfer factors defined for the specific food compartments are more or less substantially known, but there is a need to have more reliability of predictions for accidental conditions. Due to the particular course of the Chernobyl accident the following radioactive contamination of the environment differed widely from the accident scenario usually considered in radiological assessements. In this accident the release of radioactive materials took place over a longer time period and varied in rate and radionuclide composition. Some regions of Europe were contaminated several times. It was observed, that the contamination of agricultural products depended to a high degree on season and plant development at the time of radionuclide deposition.

With respect to assessment of the radiation risk due to soil-to-plant transfer of radionuclides originating from the Chernobyl accident into the foodchain it turned out, that in calculation models used several transfer parameter were unsufficiently or not considered leading to uncertainties in prediction. Before the Chernobyl accident occured, research was mainly concentrated to agricultural food production, but there was still a lack of knowledge in local, specific conditions. Transfer factors for the pathway soil-fodder-animal products are poorly defined especially for the different ruminants and in dependence on different feed and feed practices.

Little attention was given to semi-natural and natural ecosystems, which were used extensively. But there was a number of pathways leading directly to man primarily via forest food products, meat, dairy products and possibly water supplies. These areas mainly have a poor soil quality which results in a higher soil-to-plant transfer. For the aquatic pathways of radionuclides it is also necessary to have predictive models in the case of an event like the Chernobyl accident.

To improve radiation dose prediction the CEC has initiated the Post-Chernobyl radiation protection programme "Evaluation of Data on the Transfer of Radionuclides in the Foodchain" including five main items to be studied by different laboratories:

1. Impact of chemical speciation on the radionuclide transfer in terrestrial ecosystems after a core disruptive accident, especially in soils and plants.
2. Transfer paths of radionuclides in seminatural and natural ecosystems and their role in contaminating the foodchain.
3. Validation of soil-to-plant parameters.
4. Transfer of radionuclides to animals and animal products.
5. Transfer paths in aquatic systems and their importance for the contamination of the foodchain.

II. CHEMICAL SPECIATION OF RADIONUCLIDES (CEA)

1. INTRODUCTION

Following the Chernobyl accident and the fallout observed in Europe, a pertinent question was whether the radioactive substances released under very special conditions were present in the physicochemical forms in which they are generally observed. If the forms were different, to what extent did this affect their migration in the soil and in the food chains? Would existing transfer models have to be revised?

Work was undertaken to characterize the physicochemical forms of radioactive pollutants released in the event of serious accidents. The programme included laboratory simulations of a reactor core meltdown accident to reproduce a "source term" that can later be used in experimental contamination tests under controlled laboratory conditions.
The subject was approached from the standpoints : theoretical studies, experiments desiggned to reproduce accident situations, and observations following actual accidents.

2. EQUIPEMENT, METHODS AND SOURCE COMPOSITION

2.1. Equipement Summary

Three types of experimental equipement were available :

- the POLYR mockup at Cadarache
- a modified atomic absorption atomizer at the Catholic University of Louvain

- a moist aerosol generator (designed for medical use) that disperses an aqueous solution of radionuclides in the form of fine droplets at room temperature; although mentioned here as a possibility, this type of device was not tested since it does not realistically simulate a nuclear power reactor core meltdown situation.

2.2 THE POLYR MOCKUP

The mockup is shown schematically in Figure 1, and in a photo in Appendix 2. Up to 25 g of materials to be subjected to meltdown were placed in a graphite crucible with a 15 kW high-frequency induction heating system. The crucible was swept externally with argon during each experiment to prevent premature damage to the graphite.

A 1.8 m^3 tapered cylindrical Terphane vessel located above the furnace was inflated with air and water vapor heated to 60°C during the experiment. The aerosol residence time in the vessel was 15 minutes.

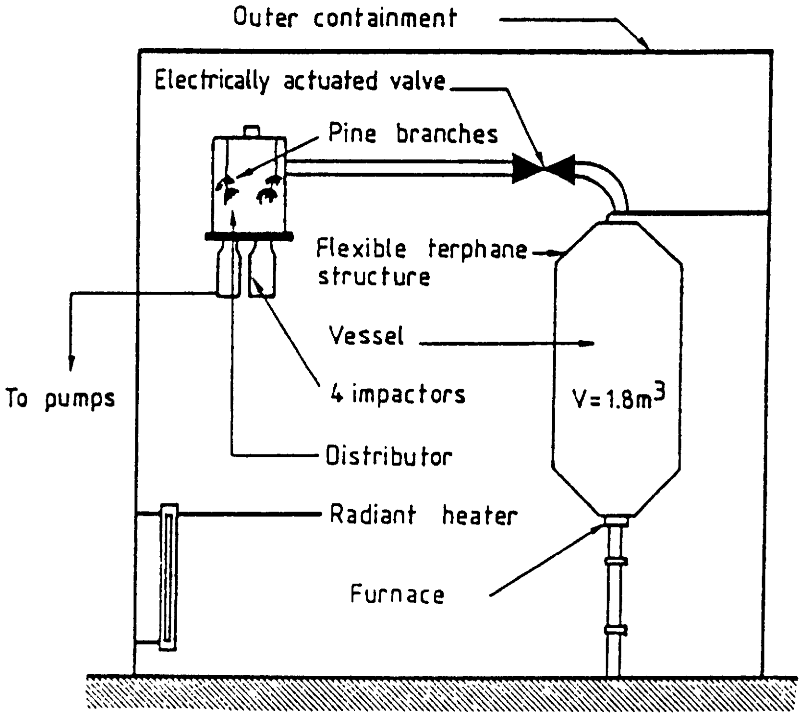


Figure 1: Overall Schematic Diagram of the POLYR Mockup

2.3 MODIFIED ATOMIC ABSORPTION ATOMIZER

Dry aerosol generation experiments were conducted with the collaboration of Professor Ronneau using a mockup based on a modified absorption furnace at the Catholic University of Louvain (UCL) in Belgium.

Aerosols were emitted from a graphite tube heated by Joule effect. The gas circulation in the chamber was modified so the contents were swept outward by a stream of argon. The chamber was cooled by a peripheral water system. Heating power was regulated by a programmable controller capable of implementing several temperature rise curves to provide greater flexibility in operation.

After emission the particles were allowed to coalesce and mature in a vinyl tent before sampling.

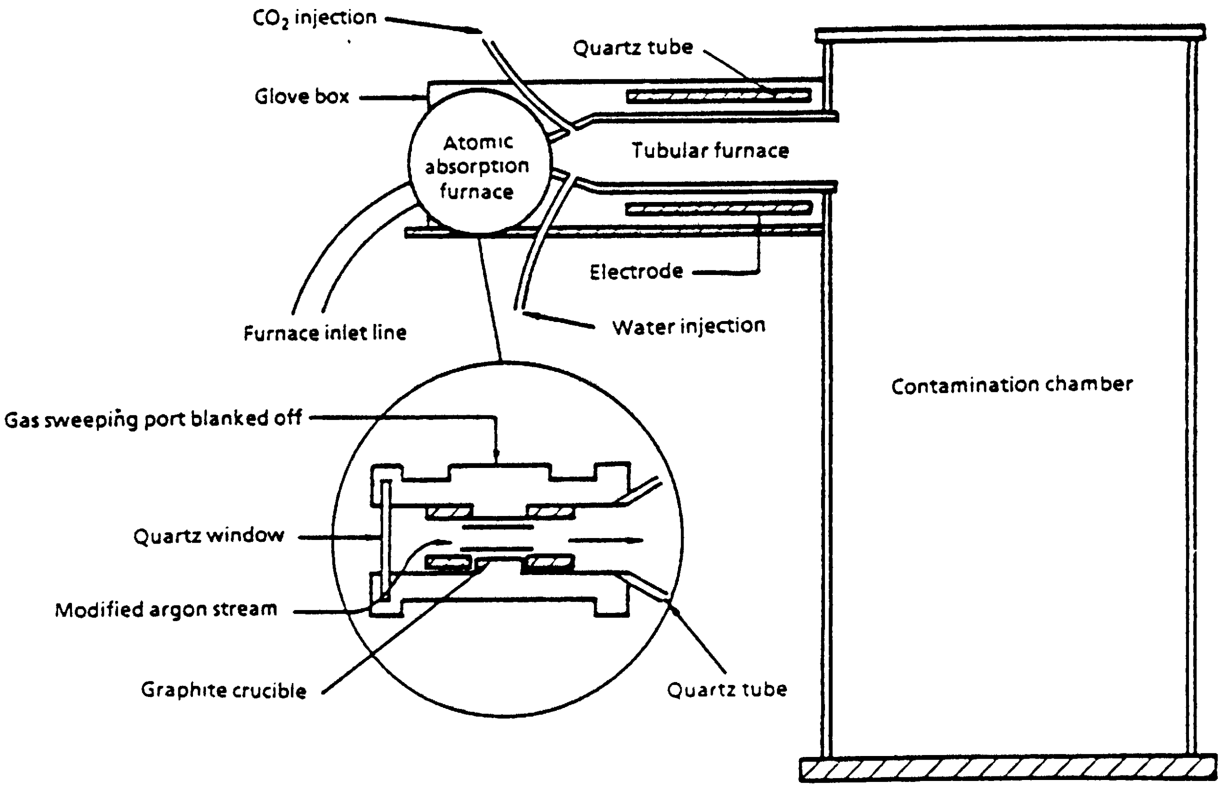


Figure 2: Schematic View of an Aerosol Generator Using a Modified Atomic Absorption Furnace

2.4 SAMPLING AND ANALYSIS METHODS

After emission and aging in the vessel, the aerosols were sampled using Andersen Mk II 8-stage cascade impactors with particle size classes ranging from 9 μm to 0.4 μm. Teflon or cellulose acetate filters were also used to obtain deposits for electron microscope observation.

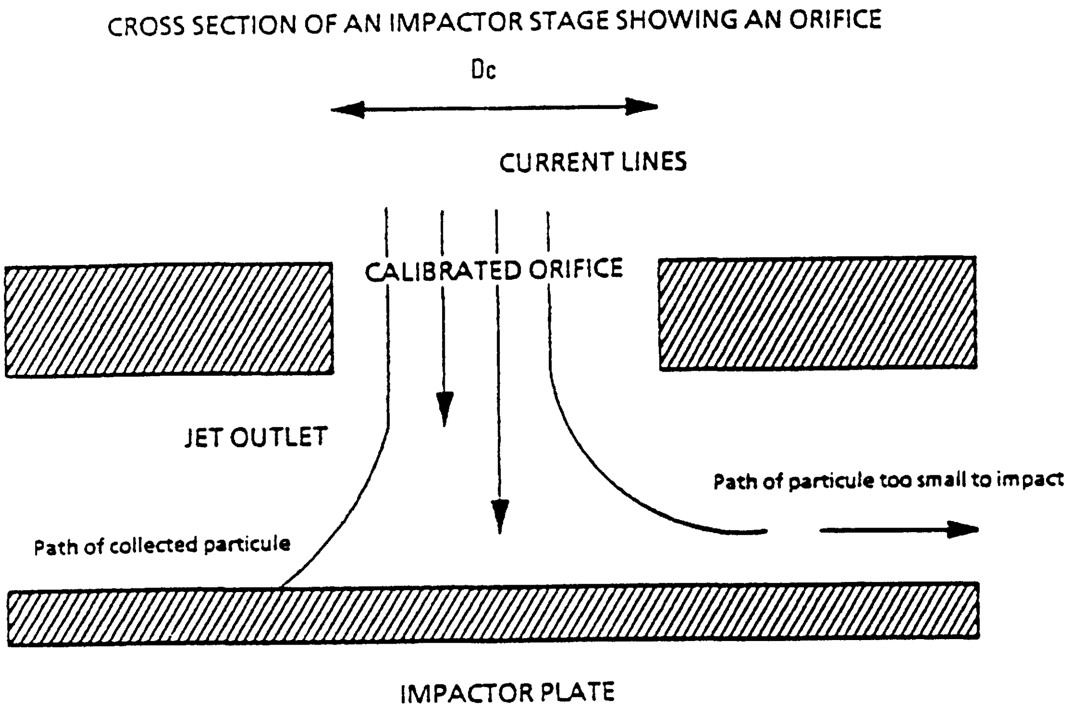


Figure 3: Cross Section of an Impactor Stage Showing an Orifice

The aerosol-laden impactor plates were then analyzed as follows:

- Particle Granulometry:

The aerosol mass on each plate was weighed to within 0.1 mg on a precision balance. Based on the mass, temperature, vector gas composition and flow rate, a computer program then determined the granular distribution of the particles in the aerosol cloud and fit them to a log-normal curve on request.

- Overall Particle Chemical Composition:

The aerosols were analyzed by Inductively-Coupled Plasma Mass Spectrometry (ICP-MS) to obtain two types of results: semiquantitative determinations identifying virtually all the elements present in the particles, and quantitative determinations of selected elements (those of the accident source term in this case). This method indicates the particle mass composition, but not the chemical form in which the elements are found.

- Identification of Chemical Species:

Considering the very minute quantities collected, the chemical species could only be determined by X-ray diffraction (using a Debye-Scherrer chamber). This only provided information on the crystallized phases, which did not predominate in the particles generated in these experiments.

- Particle Observation:

Particles were observed by electron microscopy using backscattering probes to obtain qualitative data on particle composition in a short time. Optical microscopic observations of the analyzed surface also revealed the degree of homogeneity of a given aerosol grain sample.

- Aerosol Water-Solubility:

The first experimental cycle principally involved nonradioactive nuclides, making it difficult to assess the solubility of trace elements. The solubility was determined, however, for ten elements including cesium, strontium, silver and ruthenium. Two methods were used: the aerosols were rinsed in water and selected elements were chemically determined by flame absorption; radioactive counts were also obtained for the rinsing solution of aerosols activated by irradiation.

2.5 SOURCE COMPOSITION: INITIAL MIXTURES

Powder mixtures were prepared for testing in the aerosol generators. The mixtures had to be representative of the composition of a molten PWR core: the elements selected were basically those found in the fuel and cladding tubes, control rods and stainless steel structures, and fission products liable to constitute health hazards. Sixteen elements were selected on the basis of studies conducted at the Fontenay-aux-Roses Nuclear Research Center [3]. The amounts present in a 900 MWe PWR core and the calculated amounts in the aerosols emitted inside the containment are shown in Table 1.

900 MWe PWR Inventory reduced to 16 materials				Emitted fraction	Aerosols emitted in vessel	
Elements	Weight (kg)	Abundance Type A (%)	Abundance Type B (%)		Weight (kg)	Relative abundance (%)
UO_2	80000	60.2	6.1	0.001	80	8.2(1)
Fe	20600	15.5	32	0.001	21	2.5
Zr	18326	13.8	28	0.001	18	2.1
Cr	6200	4.7	10	0.001	6	0.7
Ni	4100	3.1	6.3	0.001	4	0.5
Ag	1903	1.4	2.9	0.1	190	22.2
In	360	0.3	0.55	0.1	36	4.2
Sn	202	0.16	0.31	1	202	23.6
Cd	124	0.09	0.19	0.5	62	7.3
I	12	0.01	0.18	1	12	1.4
Cs	148	0.11	2.3	1	148	17.3
Te	26	0.02	0.4	1	26	3
Sr	238	0.18	3.6	0.1	24	2.8
Ba	69	0.05	1.1	0.1	7	0.8
Ru	192	0.14	3	0.1	19	2.2
Ce	498	0.37	0.76	0.001	1	0.1
Total	133000	100	100		856	98.9

Table 1: Theoretical Aerosol Chemical Composition

---

(1) U only

For practical reasons, all the elements could not be introduced into the mixture in pure form; some were added as compounds. Two different mixtures were tested (Table 1 and Figure 4).

2.5.1 Initial Chemical Forms

The experiment simulated the destruction of the cladding and steel structures. Zirconium found in the clads was supplied in amorphous elemental form: Zr is released above 1800°C (zirconium oxide ZrO_2 or zircon is a refractory compound with a melting point of about 4000°C). Stainless steel components (iron, chromium, nickel, etc.) were included in the initial mixture in crystallized metallic form.

Fission products are produced during normal reactor operation. In order to enhance their emission in aerosols, they were included in elemental form except for cesium, strontium and barium, which were added as carbonates (Cs_2CO_3, $SrCO_3$ and $BaCO_3$) and cerium, introduced as CeO_2.

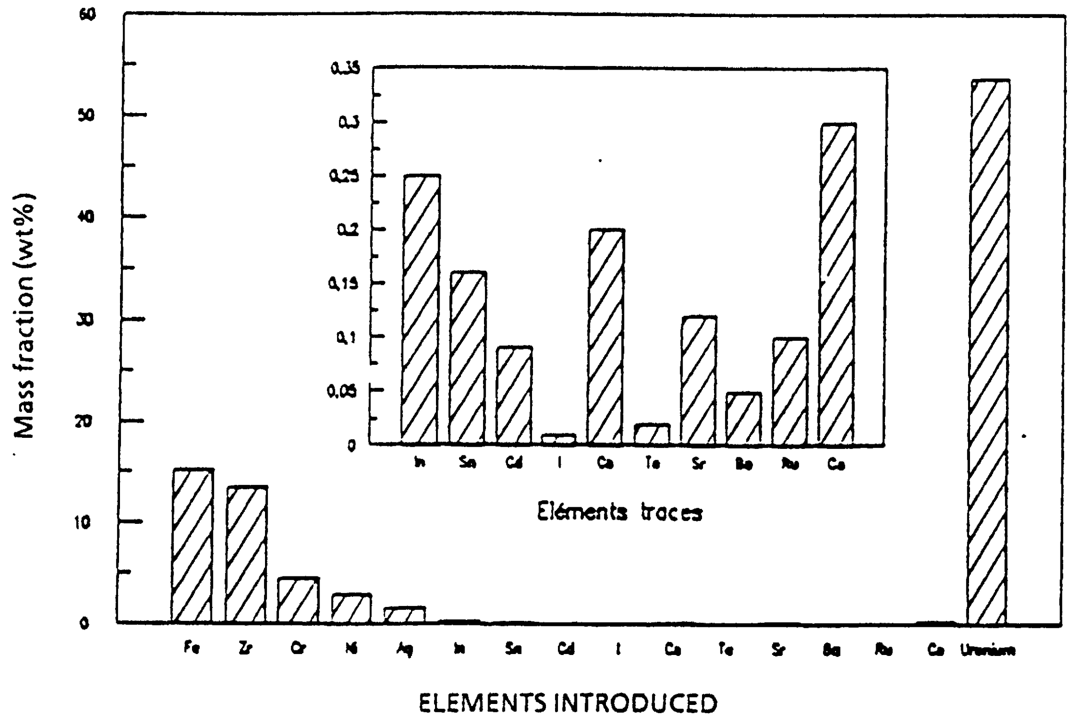
The fuel was taken into account by adding natural ^{238}U as uranium oxide (UO_2). With a melting point of about 2850°C, uranium is theoretically emitted only in very small quantities [2].

2.5.2 Mixture A (60% UO_2)

Mixture A corresponds precisely to the assumed element proportions in the core, hence the high uranium percentage.

Chemical analysis of the aerosols produced with a Type A mixture were found to deviate substantially from the theoretical composition determined by the DAS analysis, notably with a cesium deficit (melting point: 690°C) [2]. A second mixture was therefore prepared to provide more representative results.

CHEMICAL COMPOSITION OF MIXTURE A (60% UO_2)
DETERMINATED BY ICP-MS ANALYSIS



CHEMICAL COMPOSITION OF MIXTURE B (6% UO_2)
DETERMINATED BY ICP-MS ANALYSIS

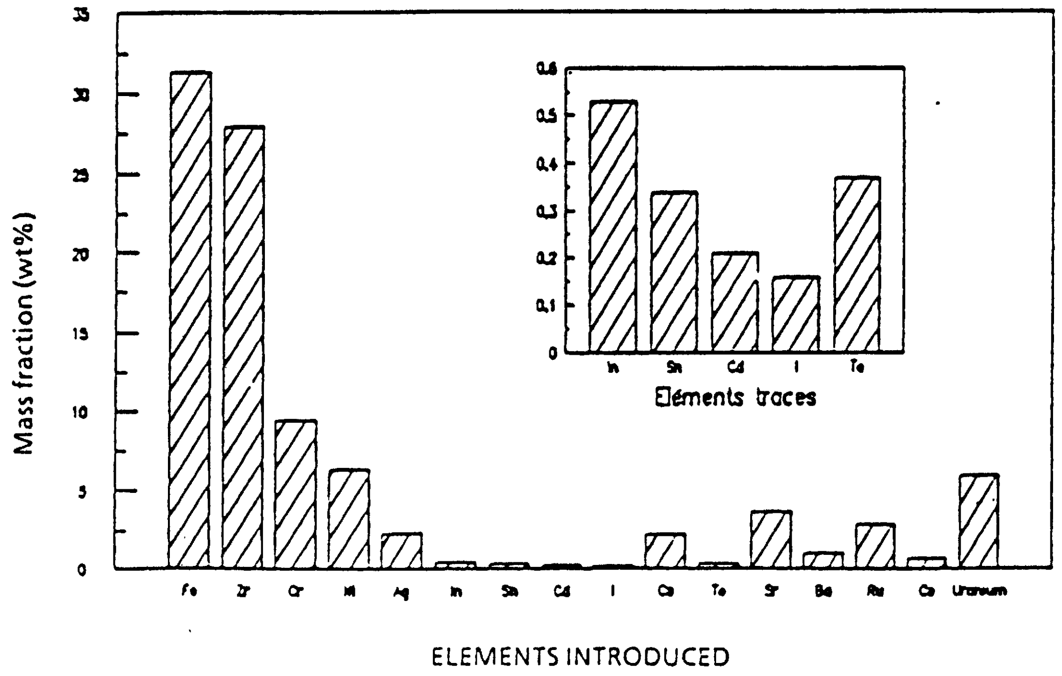


Figure 4: Composition of Mixtures A and B

2.5.3 Mixture B (6% UO_2)

Mixture B was determined with allowance for the low uranium emission. In order to favor the release of other elements, the UO_2 content was reduced to 6 wt%. The proportion of fission products with radioecological impact was increased to enhance their emission. The remainder of the weight gain corresponding the reduced uranium content was evenly divided among the other elements.

2.6 EXPERIMENTAL PROCEDURE

2.6.1 POLYR Mockup Experiments

The powder sample was placed in the crucible without further treatment.

The crucible temperature pattern depended on the selected accident scenario [4]. At any given time, every attempt was made to ensure that the crucible temperature (especially in the range above 2000°C) was as close as possible to the theoretical value.

Figure 5 compares the theoretical curve with the experimental results obtained in POLYR on Jan 18, 1989. The diagram can be divided into three sections:

- A rapid rise from 0 to 1500°C

The most volatile compounds, mainly fission products, were released during this stage. Iodine, with a vaporization point of 66°C [2] was emitted first, followed by cesium, tellurium, etc.

The temperature response of the induction-heated furnace was very fast during this phase: the actual temperature values exceeded the theoretical values. Although this did not affect the accident scenario, it required a short holding time below 1500°C to allow the theoretical curve to "catch up" before the next step.

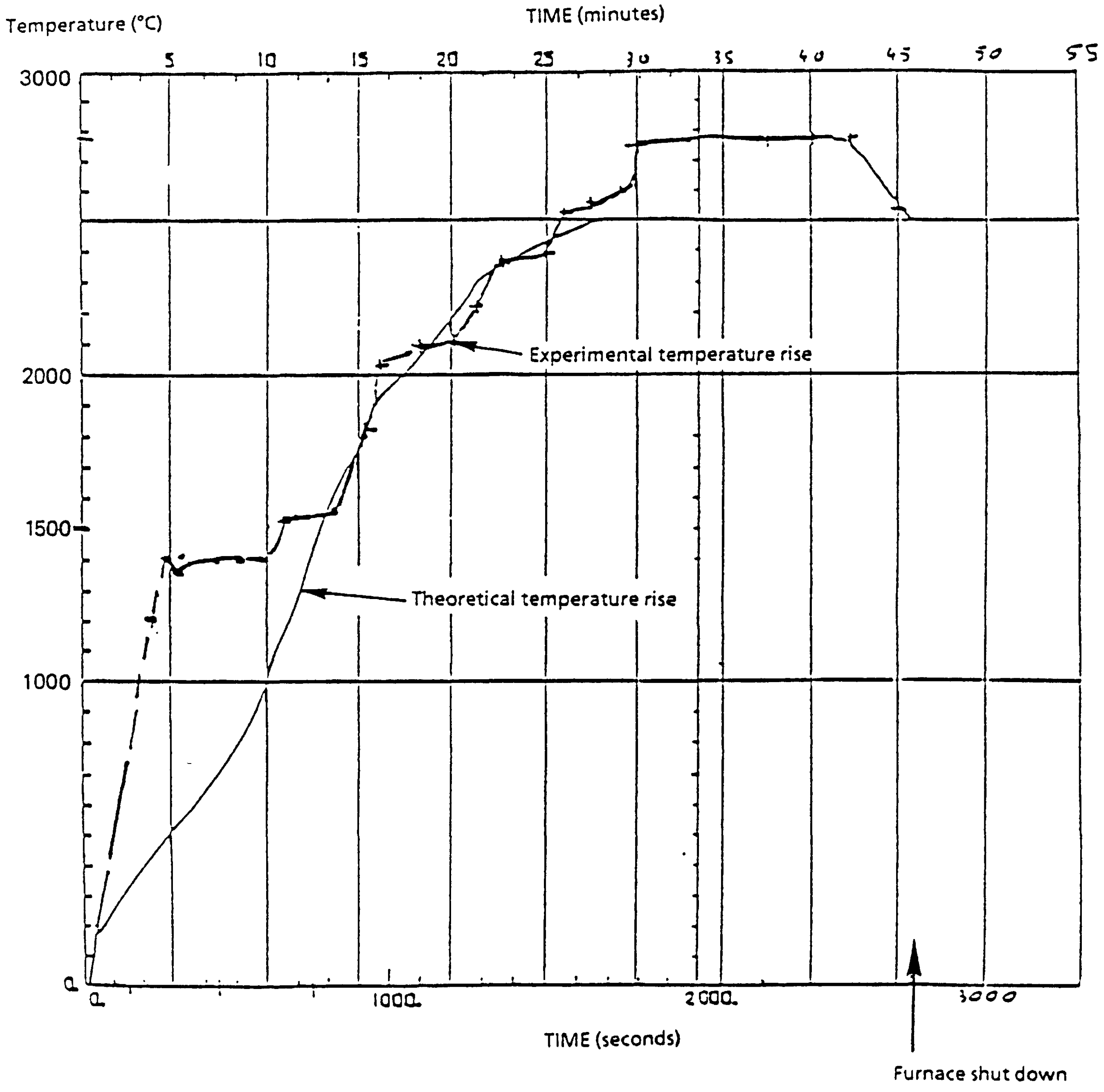


Figure 5: Temperature Rise Diagram (POLYR test on Jan 18, 1989)

- A gradual rise from 1500 to 2800°C

Most of the aerosol emission occurred in this portion of the temperature diagram. The temperature rose quickly at first, then more slowly, bracketing the theoretical curve with a stairstep temperature rise pattern (the manual power adjustment provision used to regulate the furnace temperature control did not allow a smooth, continuous temperature rise in the crucible).

- A variable residence time

While the first two steps were carefully timed (30 minutes in all) the residence time ccould be modified as necessary. Emission continued, but at a much lower rate during this phase. Most of the phenomena (coalescence and maturing by particle collisions) occurred in the Terphane vessel filled with steam and metallic aerosols. Aerosol sampling began about 15 minutes after the heater was switched off.

2.6.2 Experiments Conducted at Louvain

The powder samples were sintered at room temperature to form solid pellets 7.5 mm in diameter weighing about 300 g, which were then inserted into the atomizer tube.

The temperature was raised quickly, with a 45-second holding time at 400°C and a second 20-second interval at 2200°C. This operation was repeated several times to ensure that all the material sufficiently volatile to be emitted was in fact released. Several pellets could be heated together. Sampling with an Andersen Mk II impactor began after a 15-minute aerosol residence time in the sampling vessel.

3 RESULTS

3.1 PHYSICAL AND MORPHOLOGICAL CHARACTERISTICS OF PARTICLES: QUALITATIVE ASSESSMENT

3.1.1 Granular Distribution

The granular distribution of the particles generated is shown in Figure 6. Similar results were obtained in both test facilities (POLYR and UCL) and with both mixtures (Type A or B).

The d_{25}-d_{75} diameter range for the POLYR experiments was between 0.5 μm and 1.5 μm in most cases, with a mean mass diameter (d_{50}) of about 1 μm. These values remained relatively stable throughout the experiments, and were consistent with the calculated predictions, with post-accident observations and with figures cited in the literature [5].

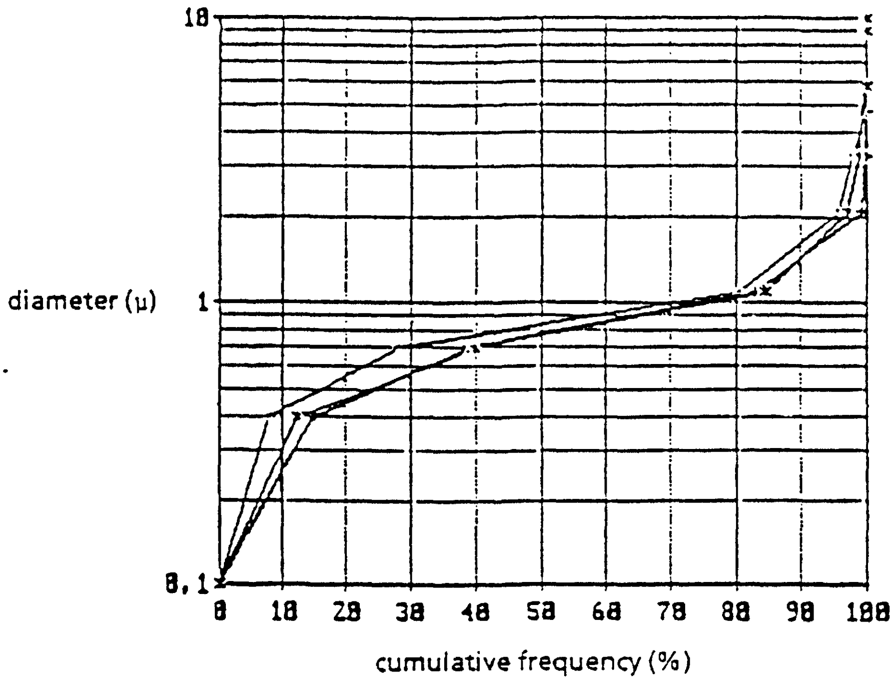
The Henry curve fitting method [6] generally indicated a log-normal distribution, except for a few instances where the graphs showed two straight-line segments: the evidently bimodal distribution in these cases was attributed either to improper sampling or, more probably, to different emission conditions depending on different high-temperature residence or aging periods in the Terphane vessel.

Date	Type of Mixture	Median Mass Diameter (μm)	Standard Deviation (%)
Jan 24, 1989	A	1.66	3.61
Jan 24, 1989	B	1.27	1.77

Table 2: Median aerosol mass diameters generated by UCL

Computer analysis of the impactor mass data provided more detailed results. The TDIMP program allows for both temperature and vapor pressure during sampling, as well as for the impactor plate response curve. Background interference can also be taken into account when granular and chemical analysis are combined. This

PRELIMINARY POLYR TESTS (JAN 18, 1989)
GRANULAR DISTRIBUTION



TESTS AT LOUVAIN (JAN 24, 1989)
GRANULAR DISTRIBUTION

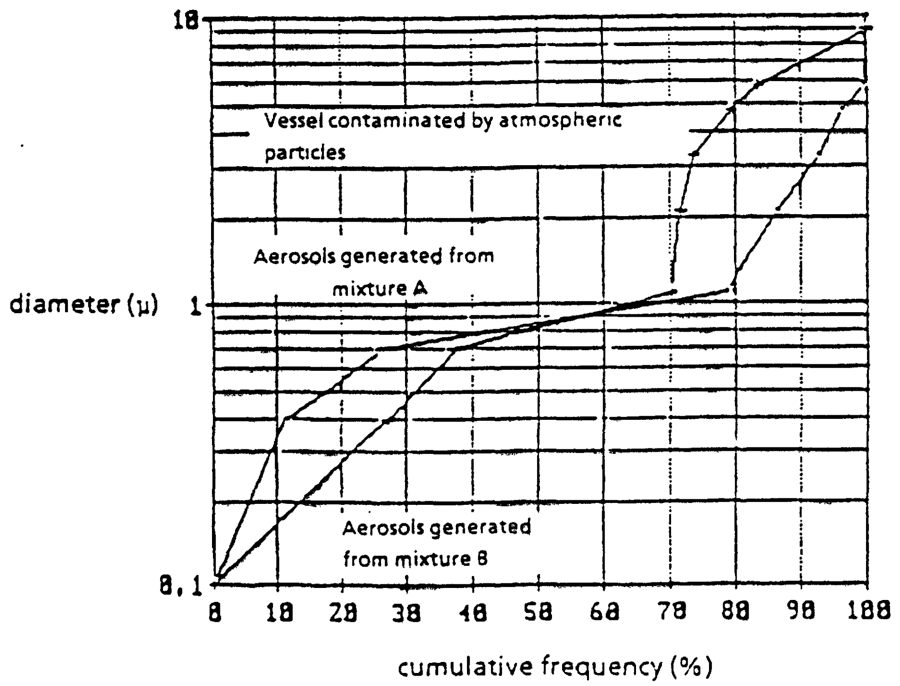


Figure 6: Aerosol Particle Granular Distributions

method produced histograms corrected for mass distribution, median aerodynamic diameter and geometric standard deviation. The relative measurement error was estimated at 15%.

Date	Type of Mixture	Median Mass Diameter (μm)	Standard Deviation (%)
Oct 25, 1988	A	2.23	2.53
Dec 15, 1988	B	1.33	1.69
Jan 18, 1989	A	1.09	1.57
Jan 20, 1989	A	1.08	1.79
Jan 24, 1989	A	0.743	2.12
Jan 27, 1989	B	1.47	1.73
Feb 01, 1989	B	1.47	1.87

Table 3: Median aerosol mass diameters generated in POLYR

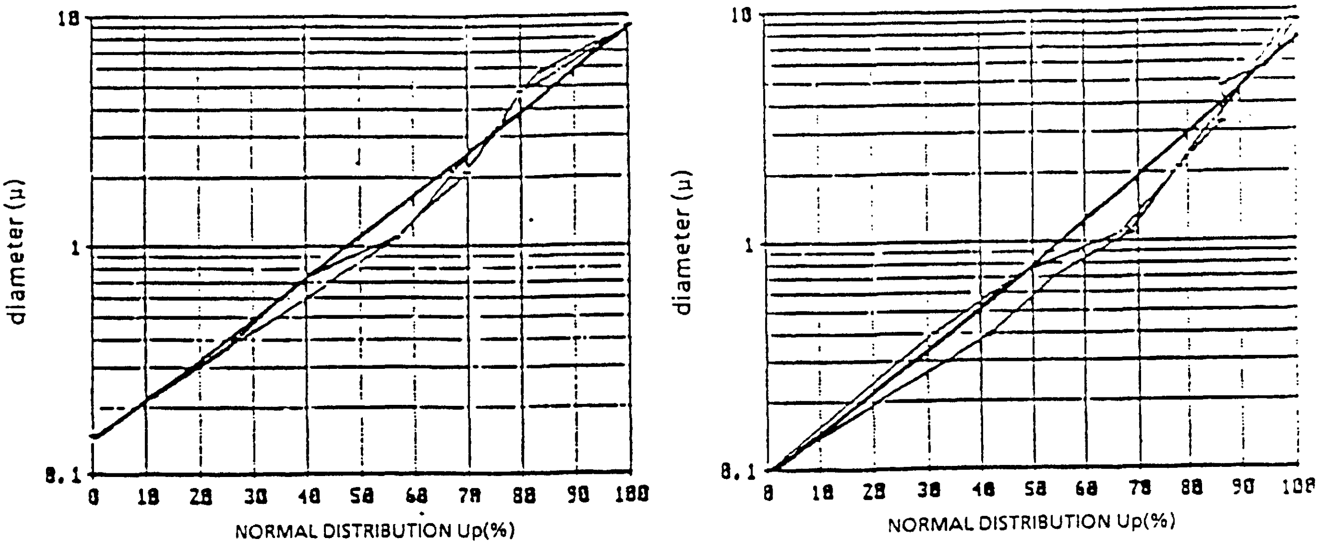
The computed results confirmed the graphic plots. For a bimodal distribution, the corrected histogram calculated over 109 bands clearly revealed two peaks. This phenomenon was even more evident when the impactor plate response curves were considered. The POLYR test on Oct 25, 1988, and the UCL test with the Type A mixture are good examples of a bimodal distribution.

The correction factors used by the program suggest that a simple statistical interpretation underestimates the emitted particle diameter, since the impactor response cannot be taken into account.

The geometric median diameter could not be estimated, since the suspended mass concentration could not be reliably determined under the experimental conditions, although the program did provide an approximation. This could be a significant parameter, since it can be used to determine the precise aerosol density involved in fallout.

3.1.2 Particle Morphology

Scanning electron micrographs are included in Appendix 2.



BIMODAL DISTRIBUTION (OCT 25,1988)

FITTED TO LOG-NORMAL DISTRIBUTION

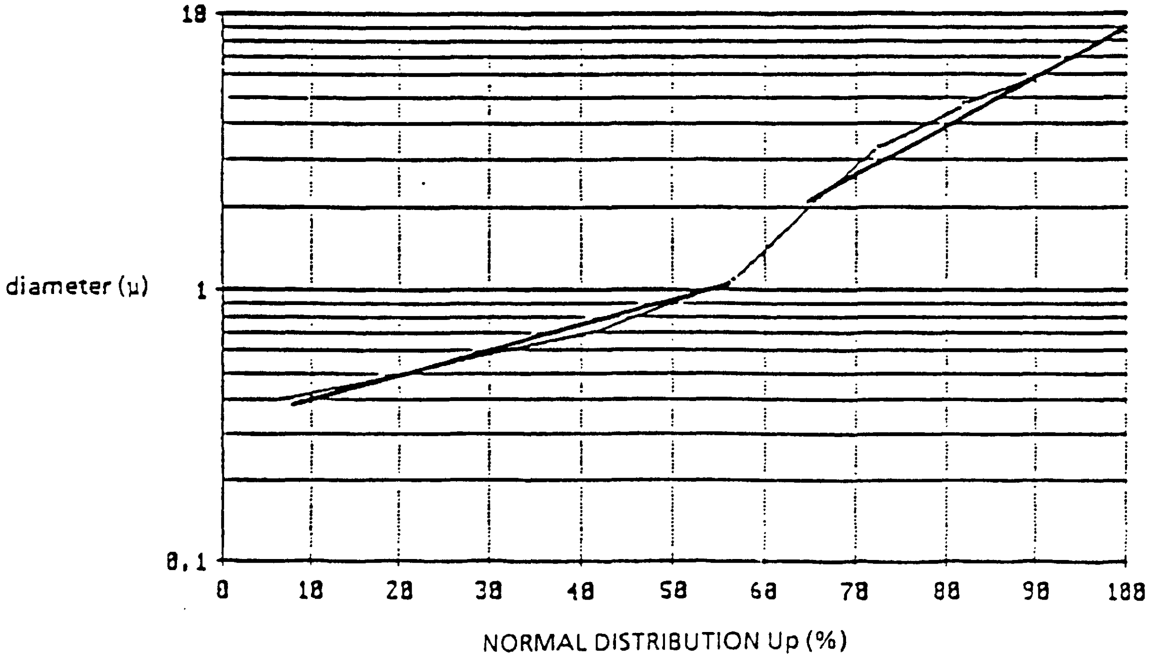


Figure 7: Log-Normal Curve Fitting

The overall appearance of the particles was remarkably consistent, and appeared not to depend on the mixture composition or on the aging time. The shape was probably related to the temperature conditions in the crucible. The particles were perfectly comparable to those found in fallout from the Chernobyl accident [7, 8, 9].

The aerosols were very heterogeneous. Examining the image field at relatively low magnification (x1000) revealed large particles (about 5 μm) and signs of other shinier, and therefore conducting, particles a few tenths of a micron in diameter.

At higher magnification (x5000) the large particles were found to comprise a compact nucleus, dense to the electron beam and thus less conducting. The surface was marked by roughly spherical protuberances that appeared smoother than the nucleus.

Observation of the small particles at high magnification revealed nothing concerning their structure; visually, they resembled the protuberances found on the large particles.

Plant specimens were impacted directly during the first test on Oct 25, 1989, in preliminary work for the experiments on contamination of crops by radioactive aerosols. Four samples (two each of Aleppo pine and Norway pine) were observed. Aerosols were found on all organs: buds, bark and needles. The particles appeared to attach themselves preferentially to the needles, with a large specific area. The size of the aerosol particles indicates that transfer to aerial organs via the cuticle would only be possible after dissolution, although a fine particle was observed trapped in a stoma (cf photo in Appendix 2).

The extent of fixation on leaves depended on their surface morphology and physiology. High humidity in the air caused the cuticle to swell, enhancing leaf absorption.

When the filters were rinsed with deionized water, the surface protuberances were much less numerous. The aerosol particle structure can be explained as follows: during the aging period, a central unit is formed by coalescence of very fine particles; this nucleus then captures other immature particles which form protuberances, which are bonded to the nucleus either by simple electrostatic forces or chemically by water-soluble compounds.

3.1.3 Preliminary Qualitative Data on Particle Chemical Composition

A rough idea of the chemical composition of the particles can be derived from backscattering electron microprobe analysis results obtained during electron microscope observations.

At relative low magnification (x750) the particle elemental composition was virtually constant regardless of the surface scanned by the electron beam: the height of the identified element peaks remained constant. Elements identified in this way included iron, silver, strontium, cesium and possibly manganese (cf spectra in Appendix 3).

Observation of the small 1 μm spherical nodules at high magnification (x9000) revealed significant heterogeneity: Fe and Ni predominated in some cases, Ag or Zr in others. Some crystallized one- or two-element particles were observed, including cesium (cesium iodide CsI, cesium hydroxide CsOH), silver (metallic or silver iodide AgI).

3.2 QUANTITATIVE RESULTS: IDENTIFIED CHEMICAL FORMS

3.2.1 Overall Chemical Composition

The overall particle composition as determined by ICP mass spectrometry is indicated below for the Type A and B mixtures tested in the POLYR mockup and for the aerosols produced at Louvain.

3.2.1.1 POLYR Test Results with Type A Mixture

The elemental percentage composition of the particles is shown in Table 4 and Figure 8.

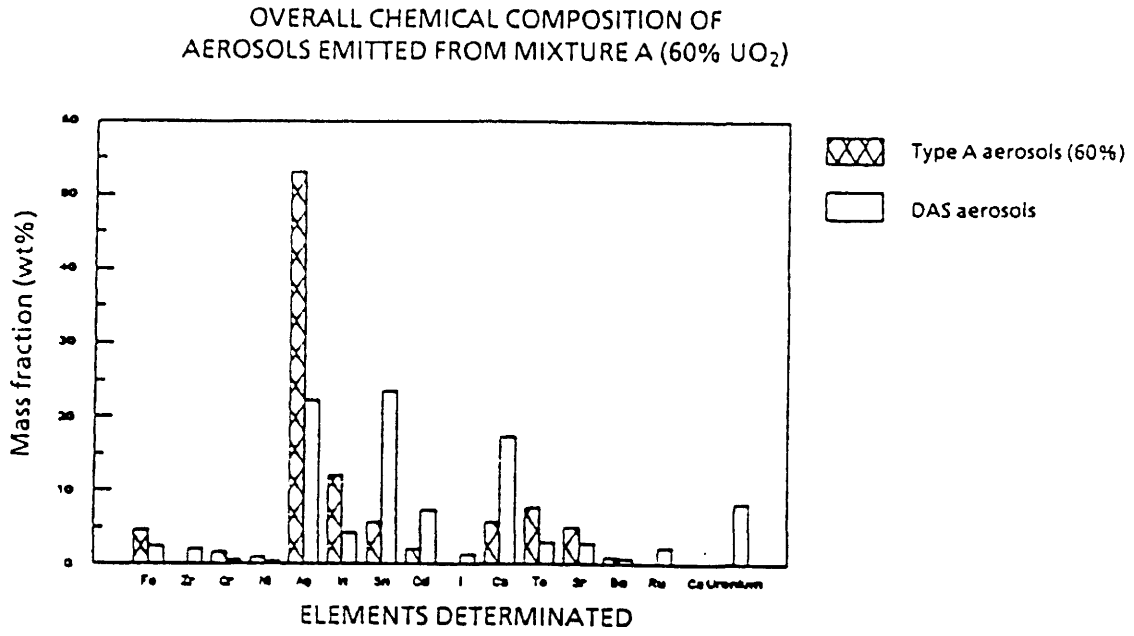


Figure 8: Relative Elemental Abundance (%) in Aerosols Generated from Type A Mixture in POLYR Mockup

Test	Oct 25, 1988	Jan 1, 1989	Jan 20, 1989	Theoretical percentage
U	1.1	0.18	0.2	8.2
Fe	7.1	5.0	16	2.5
Zr	0.88	0.04	0.02	2.1
Cr	2.5	1.8	5.7	0.7
Ni	1.4	0.95	1.9	0.5
Ag	25	57	42	22.2
In	21	11.3	14	4.2
Sn	1.6	6.2	5.8	23.6
Cd	11	2.2	2.3	7.3
I	0.03	0.01	0.31	1.4
Cs	66	18	19	17.3
Te	1.2	0.84	0.65	3.0
Sr	4.1	5.3	4.2	2.8
Ba	0.88	1.1	0.97	0.8
Ru	< 0.01	0.01	0.01	2.2
Ce	0.07	0.06	0.03	1.1
Max temp (°C)	2300	2700	2800	

Table 4: Relative Elemental Abundance (%) in Aerosols Generated from Type A Mixture in POLYR Mockup

The first experiments were conducted in a preliminary version of the test facility, resulting in contamination by elements such as silicon from the furnace heat shields or copper from the inductor. The setup was modified to eliminate this problem.

The following remarks can be made concerning only the 16 species initially placed in the crucible:

- All the elements, including uranium, were emitted.
- The results corresponded to the theoretical mass fractions [4] within a factor of 2 or 3, with a pronounced deficiency for five elements: uranium, iodine, ruthenium, tin and zirconium. The deficits can be accounted for as follows:

Uranium oxide present in the initial mixture has a melting point of 2850°C, at the upper limit of the furnace operating range. Considering that uranium accounted for 60% of the initial load, it is unlikely that higher emitted fractions will be obtained in the future.

Iodine is highly volatile, with a melting point of 66°C, and the iodine mass fraction in the crucible was very small. Gaseous iodine I_2 would no doubt be vaporized before all the other elements, forming few chemical compounds with the other 15 elements initially present.

Ruthenium was introduced as metallic powder. Its low emissivity can be attributed to its very high melting point (2460°C) and to its highly electrostatic properties: part of the ruthenium was probably lost in transferring the mixture to the crucible. Ruthenium is therefore now introduced in a more volatile form as RuO_2.

Tin boils at 2270°C, and probably escapes from the mixture at a relatively early stage before it can amalgamate with the particles.

Zirconium has a melting point of 1852°C and a boiling point of 4377°C. Its emissivity is very low, and the resulting aerosols contained less than predicted by theory. No improvement in emissivity was obtained by replacing zirconium with zircaloy.

Other elements predominated in the emitted particles:

Silver was by far the most common element, accounting for up to 42% of the total aerosol mass.

Indium was found in a relatively constant amount (10 wt%).

Iron was also found in unexpected amounts. This may be of particular importance, since iron could form complex molecules capable of trapping fission products [10].

Finally, very uniform results were noted for any given initial load with a constant silver-indium facies. Particles in which these elements predominate would have very slow dissolution kinetics in water [ ].

3.2.1.2 POLYR Test Results with Type B Mixture

The elemental percentage composition of the particles is shown in Table 5. As with the Type A mixture, uranium ruthenium, tin and zirconium were found in smaller amounts than expected, although the iodine content was not far from the predicted value.

The particles showed a general silver-indium facies, but contained satisfactory amounts of fission products with radioecological interest. The composition of this mixture is certainly suitable for future experimental contamination experiments.

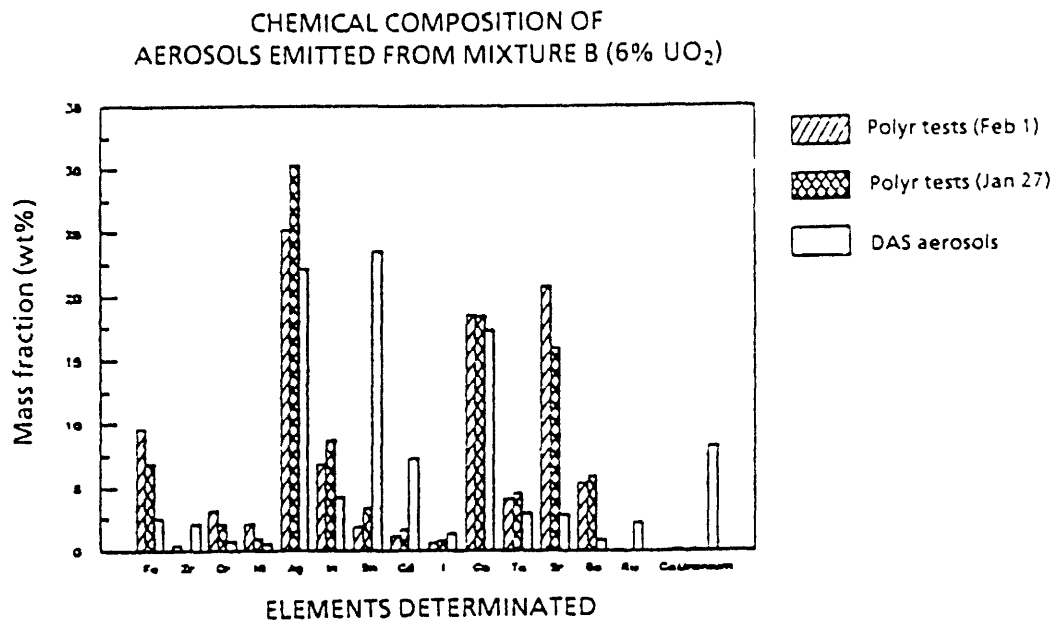


Figure 9: Relative Elemental Abundance (%) in Aerosols Generated from Type B Mixture in POLYR Mockup

Test	Dec 12, 1988	Jan 27, 1989	Feb 1, 1989	Theoretical percentage
U	< 0.01	0.02	0.06	8.2
Fe	0.8	6.9	10	2.5
Zr	0.1	0.02	0.39	2.1
Cr	0.13	2.1	3.1	0.7
Ni	0.45	0.94	2.1	0.5
Ag	14	30	25	22.2
In	5.6	8.8	7	4.2
Sn	0.25	3.3	1.9	23.6
Cd	5.2	1.7	1.2	7.3
I	1.4	0.81	0.6	1.4
Cs	66	18	19	17.3
Te	3.5	4.5	4.1	3.0
Sr	2.2	16	21	2.8
Ba	0.63	5.9	5.3	0.8
Ru	< 0.01	0.04	0.04	2.2
Ce	< 0.01	0.01	0.02	1.1
Max temp (°C)	2700	2800	2850	

Table 5: Relative Elemental Abundance (%) in Aerosols Generated from Type B Mixture in POLYR Mockup

3.2.1.3 Modified Atomic Absorption Atomizer Results at Louvain

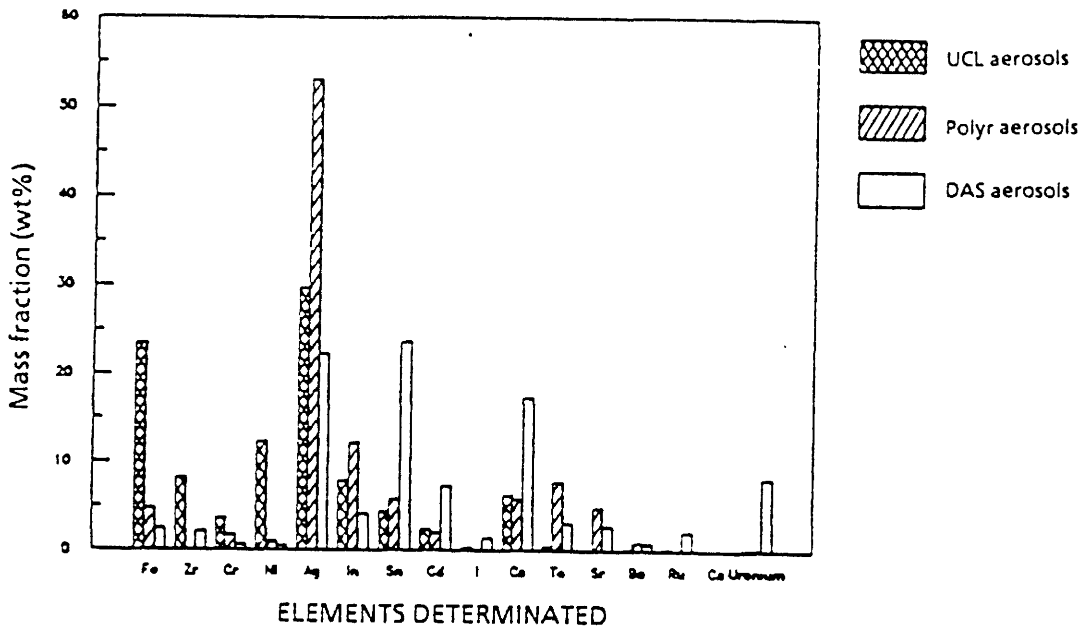
The temperature rise cycles in the UCL facility occurred much faster than in POLYR; moreover, the furnace did not open into a vessel preheated to 60°C and saturated with water vapor. Nevertheless, the overall chemical composition of the particles generated from both mixtures was similar in both facilities (Table 6 and Figure 10).

Mixture	Type A	Type B	Theoretical percentage
U	0.2	0.1	8.2
Fe	17.2	7.5	2.5
Zr	6	9	2.1
Cr	2.7	2.7	0.7
Ni	9	9	0.5
Ag	21.8	21.9	22.2
In	5.8	8.9	4.2
Sn	3.2	1.8	23.6
Cd	1.8	0.6	7.3
I	0.27	0.18	1.4
Cs	4.6	5.7	17.3
Te	0.4	2.7	3.0
Sr	0.01	16.8	2.8
Ba	0.2	4.2	0.8
Ru	0.13	0.01	2.2
Ce	0.01	0.01	1.1

Table 6: Relative Elemental Abundance (%) in Aerosols Generated from Type A & B Mixtures in UCL Furnace

The only significant difference between the aerosols generated in the two facilities is the appreciably higher zirconium fraction emitted at UCL. The Joule effect heating system using a graphite resistor could provide more uniform heat distribution in the crucible, while the induction-heated POLYR furnace would supply more heat to conductors such as silver.

COMPARISON OF CHEMICAL COMPOSITION OF AEROSOLS EMITTED FROM MIXTURE A (60% UO_2)



COMPARISON OF CHEMICAL COMPOSITION OF AEROSOLS EMITTED FROM MIXTURE B (6% UO_2)

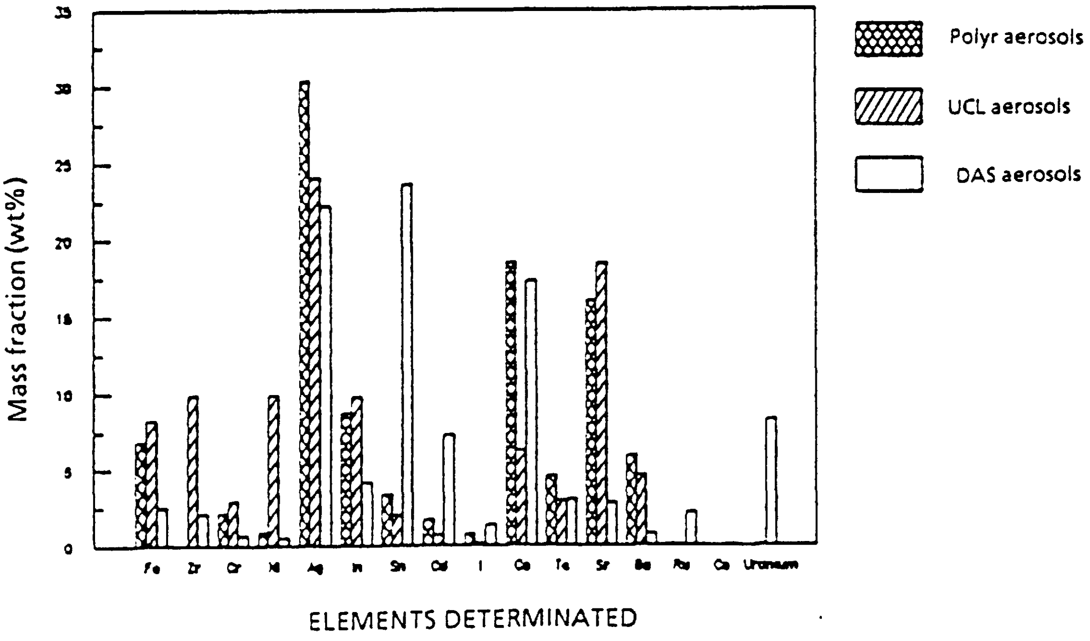


Figure 10: Comparison of Aerosols Obtained in France and Belgium

3.2.2 Identification of Chemical Forms

Crystallized chemical forms were identified after each experiment. This section summarizes the overall results.

The aerosol particles basically consisted of an amorphous, vitreous matrix, with a dark appearance that prevented chemical speciation. A number of included compounds could be identified, however:

- **Uranium** was found as beta oxide UO_2.
- **Zirconium** formed trace amounts of zircon ZrO_2.
- **Silver** appeared in well-crystallized metallic form.
- **Indium** was present in oxide form In_2O_3 as predicted.
- A complex **silver-indium iodide** phase $InAgI_4$ not previously reported was identified, as was an **indium-tellurium** alloy In_2Te_3.
- **Iron** could not be systematically characterized, but was sometimes found as magnetite Fe_3O_4 ($FeO0Fe_2O_3$); Fe_2O_3 was also identified.
- **Ruthenium** was observed as a pure metal in the experiments with Mixture B; similar particles were found after the Chernobyl accident.
- **Cesium** was present in elemental form; cesium hydroxide CsOH, which is highly water-soluble, forms rapidly in air. This compound was identified by electron microscopy (cf § 3.1.3) together with cesium iodide CsI.
- A previously unreported **cesium-cadmium nitrite** compound $CsCd(NO_2)_3$ shows the role that atmospheric nitrogen could play in the chemistry of radioactive contaminants.
- **Strontium** metal, which forms strontium hydroxide $Sr(OH)_2$ in air, was observed.
- **Chromium** was present in pure metal form.

Although many of the soluble chemical species of iodine and cesium have been described in earlier studies, some new and complementary forms were found.

3.3 PARTICLE WATER-SOLUBILITY INVESTIGATIONS

Two types of solubility tests were conducted: an overall investigation of aerosol solubility without discrimination among individual species, and an elemental solubility test after neutron activation of the aerosol particles.

3.3.1 Overall Solubility Tests

Aerosols produced in POLYR were dissolved:

- in distilled water (pH 7) to obtain a control value;
- in rain water from Cadarache for dissolution under conditions comparable to those prevailing in the natural environment;

with a sulfuric acid solution (pH 3.8) to assess the effect of industrial pollution (the pH value of 3.8 corresponds to the beginning of UO_2 dissolution);

- in a percolated soil solution obtained by leaching a clayey-sand soil column from Belleville with 10 liters of demineralized water and filtering the percolate to 0.45 µm.

An aerosol aliquot was deposited on a 47 mm 0.2 µm teflon filter which was then weighed on a precision balance (± 0.1 mg) and placed in an elution device. A micropipette was used to wet the filter with 1 ml of solution to observe the particle behavior, then an additional 9 ml; 30 minutes later, another 10 ml of leachate were added. The eluate was recovered after 8 hours on an orbital plate-type agitator. The filter was then oven-dried for 1 hour at 60°C and weighed again.

Under the test conditions, the limit beyond which elements were considered dissolved was thus arbitrarily set at 0.2 µm, i.e. well below soil filtration levels.

The overall aerosol solubility tests were conducted using particles formed from the 60% and 6% UO_2 mixture, with appreciably different chemical compositions.

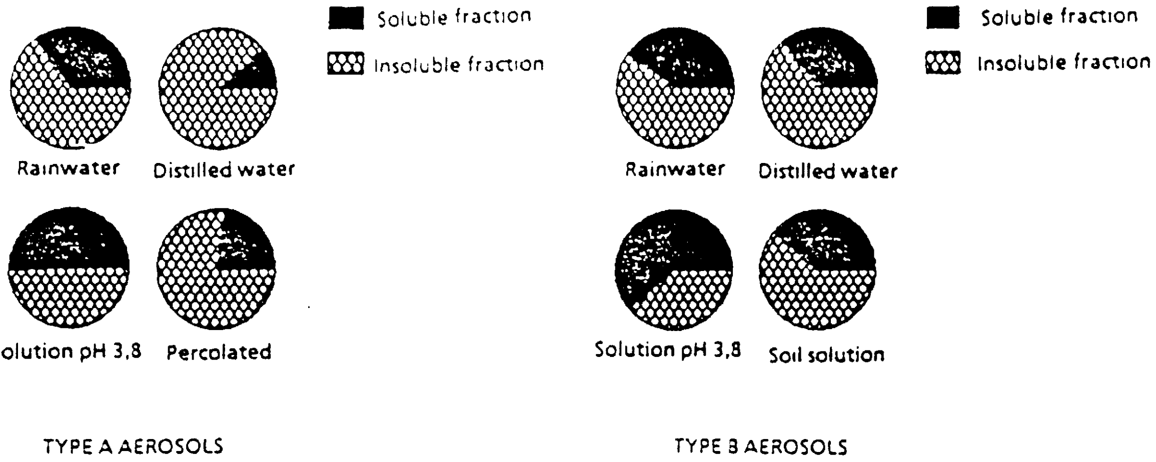
The results (Tables 7 & 8 and Figure 11) show that particle dissolution was enhanced as the pH diminished, confirming the generally alkaline nature of the aerosols, regardless of the nature of the mixture.

Solvent	Deposited Aerosol Mass (mg)	Dissolved Aerosol Mass (mg)	Soluble Element Fraction (%)	Error (%)
Distilled water	0.9	0.8 ?	11 ?	12 ?
Rain water	2.0	1.3	35	8
Acid solution (pH 3.8)	4.7	2.3	51	4
Percolate	135.6	103.9	23	< 1

Table 7: Overall solubility tests for particles generated from Type A mixture

Solvent	Deposited Aerosol Mass (mg)	Dissolved Aerosol Mass (mg)	Soluble Element Fraction (%)	Error
Distilled water	3.3	2.1	36	5
Rain water	4.8	2.8	41	3
Acid solution (pH 3.8)	5.1	2.0	61	5
Percolate	3.1	1.9	39	5

Table 8: Overall solubility tests for particles generated from Type B mixture



TYPE A AEROSOLS

TYPE B AEROSOLS

Figure 11: Overall Solubility Tests (All Elements)

This was not at all unexpected, considering the initial composition and the physicochemical conditions of aerosol formation. A simple means of assessing the alkalinity of the aerosols was to place a particle aggregate in contact with litmus paper moistened with distilled water: the pH ranged from 8.5 to 10.5 depending on the initial mixture.

Complete dissolution was never obtained. This suggests that, as following the Chernobyl accident, insoluble species were formed. The overall solubility of the particles in the natural environment would probably range from 20% to 40%.

The high particle solubility in sulfuric acid shows that the consequences of a serious accident would be more significant in areas either heavily industrialized or subjected to industrial pollution.

Electron microscope observations (§ 3.1.2) showed that the particles consisted of a nucleus surrounded by various protuberances. The photos in Appendix 2 show that these protuberances virtually disappeared after rinsing in water, and no doubt represented the most soluble fraction.

A precipitate was formed during dissolution tests with a Type B aerosol (generated from a 6% UO_2 mixture) in a soil percolate. This floculation phenomenon could be due to shifting chemical equilibria, notably for the carbonates, due to the particle alkalinity. Neutral complexes were thus formed from iron oxides present in the soil solution, or even from the magnetite revealed in the particles by X-ray diffraction, for which the adsorption capacity was investigated by Gandon [13].

In any case, if the same phenomenon occurred in the natural environment it might limit the biological availability of the radiocontaminants trapped in these colloids inasmuch as plant uptake through the roots is limited to ionic chemical species. Radionuclide migration in the soil would also be reduced since they are included in colloids measuring between 0.001 and 0.1 μm, much larger than a simple hydrated ion.

3.3.2 Elemental Solubility Tests

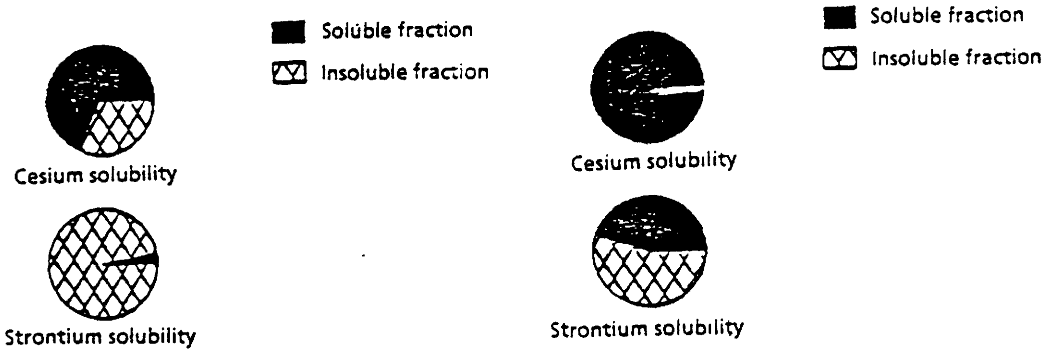
Some elemental solubility tests were performed with stable isotopes, and determined by spectrophotometry. The very small aerosol mass, and the minute quantities of cesium and strontium used, made it difficult to estimate the error. Table 9 and Figure 12 therefore only include results considered reliable.

The aerosol aliquots were weighed to within 0.1 mg, then divided in two: one fraction was left in contact with 10 ml of distilled or rain water for about 30 minutes, then filtered to 0.01 μm and analyzed; the other fraction was dissolved in 13N nitric acid and analyzed in the same way. The ratio of the two results corresponded to the soluble fraction.

Other tests at Cadarache revealed the solubilization kinetics of cesium generated from mixture A. The immediate solubility level of about 40% rose to 80% after one day, and over 90% after 40 days.

Type of Mixture	Cesium Solubility		Strontium Solubility	
	Distilled water	Rain water	Distilled water	Rain water
A	69	-	2	-
A	81	-	0	-
A	55	-	5	-
B	99	61	45	21

Table 9: Elemental solubility (%) of cesium and strontium aerosols



TYPE A AEROSOLS

TYPE B AEROSOLS

Figure 12: Element Solubility Tests

These tests show that:

- **Cesium** is found in relatively soluble forms. With mixture A, which most closely approximates the material proportions in a reactor core, the solubility ranged from 50% to 80%, similar to the figure of 75% measured for fallout from the Chernobyl accident. Aerosols generated from mixture B were almost totally soluble (more than 90%), confirming the chemical speciation results which included compounds such as CsOH or CsI.

- **Strontium** was moderately soluble; some salts of this alkaline earth such as $SrCO_3$ have a low but positive solubility constant.
- The fact that no solubility results could be obtained for **ruthenium** tends to demonstrate that it is insoluble. The pure elemental form observed is in fact insoluble, as is the oxide RuO_2 which is likely to form.

Other gamma spectrometry tests following neutron activation at UCL confirmed the findings for mixture B, i.e. the virtually complete cesium solubility. Results are not yet available for mixture A.

4. CONCLUSION

Considerable progress has been made since physicochemical speciation work began on radioactive pollutants released in accident situations.

The particle nature of the emission has been established, and the particles have been described and characterized. The mean grain size was found to be about 1 µm. Most often, the particles showed a compact nucleus surrounded by smaller protuberances easily eliminated by rinsing in water.

At high magnification (x 9000) the particles are clearly heterogeneous, enriched in certain metals such as silver or indium, and sometimes even pure substances such as ruthenium or cesium hydroxide. At a larger scale (x 1000) the deposits appear much more uniform.

The concentration of sixteen elements were determined in the aerosols. Iodine was quickly released, and was thus found only in very small quantities in the aerosols. The chemical forms predicted by theoretical studies (CsI, CsOH, metallic Ru and Ag) were found, together with some unexpected compounds such as a cadmium-caesium nitrite.

The solubility of fission products with radioecological impact appeared similar to the solubility generally observed for the elements involved, and depended on their chemical properties. Ruthenium, for example, is virtually insoluble, like metallic silver. Strontium shows low or moderate solubility, depending on the relative amounts of the element and water. When cesium is generated from uranium-rich mixtures it is moderately soluble, as observed in fallout from the Chernobyl accident; when the initial mixture contains little uranium, however, cesium is almost totally soluble.

Short-term transfer factors to plants following an accident were found to be lower than those based on standard commercial sources because of the lower solubility values. As the particles break up and the elements are subjected to environmental action, their longer term behavior will probably more closely resemble their more common chemical behavior determined by the lowest possible energy states.

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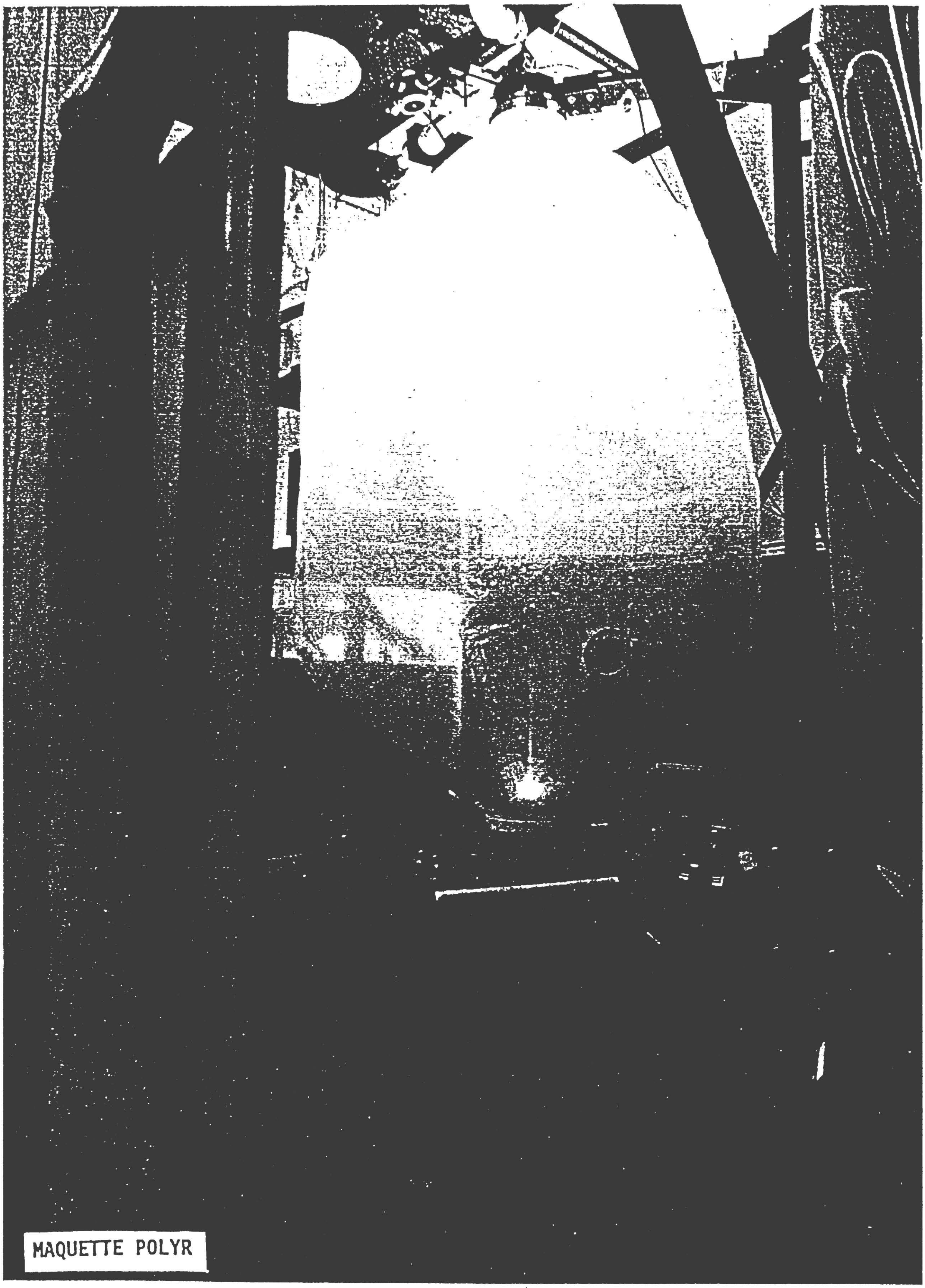
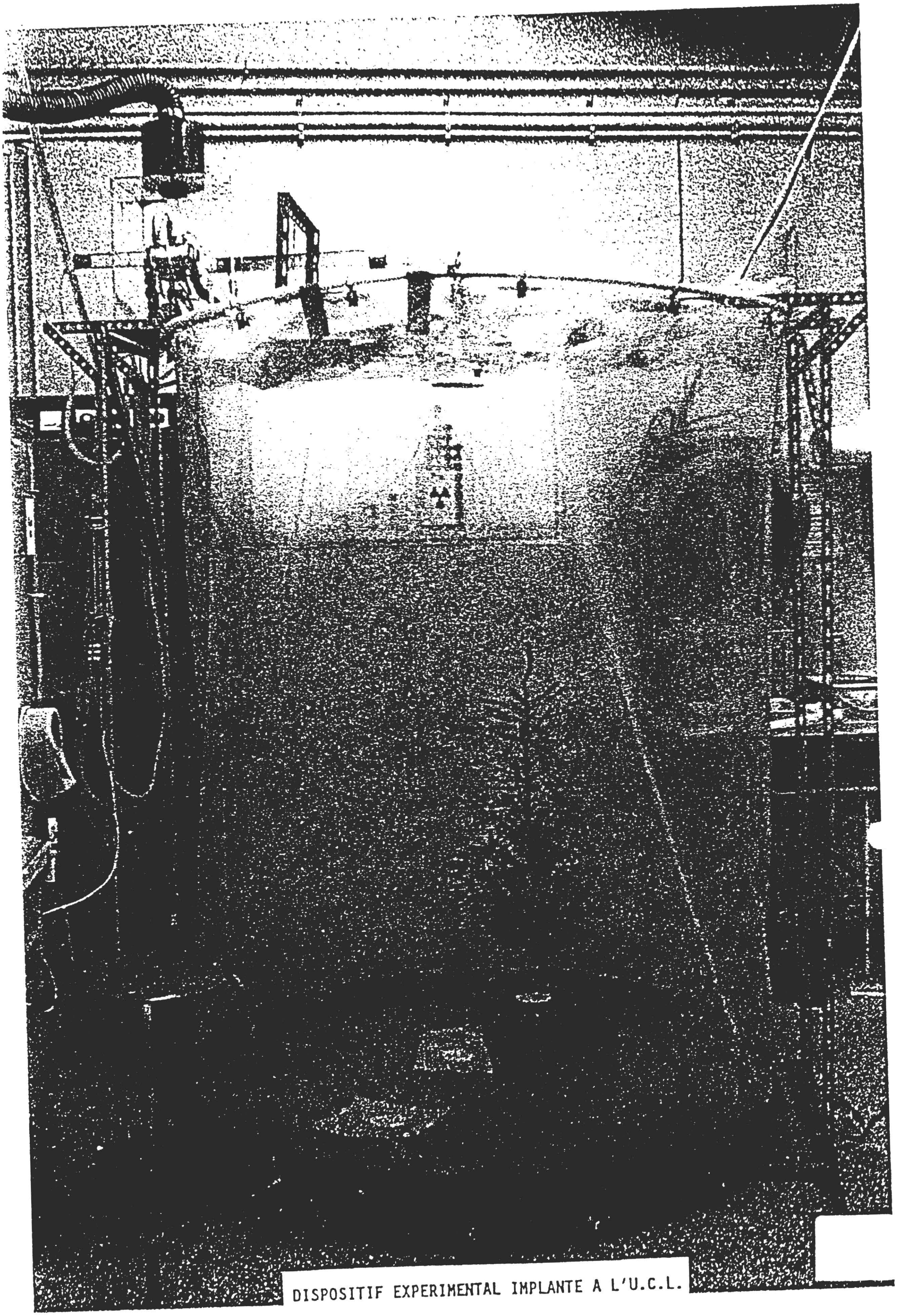
APPENDIX 1

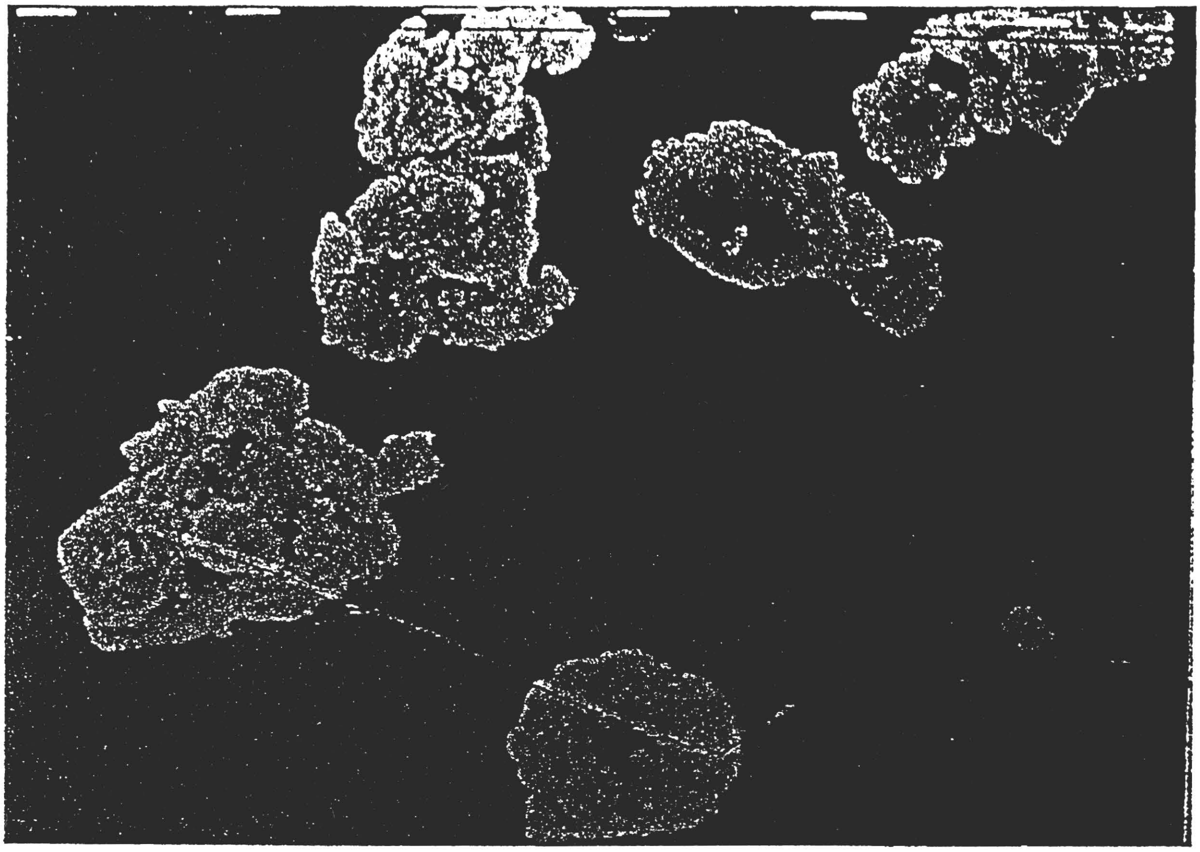
Table A1: Dissolution Water Analysis Results

Parameter	Unit	Rain water (Cadarache)	Soil percolate (Belleville)
pH		6.30	7.13
Conductivity	µS	25	326
Ca^{2+}	$mg*l^{-1}$	3.7	37
Mg^{2+}	$mg*l^{-1}$	0.5	2.3
Na^{+}	$mg*l^{-1}$	0.6	3.4
K^{+}	$mg*l^{-1}$	0.6	9.0
Cl^{-}	$mg*l^{-1}$	4.3	11.7
HCO_3^{-}	$mg*l^{-1}$	5.4	167
SO_4^{2-}	$mg*l^{-1}$	< 1	10
Fe	$mg*l^{-1}$	< 0.01	-
Silica	$mg*l^{-1}$	< 0.01	14.6
$KMnO_4$ Oxidizability	$mg\ (O_2)*l^{-1}$	< 0.1	91

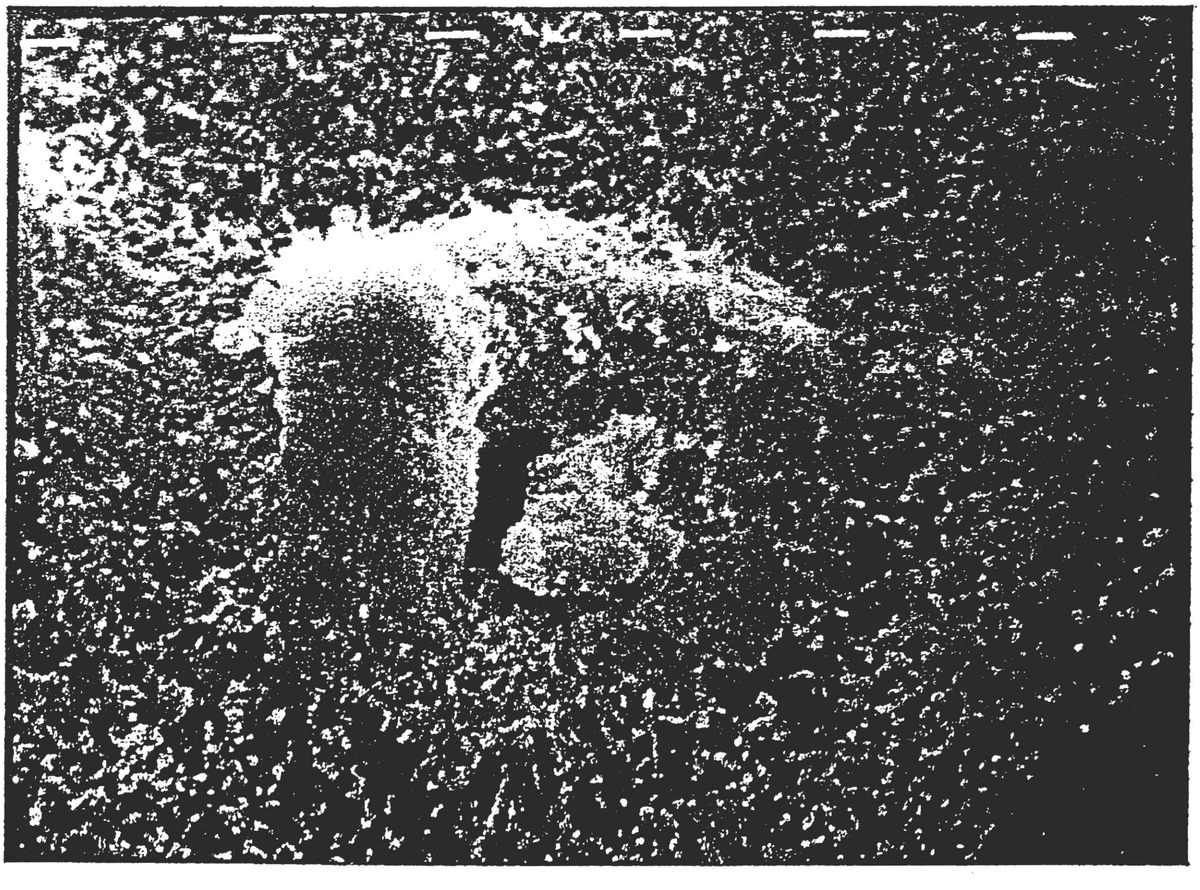

MAQUETTE POLYR



DISPOSITIF EXPERIMENTAL IMPLANTE A L'U.C.L.

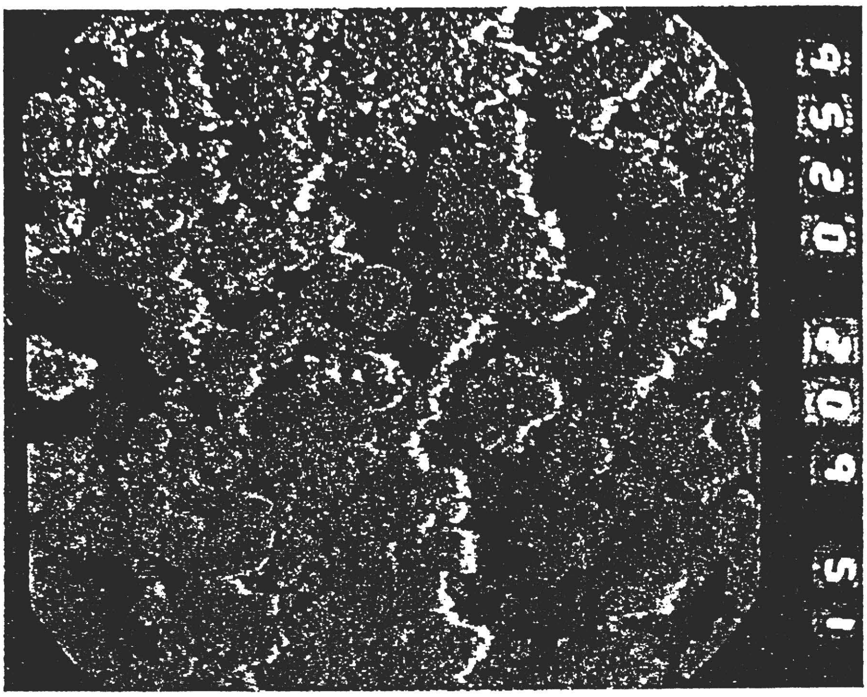


Collected polymetallic aerosols (1 µm graduations)

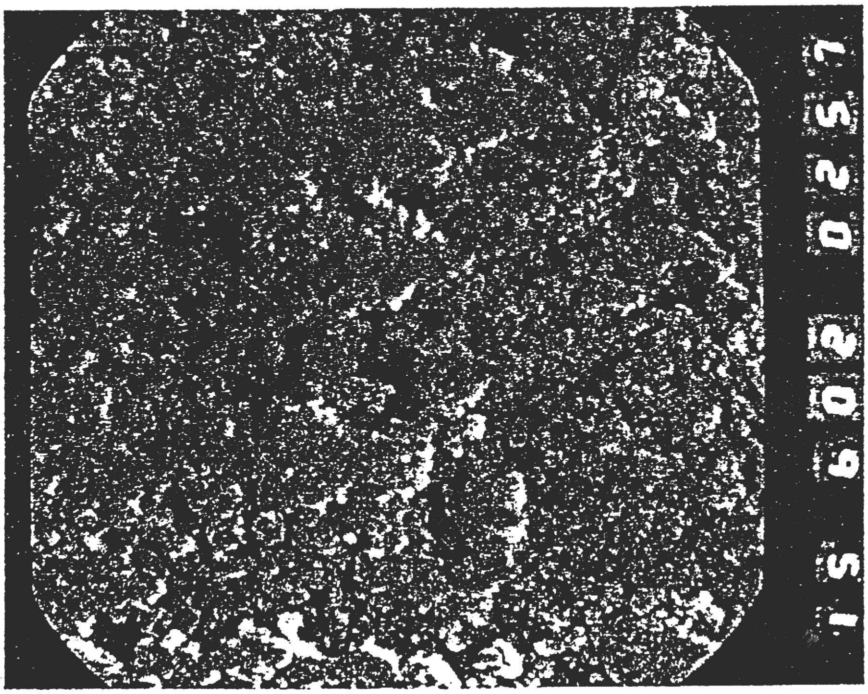


Lamellar particle (probably caesium) measuring about 10 µm trapped in a pine stoma.
Note the fluffly cuticle due to the presence of was and polyphenols.
The two guard celles responsabile for stomatal opening are clearly visible.

Aerosols generated using the experimental device at UCL (Belgium)



Aerosol A (SEM image x 6000)



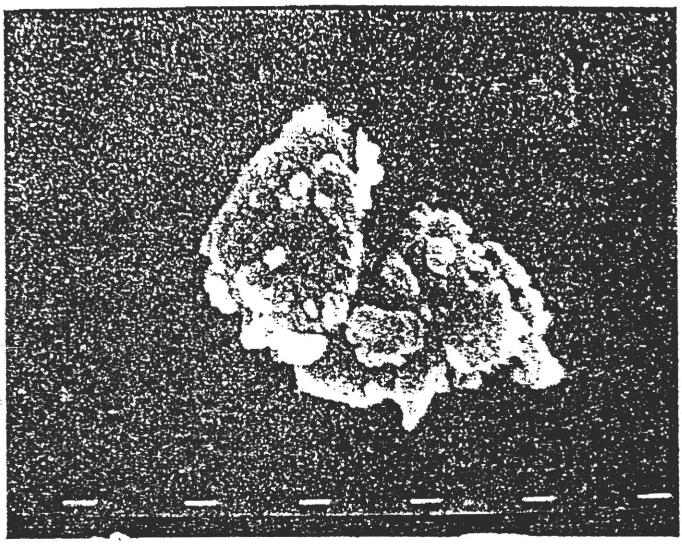
Aerosol B (SEM image x 600)

The images are unresolved due to a different preparation process : the aerosols were applied on the substratum with a spatula, then observed without fixing. This explains the fleecy appearance of the image field.

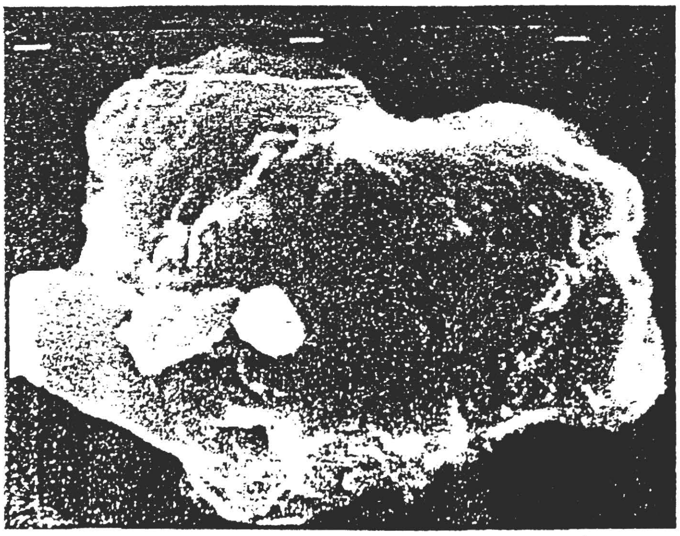
APPENDIX 2

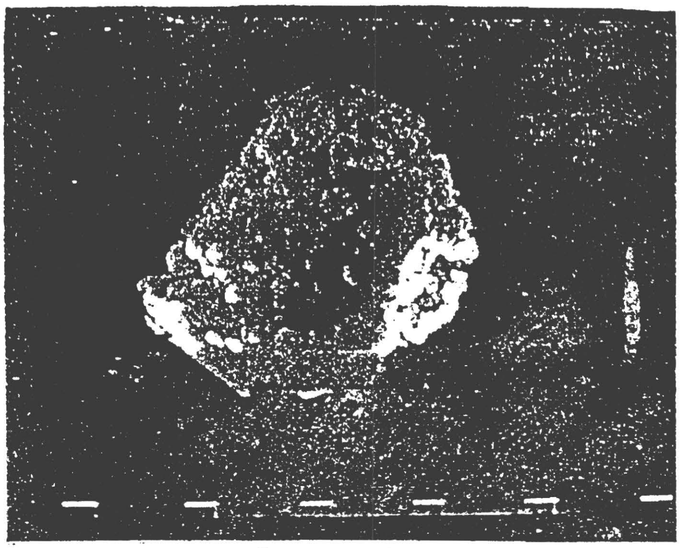
Superficial Appearance of Particles Before and After Dissolution in Deionized Water

Unrinsed particle : The two particles visible in the scanning electron microscope image include surface globules tending to be electrically conducting (i.e. rich in metals) that appear white in the photo. These are fine particles captured by larger nuclei during aging.

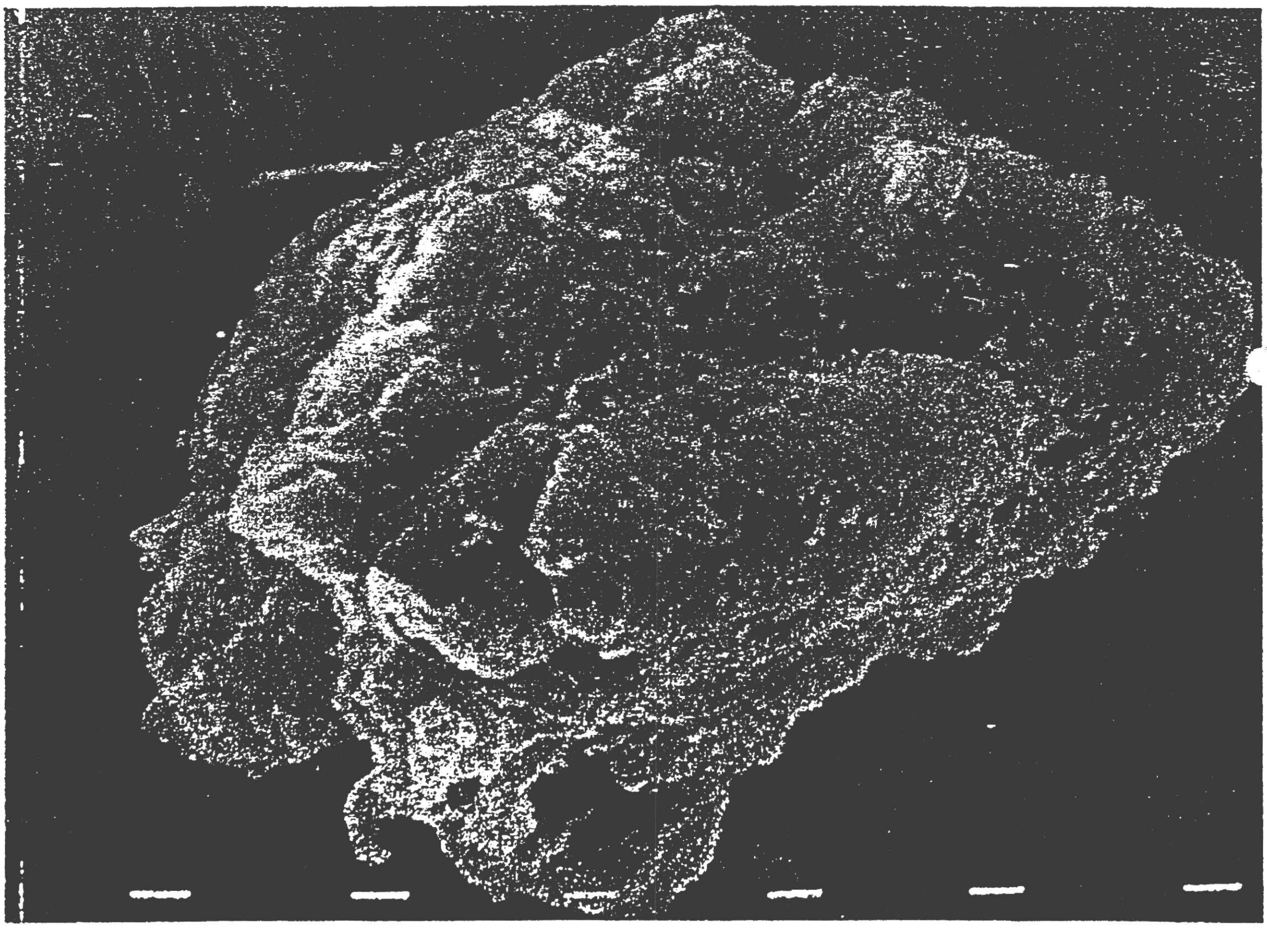


Rinsed particle : The surface globules were much less numerous : rinsing in dionized water thus entrains the surface globules, either by simple mechanical effect in the case of electrostatic bonding, or by chemical effect in the case of ionic or covalent bonding involving water-soluble compounds (1 μm graduations).

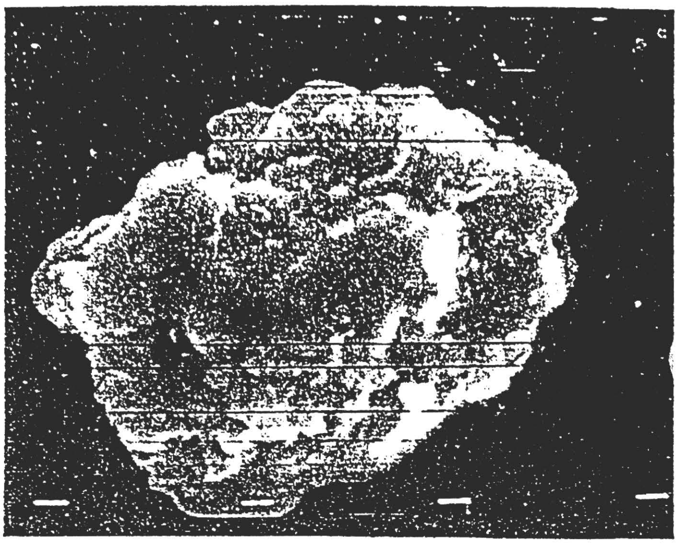




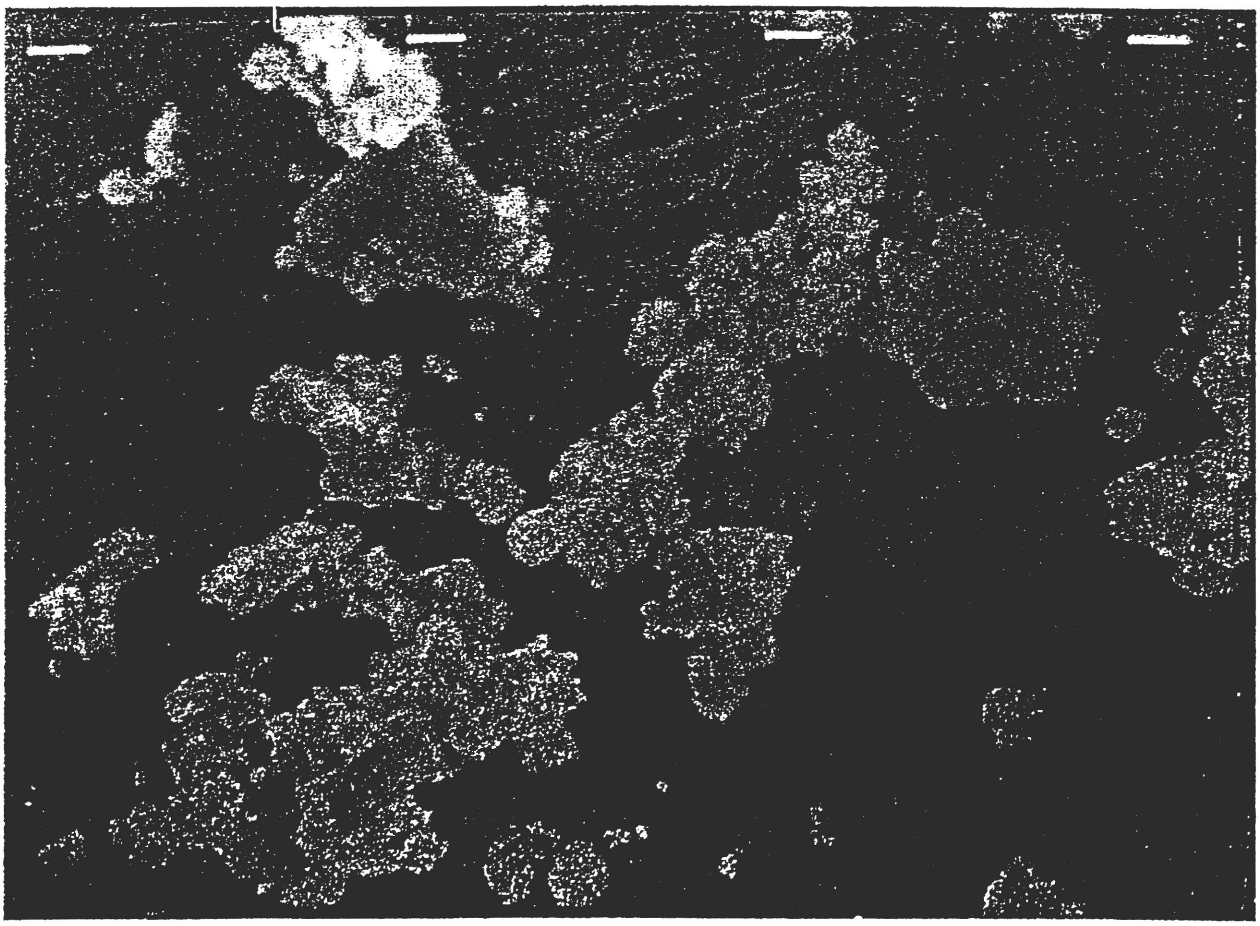
5 μm particle : visibly an aggregate of finer particles, as shown by the many globules at the surface, measuring a few tenths of a micron.



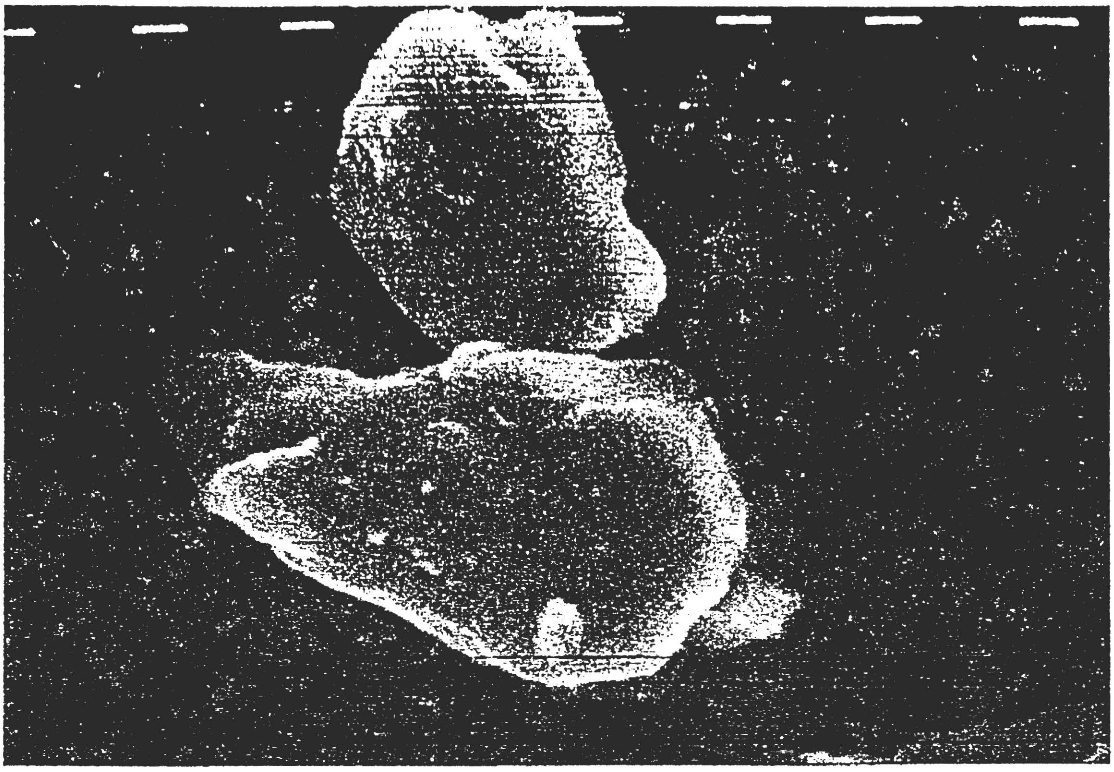
350 μm polymetallic particle (the largest observed to date).



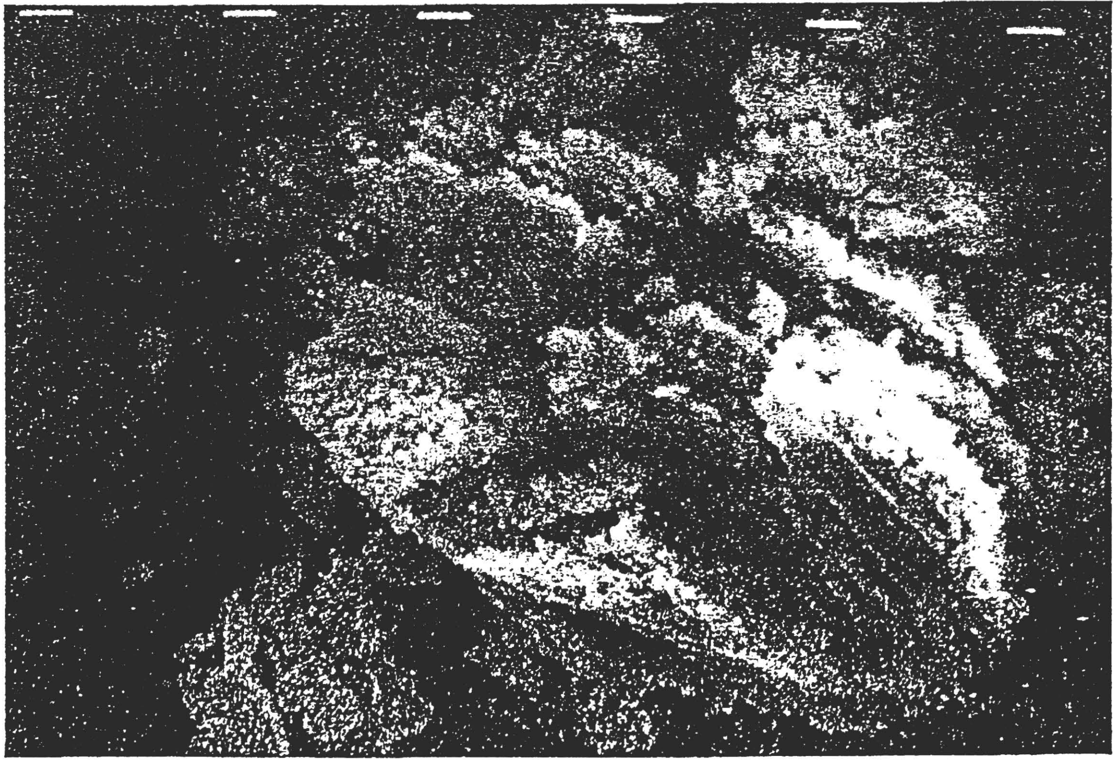
7 μm polymetallic globule : the shiny surfaces (white in the photo) reveal local enrichment of a highly conducting metal (e.g. silver). Streaks were due to a video coupling fault.



Overall SEM image. Note the wide morphology variations among particles of similar dimensions. The darkest particle, with sharp edges in the upper quarter of the field is probably caesium iodide (5 μm graduations).

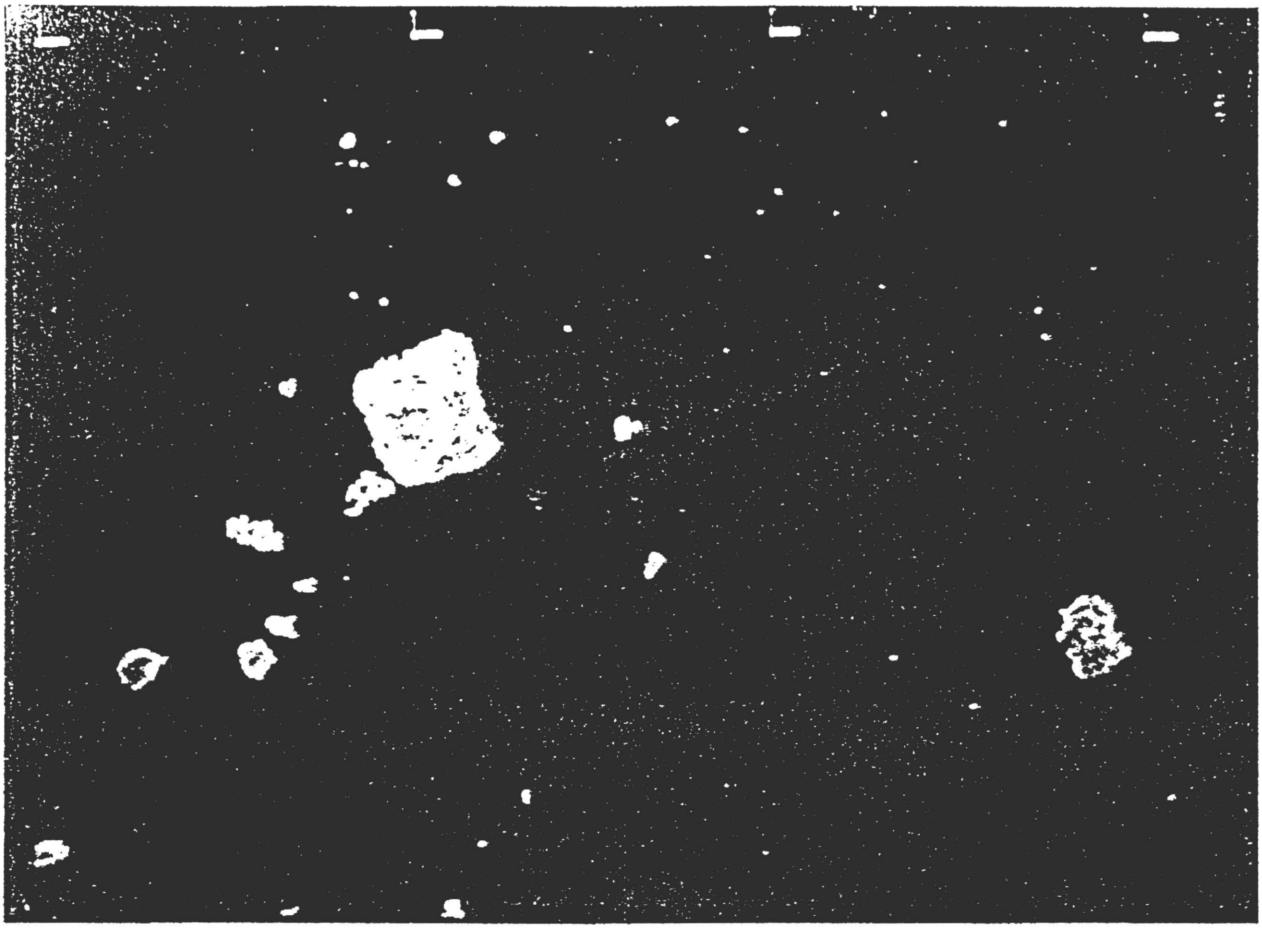


Lamellar particles of CsI (3 and 7 μm)
(note the Shinier polymetallic globule at the surface of the largest particle)



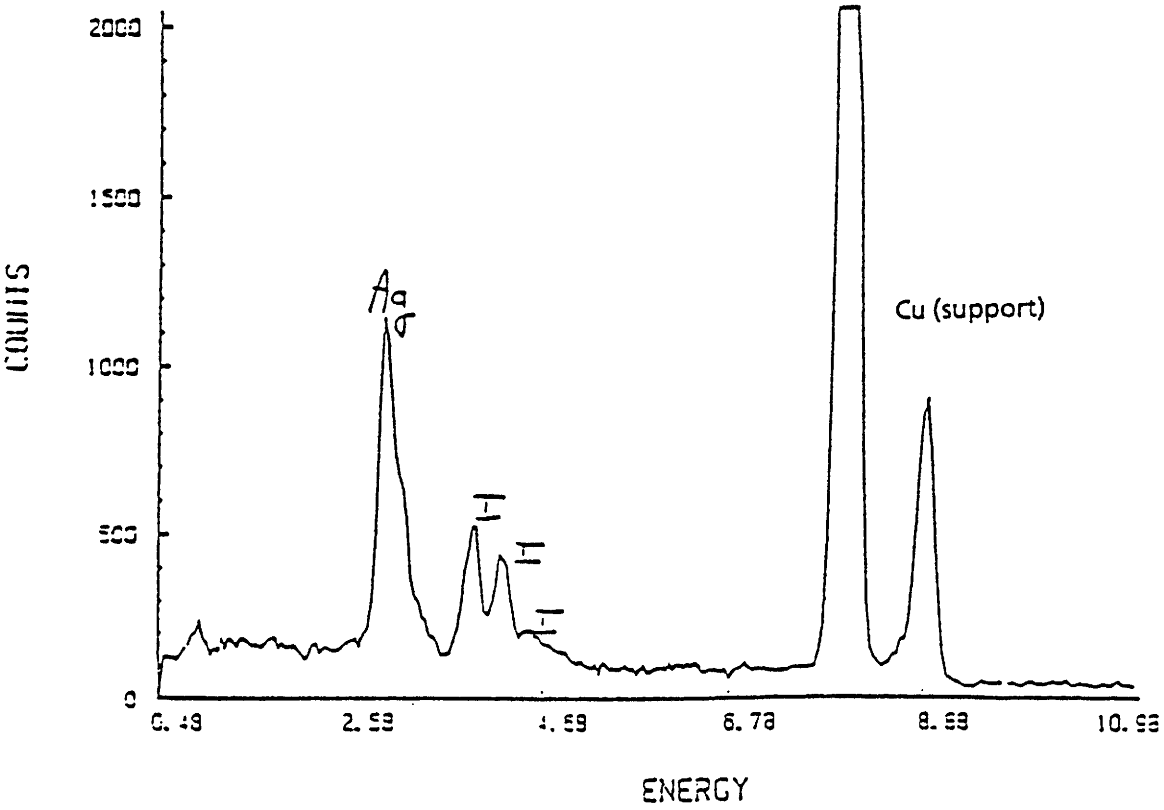
Particles aggregate, diameters inferior to 1 μm
(graduation = 1 μm)

Cubic particle, probably CsOH, measuring about 3 μm

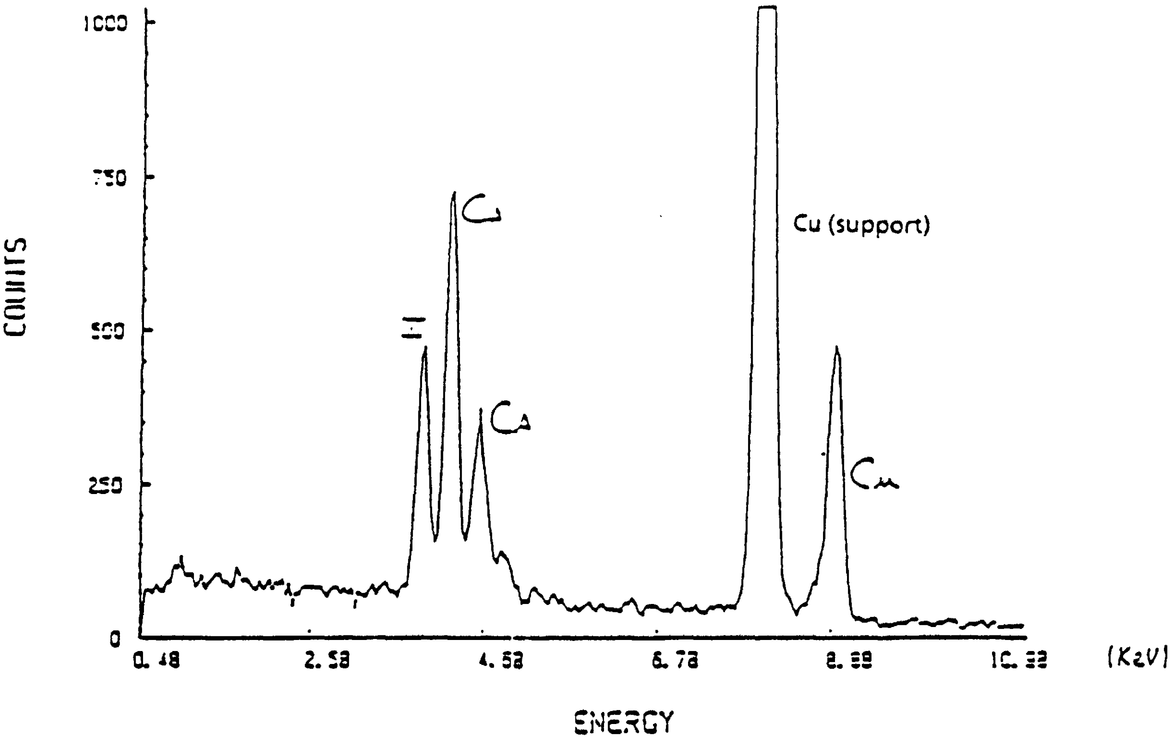


APPENDIX 3

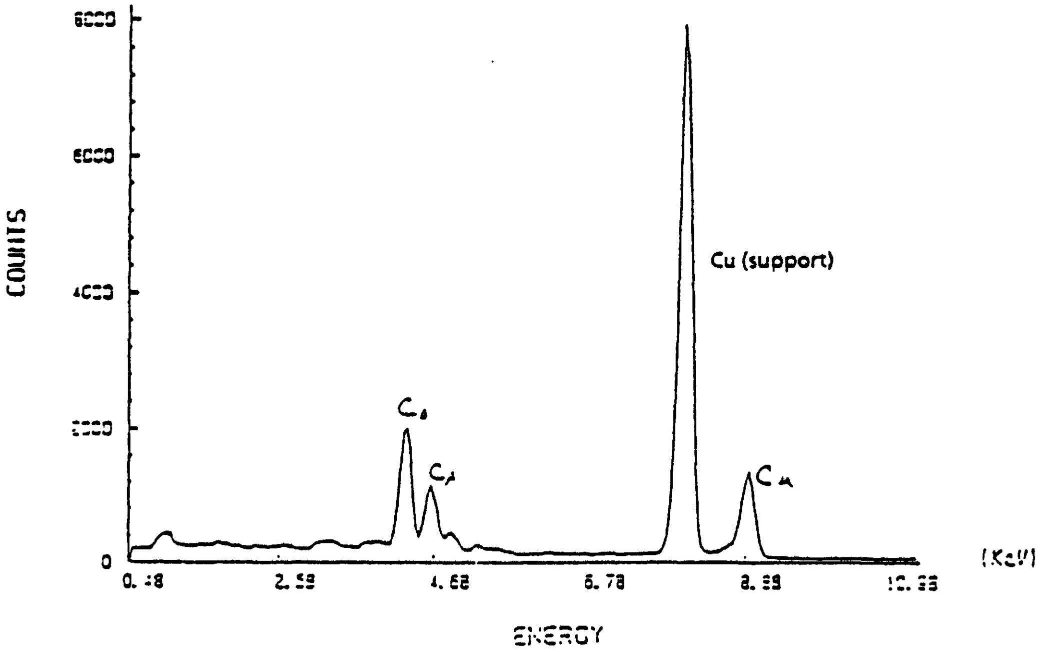
Backscattering electron microprobe
Analysis spectra - Mixture B



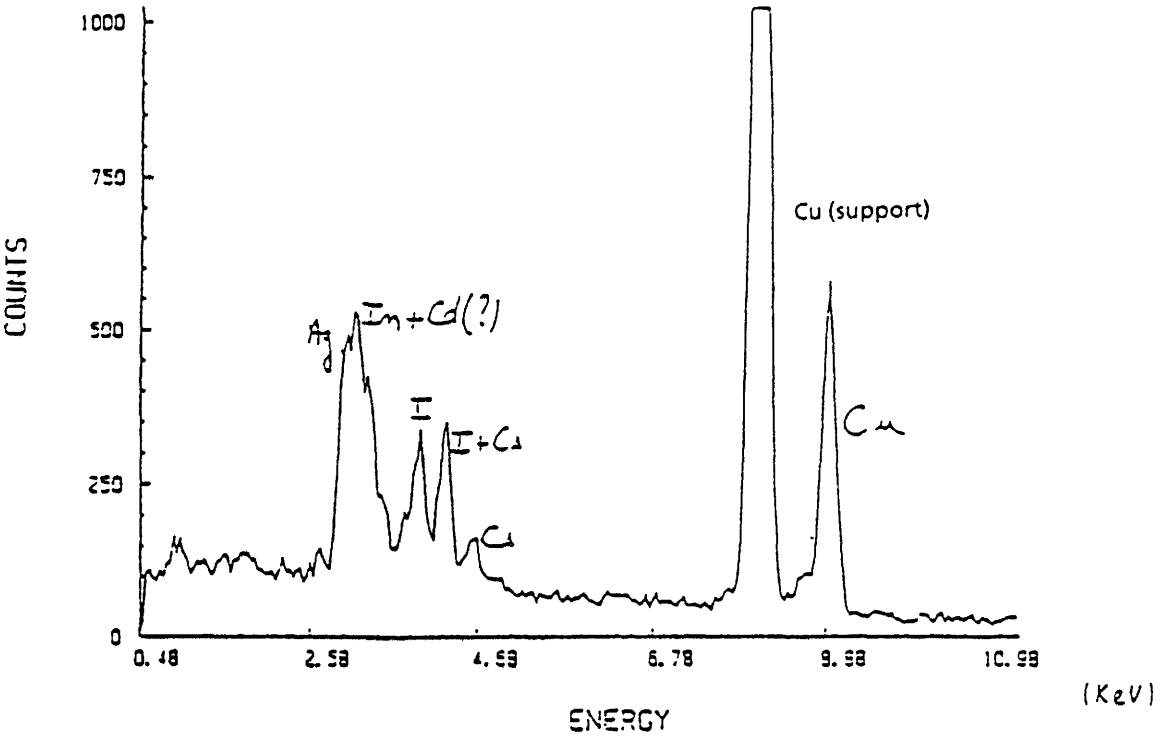
Silver Iodide Particle (AgI)



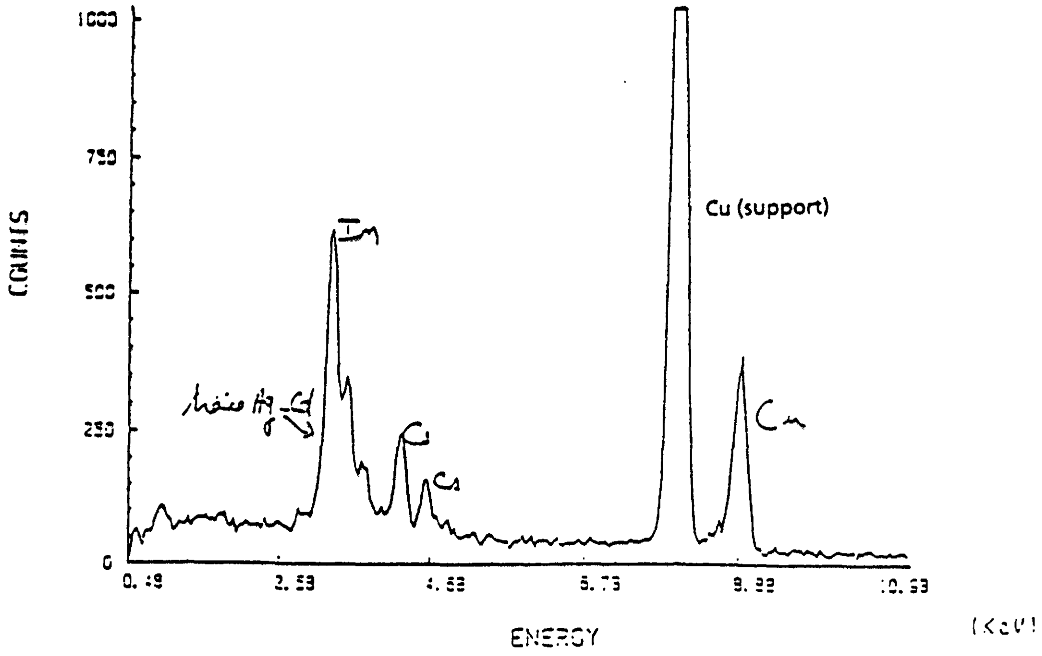
Caesium Iodide Particle (CsI)



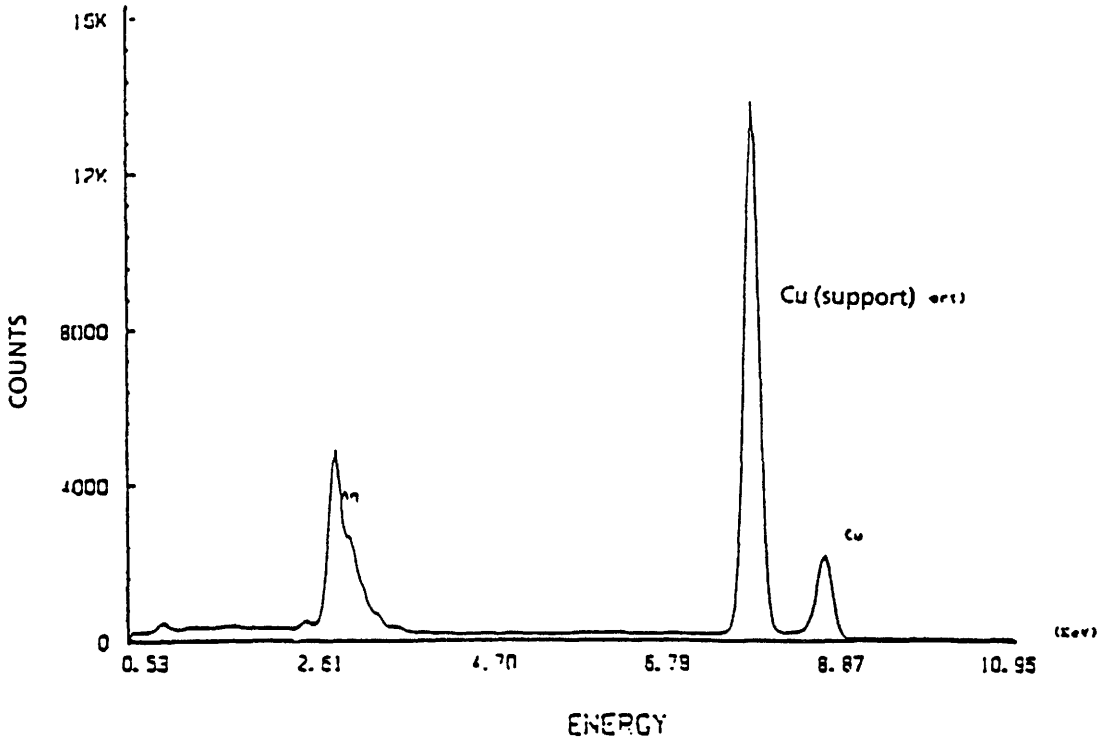
Caesium Hydroxyde Particle (CsOH)



Complex Particle : Size 0.3 Micrometer

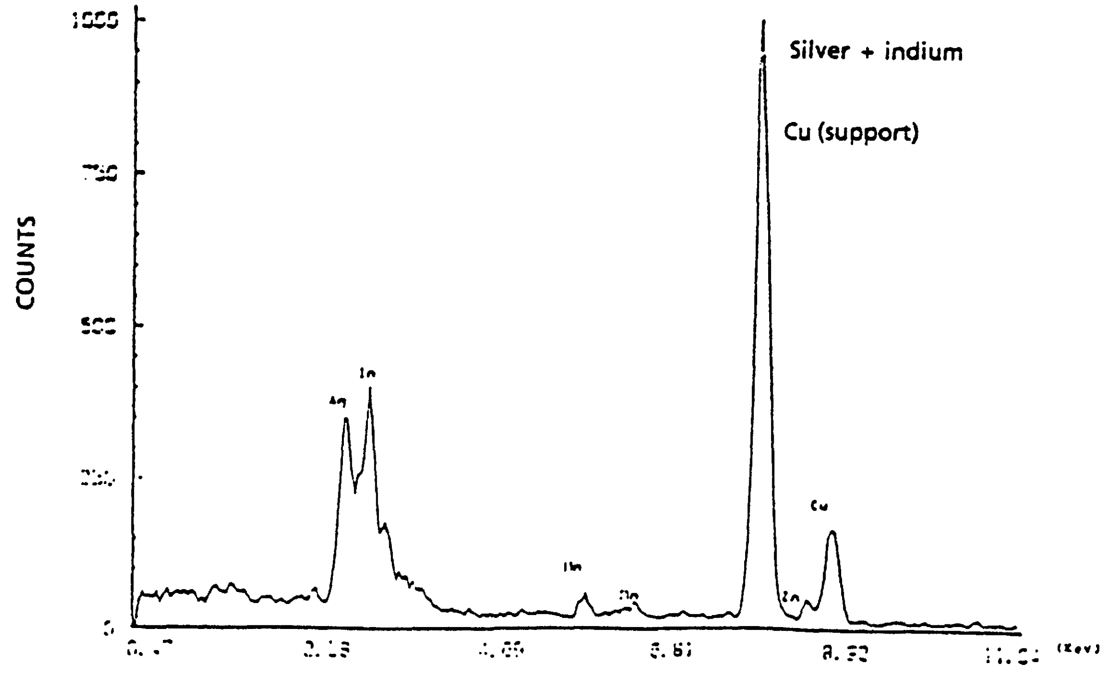


Complex Particle with Co and In



Small Silver Sphere : Probably Metallic Form

Spectre de microanalyse



Spectra from Mixture A

III. SOIL-PLANT TRANSFER (RIVM, ITE, KFA, ENEA)

1. SEMI-NATURAL ECOSYSTEMS

1.1 INTRODUCTION

A wide range of natural ecosystems have been contaminated by fallout from the Chernobyl accident. The association of caesium deposition with high rainfall means that the areas affected by the fallout are often those in countries which experience Arctic, Boreal and Atlantic climates. This high correlation between rainfall and deposition also means that much of the material has been deposited in upland areas where agriculture is of a marginal nature or natural plant communities dominate. Many of these plant communities are exploited by Man with minimal or no management. Where other climatic regimes are involved, such as the Central European type, these are frequently modified by the mountainous nature of the terrain.

The ecosystems affected are predominantly by those developed under wet, cool climates. The vegetation is often bog, heathland or coniferous forest. Slow decomposition rates and the growth of species such as *Sphagnum* allows the development of highly organic soils. The dominant angiosperms are often members of the Ericaceae and comprise members of the genus *Vaccinium, Empetrum, Oxycoccus* and *Erica*. Other species of these habitats are *Eriophorum* and *Carex spp.* which together with lichens such as *Cladonia rangiferina* and *Cetraria nivalis* are potential foodplants for grazing animals. In more southern areas of Europe the effect of altitude is such that that a Boreal forest of beech and beech-fir dominates. There is a range of vegetation. The extremes are from acidic soils with species such as *Vaccinium myrtillus, Melampyrum sylvaticum* and *Luzula spp.* to the base-rich areas of the Dolomites which support a wide variety of herbaceous species.

Pathways back to Man from the natural and semi-natural systems are varied both in type and importance. The main pathway is by way of animals via meat and dairy products. The meat pathway can dominate, as is the case in parts of the Scandinavian countries, where the Laplanders rely to a large extent on reindeer. On the other hand contaminated animals and birds which are luxury foods, such as the red grouse in the UK, may play a part only in the diet of a few individuals. Dairy products, particularly those from goats, can form an important pathway back to man where grazing occurs on contaminated natural systems or hay is gathered from natural pastures. Direct use of the ecosystem only occurs when native fruits are collected. Again this type of exploitation tends to occcur in the more northerly countries of Europe. Important pathways in some countries can be by way of wild fungi which are collected in large quantities for food and flavouring.

Natural and semi-natural ecosystems are therefore important in the passage of the Chernobyl fallout back to man. This is particularly so in the areas where climate limits agriculture and Man has to rely on natural systems to provide food and grazing for his animals. Whilst not within the remit of the present work it should not be forgotten that many of the areas considered in this work also serve as catchments for the supply of domestic drinking water.

1.2 STATE OF KNOWLEDGE PRIOR TO THE CHERNOBYL ACCIDENT

a) Natural and semi-natural ecosystems

Natural ecosystems are those which have evolved without direct intervention by man; they are usually remote from human settlements and have soils that are unsuitable for the production of food crops. In Europe they include mires, tundra, uplands above the snow-line and exposed or inaccessible areas of coast, uplands and bogs.

Semi-natural ecosystems are those which have been exposed to minimal intervention by man. Any ecosystem that has been drained, limed or fertilized cannot be defined as semi-natural. Intervention by Man usually takes the form of regulated grazing by farm animals such as sheep, hunting deer and game birds for meat, and coppicing or minor felling activities in native forests. In Europe semi-natural ecosystems include some upland grasslands, some areas of tundra, native forests, heathlands, scrub, peatland mires and salt marshes.

b) Information available on the behaviour of radiocaesium in soils

Five important factors which affect the mobility and/or accumulation of radionuclides in ecosystems are climate, soil characteristics, plant and animal populations, and characteristics of specific radioisotopes. The behaviour of radiocaesium in soils typical of natural and semi-natural ecosystems appears to be dominated by climate and soil type as well as the chemical and physical form of radiocaesium.

In exposed natural ecosystems such as areas of tundra or much of the Alps there is little or no soil and surviving plant forms are mainly limited to mosses and lichens. Less extreme semi-natural ecosytems typical of Europe include large tracts of upland covered by loamy soils in drift over hard rock, with a wet, peaty surface and a thin iron-pan; raw, upland peats with associated peaty-topped shallow and stony mineral soils; and brown podzolic soils that may be loamy and clayey over relatively impermeable clayey sub-soils. Often these soils have poor drainage, experience seasonal waterlogging, have a high percentage of organic matter and a low mineral content.

Both potassium and caesium are are Group I alkali metals and it has been frequently suggested that they may act as analogues due to similarities in their chemical and physical characteristics. Potassium soil concentrations have been found to affect the availability of radiocaesium for plant uptake (For example: Nishita et al., 1960; Cline, 1962; Evans & Dekker, 1965). However, potassium does not always behave in an analogous manner to caesium in many systems. The addition of potassium to soils already containing high concentrations has no effect on plant uptake of ^{134}Cs and ^{137}Cs whereas the addition to soils with low concentrations of potassium can reduce plant uptake of radiocaesium (Nishita et al., 1960). Solubility of radiocaesium in soil water is not just dependent on potassium concentrations in the soil; it is also affected by the relative concentrations of other pollutants and nutrient elements in the soil system (Handley & Overstreet, 1961).

It is generally accepted that the availability of radiocaesium for plant uptake decreases with increasing clay mineral content (For example: Cummings et al., 1969; Garten & Paine, 1977; Pinder et al., 1980; Kuhn et al., 1984). However studies have also shown that radiocaesium is more readily desorbed from some clay minerals than from others. Illite clays provide stronger binding sites than kaolinite clays (For example: Schulz et al., 1960; Sawhney, 1964; Brisbin et al., 1974; Sharitz et al., 1975; Evans et al., 1983; Cawse & Horrill, 1986) Table 1. Caesium ions have relatively greater bonding energies than potassium ions so it is probable that they will displace potassium ions when there are a limited number of sorption sites within the soil (Brisbin et al., 1974).

The presence of clay minerals in upper soil layers also appears to inhibit the movement of radiocaesium down the soil profile (Table 1) by leaching processes (Squire & Middleton, 1966; El-Fawaris & Knaus, 1984). Even when soils have a high percentage of clay minerals that readily adsorb radiocaesium, the adsorption processes are strongly affected by soil pH. Strongly acidic soils appear to act as a catalyst for caesium adsorption processes (Prout, 1958; Greenland, 1974).

Soil mineral type	**Observations**
Hydrobiotite	-caesium adsorbed irreversibly (Coleman et al., 1963; Klobe and Gast, 1970)
Illite	-almost irreversible adsorption of caesium in sediments with a high percentage of illite (Lomenick and Tamura, 1965)
Kaolinite	-little adsorption of caesium (Schulz et al., 1960)
Micas	-caesium adsorbed irreversibly (Coleman et al.,

	1963; Klobe and Gast, 1970) -adsorption of caesium reaches equilibrium with soil solution very quickly (Sawhney, 1966)
Montmorillonite	-adsorption of caesium reaches equilibrium with soil solution very quickly (Sawhney, 1966) -very little fixation of caesium observed (Schulz et al., 1960; Sawhney, 1964, 1966) -relatively high cation exchange capacity (Cawse, 1983)
Vermiculite	-variable behaviour reported (Coleman et al., 1963; Klobe and Gast, 1970; Schulz et al., 1960)

Table 1: Reported behaviour of radiocaesium in the presence of soil clay minerals

Soil organic matter is an active adsorber of radiocaesium (Brisbin et al., 1974). It temporarily immobilises radiocaesium in the soil before decomposition processes release it back into the soil system. It is then available for transfer to detritivores and microbial decomposers, for uptake by plants from soil solution, or it may be leached down the soil profile before being immobilised again by cation exchange processes.

Several studies have shown that where there is a high percentage of organic matter in upper soil layers relatively high radiocaesium concentrations remain at the top of the soil profile for many years (Pegoyev & Fridman, 1978; Cawse, 1983; Cawse & Horrill, 1986; Nifontova et al., 1986). Where extremes of climate, such as are found in the Arctic, lead to slow decomposition rates this effect is even more pronounced (Svoboda & Taylor, 1979).

Many of the soils that received a relatively high Chernobyl fallout have a high organic matter content so it is not surprising that the Chernobyl radiocaesium has remained available for plant uptake for several years. Most of the models on the behaviour of radionuclides in the terrestrial environment that were were available at the time of the Chernobyl accident were based on data for well mixed agricultural soils, not for upland soils with little agricultural potential. They therefore predicted that the radiocaesium would be pass into the soil and soon become unavailable for plant uptake even though there were indications in published literature that this would not be the case.

c) Radiocaesium in plants pre-Chernobyl

Important factors affecting plant accumulation and uptake of radiocaesium include properties of the soil and soil water, climate, season, physical and biochemical characteristics of the plant and the form rate and manner of radiocaesium contamination. The behaviour of radiocaesium in plants appears to be determined by a combination of all these factors.

Natural and semi-natural upland ecosystems have only survived because the soils that support them are unsuitable for intensive agricultural use, and/or because they are remote from areas suitable for settlement by man. This means that they support a very different range of natural species from those that are typical of lowland areas. Species that are out-competed in less extreme environments can survive in more exposed upland areas. They include a wide range of mosses and lichens, reeds, rushes, grasses, herbaceous plants and ericaceaous species in the temperate zone; and beech and beech-fir forests in areas nearer to the Mediterranean.

There is a growing body of data showing that it is impossible to make universal generalisations about plant uptake of radiocaesium even for species growing in close proximity to one another. Some plants have been found to accumulate radiocaesium more readily than others in both natural and semi-natural ecosystems (Table 2). Even within species there can also be significant differences in radiocaesium uptake; these differences appear to dependent on external environmental factors such as season, soil characteristics, species growth strategy and the physical structure of the plant.

Species	^{137}Cs Bq kg^{-1} (Dry weight)	Observations
Typha latifolia	4100	Samples taken from a bog at Ol'Khovskoe that had received radioactive effluent from the Beloyarsk nulear power station.
Carex caespitosa	7200	
Lemna spp.	47000	
Typa lattifolia	19	Samples taken from a control bog at Lake Peschanoe. (Nifontana et al, 1986)
Carex caespitosa	410	
Pleurozium schreberi	680(green)	^{137}Cs concentrations from global atmospheric fallout, Jay Park, Vermont, July 1977.
Dicranum fuscescens	570(brown)	
	430(decomp)	
	530(green)	
Sphagnum spp.	220(brown)	
Carex stricta	170	Sample taken from submountain marsh, Vermont. (Svoboda and Taylor, 1979)

Moss	810	Contamination attributable to global fallout; Hourglass Creek, USA. (Hubbard and Strifler, 1973)
Equisetum fluviatile	96 410	Concentrations varied seaonally; highest values at beginning of growing season.
Carex rostrata	37	All samples taken from Lake Ulkesjon, Sweden
Nuphar luteum	230	(Carlsson and Liden, 1978)
Andropogon spp.	17,000 85,000	First value for each species mean for samples from floodplain Second value mean for samples from islands that were underwater during period of low-level radioactive effluent discharge to Steel Creek, Aiken, USA. (Anderson et al, 1973)
Alnus serrulata	14,000 37,000	
Myrica cerifera	20,000 56,000	
Salix nigra	19,000 59,000	
Potamogeton spp.	70,000	Pond weeds growing in a reservoir used to contain low-level radioactive effluent. (Rickard et al, 1981)
Elodea	37,000	

Table 2: Reported concentrations of radiocaesium in plant species typical of pre-Chernobyl contaminated natural and semi-natural ecosytems.

Experimental and monitoring data indicate that important factors affecting the retention and recycling of radiocaesium in natural and semi-natural ecosystems include:

- low soil potassium concentrations (Cline, 1962; Evans & Dekker, 1965)
- a relatively high soil moisture content for most of the year (Evans & Dekker, 1965)
- low soil pH (Evans & Dekker, 1965)
- slow decomposition of organic matter (Cline & Rickard, 1972)
- low concentrations of clay minerals (Rogowski & Tamura, 1970; El-Fawaris & Knaus, 1984; Garten & Paine,1977)

1.3 METHODS

1.3.1 Sampling areas

Two approaches have been used to investigate the behaviour of radionuclides in natural and semi-natural ecosystems. The first has been to use three study areas within Europe where heavy contamination has occurred and the second to intensively study the individual species from a number of sites within one contaminated study area.

i) Europe

The three European areas used for studies were in northern Italy (Friuli-Venezia Giulia region in the north-east), Norway (Jotunheim reserve) and Scotland (Loch Laggan, Highland region). The detailed contaminated study was carried out in Cumbria, England , where within a few tens of kilometres of each other eleven sites were sampled intensively for individual plant species.

The approach at the European sites has been to sample both the soils and vegetation from at least three ecosystems and at the Cumbrian sites to concentrate on the variation within individual species. Ecosystems studied included conifer, beechwood and alpine meadow communities in Italy, dwarf shrub, birchwood and mire in Norway and heathland, upland pasture and birchwood in Scotland. In Cumbria, heathland, bog and acid grassland vegetation were sampled. In the majority of these habitats the soils were wet, acidic and organic in nature. The Italian sites , where a rich brown-earth soil derived from base-rich rocks was present were the main exception.

ii) Cumbrian area

The objective of the study in Cumbria was to collect samples of plant species from the same habitat in order to assess the variation in caesium activity both within and between sites. The habitats used included acidic grassland, bog, dry and wet heath and mire (Table 3). In all one hundred and thirty samples of thirty nine plant taxa were collected from eleven sites. Soil samples were collected from under all vegetation types usually to a depth of 20 cm but often less if rock was encountered. All sites were subject to some degree of sheep grazing and thus there was a direct pathway to man in the sheepmeat. All samples were collected over a period of 18 days in August and September of 1988 within an area where the deposition due to Chernobyl consistently exceeded 500 Bq m^{-2}.

1.3.2 Sampling techniques

i) Vegetation

Bulk vegetation samples were collected from 1 m^2 areas centred on the soil pits. Samples of individual plant species were collected from the immediate vicinity of the sampling points. Vegetation samples were dried at 85° C and ground in a knife mill.

Type	Soil	pH	Typical plants
Bog	Wet deep peat Standing water	3.2-4.7	*Sphagnum, Trichophorum, Eriophorum, Molinia Erica tetralix*
Flush	Unhumified peat	4.2-4.6	*Sphagnum, Juncus effusus*
Wet heath	Wet sandy peat	3.9	*Calluna, Erica tetralix*
Peaty grassland	Wet deep peat	3.7-4.7	*Nardus, Juncus effusus*
Till grassland	Humic silt/clay	3.6-4.9	*Agrostis, Nardus Anthoxanthum odoratum*
Dry Heath	Shallow peat on clay, silt or rock	3.8-5.0	*Calluna, Erica cinerea Vaccinium myrtillus*
Enriched wetland	Shallow peat over rock	6.0-6.9	*Juncus spp., Sphagnum*

Table 3: The broad categories of habitats and their characteristic plants.

Soils - Europe

Soil samples were collected by excavating 20 x 20 cm soil blocks, sectioned changes in the profile or, in the case of uniform soils, at 5 cm intervals. Soil samples were dried at 30° C then sieved through a 2mm mesh.

Soils - Cumbria

Soils were treated differently from the European sites as it was not possible to perform such detailed analysis a large number of samples. Where no horizon was distinguishable the soil was sampled to a depth of 20 cm. If a distinct horizon was present the soil was split at this point and the caesium isotopes determined for each horizon.

1.3.3 Chemical and physical analytical techniques.

A number of chemical parameters were determined, generally according to the methods of Allen et al. (1989). Cation exchange capacity was determined by ammonium saturation of the exchange sites, followed by displacement of the adsorbed ammonium, using potassium, and colourimetric analysis. Exchangeable cations were determined in ammonium acetate extracts, followed by flame photometric analysis of sodium and potassium and atomic absorption analysis of calcium and magnesium. Exchangeable ammonium was determined colourimetrically in potassium chloride extracts. Carbonate was determined by reaction with hydrochloric acid and back-titration. Total organic carbon was determined by dichromate oxidation and back-titration of excess oxidant with iron(II).

The soils were fractionated into sand (63μm-2mm), silt (2-63μm) and clay (<2μm) components. The method used is based on that of Livens & Baxter (1988). The soil is dispersed in distilled water using a high-speed mixer, then sieved mechanically through 2 mm and 63 μm sieves. Each sieve is thoroughly washed and the contents dried at 35° C. The <63 μm fraction is separated by suspension in distilled water and settling (Tanner & Jackson, 1947). After 7 hours 47 minutes, all material coarser than 2 μm has settled 10 cm or more, so the top 10 cm of the suspension contains only particles <2 μm in size. This is siphoned off and the procedure repeated until a clear supernatant remains after settling. At this stage, the remaining material comprises the silt fraction (ie particles from 2 to 63 μm in size). The clay and silt fractions are concentrated by high-speed centrifugation, then dried at 35° C.

There are both advantages and disadvantages to this technique. The advantages include the ability to cope with very large samples (< 100g) and the physical isolation of individual fractions for subsequent radiometric and other analysis. In spite of the use of very large volumes of water in the separations, leaching of caesium is not found to be a problem. This has been verified both by counting the supernatant solution and from caesium budget calculations. The main drawback of this technique is its limited dispersion efficiency, particularly for highly organic or carbonate-rich samples. Conventional particle sizing techniques usually incorporate peroxide and acid treatments to improve dispersion and frequently use dispersing agents such as Calgon as well. Such procedures are unacceptable here, where quantitative recoveries are essential. Furthermore, they are often intended to deal only with small samples and may well use indirect methods (eg density measurements) in the actual analysis.

Caesium isotope activities were determined in all the samples by gamma spectrometry using appropriately calibrated detectors. Spectra were accumulated on 4096 channel MCAs and analysed using Canberra Apogee software.

1.4 RESULTS

1.4.1 Soil Physico-Chemical Properties

i) Site Characteristics

Altogether, ten sites were sampled. Brief details are given in Table 4. Four sites are in north-east Italy, three are in central Norway and three in the Scottish Highlands.

Deposition of both caesium isotopes varies widely, obviously reflecting the range of local climatic conditions at the various sites. Movement of caesium down the profiles is quite variable, with ^{134}Cs confined to the top 15 cm of the 40 cm deep Glen Shirra

Site	Ecosystem	Soil Type	Radiocaesium deposition Cs-134 (Bq/m2)	Cs-137 (Bq/m2)
Tarvisio (Italy)	Conifer Wood	Podzol	1740	11500
Pozzis (Italy)	Beech Wood	Brown Earth	7020	40500
Sella Nevea (Italy)	Conifer Wood	Brown Earth	11500	65500
Stolvizza (Italy)	Alpine Meadow	Calcareous Brown Earth	8760	54000
Geiteseter (Norway)	Open Hillside	Ranker	8200	44100
Stuttgonglia (Norway)	Aspen Wood	Brown Earth	8580	51700
Griningsdalen (Norway)	Fen/Mire	Peat	980	5450
Sherrabeg (Scotland)	Birch Wood	Brown Earth	1350	10100
Glen Shirra (Scotland)	Heather Moor	Podzol	1710	930
Creag Meagaidh (Scotland)	Open Hillside	Ranker	560	2850

Table 4: Site characteristics

podzol and the top 8 cm of the 27 cm Tarvisio podzol profile, but reaching 25 cm in the Pozzis (25 cm deep), Sella Nevea (27 cm deep), Geiteseter (30 cm deep) and Stuttgonglia (30 cm deep) soils. ^{137}Cs derived from weapons fallout is distributed much further down all the soil profiles although the large Chernobyl-derived component is held further up the profiles (Figure 1).

The soils themselves cover a wide range of soil types with correspondingly varied physical and chemical properties, but represent many of the main soil types encountered in upland areas of Europe. Their physico-chemical characteristics obviously reflect the nature of the material from which they are derived, together with human influences and the effects of vegetation type (Table 5). For example, the Italian soils have measurable, if not generally high, carbonate contents in spite of an often low pH, whereas the Norwegian and Scottish soils contain barely measurable quantities of carbonate. This reflects the underlying geology, which is largely dolomitic limestone at the Italian sites and medium to high grade metamorphic rock in Scotland and Norway. The presence of conifers or ericaceous plant species is reflected in the nature of the surface horizons of the soil. Thus, for example, the Italian conifer woods (Sella Nevea and Tarvisio) and the Scottish podzol all have highly organic surface layers, with correspondingly low pH values and very high cation exchange capacities. In contrast, the Italian meadow (Stolvizza) has a very high pH and carbonate content, reflecting its very different vegetation and use.

ii) Particle size relationships

Particle size fractionation of the soils generally shows the same distribution. Some examples are presented in Table 6, together with concentration ratios (Activity in an individual fraction/ Activity in the bulk soil) for the caesium isotopes in the different fractions. The majority of the material being in the sand (63μm-2mm) fraction. This comprises between 74.9% (Sella Nevea) and 95.6% (Stolvizza meadow). The silt (2-63μm) fraction generally accounts for over 10% of the soil, but ranges from 0.9% (Sherrabeg birch wood) to 23.8% (Sella Nevea conifer). The clay (<2μm) fraction is generally in the range 0.5 to 2.0%. There are two exceptions. The Sherrabeg birch wood contains a substantial proportion of fine mica derived from the underlying schist.

SITE	pH	Organic C (%)	CEC (meq/100g)	Carbonate (%)
Tarvisio (Conifer)	3.7	32.5	90.8	2.3
Sella Nevea (Conifer)	4.1	40.7	91.6	2.6
Stolvizza (Meadow)	7.1	17.9	46.6	18.0
Stuttgonglia (Aspen wood)	4.6	8.5	13.7	<0.4
Glen Shirra (Podzol)	4.0	44.0	99.6	<0.4

Table 5: Examples of some physico-chemical properties of the surface horizons of some sample soils.

Altogether, 12.7% of this soil is in the <2μm fraction. The other exception is the Stolvizza meadow soil from which no clay-size material was isolated. This is so remarkable, given the obvious fertility of the soil, that the fractionation was repeated using the technique described by Avery & Bascombe (1982) to check that this result was not an artifact arising from the unusual nature of the soil. This procedure is a standard particle size analysis technique and involves pretreatment with hydrochloric acid, then hydrogen peroxide, to remove carbonate and organic matter respectively. Following these treatments, it was still not possible to isolate any clay-sized material from this soil, implying that it really does contain negligible amounts of clay.

SITE	SAND (%)	SILT (%)	CLAY (%)
Pozzis	81.6	16.3	2.1
(Beech wood)			
CR Cs-134	1.25	0.62	1.19
CR Cs-137	1.33	0.69	1.22
Geiteseter	82.1	17.0	0.9
(Open hillside)			
CR Cs-134	0.60	0.56	3.00
CR Cs-137	0.56	0.62	3.27
Stuttgonglia	72.3	27.0	0.7
(Aspen wood)			
CR Cs-134	1.63	0.54	3.57
CR Cs-137	1.64	0.62	3.65
Sherrabeg	86.4	0.9	12.7
(Birch wood)			
CR Cs-134	1.00	0.52	1.68
CR Cs-137	0.99	0.63	1.80

Table 6: Particle size distribution of some of the sample soils

Within these size fractions there is always a concentration of caesium into the clay-size material. The concentration ratios observed for this fraction range from 1.22 (Pozzis beech wood) to 3.65 (Stuttgonglia aspen wood). There are two distinct patterns observed in the coarser fractions. Whilst the silt fraction generally has a concentration ratio less than one (as low as 0.31 in the Stolvizza meadow), showing that it is not preferentially taking up caesium, the sand fraction displays a range of of concentration ratios spanning 1.00. They range from 0.60 (Geiteseter open hillside) to 1.63 (Stuttgonglia aspen wood).

There is no obvious relationship between the concentration ratio in the sand fraction and the organic content, or any other parameter, which might account for the observed differences in concentration ratio. There is little difference in the concentration ratio values for ^{134}Cs and ^{137}Cs, although the ratio for ^{137}Cs in the clay fractions is usually slightly higher. The exception is the Glen Shirra podzol, where the concentration ratio for ^{134}Cs in the clay fraction is 12.7, but that for ^{137}Cs is only 3.26. This may well reflect the predominance of weapons fallout caesium, rather than Chernobyl-derived material, in the mineral horizons of this soil.

By virtue of its dominance, the sand fraction generally holds the bulk of the caesium inventory in these soils, typically between 70 and 95%. Although the silt fraction comprises a sizeable part of many of the soils, its generally low concentration ratio means that it it is relatively unimportant as a sink for caesium. The low abundance of clay-size material in most of the soils means that it, too, is relatively unimportant as a casium sink, in spite of the concentration ratios generally being greater than 1.00. The major exception is the Sherrabeg birch wood soil, where there is a relatively high clay content and this, together with concentration ratios of 1.68 for ^{134}Cs and 1.80 for ^{137}Cs means that it holds 21% of the ^{134}Cs and 23% of the ^{137}Cs inventories in this soil. As was to be expected the range of soil concentrations was large and the spatial variability within sites was appreciable. ^{137}Cs concentrations ranged from 70 to 1650 Bq kg^{-1} dry weight. Higher concentrations were often found in the low density organic soils. The degree of spatial variability may be judged by the results from three areas of bog at Hesk Fell, Cumbria where within a few metres values for ^{137}Cs of 170, 280 and 850 Bq kg^{-1} were obtained although the bulk density was almost identical. Mineral and peat soils at the same site contrasted markedly. At Gawthwaite, Cumbria the activity in mineral soil was < 100 Bq kg^{-1} whilst two peaty sites gave 580 and 700 Bq kg^{-1} respectively.

Location and species	^{137}Cs	^{134}Cs
ITALY		
Stolvizza (Alpine meadow)		
Arrhenatherum elatius	1110	210
Galium sp.	650	130
Ranunculus sp.	760	149
Stachys alpina	140	30
Trifolium dubium	910	170
Sella nevea (Upland conifer)		
Vaccinium myrtillus-	6680	1220
Venatrum lobelianum	2590	520
Rhytidiadelphus triquetrus	8770	1700
Pozzis (Beechwood)		
Primula vulgaris	2680	520
Aposeris foetida	5370	980
Tarvisio (Conifer woodland)		
Vaccinium myrtillus	1180	210
Pyrola sp.	4720	870
Dicranum majus (tops)	9140	1720
Dicranum majus (bases)	7190	1420
NORWAY		

Geiteseter (Dwarf shrub community)

Bryophytes+lichen under juniper	52500	10500
Juniperus communis (shoots)	890	180
Vaccinium vitis-idea	630	120
Geranium sp.	150	30

Griningsdalen (Mire)

Carex sp.	1050	210
Menyanthese trifoliata	200	40
Betula sp.	250	50

Stuttgonglia (Birchwood)

Juniperus communis (shoots)	680	130
Geranium pratense	100	20
Vaccinium myrtillus	1140	210
Melampyrum pratense	290	50
Trifolium repens	520	100

– –

Besstrond (rocky outcrop)

Lichen thallus samples

1	3540	680
2	24670	4860
3	12880	2550
4	16950	3360
5	17370	3380
6	22150	4440
7	3510	670

SCOTLAND

Sherrabeg (Birchwood)

Agrostis tenuis	780	140

Deschampsia caespitosa	480	80
Dryopteris felix-mas	300	50
Holcus mollis	230	40
Hylocomium splendens	1810	330
Rhytidiadelphus loreus	5856	2730
Creag Meagaidh (Montaine grassland)		
Calluna vulgaris	2880	490
Deschampsia caespitosa	390	60
Empetrum nigrum	760	130
Juncus squarrosus	600	90
Nardus stricta	330	60
Molinia caerulea	600	100
Trichophorum caespitosum	520	70
Vaccinium myrtillus	1430	200
Polytrichum formosum	1160	190
Rhacomitrium lanuginosum	3290	600
Sphagnum rubellum	4510	820
Glen Shirra (Heathland)		
Calluna vulgaris	1860	330
Hypnum cupressiforme	5800	1170

Table 7: Caesium concentrations in individual species from the European areas Bq kg^{-1} dry weight.

1.4.2 Concentrations in individual species

i) European

The vegetation results from the European sites are presented in Table 7. There is considerable variation between species even from the same habitat. However, some generalisations can be made.

The lower plants (bryophytes and lichens) are clearly different from the others on account of their mode of nutrition. They trap aerosols and absorb ground water largely by capillary action rather than by internal translocation. In all instances the lower plants contained much higher concentrations of the caesium isotopes. Lichens in particular often contain concentrations orders of magnitude greater than flowering plants. Samples from Norway often reached in excess of 20,000 Bq kg^{-1} of ^{137}Cs. This is particularly relevant in the high latitudes where lichens are an important component of winter feed for animals such as reindeer. Bryophytes do not form an important component of the food of grazing animals although they may be eaten incidentally with other plants. The high concentrations in these plants, however, does mean that radiocaesium is held at or near the soil surface and may be released into the system when the plants die for recycling via the roots of the higher plants.

There is much evidence from other work that the ericaceous species accumulate radiocaesium to a greater extent than other plant species. This has proved to be the case for many of the sites in the present study. If *Vaccinium* is considered it will be seen that in all three European sites it is one of the species giving the highest concentrations of radiocaesium with values ranging from 630 Bq kg^{-1} in Norway to 6700 Bq kg^{-1} in a conifer wood in Italy. It was only possible to obtain the ericaceous shrub *Calluna vulgaris* from the Scottish site but concentrations were high at 2880 and 1860 Bq kg^{-1}.

Radiocaesium concentrations in the higher plants generally lay in the range 100 - 1000 Bq kg^{-1} the highest values being found at the Italian sites. Of note is the beechwood near Pozzis, where the

Species	N	Activity ^{137}Cs				^{134}Cs		Range of Conc.ratio
		Max	Min	Mean	S.E.	Max	Min	
Juncus effusus	14	1341	14	378	116	259	-	0.05-7.53
Nardus stricta	13	868	10	237	86	186	-	0.01-2.48
Calluna vulgaris	7	4464	39	915	598	999	9	0.70-6.37
Pteridium aquilinum	7	700	11	236	96	147	-	0.05-1.14
Juncus squarrosus	6	1989	101	832	301	429	30	0.10-5.68
Erica cinerea	5	93	28	52	12	21	-	0.14-0.53
Erica tetralix	3	2324	179	1212	620	553	40	0.30-5.94
Vaccinium myrtillus	4	220	78	141	33	44	12	0.24-0.36
Polytrichum commune	10	4722	339	1589	410	1176	82	0.41-23.5
Sphagnum spp.	10	5918	411	1992	551	1444	93	0.70-17.1

Table 8: Range of radiocaesium activities for species of frequent occurence (Bq kg^{-1} dry weight)

sparse ground vegetation consisted mainly of *Primula vulgaris* and *Aposeris foetida*. Here despite the base-rich nature of the soil both species had concentrations of ^{137}Cs in excess of 2600 Bq kg^{-1} in their tissues. Radiocaesium concentrations between species on the same site within a few centimetres of each other can be very differnt. This is best illustrated by the site at Creaaig Meagaidh in Scotland. Here eight higher plants were all sampled from the same area. They range in concentration was from 330 to 2870 Bq kg^{-1} with the highest values being associated with the ericaceous plants *Calluna vulgaris* and *Vaccinium myrtillus*.

Plant species	Habitat	^{137}Cs	^{134}Cs
Sphagnum spp.	Raised Bog	5918	1444
Polytrichum commune	Raised Bog	4722	1176
Calluna vulgaris	Wet Heath	4464	999
Carex spp.	Peaty grassland	4045	751
Sphagnum spp.	Valley Bog	3992	966
Trichophorum cespitosum	Raised Bog	2916	623
Polytrichum commune	Valley Bog	2514	601
Trichophorum cespitosum	Valley Bog	2381	494
Sphagnum spp.	Valley Bog	2369	576
Erica tetralix	Raised Bog	2324	553
Sphagnum spp.	Fell flush	2141	455
Polytrichum commune	Valley Bog	2116	506
Juncus squarrosus	Valley Bog	1989	429
Molinia caerulea	Raised Bog	1561	374
Polytrichum commune	Grassland	1461	334
Polytrichum commune	Grassland	1428	308
Molinia caerulea	Valley Bog	1418	318
Juncus squarrosus	Valley Bog	1341	259
Sphagnum spp.	Fell flush	1300	316

Table 9: Plant species showing highest recorded activity (Bq kg^{-1} dry weight)

It is important to note that, at the three European sites, high concentrations of radiocaesium are not necessarily always associated with plants growing on highly organic soils. in addition the concentrations of radionuclides in species growing on the same site can be highly variable, which has important implications when the selective habits of the grazing animal are considered.

ii) Cumbria

Radiocaesium concentrations in vegetation varied widely both between and within species as did the transfer factor. The twenty plant samples showing the highest radiocaesium activity are shown in Table 8 with those of most frequent occurrence in Table 9. Those species growing on organic waterlogged soils contained the highest levels of caesium activity. As was the case in the European samples, the bryophytes often contained the greatest amounts. The range of concentrations found within individual species was large but there is some evidence that the ericaceous species accumulate radiocaesium to the greatest extent. Of the higher plants *Calluna vulgaris* and *Erica tetralix* with ^{137}Cs concentrations of 4460 and 2320 Bq kg^{-1} respectively have the highest maximum concentrations.

Ericaceous species were investigated in more detail as it was possible to compare their performance in a number of habitats. The data are presented in Table 10. Within each site the Chernobyl deposition would be identical for all species. It can be seen that, with the exception of one site, *Calluna vulgaris* consistently yields the highest activities. The other species usually follow in the order *Calluna* > *Vaccinium* > *Erica* which is in agreement with the findings of other workers (Bunzl & Kracke, 1984).

Site	Species	^{137}Cs	^{137}Cs Soil
Grasmoor	*Calluna vulgaris*	469	669
NGR NY167198	*Vaccinium myrtillus*	220	
360m	*Erica cinerea*	93	
Hindscarth	*Calluna vulgaris*	366	730
NGR NY218177	*Empetrum nigrum*	107	
490m	*Vaccinium myrtillus*	78	
	Erica cinerea	33	
Yewbarrow	*Calluna vulgaris*	227	76
NGR NY16709	*Erica cinerea*	40	
85m			
Robinson	*Calluna vulgaris*	126	500
NGR NY193176	*Vaccinium myrtillus*	96	
335m	*Erica cinerea*	65	
Gawthwaite	*Calluna vulgaris*	39	98
NGR SD263848	*Vaccinium myrtillus*	170	713
200m	*Erica cinerea*	28	98

Table 10: Comparative contamination of ericoid species on dry highland sites (Bq kg^{-1} dry weight)

1.4.3 Soil/Plant transfer

i) Transfer to bulk vegetation

Examples of the transfer factors from soil to the standing vegetation crop are presented in Table 11. They are expressed both in terms of area (Bq/m^2 in veg/ Bq/m^2 in top 10 cm of soil) and in terms of weight (Bq/kg in veg/ Bq/kg in top 10 cm of soil). Expression in terms of area is more useful for many purposes since it removes the effects of vegetation yield and soil density. However, for comparison with the results for individual plant species (see later), it is necessary to express the transfer factors on a weight basis.

A considerable range of transfer factors is observed. The lowest is that at the Stuttgonglia aspen wood site, whilst the highest are found at the Scottish sites.

A preliminary statistical analysis of the data has been carried out. The transfer factors for the caesium isotopes have been correlated with a number of soil parameters (CEC, pH, % Sand, % Silt, % Clay, carbonate, organic carbon content, exchangeable ammonium, sodium, potassium, calcium and magnesium). Whilst a number of very good correlations arise between various soil parameters, no parameter is well correlated with the transfer factor. However, a few points do arise. For example, the transfer factors are poorly negatively correlated with % Clay, % Carbonate and pH. They are poorly positively correlated with organic carbon and cation exchange capacity. Interestingly, there is almost no correlation between the transfer factors and exchangeable potassium. Better correlations exist with exchangeable sodium and ammonium. Nevertheless, it must be emphasised again that all the correlations are relatively poor, with the highest correlation

SITE	TRANSFER FACTOR (AREA BASIS)		TRANSFER FACTOR (WEIGHT BASIS)	
	Cs-134	Cs-137	Cs-134	Cs-137
Pozzis (Beech wood)	1.18E-03	1.07E-03	2.84E-01	2.71E-01
Stolvizza (Meadow)	5.41E-03	4.27E-03	4.36E-01	3.58E-01
Stuttgonglia (Aspen wood)	6.95E-04	6.57E-04	2.16E-01	1.99E-01
Sherrabeg (Birch wood)	4.61E-03	3.35E-03	1.12E+00	8.79E-01
Creag Meagaidh (Ranker)	2.96E-02	2.54E-02	1.55E+00	1.44E+00
Glen Shirra (Podzol)	2.04E-02	1.72E-02	2.14E+00	1.95E+00

Table 11: Examples of soil-plant transfer factors for bulk vegetation

coefficients between transfer factors and any soil parameter being in the range 0.5 to 0.6. It is possible that a more complex relationship (other than the simple linear model used in determining correlations) exists between the transfer factors and some soil parameter(s). This has yet to be tested. Alternatively, if there is a dominant controlling mechanism at all, it is not reflected in any of the physico-chemical properties of the soils which have been determined. The most obvious mechanisms of this type are the biological processes which operate in the soil.

ii) The transfer of caesium isotopes to individual plant species

The transfer factor for the caesium isotopes to the dominant plant species at each site are presented in Table 12.

ITALY		TRANSFER FACTOR	
		Cs-134	Cs-137
Tarvisio	Vaccinium myrtillus	7.24E-01	6.63E-01
(Conifer)	Pyrola sp.	3.00E+00	2.65E+00
	Dicranum sp. (tops)	5.93E+00	5.13E+00
	Dicranum sp. (bases)	4.90E+00	4.04E+00
Pozzis	Primula vulgaris	2.89E-01	1.49E+00
(Beech)	Aposeris foetida	5.44E-01	2.98E+00
Sella Nevea	Vaccinium myrtillus	9.84E-01	9.78E-01
(Conifer)	Veratrum lobelianum	3.79E-01	3.79E-01
	Rhytidiadelphus triquetrus	1.37E+00	1.28E+00
Stolvizza	Arrhenatherum elatius	4.20E-01	3.65E-01
(Meadow)	Galium sp.	2.60E-01	2.14E-01
	Ranunculus sp.	3.00E-01	2.50E-01
	Stachys alpina	6.00E-02	4.61E-02
	Trifolium dubium	3.40E-01	2.99E-01
NORWAY			
Geiteseter	Mixed bryos & lichen	2.33E+01	2.12E+01
(Open hillside)	Juniper (shoots)	4.00E-01	3.59E-01
	Vaccinium	2.67E-01	2.54E-01
	Large geranium	6.67E-02	6.05E-02
Stuttgonglia	Juniper (shoots)	2.95E-01	2.83E-01
(Aspen wood)	Small geranium	4.09E-02	4.17E-02
	Vaccinium	4.77E-01	4.75E-01
	Melampyrum pratense	1.16E-01	1.21E-01
	Trifolium repens	2.27E-01	2.17E-01
Griningsdalen	Carex	1.31E-01	1.20E-01
(Mire/Fen)	Menyanthese trifoliata	2.38E-02	2.18E-02
	Betula	3.00E-02	2.75E-02

SCOTLAND

Sherrabeg	Holcus mollis	1.12E+00	8.79E-01
(Birch wood)	Deschampsia caespitosa	3.62E-01	3.15E-01
	Agrostis tenuis	7.62E-01	6.56E-01
	Dryopteris felix-mas	4.48E-01	4.10E-01
	Hylocomium splendens	3.12E+00	2.48E+00
	Rhytidiadelphus loreus	2.60E+01	8.02E+00
Glen Shirra	Calluna vulgaris	1.91E+00	1.75E+00
(Podzol)	Hypnum cupressiforme	6.86E+00	5.47E+00
Creag Meagaidh	Deschampsia caespitosa	4.41E-01	4.34E-01
(Ranker)	Empetrum nigrum	9.63E-01	8.68E-01
	Nardus stricta	4.04E-01	3.76E-01
	Trichophorum caespitosum	5.44E-01	5.95E-01
	Vaccinium myrtillus	1.50E+00	1.62E+00
	Calluna vulgaris	3.60E+00	3.27E+00
	Juncus squarrosus	6.69E-01	6.76E-01
	Molinia caerulea	7.21E-01	6.77E-01
	Polytrichum formosum	1.40E+00	1.31E+00
	Rhacomitrium lanuginosum	4.40E+00	3.73E+00
	Sphagnum rubellum	6.02E+00	5.13E+00

Table 12: Transfer factors to individual plant species

1.5 DISCUSSION

Soil/plant transfer factors are highest for the bryophtes and lichens. This is to be expected in view of their mode of nutrition, although these values are not truly comparable with those of the higher plants. Nevertheless, these species can be a significant component of many of the vegetation types studied and the part they play in holding radionuclides in the upper parts of the system is important. Maximum transfer to Man by this route will occur in northern countries where, at certain times of the year, lichens can be a major item in the food of grazing animals.

Transfers to both bulk vegetation samples and individual species are larger for ^{134}Cs than ^{137}Cs. This indicates a greater availability of the Chernobyl derived caesium and the existence within the soil caesium pool of a less plant-available pre-Chernobyl component. On the longterm this situation may change, also the Chernobyl derived caesium may become less available with time. Within a site there is often a high degree of variability between species but with a few exceptions the ericaceous shrubs such as *Vaccinium myrtillus* and *Calluna vulgaris* gave the highest transfer factors. It was not possible to sample fungi in the present study but the concentrations in the fruiting bodies can be high and lead to high levels in grazing animals such as moose (Zach et al., 1989).

Direct comparisons of ecosystems are difficult due to the varying combinations of species and the localised variations in the Chernobyl fallout. However, with some exceptions, both in the northern European and the more intensive Cumbrian sampling, the higher plant activities have been found in association with the damp, organic soils with lower activities associated with the drier mineral soils. The calcareous soils at the Italian sites are very different in character to the mineral soils at the northern European sites, and casium remains highly mobile in them. Further investigations into the soil geochemistry are in progess to investigate the high mobility of the Chernobyl material in these soils. Caesium behaviour in the Italian podsols, even though they are developed over calcareous rock, is similar to that in the northern European ones. This is due to the control of caesium behaviour by the upper inorganic horizons of the soil, rather than the deeper layers whixh is where the Italian podzols differ from the others.

In most instances there is poor correspondance between soil and plant concentration. This leads to the wide range of concentration factors found (Table 11) and obviously leads to difficulties in attempting to predict the levels of radiocaesium in vegetation from that in the soil. It is therefore the mobility of the radiocaesium in the soil rather than its absolute amount which is important.

1.6 CONCLUSIONS

In the natural and semi-natural ecosystems studied there are important pathways back to man. Radiocaesium uptake by vegetation is variable and, within a site, depends on the individual species concerned. It is clear that, within Europe, there were still differences between Chernobyl-derived and fallout-derived caesium at the time of sampling. Soil-plant transfer factors are consistently larger for ^{134}Cs reflecting a greater plant availability of Chernobyl derived material. In some of the soil profiles, notably the podzols, Chernobyl-derived material is held much further up the profile than that from weapons fallout and is thus more likely to be found in the rooting zone. In the longterm the availability of Chernobyl derived caesium may decrease. Activity of ^{137}Cs tends to be more enhanced in the clay sized fractions and this may well reflect the ageing of weapons fallout. There appears to be no one dominant physico-chemical process in these natural and semi-natural ecosystems but it is possible that the soil biology has a strong influence on caesium mobility.

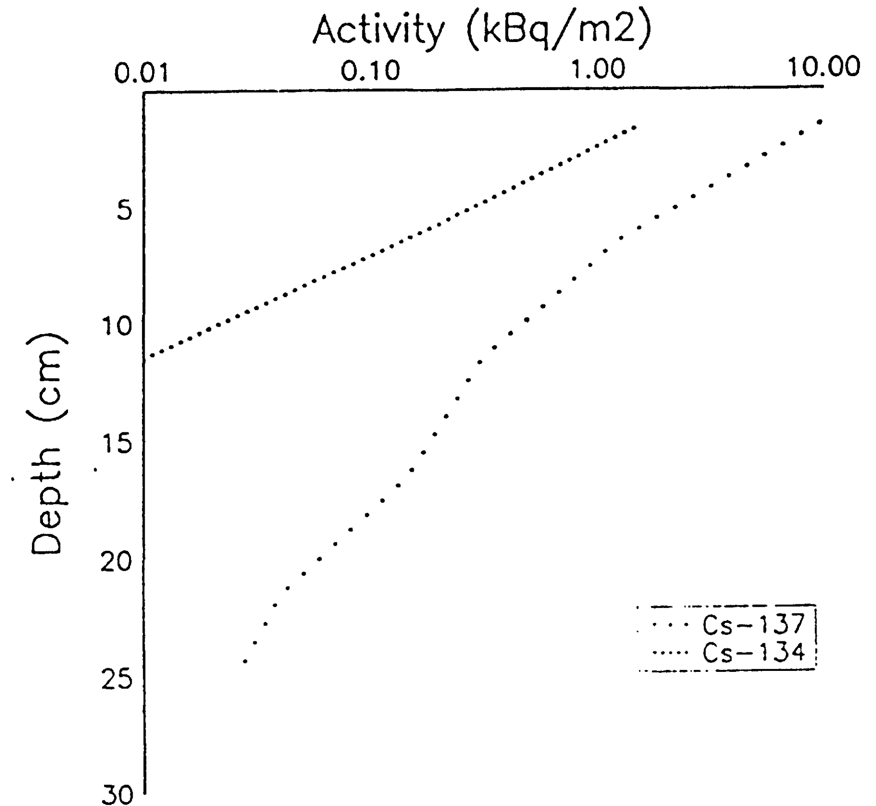


Fig. 1a: Caesium distribution in Tarvisio podzol profile

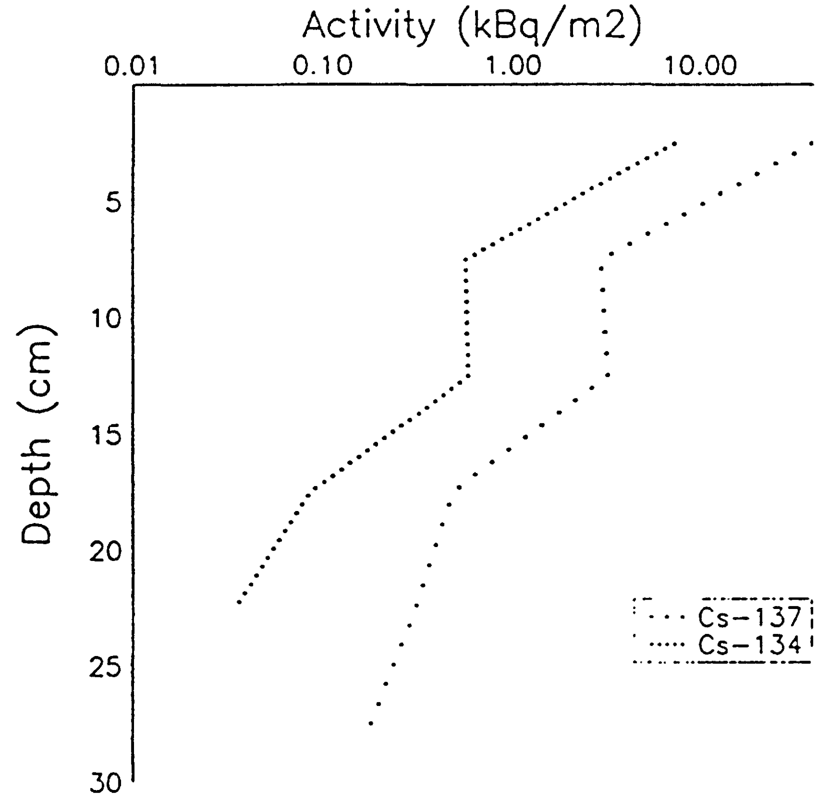


Fig. 1b: Caesium distribution in Stuttgonglio brown earth profile

Figure 1: Caesium distribution in soil profile

2. AGRICULTURAL ECOSYSTEMS

2.1 STATE-OF-THE-ART

2.1.1 Hypothesis

Radionuclides released after the accident in the Chernobyl nuclear power plant in April 1986 caused a widespread contamination of the environment in European countries (CCRX, 1989). After several months, when effects of short-lived radionuclides had disappeared, the transfer of cesium isotopes through the food chain appeared to be a major pathway for future doses to the human population (UNSCEAR, 1988). Besides large regional differences in deposited amounts of radionuclides, regional effects were also observed for the transfer of Cs from soils to crops. A matter of concern was the high uptake from upland soils in northern parts of Europe (data Horrill 1986 in: IUR, 1990). The hypothesis was raised that Cs originating from Chernobyl might be more easily transferred from soil to plant than Cs from other sources (fallout from nuclear weapons, discharges from nuclear industry or artificial addition to soils).

2.1.2 Objectives

Main aim of this study was to examine whether soil-to-plant transfer of Cs originating from Chernobyl is higher indeed than that from other sources. A comparison of both groups of transfer data is complicated because the variability in soil-to-plant transfer factors is usually large (Coughtrey & Thorne, 1983). The evaluation should therefore be based on many measurements. Furthermore, differences in environmental conditions between the locations where transfer was measured should be taken into consideration because these conditions influence the transfer. This implies that effects of in this context important environmental factors had to be distinguished and quantified. Therefore, a second aim was to study influences of environmental parameters on soil-to-plant transfer of Cs. From this study correction factors were derived which can be used for the comparison of transfer factors determined under different environmental conditions.

2.1.3 Literature review

Introduction

Soil-to-plant transfer of radionuclides, particularly of radioactive Cs and Sr isotopes, has been the subject of numerous studies, documenting a multiplicity of factors conditioning transfer. Furthermore a number of comprehensive reviews are devoted to the transfer of Cs (Coughtrey & Thorne, 1983; Fredriksson et al., 1966; Koranda & Robison, 1978; Ng et al., 1982; Nielsen & Strandberg, 1988). Apart from some references to Sr this study will mainly be confined to Cs.

Since the soil-plant relationship is directly controlled by two groups of factors, i.e. factors affecting the specific genetically-determined nutrient uptake potential of the plant and those affecting the availability of the mineral in the soil, these groups of variables will be discussed separately.

Differences in TF_{SP} among plant species and varieties

For an evaluation of genotypic differences in uptake potential, various species should be grown under similar conditions (a.o. Evans & Dekker, 1968). In most cases, however, either soil-plant combinations had to be compared, or crops grown on soils contaminated in different ways (Bunzl & Kracke, 1989; Frissel & Koster, 1987; Ng et al., 1982; Steffens et al., 1988). For Cs the major conclusion is: its transfer to vegetable crops is generally higher than to forage crops, which in turn is generally higher than the value found for cereals. TF_{SP} values measured by Evans & Dekker (1968) -on one specific soil- varied between 0.004 (*Linum usitatissimum*) and 0.47 (leaves of *Beta vulgaris*). Frissel & Koster (1987) distinguished five crop classes with a geometric mean TF_{SP} of about 0.015 for the class with lowest TF_{SP} (cereals) and of 0.20 for the one with the highest TF_{SP} (vegetables). Within more restricted groups of plants, e.g. families or genera, variations of a factor 2 to 3 were observed. For different varieties of the same species the recorded variability in accumulation of strontium is of the same order of magnitude (Rasmusson et al., 1963).

Internal distribution and redistribution

The above-mentioned difference between types of crops is partly due to a comparison of different organs or tissues, since Cs is inhomogeneously distributed within the plant. Per unit dry weight fruits and seeds accumulate less Cs than roots and leaves (Evans & Dekker, 1968; Frissel & Koster, 1987; Nishita et al., 1961). Coughtrey & Thorne (1983) concluded on the average 50% of root-absorbed Cs to be translocated to the shoot. As compared to Sr, Cs is highly mobile, whether absorbed by the roots (Handley & Babcock, 1972) or after foliar contamination (Middleton, 1958). It can be translocated out of the leaves towards either new grown leaves, fruits or storage organs (Russell, 1963). The final distribution pattern depends on the stage of growth at which contamination occurred and the time elapsed between contamination and sampling (Middleton & Squire, 1963). Due to its higher mobility Cs is usually more uniformly distributed than Sr (Nishita et al., 1961).

Transfer as a function of plant age

Few studies on the accumulation of radionuclides throughout the growth period are available, since main interest was focussed on concentrations in mature crops. Interference of external factors, such as rainfall and temperature, or an inhomogeneous distribution of the contaminant in the rooting zone hampered a correct interpretation of field experiments on this matter (Bergamini et al., 1970; Bunzl & Kracke, 1989; Coughtrey et al., 1990; Kirton et al., 1990). In field studies the Cs content of plants was shown to fluctuate throughout the growth period. Changes of a factor 2 to 3 are observed (Coughtrey et al., 1990; Kirton et al., 1990) but 7 to 10-fold changes have been described as well (Bunzl & Kracke, 1989).

Experiments under controlled conditions demonstrated that the Cs concentration in hydroponically grown crops continuously increased with age. For shoots of *Brassica oleracea* and *Salsola kali* a 2 to 3-fold increase in TF_{SP} was observed after 9 respectively 12 weeks of growth (Routson & Cataldo, 1978; Weaver et al., 1981). Bergamini et al. (1970) found that the transfer for *Trifolium pratense* changed markedly during flowering periods due to the high uptake potential of the reproductive organs.

Uptake processes - interactions

Results of short term experiments on the inhibiting effect of a variety of monovalent cations upon Cs ad- and absorption by excised roots showed that inhibition was proportional to the cation concentration, K being most effective in lowering sorption (Handley & Overstreet, 1961). In entire plants growth and physiological activity, however, may cause different results. Jackson et al. (1965) e.g. showed NH_4^+ to increase uptake, whereas it reduced sorption by excised roots (Handley & Overstreet, 1961). The so called observed ratio, i.e. $(Cs/K)_{plant}/(Cs/K)_{solution}$, used to compare the relative rates of uptake of Cs and K, appeared to depend both on the ratio and concentrations in the nutrient medium (Cline & Hungate, 1960; Middleton et al., 1960; Nishita et al., 1962a). In most cases the observed ratio was smaller than unity, indicating a more pronounced uptake of K. Cline (1962) and Nishita et al. (1962a) observed only limited effects of K on uptake of Cs over wide concentration ranges. In general uptake was reduced by increased levels of K, when substrate K was low. Furthermore it was shown that Cs and K were not similarly distributed over the different crop parts (Souty et al., 1975).

Experiments on Cs uptake from soils mostly confirm the above-mentioned results from experiments with nutrient solutions. Plants grown on soils with a high available K content were low in Cs (Evans & Dekker, 1966; Fredriksson et al., 1966; Fredriksson, 1970a; Reitemeier & Menzel, 1960). Therefore addition of K was suggested to be an effective countermeasure against Cs uptake when K levels in soil are low. Bunzl & Kracke (1989) observed K and Cs to vary accordingly in two grass species, resulting in a nearly constant ratio of Cs to K during the year. In conclusion, absorption of Cs by plants bears no constant relationship to that of K in the close manner shown by Ca and Sr (Russell, 1963).

Uptake processes - concentration effects

Environmental assessment models treat plant uptake of elements as a simple concentration ratio of concentration in plant tissue to total concentration in the soil. This simplification assumes that plant and soil concentrations are linearly related. However, from soil fertility studies it is well-known that the uptake of many essential and non-essential elements does not follow a linear relationship. Different mechanisms and models for the observed uptake patterns have been developed and studied in relation to uptake of radionuclides (Borstlap, 1981; Epstein, 1966; Nissen et al., 1980; Sheppard & Sheppard, 1985; Sheppard & Evenden, 1988; Simon & Ibrahim, 1987). Both linear and non-linear relations have been described for a variety of nuclides, on soils as well as on nutrient solutions (Nishita et al., 1962a; Routson & Cataldo, 1978; Sheppard & Sheppard, 1985; Sheppard & Evenden, 1988; Simon & Ibrahim, 1987; Van Loon et al., 1989). Different explanations are possible:

1. For some plant types and elements it was suggested that the radionuclide concentration in the soil should surpass a threshold before uptake occurs (Nishita et al., 1962a; Simon & Ibrahim, 1987). Furthermore the effect of an addition of stable Cs to the soil on the uptake of radiocesium was shown to depend on the sequence of addition of the isotopes (Wallace et al., 1982). Both observations seem to be caused by soil sorption characteristics -i.e. specific and irreversible adsorption- since partitioning between soil solid and liquid phase depends on the total soil concentration (Nishita et al., 1962b; Sheppard & Evenden, 1988). As a result, comparison between the concentration of an element in the soil liquid and that in the plant will be necessary for a separate analysis of plant uptake processes (Sheppard & Evenden, 1988).

2. The uptake pattern depends on the concentration range studied. When applying trace amounts of Cs and Sr to soils Routson & Cataldo found that transfer factor values were independent of the concentration, covering more than three respectively five orders of magnitude, and were constant for a given plant age (Routson & Cataldo, 1978). Application of macro-amounts of Cs resulted in a non-linear relationship due to saturation of metabolic processes and possibly plant intoxication (Nishita et al., 1962a). Linearity was argued to be related to the fact that soil-added radioisotopes of Cs (and Sr) trace large endogenous elemental pools of their stable counterparts or chemical analogues, thus without changing the total concentration. Furthermore linearity for non-essential elements would be due to non-interference up to the toxicity treshold (Sheppard & Sheppard, 1985).

Climatic effects

Hardly any information was available on the effect of meteorological or seasonal factors on soil-to-plant transfer of Cs. Theoretical studies concentrated on the consequences of the moment of an atmospheric release on the ratio between indirect (root uptake) and direct contamination of plants (interception) as a function of development of the standing crop (Boeri, 1988; Simmonds, 1985). Studies on fallout from nuclear weapon tests in the early sixties tackled comparable questions (a.o. Csupka et al., 1967).

Only few, recent field studies allowed for an indirect and partial quantification of the effect of meteorological factors. Interpretation was, however, difficult e.g. because climatic changes may affect plant physiological processes as well as chemical equilibria in the soil system. Comparable fluctuations in transfer throughout the growth season, at different locations or on different soils and correlated variation in the concentration of different nuclides in the plant suggested climatic factors to be of importance (Lembrechts et al., 1990; Mascanzoni, 1988; Sandalls et al., 1989). Time dependent changes in concentration of a factor of 2 to 3 are commonly observed (Kirton et al., 1990;

Sandalls et al., 1989) but higher values, up to a factor of 11, have been described as well (Bunzl & Kracke, 1989; Steffens et al., 1988). Resuspension, rain splash, weathering losses, etc. are other aspects related to meteorological conditions which influence transfer (Coughtrey & Thorne, 1983).

Effects of soil pH

The effect of soil pH on the transfer of radiocesium into plants is very often descripted as the most important soil parameter, although in most cases soil pH is only a substitute for several other soil parameters, especially for the cation fraction. Nevertheless, soil pH is a parameter which is easy to determine and seems to be a fairly good estimation parameter for TF_{SP}.

Adriano et al. (1984) tested the effect of liming a soil to pH 6.8 on TF_{SP} using clover, soybean, grass, wheat and corn leaves grown on 3 different soils. In nearly all cases he found a significant decrease of radiocesium in plants due to the effect of increasing pH. Kühn et al. (1984) illustrated a close non-linear relationship between TF_{SP} and pH of soil. Kerpen (1986) investigated the transfer of radiocesium from small pots into cereals and demonstrated a significant correlation between pH and plant uptake of ^{137}Cs.

Even Boikat (1982) found a correlation between pH and TF_{SP} in small pots. Schuller et al. (1988) carried out a stepwise multiple regression and was able to explain 67 % of the variance of TF_{SP} by soil pH. They found, that all the other soil parameters contributed only few percent of the variance.

Effect of potassium content in soil

Due to the similar chemical behaviour of potassium and cesium, the content of potassium in soil has an important influence on the TF_{SP} of cesium.

Menzel (1954) and Nishita et al. (1960) carried out important investigations on the question of the influence of potassium content in soil on the plant uptake of radiocesium. Both authors found a very close relationship between the uptake of Cs and the K content in soil. Especially on soils with a low level of available K, the uptake of Cs was extremly high. The higher the K content in soil, the lower is the effect of an additional amount of K.

Fredriksson et al. (1966a) performed a study of plant uptake of ^{137}Cs under field conditions from three different soils at three different rates of potassium level. Plant uptake was found to be decreased substantially with an increased rate of potassium fertilization. In his work 1970 Fredriksson found a correlation between ^{137}Cs content in plants and K content in soil, which is due to clay content, too.

These results were similar to results of Orlovius et al. (1987), who showed that the increase of ^{137}Cs content in plants originating from Chernobyl fallout was extremly high on soils with a low content of K. On an extreme K deficiency site of a grassland ^{137}Cs in plants was reduced by more than 60 % due to a K fertilization, while on the better supplied sites ^{137}Cs was reduced only by 30 %.

Effect of clay, silt and sand content in soil

The grain size of the soil is of importance for the TF_{SP} because of the capability of clay minerals to exchange and to fix cesium.

Already in 1940 Schachtschabel investigated the important role of clay minerals for ion exchange. Sawhney (1964,1966) studies sorption and fixation of cesium by different soil minerals. Differences of sorption capacity between montmorillonite, kaolinite, vermiculite and micas are shown.

Shalhevet (1973) demonstrated differences between the fixation of ^{137}Cs in interlayer spaces and edges fixation. The lower content of illitic clay minerals and the higher content of kaolinites is one reason for the high TF_{SP} on tropical and sub-tropical soils (Fredriksson 1970b).

If clay minerals are missing, sorption may be taken over by the soil organic matter. Often an increase of soil organic matter is combined with a decrease of clay minerals. Bell et al. (1988) show an increase of ^{137}Cs content in heather due to an increase of organic matter (10 %, 50 %, 90 %) and a decrease of clay content (18 %, 10 %, 2 %) with data of Mills. The low content of clay minerals combined with a high content of soil organic matter might be the reason for the high transfer factors for grass grown on organic soils in northern europe (Eriksson, 1988).

Influence of soil organic matter

Although the sorption capacity of organic matter is sometimes higher than the sorption capacity of clay minerals, adsorbed ions may be exchanged by other ions. If cesium ions are adsorbed on soil organic matter, the fixation in the interlayer spaces of clay minerals is reduced and the ions may remain in an exchangeable form.

Barber (1964) investigated the uptake of ^{137}Cs by ryegrass on six different soils with wide variations in soil organic matter (1.3 % O.M. - 9.95 % O.M.). He found a high positive correlation between the uptake of ^{137}Cs and the content of soil organic matter. No relationship was determined between uptake and percentage of clay and exchangeable potassium.

Fredriksson (1966) studied the uptake of ^{137}Cs into hay from pot experiments as well as in a field survey. He found a very close relationship between ^{137}Cs uptake and content of organic matter in soil as it increases with an increasing content of organic matter. The mean contamination with ^{137}Cs in hay from the soil with the highest content of organic matter was 30 % higher than in the hay from the soil with lowest content.

Similar results are found by Abbazov et al. (1979). The positive correlation coefficient between ^{137}Cs accumulation in wheat and rice grain and the humus content ($r=0.63$) is only negligible lower than the correlation to exchangeable potassium ($r=-0.75$).

Effect of soil moisture

The effect of soil moisture on the uptake of ^{137}Cs into plants is uncertain. One possibility for getting further information about the influence of increasing soil moisture on TF_{SP} is to study the passage of radiocesium from soil into soil solution. Kulikov et al. (1973) showed, that the influence of soil moisture becomes more and more important with increasing soil moisture. On a low level of soil moisture content the influence is neglectable, while ^{137}Cs passage into soil solution increases on a medium level. On the other hand, an extremely high level of soil moisture (coefficient of hydration > 2.0) decreases the concentration of ^{137}Cs in soil solution.

Shalhevet (1973) carried out 3 different greenhouse experiments with natural soils as well as with mixtures of fine sand and vermiculite in a split root experiment. In the two split root experiments Shalhevet found a significant reduction in radiocesium uptake with decreasing moisture content. In the third experiment he found a significant increase of ^{137}Cs uptake with decreasing soil moisture content. This effect was especially high on the soil with predominant kaolinit content. On the other soil with a high content of illit, this effect was less obvious.

Karavaeva et al. (1979) investigated the possibility of a vertical movement of ^{137}Cs due to different regimes of soil moisture, which is also important for the possibility of plant uptake. No effect of different moisture contents was noticed even when the soil was established on 100 % of moisture capacity.

2.1.4 Data sources

Data on transfer of Chernobyl originating Cs from soil to plants are relatively scarce as most measurements were executed shortly after deposition, when interception of radionuclides by the vegetation dominated over uptake from the soil. Furthermore, in many surveys the Cs contamination of soils and crops was measured according to different schemes and consequently not at the same location, whereas a combination of both measurements at the same location is necessary to evaluation soil-to-plant transfer. The EC data base in Ispra (Italy), the IAEA data base on the Chernobyl accident and the post-Chernobyl data base of the nordic countries provided therefore no relevant information for this study (pers. comm. management of the databases). The regular literature shows the same difficulty and as a result hardly any useful data are found.

The major part of the about 500 Chernobyl transfer data used in this study are, as a consequence, from special experimental studies by institutes participating in the post-Chernobyl research program of the European Community. Data were collected by:

- Belli and Sansone (Italy)
- Bilo (FRG)
- Fagniart (Belgium)
- Horrill (UK)
- Mac Neill (Ireland)

In addition, a considerable part of the data was obtained from Eriksson et al. (Sweden).

All transfer factors together with extensive information on experimental and environmental parameters have been collected in a data base by the International Union of Radioecologists (IUR, 1990). The data base also contains about 1200 transfer factors for Cs originating from non-Chernobyl sources of contamination, i.e. fallout from nuclear weapons tests of the sixties, discharges from nuclear industry or artificial (experimental) additions to the soil. It includes results from pot, lysimeter and field experiments, by:

- Boikat and Fink (FRG): pot, artificial contamination
- Cawse (UK): field, environmental contamination
- Frissel and Pennders (Netherlands): field, environmental contamination
- Gatti (Italy): field, artificial contamination
- Grogan, Minski and Bell (UK): pot, artificial contamination
- Haak (Sweden): field, environmental or artificial contamination

- Haisch, Capriel and Stark (FRG): lysimeter, artificial contamination and field, environmental contamination
- Ham (UK): lysimeter, artificial contamination
- Heine and Wiechen (FRG): field, environmental contamination
- Kirchmann and Fagniart (Belgium): field, artificial contamination
- Kuehn, Handl and Schuller (FRG): field, environmental contamination
- Mascanzoni (Sweden); lysimeter, artificial contamination
- Popplewell, Ham and Stather (UK): pot, environmental contamination
- Roussel et al. (France): field, environmental contamination and lysimeter, artificial contamination
- Steffens, Mittelstaedt and Fuehr (FRG): lysimeter, artificial contamination
- Stoutjesdijk, Sinnaeve, van Ginkel and Lembrechts (Netherlands): lysimeter, artificial contamination
- Szabova (CSSR): field, environmental contamination

The data have in common that they (1) present a realistic quantification of Cs transfer under prevailing environmental conditions and (2) provide specific information on environmental parameters together with each transfer factor.

2.2 METHODS

2.2.1 Experimental studies

2.2.1.1 Field Studies - Description of sampling areas

South and West Germany

After the reactor accident in Chernobyl, very different soil contaminations due to different radionuclides were observed in the Federal Republic of Germany. These differences primarily depended on the precipitation quantities which varied considerably from place to place. Thus in Munich from 29.4. to 11.5.1986 a radioactivity of approx. 20 kBq/m^2 ^{137}Cs was deposited, whereas for the same period in Berlin merely one tenth of this value was recorded (Radiation Protection Commission of the Federal Republic of Germany (1987): Effects of the Reactor Accident in Chernobyl on the Federal Republic of Germany. SSK Publication, no. 7). Soil contamination maxima were observed with the tracer nuclide ^{137}Cs in Upper Swabia, Lower Bavaria and in the Berchtesgaden district.

Questions arising in the post-Chernobyl programme of the EC (Evaluation of Data on the Transfer of Radionuclides in the Food Chain) indicated that a study of transfer factors in various soil types from different areas would be of particular interest. In particular, a comparison of transfer factors in soils of the same type in various regions should provide information about the possibility of forecasting transfer factors for certain soil types. Due to the level of radioactive depositions after the reactor accident at Chernobyl, it seemed possible to implement the study of transfer factors by various plants on different soils under "natural" conditions.

In order to discover the differences in transfer factors between various soil types it was necessary to locate a region with clearly differentiated soil types. On the other hand, the distribution of these soil types should not be too extensive in order to eliminate other parameters such as climatic effects. The transfer factors for this region thus determined should then be compared with transfer factors of the same soil types from a different region. Apart from this requirement made on the soil types, a second aspect was the presence of a high soil contamination. Only in more highly contaminated areas can Chernobyl fallout be determined by a reasonable expenditure of time and analysis work.

-Upper Swabia

The above-mentioned requirements to be made on the working area, high soil contamination with $^{134/137}$Cs as well as clearly differing soil types, were found to be present in and around the village of Tannheim in Upper Swabia. On the one hand, the Tannheim area represented a highly contaminated region ($\approx$ 40,000 Bq/m^2 ^{137}Cs), on the other hand the fluvially characterized geomorphology led to recent, homogeneous soil types which are clearly delimited with respect to each other. The complete glacial series can be found on a molasse basement superimposed by the Ice Age. Next to the terminal moraine with a more or less distinct loess veil is found a fill terrace created by the River Iller and also a recent river flood plain. While the topography of the terminal moraine displays high relief intensity, the lower terrace is almost flat. The meadow on the flood plain is interspersed in places with abandoned river courses and gravel ridges which are generally visible in the terrain as a slightly ondulating surface. The transitions of the respective geomorphological units can be seen in the field as clear steps. The transition from the meadow to the lower terrace can generally be recognized as a step in the terrain 2 - 4 m in height, while the rise towards the terminal moraine is as much as 40 - 50 metres.

Various types of soil have been formed on the respective geomorphological units in accordance with the rock formation, climate, vegetation and topography. Whereas the soils are exceptionally uniform on the almost level lower terrace, several soil types are present within one geomorphological unit on the meadows and moraine.

Stony to acid brown soils (Braunerde) have been formed on the lower terrace on silicatic Alpine gravels, which also contain carbonate in places. This soil type is generally present covering the entire lower terrace. Only at the transition to the terminal moraine have hydromorphous soil types been formed at some locations by the corresponding topography and slope water. Utilization as pasture causes fens to form here, which are in part degraded by amelioration.

In the meadow situation, alluvial soils are generally found (allochthone braune Auenböden, allochthone Vega). Due to the extremely limy initial substrate, a subdivision into calcareous alluvial soil (allochthone Kalkvega) is possible. The sedimentation of the fluvial colluvium is in part so low that the Ap horizon lies directly on the gravel. In this case the soil is to be regarded as rendzina or pararendzina.

On a limy layer of decomposed rock with a loess veil on the terminal moraine, soils have resulted influenced by the perched water. The processes of lessivage and pseudogley formation have led to soils ranging from pseudogleys to degraded brown soil (Parabraunerde)-pseudogleys under the given climatic conditions. The formation of pseudogleys varied greatly due to the great differences in relief on the terminal moraine. Thus, for example, degraded brown soils and brown soils can be observed on the hill tops and high slopes.

Representative soil types in this highly contaminated region around Tannheim in Upper Swabia can therefore be regarded as alluvial soils on the flood plain, brown soil on the lower terrace and pseudogley on the moraine.

The classification according to the soil map of the FAO (1974, 1981) is Calcaric Fluvisol for the alluvial soil and Cambisol for the brown soil on the terrace. The soils with a perched water table range between Luvisol and Planosol. Due to the fact that in most cases the influence of the perched water table was more obvious than the influence of illuvial accumulation of clay, the typical soil of the terminal moraine was classified as Planosol with a medium base saturation.

Tables 13 to 15 describe three sampling points representative of these soil types. The section designations follow the recommendations made by the Soil Science Working Group of the Regional Geological Offices of the Federal Republic of Germany (AG Bodenkunde, 1982).

Depth(cm)	Horizon	Description
0-30	Ap	Dark brown (7.5 YR 3/3), weakly fine-sandy, humus silt with numerous micaceous minerals. Very weak pseudogley formation can be recognized in places. Weakly penetrated by fine roots, strong carbonate reaction.
30-75	M_1	Brownish-grey (7.5 YR 4/2) to grey-brown (7.5 YR 5/3) weakly fine-sandy, weakly humus silt with numerous micaceous minerals. Very strong to extremely strong carbonate reaction.
>75	Cv	Brownish-grey (7.5 YR 4/2), somewhat more densely stratified. Very weakly humus, silty fine sand with numerous well rounded limy pebbles. Extremely strong carbonate reaction

Table 13: Section designation of sampling point OS33 (Calcaric Fluvisol) in Upper Swabia on the flood plain (easting 3583.8; northing 5319.6; topographical map TK 7926).

Depth(cm)	Horizon	Description
0-25	Ap	Dark brown (7.5 YR 3/3), humus, weakly clayey-silty sand with numerous poorley rounded stones. Weakly penetrated by fine roots. Very weak carbonate reaction.
25-50	Bv_1	Brown (7.5 YR 4/3), weakly humus, weakly clay-silty fine sand with numerous poorley rounded stones. Very weakly penetrated by fine roots. No or very weak carbonate reaction.
50-70	Bv_2	Partly brown (7.5 YR 4/4), partly light brown (7.5 YR 5/6), very weakly humus, silty fine

		sand with numerous poorley rounded stones. Moderately dense, weak carbonate reaction.
>70	Bv/Cv	Light brown (7.5 YR 5/6), decomposed rock, partly merging into loamy fine sand. Moderately dense, medium carbonate reaction.

Table 14: Section designation of sampling point OS29 (Cambisol) in Upper Swabia on the terrace (easting 3581.5; northing 5320.4; topographical map TK 7926).

Table 16 shows the most important soil parameters for these three typical sections. All values refer to the air-dry fine soil of the Ap horizon.

In taking samples of soil and plant material from the respective geomorphological units, care was taken that the soils corresponded to these typical profiles as uniformly as possible.

-Lower Bavaria

Apart from the region in Upper Swabia described above, the attempt was made to find similar soil types in a different, equally highly contaminated region. Due to the fact that the area around Tannheim is characterized by a river originating in the northern limestone Alps, and also the Flysch zone of the Alpine foothills, it was reasonable to suppose that similar surface forms and soil types would be found in the valleys of the Isar and Inn.

Due to the similarly fairly high soil contamination in Lower Bavaria, the valley of the Isar between Landau an der Isar and Plattling was selected as the reference area. The Chernobyl fallout there was between 40,000 and 50,000 Bq $^{137}Cs/m^2$.

Depth(cm)	Horizon	Description
0-22	Ap	Dark brown (7.5 YR 3/3), humus, fine-sandy silt with numerous root residues and small stones (2-5 mm). Very occasionally weakly perceptible pseudogley mottling. Medium to moderately hight, macroscopically perceptible biological activity, no carbonate reaction.
22-55	Sw	Moderately dense, traces of humus, grey-orange to strong-orange brown (7.5 YR 6/3 - 6/8) fine sandy silt (loess loam) with high stone content. Occasionally pseudogley mottling.
55-70	Sw/Sd	Dense, strong orange-brown (7.5 YR 6/8) silty fine sand with very high stone content. Moderate proportion of pseudogley mottling.
>70	Cv/Sd	Very dense, gritty-sandy silt with high stone content. Clear, very extensive pseudogley mottling, mainly 7.5 YR 6/8 - 5/8.

Table 15: Section designation of sampling point OS25 (Planosol) in Upper Swabia on the moraine (easting 3579.5; northing 5318.6; topographical map TK 7926).

Examinations of the terrain in May 1989 indicated that this typography is similarly characterized by fluvial influence but displayed soil types which could not be compared to those of the Iller Valley.

	Calcaric Fluvisol	Cambisol	Planosol
	OS33	OS29	OS25
Clay(%)	4.20	13.90	11.70
Fine silt(%)	5.35	13.70	12.40
Medium silt(%)	15.80	12.55	19.70
Coarse silt(%)	31.55	15.70	21.95
Fine sand(%)	33.75	18.25	13.50
Medium sand(%)	8.50	17.15	12.40
Coarse sand(%)	0.85	8.75	8.35
N_{tot}(%)	0.235	0.140	0.195
C_{org}(%)	2.47	2.65	2.35
C/N	10.51	11.00	12.05
pH($CaCl_2$)	7.40	5.25	5.75
Na^+(meq/100g)	0.01	0.01	0.20
K^+(meq/100g)	0.60	1.15	0.13
Ca^{++}(meq/100g)	15.08	6.90	11.20
Mg^{++}(meq/100g)	1.38	0.91	1.26
Total Exch. Bases (meq/100 g)	17.07	8.97	12.79
CEC (meq/100 g)	17.07	14.80	23.20
Percentage Base Saturation (%)	100.0	60.61	55.13

Table 16: Some soil parameters for the three sampling points described (Upper Swabia).

In a distinctive flood plain topography, soils are formed on Holocene sediments, which may be classified as gleys. On recent Holocene accretions, turfy moulder gleys and fen soils may also be observed, whereas in the older Holocene meadows eutrophic brown soils can be found on sands. In contrast to the lower terrace of the Iller, the lower terrace of the Isar is covered with loess here so that deep degraded brown soils are formed.

These two geomorphologically comparable topographies have therefore formed completely different soil types on similar terrace levels. It therefore did not appear meaningful to take samples in the Isar Valley in order to compare transfer factors.

-North Rhine-Westphalia

Soil types were found in North Rhine-Westphalia (NRW) comparable to the alluvial and degraded brown soil-pseudogleys of Upper Swabia. For this reason, soil samples were similarly taken from these soils and also samples of winter cereals, and a section designation made of the sampled soils. Due to pressure of time during the winter cereal harvest taking place at the same time in Upper Swabia and NRW, it was only possible to obtain eleven samples.

The weak soil contamination of approx. 1000 Bq $^{137}Cs/m^2$ meant that the plant contamination was too low for transfer factors to be determined for these areas. It was only possible to significantly detect ^{137}Cs (0.14 Bq/kg) in one sample. Only the $^{134/137}Cs$ contents for the soils are therefore given in Chapter 2.3.1.1.

Friuli-Venezia Giulia plain (Northeast of Italy).

After Chernobyl accident, the Friuli-Venezia Giulia region, located in the North-Eastern part of Italy, was more affected than other regions of Italy by the radioactive fallout. Therefore the Italian Directorate for Nuclear Safety and Health Protection (ENEA-DISP) and the Regional Centre for Agricultural Experimentation of Friuli-Venezia Giulia region (C.R.S.A.) have developed a multi-years radioecological programme to study the principal components of terrestrial and aquatic ecosystems.

Activities in the agricultural environment started in May 1986 and required a preliminary stage to determine:

- the ^{137}Cs soil deposition in the Friuli-Venezia Giulia region;

- the principal parameters necessary to characterize the complex agricultural environment (physical and chemical parameters of soils, agricultural practices, etc.).

In this paragraph the principal features of soils in the Friuli plain and the values of ^{137}Cs soil depositions for the regional territory will be described.

The Friuli plain covers an area of about 300,000 hectares. It extends from the foot of the Carnian and Julian Alps to the Adriatic sea, with a special connection to the Tagliamento river Moraine Amphitheatre. Figure 2 shows the 7 geographical zones in which the Friuli plain is traditionally subdivided. This area is bounded by the Livenza river and the Veneto region boundary to the south-west and by the Yugoslav frontier and the Gorizia and Trieste karst to the east. On the basis of a regional soils study (Comel A. et al., 1982), C.R.S.A. has divided the plain into 13 principal soil classes according to their origin, prevalent composition and agricultural characteristics (Tab.17). The sampling stations have been located in 11 of these classes that represent 96% of Friuli plain.

DESCRIPTION OF THE SOIL CLASSES		CLASSES
Weathered aluvial soils of the High Plain	Weathering depth 30-40 cm	1
	Weathering depth 40-70 cm	2
	Weathering depth over 70 cm	3
Alluvial soils	Predominantly gravelly	4
	Predominantly sandy	5
	Predominantly clayey	6
Alluvial soils influenced by rising groundwate	Predominantly gravelly	7
	Predominantly sandy	8
	Predominantly clayey	9
Soils of the low-lying parts of moraine amphitheatre		10
Soils of the moraine hills		11
Soils derived from weathered coastal sands and dunes		12
Recently reclaimed soils from the lagoon border zone		13

Table 17: Simplified soil classification of the Friuli plain

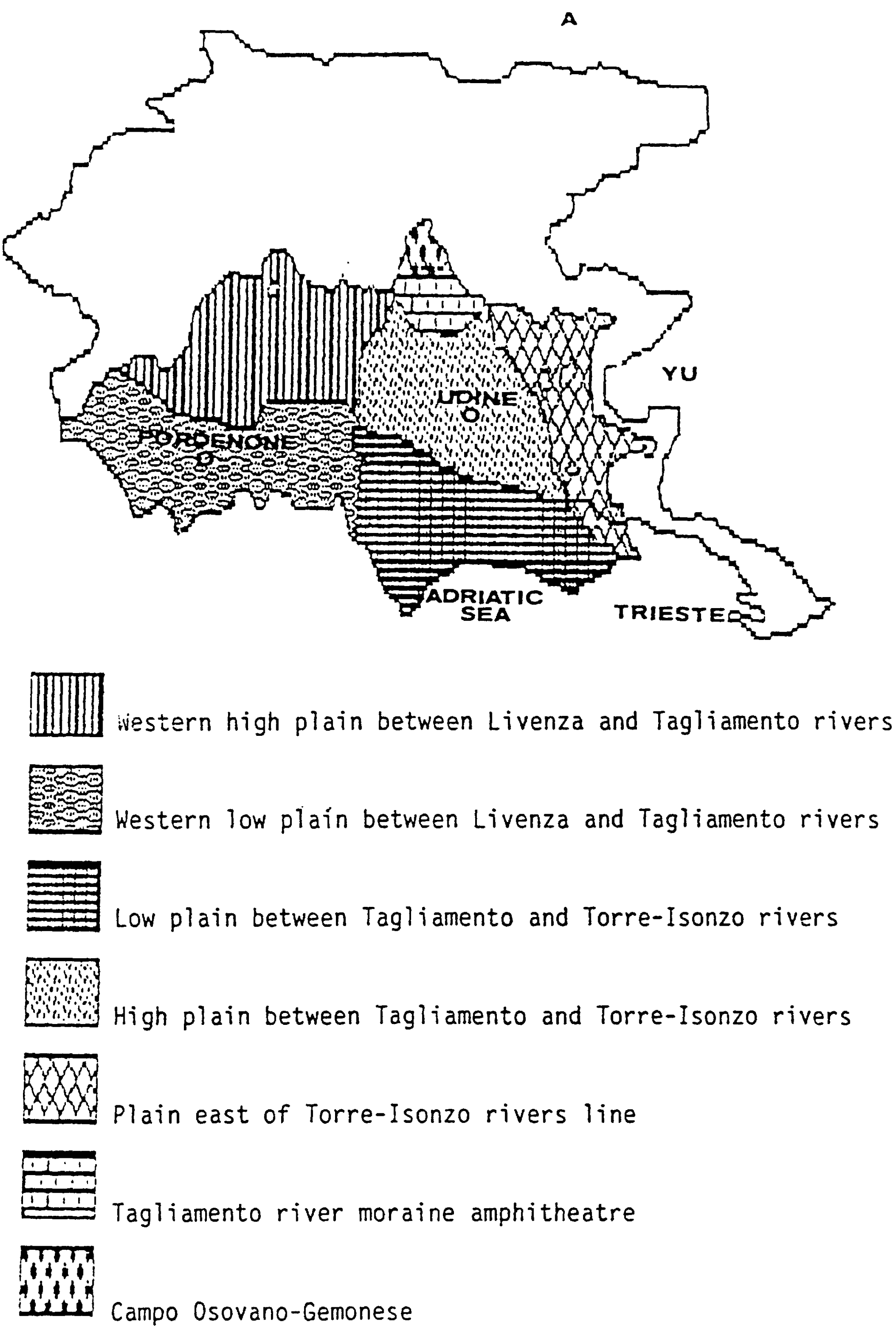


Figure 2: Geographical subdivision of Friuli plain

Table 18 shows the correspondence between these classes and the FAO/UNESCO international classification system. Each of the geographical zones in which the Friuli plain was divided (Fig.2) contains more than one of the 13 soil classes identified.

Class	FAO/UNESCO classification	Nature
1	Dystric Ferralic Cambisols	Clay
2	Eutric Ferralic Cambisols	Clay
3	Eutric Ferralic Luvisols	Clay
4	Dystric Fluvisols and Dystric Calcaric Cambisols	Gravel
5	Eutric Fluvisols, Eutric Calcaric Fluvisols and Humic Cambisols	Sand/Clay
6	Eutric Fluvisols, Calcaric Cambisols	Clay-Silt
8	Eutric Fluvisols, Gleysols	Sand-Silt Clay
9	Humic Cambisols, Gleysols	Clay
10	Eutric Fluvisols, Dystric Fluvisols, Gleysols, Histosols	Silt
11	Dystric Lithosols, Rankers, Rendzinas	Gravel
12	Solonchaks, Solonetz, Histosols, Phaeozems	Clay

Table 18: Correspondence between soil classification scheme used for the Friuli plain and the FAO/UNESCO system; predominant particle size in each class.

The description that follows, will present the principal soil characteristics of the following area: the High Plain, the Low Plain and the Tagliamento river Moraine Amphitheatre.

The soils of the Friuli plain have a common origin, comprising or being derived from alluvial deposits of fluvial or fluvial-glacial origin. In the northern part, where the currents are faster flowing, there is a zone composed of coarse, gravelly material, called the "High Plain". Further south, is present a zone composed mainly of sand and clay called the "Low Plain". After the withdrawal of the surface water, these zones have been exposed to more or less prolonged weathering and now show distinctive surface modifications.

In the FAO/UNESCO classification the term "Fluvisols" is used for soils that have developed on top of recent alluvial deposits, while the term used for older soils may depend on the specific characteristics to be emphasized. Many such older soils are classified as "Cambisols", soils in which mild weathering of the bed-rock has produced changes in colour, structure and consistency. On the basis of their agricultural potential, soils subject to periodic flooding or largely composed of gravel are called "Dystric" (a synonym for infertile). The term "Eutric", on the other hand, means fertile and is used, for example, for soils with a certain width of finer particles (sandy or clayey) that allow consistent plant cover to develop. Thus some zones of the western High Plain formed by the Meduna and Cellina rivers and composed mainly of calcareous-dolomitic gravel can be classified as Dystric Calcaric Fluvisols.

Then there are soils that are still gravelly but with a thin (about 10 cm), blackish, humus-rich surface layer consisting of organic matter accumulated from a thin steppe-like vegetation. The layer below is greyish in colour, due to the mixing of organic particles with the white subsequent gravels. This second layer is of variable thickness (20-40 cm) and rests on practically unweathered original calcareous-dolomitic gravel. Soils of this type in the High Plain are classified as Dystric Calcaric Cambisols. An improvement in the agricultural quality of soils can be observed in zones where there is a layer of finer particles on the surface, capable of holding rain water for a significant length of time. This encourages rapid demolition of organic matter and causes an accumulation of inorganic residues, in particular ferro-aluminium hydrates that impart a reddish colour to the soil. Soils of this type, of alluvial origin but with a surface horizon rich in ferro-aluminium compounds, are called Ferralic Cambisols. The addition of adjective Eutric or Dystric defines their greater or lesser thickness.

These changes in the physical and chemical conditions of the soil may lead to a progressive under saturation of the circulating water and thus produce a certain instability of the colloids, which tend to migrate to the layers below. Here they find an environment richer in basic elements and reprecipitate, giving rise to an accumulation of illuvial or B horizon (that is, formed of deposits of substances that have been leached from higher up) which is typical of Luvisols.

Proceeding in the direction of the sea, and therefore referring mainly to the Low Plain soils, we find that finer material of alluvial origin often obstructs underground water flow and the water is therefore forced to the surface. Where the soil is more or less constantly soaked in this way it takes on a typical marsh appearance, with a humic layer that may range from a few centimetres to over a metre in thickness and with vegetation composed of sedges and reeds. These are called Mollic Gleysols (gley = muddy mass). If groundwater rises to the surface only occasionally, the effect on the soil is weaker. Such soils are infertile and are therefore classified as Dystric Calcaric Cambisols.

The sandy-clayey soils of the Low Plain that are not affected directly by groundwater are nevertheless rather impermeable and permit only slow infiltration of rainwater. At the point where it is halted, this water may give rise to deposits of calcium or iron salts, depending on the substances present in the surface layer. Soils with these characteristics are classified as Calcic Cambisols, or else Humic Cambisols if there is a marked surface humus-rich layer.

In the zone nearest to the sea the long-established marsh vegetation has directly or indirectly influenced soil development.

An organic surface layer has formed that may be peaty (when the plant remains are still recognizable) or humic (if the remains are reduced to a mass of humus-rich soil). These soils are called Histosols and Phaeozems respectively.

Salinity may also influence soil composition to a more or less pronounced degree. Thus saline deposits may form on the surface or at other levels, with saturation of colloidal compounds. Solonchaks are very saline soils and Solonetz those with some residues of salinity.

The Tagliamento Moraine Amphitheatre deserves a separate discussion. This is a transitional environment situated between the central Friuli plain and the mountains. It is an area of particularly varied topography, with hills and plains formed of predominantly calcareous-dolomitic gravelly material with marked variations in soil types. Where erosion has exposed the moraine, the soils are classified as Lithosols, soils that are more or less the same as the bed-rock. The term Rankers is used for the slopes, with a surface horizon rich in organic matter and the layer below in an advanced state of decalcification. Rendzinas comprise a humus-rich surface horizon resting on very calcareous bed-rock.

The soils of the big inter-moraine valleys and those of the wide plain adjacent to the Tagliamento Moraine Amphitheatre (the Campo Osovano-gemonese) are similar to the recent alluvial soil of the Friuli plain and are therefore included in the corresponding classes discussed above.

The ^{137}Cs deposition in the Friuli-Venezia Giulia region has been estimated from precipitation data recorded by the regional rainfall network (composed by 135 meteorological stations) and from the ^{137}Cs air concentration data, both measured daily (Belli M. et al., 1988). Wet depositions were estimated taking into account the following hypotesis:

-uniform air distribution of ^{137}Cs;

-height of rain clouds: 1000 m.

The daily precipitation values were interpolated on a grid of 20*20 rectangles, each of 6.75 km by 6 km. ^{137}Cs dry deposition was also estimated using a ^{137}Cs deposition velocity of 10^{-3} m/s. The dry deposition contribution was 1.9 kBq/m^2. Figure 3 shows the ^{137}Cs total washout following the passage of the contaminated cloud over the italian territory (from 30 April 1986 to 09 May 1986). In the high plain the ^{137}Cs depositions ranged from 5 to 15 kBq/m^2, with values until 20 kBq/m^2 in the area of Campo Osovano-gemonese and Tagliamento moraine anphitheatre. In the low plain the ^{137}Cs depositions ranged from 2 to 10 kBq/m^2, with values less than 2 kBq/m^2 in the area where the weather was dry during the same period.

On the basis of soil characteristics and ^{137}Cs deposition, 26 sampling stations have been selected to evaluate the ^{137}Cs transfer factors from soil to forage crops. Table 19 gives indications on these sampling stations grouped according to the above mentioned pedological classes. In this table the area of the Friuli plain covered by each soil class, the estimated ^{137}Cs deposition and the existing fodder crops are indicated.

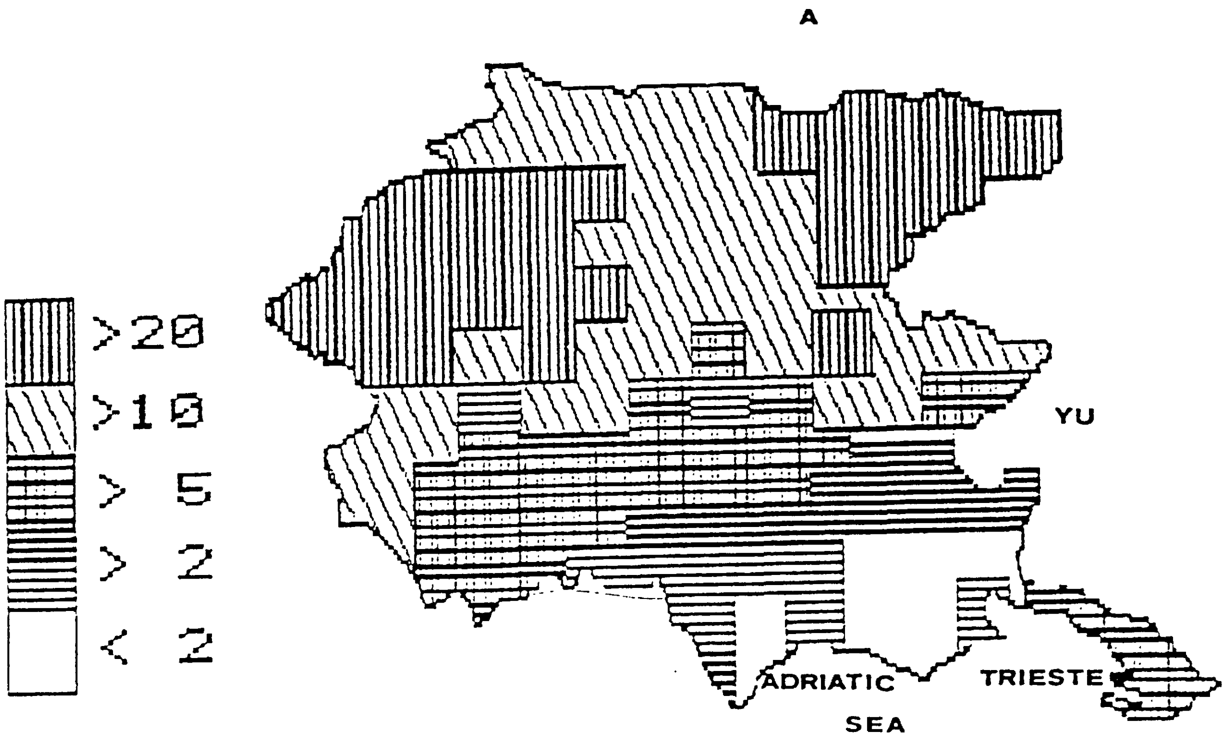


Figure 3: ^{137}Cs wet deposition (kBq/m^2)

Soil class	% on the plain surface	Sampling stations	Fodder crops	^{137}Cs (kBq/m^2)
1	16.4	03ab 04ab 11ab	alfalfa alfalfa alfalfa	10÷15 10÷15 5÷10
2	16.2	02ab 02ac 09ab 12ab 14ab	alfalfa alfalfa alfalfa alfalfa alfalfa	10÷15 10÷15 2÷5 5÷10 2÷5
3	1.7	08ab	alfalfa	5÷10
4	19.	57ab 32ab 58ab 33ab	alfalfa alfalfa alfalfa alfalfa	5÷10 2÷5 2÷5 < 2
5	17.2	15ab 34ab	alfalfa alfalfa	2÷5 < 2
6	13.6	13ab 38ab 29ab 37ab	alfalfa alfalfa alfalfa alfalfa	10÷15 5÷10 2÷5 2÷5
8	1.7	51ab	alfalfa	2÷5
9	1.6	27ab	meadow	15÷20
10	4.3	24ab 39ab 48ab	alfalfa meadow alfalfa	15÷20 5÷10 5÷10
11	3.5	19ab	alfalfa	15÷20
12	0.6	44ab	alfalfa	< 2

Table 19: Sampling stations selected in the Friuli plain.

2.2.1.2 Lysimeter Experiment

After the reactor accident at Chernobyl, a drastic increase of radioactivity was recorded in sewage sludges, which was attributed to the runoff of contaminated precipitation water from sealed surfaces. In the municipal sewage works at Tannheim, the contamination of the sludge was up to 10,218 Bq ^{137}Cs and 2733 Bq ^{134}Cs/kg dry solid matter (1.8.88). Sewage sludge is generally used in the area as fertilizer since it is poor in heavy metals and extremely rich in nutrients. Due to contamination of the sewage sludge an increase in soil radioactivity was to be expected in this case, and due to root transfer possibly also a rise in radioactivity in the plant. For this reason, the Institute of Radioagronomy at the Nuclear Research Centre Jülich started a long-term lysimeter experiment in 1989 to study whether radioactivity in cultivated plants increases due to the application of sewage sludge.

The experiment consists of 4 lysimeters whose soil was removed from an arable or pasture site. The soil is a podzolic gleysol with a sand fraction of 88 % in the Ap horizon. The other soil data (pH value 5.4, Total Exchangeable Bases 5.95 meq/100 g, CEC 11.00 meq/100 g) also lead one to expect high transfer values. This soil was selected so that any recommendation for or against the use of sewage sludge would be based on a "worst-case model".

In March 1989, the maximum permissible quantity according to the German Sewage Sludge Ordinance of 1 kg dry solid matter/m^2 was applied to two arable and two pasture lysimeters. With a dry solid matter proportion of 5 %, the quantity corresponded to 20 l of sewage sludge/m^2. After applying the sewage sludge, the lysimeters were sprinkled with 5 mm of precipitation to rinse off any sewage sludge adhering to the grass. Four further lysimeters functioned as controls without sewage sludge fertilization. The radioactivity applied with the sewage sludge amounted to 12,951 Bq $^{134+137}$Cs/m^2. After a few days the sewage sludge was worked in on the arable lysimeters to a depth of 0-10 cm. The control lysimeters were fertilized with complete fertilizer corresponding to the nutrient content of the sewage sludge in order to achieve comparable conditions, at least with respect to the macronutrients nitrogen, phosphorus and potassium.

Potatoes were planted on the arable lysimeters in April 1989. During June all the lysimeters were sprinkled at irregular intervals with 5 mm of water in order to compensate for the dry period. The potatoes were harvested at the end of June. In August, lettuce was planted on the arable lysimeters after N fertilization. The lettuce was then harvested in September 1989.

The pasture lysimeters were mown on May 19 and July 19, 1989. After each cut the lysimeters were fertilized with N and also irrigated again, corresponding to 5 mm of precipitation. The grass was dried in the sun and then chopped in a food processor (Moulinette).

After harvesting, the potatoes were cleaned intensively with water and after drying were cut into slices. After determining the plant fresh mass the potatoes were sealed into PVC foil and deep frozen. The lettuce was similarly washed after harvesting and was frozen after the fresh weight had been determined.

2.2.1.3 Sampling and Preparation of Soil and Plant Material

South and West Germany

Samples to determine the transfer factors for winter cereals in the three soil types described were taken immediately before the cereal harvest. It was thus possible to store the cereal samples without further drying. Samples of cereals in the field were taken with the aid of electrically operated shears, maintaining a minimum distance of approx. 10 m from the edge of the field. The entire plant, without roots, was harvested on a surface area of 1 m^2. During harvesting a minimum cutting height of 20 cm was observed in order to avoid carrying over soil particles clinging to the plant. Additional ears were harvested on a further square metre in order to obtain sufficient sample material for the measurements. The plant material was placed into paper bags and stored.

The soil samples were then taken on these surfaces after harvesting. The samples were taken by a hand drill of the HUMAX SH 300/35 type. The samples were taken in the form of soil cores 35 mm in diameter and 30 cm in length protected in a PVC sleeve. This sleeve is then closed by a lid and the soil cores are ready for transportation. Depending on the stone content of the soil, between 5 and 10 soil cores were taken per sampling point.

The section designation was similarly carried out on these areas by means of a hand drill of the Pürckhauer type, 1 m in length. However, due to the slight depth of the soil it was frequently impossible to take samples as deep as 1 m.

All soil samples ready for measurement were encased in PVC sleeves 30 cm in length. In the laboratory the sleeves were divided into the segments 0-20 cm and 20-30 cm. The soil cores were broken up slightly in a mortar and then dried in a drying chamber at 50°C until a constant weight was achieved. The material was then further manually reduced and sieved to < 2 mm.

The winter cereals obtained during the field work were similarly dried off at 50°C. The ears were threshed in a threshing machine and the grain filled into bottles. The straw was chopped in a rotary mill supplied by the Retsch company to approx. 1.5 mm.

The deep-frozen plant material from the lysimeter experiments was dried in a freeze drier of the DURA-DRY type. After drying the plant material was reduced in a food processor and homogenized, and then placed into a 1-l Marinelli beaker.

Northeast of Italy

Undisturbed soil and forage crop samples were collected at the 26 sampling stations from September 1986 to October 1987. The forage crops include alfalfa and natural meadow grass. The alfalfa was planted between March 1983 and March 1986 and the soil was not tilled afterwards.

During 1986 the forage crops were collected once in September, five months after Chernobyl accident. In 1987 alfalfa was sampled monthly at each cut from May to October, while natural meadow grass was sampled in June and September.

Soils were collected at each sampling station in autumn 1986 and 1987. During 1986 each soil sample has been collected over an area of 900 cm^2 to a depth of 5 cm. In 1987 soil samples have been collected over an area of 900 cm^2 at the following depths: 0-5 cm and 5-10 cm. The error associated to this sampling method is about 20%.

The soil samples taken were air dried, sieved to 2 mm and carefully mixed.

1 m^2 of crops was cut at 2 cm above ground and dried at 60° C.

In addition, in all collected soil samples the following parameters have been determined:

- percentage of fine soil (particles diameters less than 2 mm. Percentage expressed over dry weight of whole sample);
- percentage of sand (particles diameter between 2 and 0.05 mm. Percentage expressed over fine fraction of sample);
- percentage of silt (particles diameter between 0.05 and 0.002 mm);
- percentage of clay (particles diameter less than 0.002 mm);
- soil pH in water solution and in KCl 1N.
- organic matter content indicated as weight percentage (organic carbon % (Walkley-Black method) by 1.72);
- extractable phosphorus expressed in mg/kg (Olsen method);
- extractable potassium, magnesium, calcium expressed in mg/kg (Neutral ammonium acetate 1 N method);

- Cation exchange capacity in meq/100 g (Sodium saturation method - Sodium acetate at pH 8.2).

2.2.1.4 Measurement of radiocesium

Germany

The gamma radioactivity was measured with the aid of a lead-shielded Ge(Li) detector and a multichannel analyser (MCA). The measured spectrum was evaluated on a PDP-11/84 using the SPECTRAN$^+$ program package.

The sample material was placed into a Marinelli beaker with a lid for measurement purposes. The soil samples were measured in a 0.5-l Marinelli beaker, whereas due to the lower specific radioactivity the plant material was placed into a 1-l Marinelli beaker.

The measuring time for the soil samples varied between 3 and 12 hours. The plant samples had to be measured for up to 72 hours in order to reduce the measuring error. In the analysis of soil samples for ^{137}Cs this error was in the region of 1 % and for ^{134}Cs about 3 %. The error for the plant samples varied between 5 and 10 % for ^{137}Cs, depending on the radioactivity. No calculated measuring error can be given for ^{134}Cs in the plant samples since the program was only able to identify ^{134}Cs in plants on rare occasions. The error can be estimated as being between 20 and 30 %.

Italy

Caesium isotopes were determined in all samples by direct gamma spectrometry, counting weighted amounts of sample contained in a 1 l Marinelli beaker, using a HPGe-detector and a multichannel analyser.

2.2.2 Statistical studies

2.2.2.1 Application of the relative comparison method

The choise of the method to evaluate a large amount of soil-to-plant transfer factors (= TF_{SP}) depends on the aim of the study, the nature of the data and the way they have been sampled (Sokal & Rohlf, 1981). One method is multiple linear regression analysis, amongst other methods used by the IUR Working Group to derive best estimates and confidence intervals for TF_{SP} of non-Chernobyl Cs under different environmental circumstances (Heisterkamp & Koster, 1986; Keen, 1984; Frissel & Koster, 1987).

Aim of this study is to compare Chernobyl and non-Chernobyl transfer data and to quantify environmental influences relevant for this comparison. Application of multiple linear regression is for this purpose hindered by the following factors:

- Data in the collection are clustered, each cluster relating to specific environmental and experimental conditions. Due to the presence of these clusters effects of many parameters influencing the TF_{SP} are interdependent (Heisterkamp & Koster, 1986).
- Data have not been sampled at random. The schemes for experiments of the investigators were limited, no complete random programme could be carried out (Keen, 1984).
- Several parameters are only partly known (K^+ content, cation exchange capacity, soil moisture, etc.).

An evaluation of the effect of a given parameter on TF_{SP} may therefore be thoroughly influenced by the other (unknown) parameters.

For the evaluation of environmental influences on TF_{SP} by use of the IUR data file a special procedure has been developed, which takes the aforementioned factors into account. Advantages of this procedure, called the 'relative comparison method', are:

- Effects of interactions are eliminated or drastically reduced.

- Deviating measurements can be recognized soon through preliminary treatment of observations. Such measurements appear often to be related to extreme ecological conditions and are thus less suitable for a comparison study.
- It is independent of the underlying distribution of data. Both a normal and a non-normal distribution is allowed.

Disadvantages are:

- The method is time consuming.
- The output is related to so called reference values. Deviations in these values will influence the final result.
- Data should be comparable to any other data in the file. Those which are not comparable, such as those measured under extreme conditions, can not be used.

2.2.2.2 Description of the relative comparison method

The relative comparison method may be used to analyse data files in which (1) data are clustered according to groups of parameters and (2) interactions between parameters are expected and unsufficient information is available to determine the effects of these interactions. The method is based on clustering and relativization of data. An example of an analysis of the effect of soil organic matter (=OM) on TF_{SP} is described to explain the method.

Preliminary analysis

1. First of all, parameters are selected which are assumed to have major influence on TF_{SP}, e.g. an effect of more than a factor 2. This selection can be based on literature study, rough analysis of cross sections of the data base or simple regression analysis. Major parameters are e.g. plant species, soil type and time lag.

Clustering of data

2. The data are clustered according to the parameters selected in step 1. Within each cluster these parameters should be constant or

show only minor variation. In Table 20 data are clustered according to crop species, type of soil and time lag. The different clusters are separated by broken lines.

3. The observed range of data for the parameter being studied (OM) is divided over classes which will present columns in a new table (Table 21, OM classes 0.5-1%, 1-3%, 3-5% and 5-7%). Subsequently data of each cluster are distributed over the classes and, when two or more data are present in the same class, averaged. As a result each cluster will be represented by a single **row** (Table 21). TF_{SP} values from observations 1, 2 and 3 in Table 20 refer to the same class of OM (0.5-1%). In Table 21 the geometric mean TF_{SP} value (0.049) of these observations is presented. A geometric mean is calculated instead of an arithmetic mean because TF_{SP} values are usually lognormally distributed. The single or mean TF_{SP} values in Table 21 will be called **cell values.**

When all measurements of a cluster refer to the same class they are not used because the cluster does not allow to draw conclusions on the effect of the parameter. This is the case for observations 10 and 11 in Table 20, both with an OM content of 0.8%.

The effect of the parameter -OM in this case- on the cell values can now be derived by comparing cell values within each row in Table 21. To determine the overall effect of the parameter on TF_{SP} values, i.e. the effect over all rows, a system of relativization has to be applied which is described in the next steps.

Relativization of data

4. First a so called **reference class** is selected. Thereafter all cell values of Table 21 will be expressed as a fraction of the values in this reference class. The choice of a reference class depends on the structure of the table. A primary requirement for selection is that a reference class contains as many cell values as possible over the different rows. Within a particular row the cell value of the reference class is called **reference cell value.**

NO	CROP	SOIL	TL	OM(%)	TF_{SP}
1	POTATO	C	0	0.5	0.072
2	POTATO	C	0	0.5	0.036
3	POTATO	C	0	0.6	0.045
4	POTATO	C	0	4.2	0.076
5	POTATO	C	2	1.1	0.024
6	POTATO	C	2	3.8	0.032
7	SPINACH	L	2	2.3	0.41
8	SPINACH	L	2	3.7	0.53
9	SPINACH	L	2	5.3	0.60
10	SPINACH	L	1	0.8	0.096
11	SPINACH	L	1	0.8	0.021
12	WHEAT	C	5	2	0.001
13	WHEAT	C	5	5.6	0.003

Table 20: A TF_{SP} data file clustered in five groups. Parameters crop (potato, spinach, wheat), soil (clay, loam) and time lag (TL = 0, 1, 2 or 5 years) are constant within each group. OM=soil organic matter content.

The choice of more than one reference class is possible. In that case the mean of the cell values in the reference classes serves as reference cell value. The advantage of using more reference classes is that more values are used to determine the reference cell value, and thus errors are minimized. Therefore, where possible more reference classes should be selected, being adjacent to each other and containing as many cell values as possible. In Table 21 OM classes 1-3 and 3-5% are selected as reference classes.

CROP	SOIL	TL	CLASS ORGANIC MATTER (%) 0.5-1	1-3	3-5	5-7
POTATO	C	0	0.049		0.076	
POTATO	C	2		0.024	0.032	
SPINACH	L	2		0.41	0.53	0.60
WHEAT	C	5		0.001		0.003

Table 21: TF_{SP} values in each cluster of Table 20 are distributed over four classes of OM. The mean or single TF_{SP} values are called cell values.

5. The **reference cell value** is calculated in each row. Next all cell values are divided by the reference cell value of the respective row. The cell value for spinach in OM class 1-3% , 0.41 (Table 21), is divided by its reference TF_{SP}, 0.47. The result, a relative cell value of 0.87, is shown in Table 22.

CROP	SOIL	TL	CLASS ORGANIC MATTER (%) 0.5-1	1-3	3-5	5-7	REFERENCE CELL VALUE
POTATO	C	0	0.64		1.00		0.076
POTATO	C	2		0.86	1.14		0.028
SPINACH	L	2		0.87	1.13	1.28	0.47
WHEAT	C	5		1.00		3.00	0.001

Table 22: Result of relativization. Values of Table 21 are expressed as a fraction of the mean cell value of OM classes 1-3 and 3-5% (=reference cell value).

Generalization of observed effects

6. The overall effect of a parameter on TF_{SP} is determined by calculating the medians of the relative cell values per class (column). Furthermore a non-parametric confidence interval is calculated to indicate the variation among cell values. If there are ten or more cell values per class the confidence interval is made up by the 10% and 90% points (Sokal & Rohlf, 1981). If there are less than ten cell values per class (Table 23) such a confidence interval has no sense and the total range of the cell values (minimum to maximum) is given instead. The significance of the overall influence of a parameter can be tested by non-parametric statistical procedures (test of Kruskal and Wallis, Spearman's test, Wilcoxon's signed-ranks test, sign test (Sokal & Rohlf, 1981). The result of this testing is indicated by P, the significance level. If $P<0.10$ the observed effect is assumed to be significant, the smaller P the more significant the effect. If $P>0.10$ the effect is regarded as not significant.

Detailed evaluation

7. To evaluate whether a parameter interacts with other - important - parameters the median cell value per column is calculated for a selection of rows in which the parameter being studied has one particular value. This median can then be compared with that of another selection of rows, in which the particular parameter has a different value. For example, median cell values per class of OM (Table 22) can be calculated for clay and for loam separately. If the resulting trend in median values with OM differs between clay and loam, it may be concluded that the influence of OM on TF_{SP} interferes with that of type of soil. In the example numbers of cell values are too small to draw conclusions on interaction between soil type and OM.

RESULT	CLASS ORGANIC MATTER (%)			
	0.5-1	1-3	3-5	5-7
N	1	3	3	2
MEDIAN	0.7	0.9	1.1	2.1
MIN TO MAX		0.9-1.0	1.0-1.1	1.3-3.0

Table 23: Median relative cell value (=relative TF_{SP} value), with minium- maximum range, in relation to soil organic matter content. N=number of cell values.

2.3 RESULTS AND DISCUSSION

2.3.1 Experimental studies

2.3.1.1 Field study in South and West Germany

Radiocesium in Soil Samples

All the following radioactivity data and transfer factors have been decay-corrected to August 1, 1988.

In 1988 samples were taken from a total of 22 sites in Upper Swabia, which were then allocated to the three soil types already described. Taking all the sites together, an average of 186.07 Bq ^{137}Cs/kg (s = 53.7) resulted with a minimum of 45.11 Bq ^{137}Cs/kg and a maximum of 296.65 Bq ^{137}Cs/kg. An isotopic ratio for ^{137}Cs/^{134}Cs of 3.42 was calculated from the average ^{134}Cs content of 54.79 Bq/kg (minimum 11.72 Bq/kg, maximum 88.74 Bq/kg).

A deposition per unit area of 46,000 Bq/m^2 for ^{137}Cs and 14,000 Bq/m^2 for ^{134}Cs can be calculated from these averages based on an average ploughing depth of 20 cm and a volume weight of 1.25 kg/l. This corresponds to the values given by the Federal Health Office for this region.

In the samples taken near Zülpich and Düren, North Rhine-Westphalia (NRW), the average ^{137}Cs content is about 10.06 Bq/kg (minimum 6.44 Bq/kg, maximum 12.38 Bq/kg). This is contrasted with an average ^{134}Cs content of 1.55 Bq/kg (minimum 0.63 Bq/kg, maximum 2.36 Bq/kg). At 7.13 the average isotopic ratio is clearly higher than in Upper Swabia.

In 1989, samples were taken once again in the same region of Upper Swabia. Of a total of 30 samples, 25 soil samples have been measured to date. In these samples, with a standard deviation of 45.35, an average value of 160.01 Bq ^{137}Cs/kg (minimum 101.46 Bq/kg, maximum 277.38 Bq/kg, reference date 1.8.1989) has been found, contrasted with an average of 37.63 Bq/kg (s=10.88) for ^{134}Cs. Due to decay, the isotopic ratio is extended to 4.25.

Radiocesium in Plant Samples

An average ^{137}Cs contamination of 0.75 Bq/kg of grain was determined from a total of 13 winter barley samples (minimum 0.11 Bq/kg, maximum 3.26 Bq/kg). The average value for ^{137}Cs is 0.49 Bq/kg, although no ^{134}Cs was determined for some samples. The maximum is 1.00 Bq ^{134}Cs/kg. At 0.26 Bq ^{137}Cs/kg the average is clearly lower for the 12 winter wheat samples. A minimum of 0.12 Bq/kg is contrasted with a maximum of 0.58 Bq/kg. The average ^{134}Cs content is to be found at 0.16 Bq/kg. The radiocesium contamination attributable to the reactor accident at Chernobyl must be placed in the perspective of the natural ^{40}K content in winter cereals, which in Upper Swabia is in the region of 140 Bq/kg.

Transfer Factors for Cereals on Different Soil Types

The transfer factors for winter barley fluctuate for ^{137}Cs by a factor of 42 from 0.00062 to 0.026. The arithmetic mean is 0.0049. The maximum of 0.026 is identical to the maximum for ^{137}Cs, whereas the ^{134}Cs minimum of 0.0030 is clearly higher than for ^{137}Cs. The higher minimum raises the average to 0.0069.

For winter wheat, the transfer factors merely fluctuate by a factor of 3 between 0.00060 and 0.0020 (average 0.0013). No transfer factors are given for ^{134}Cs since the plant contamination was too weak. Table 24 shows the transfer factors in relation.

Sample	Wheat(W) Barley(B)	^{137}Cs	^{134}Cs	Soil type
OS4a	B	0.00128	-	Planosol
OS4b	B	0.00181	-	Planosol
OS7a	B	0.00235	-	Cambisol
OS12a	B	0.00139	-	Cambisol
OS13a	B	0.00125	-	Cambisol
OS16a	B	0.00102	-	Cambisol
OS18a	B	0.000747	-	Cambisol
OS21a	B	0.00565	0.00556	Fluvisol
OS23	B	0.000617	-	Fluvisol
OS24	B	0.00786	0.00693	Planosol
OS25	B	0.0125	0.0122	Planosol
OS26	B	0.0257	0.0264	Planosol
OS27	B	0.00215	0.00304	Cambisol
OS28	W	0.00197	0.00253	Cambisol
OS29	W	0.00125	-	Cambisol
OS30	W	0.00125	-	Fluvisol
OS31	W	0.00067	-	Cambisol
OS32	W	0.00167	-	Cambisol
OS33	W	0.000699	-	Fluvisol
OS34	W	0.000847	-	Planosol
OS35	W	0.00167	-	Planosol
OS36	W	0.00158	-	Planosol
OS37	W	0.00142	-	Planosol
OS38	W	0.00060	-	Fluvisol

Table 24: Transfer factors for ^{134}Cs and ^{137}Cs in winter cereals grown on soil types in Upper Swabia, relative to the plant fresh mass (approx. 88 % dry solid matter).

If one summarizes the transfer factors of the soil types described in 2.2.1.1 then an average transfer factor of 0.00069 results for the calcaric Fluvisol (Auenvega) at a standard deviation s of 0.00011. The corresponding values for the Cambisol (Braunerde) are 0.0014 (s = 0.00055) and for the (Luvi-) Planosol 0.0067 (s = 0.0087).

Discussion

The soil samples obtained in the Iller Valley (Upper Swabia) over a distance of approx. 20 km vary from 45.11 Bq ^{137}Cs/kg to 296.65 Bq ^{137}Cs/kg. While the maximum represents the end of an approximate normal distribution - the next highest values are at 277 Bq/kg and 254 Bq/kg - the minimum is a statistical outlier. The second lowest value at 107 Bq/kg is more than twice as high. This minimum was found at the most southerly sampling point so that the boundary of the major contamination area was probably reached here. The small-scale differences in deposition are due to the locally restricted precipitation events in the week from April 30 to May 5, 1986. In the Tannheim area during this period the precipitation was up to 45 mm. The ranges of the other samples can be attributed to inhomogeneities during ploughing. Distribution of the ^{134}Cs values is, with only slight deviations, similar to the distribution of the ^{137}Cs values.

The average isotopic ratio of 3.42 (1.8.1988) is close to the isotopic ratio of 3.66 given by the Radiation Protection Commission of the Federal Republic of Germany for the Munich area.

Due to the weaker deposition, the soil samples taken in NRW are significantly less contaminated. Due to the low $^{134/137}$Cs deposition after the Chernobyl accident, the earlier nuclear weapons fallout has an effect on the isotopic ratio here. At 7.13, the average isotopic ratio was clearly higher than could have been expected due to the reactor disaster alone.

The transfer factors soil/cereal are the same for both cesium isotopes. This becomes clear in the identical maximum of 0.026 determined in one sample for both isotopes. The differences in the smallest transfer factor (0.0030 for ^{134}Cs, 0.00062 for ^{137}Cs) can be explained by the fact that ^{134}Cs was no longer detected in the plant below a transfer factor of approx. 0.002. This also explains the apparently higher average.

It is surprising that even within individual soil types the transfer factors are subject to large variations. For this reason an F test was implemented to examine the variances of the individual soil types with respect to the question of whether a comparison of the averages of the individual soil groups can be justified. However, the results of the F test (levels of significance 2.56 %, 0.01 % and 0.01 %), do not permit a direct comparison of the average values. For this reason, an approximated t test was carried out after SATTERTHWAITE. This t test shows that only the averages of the Fluvisol and the Cambisol cannot be significantly differentiated from each other (t value 4.33). The averages of Fluvisol and Planosol, and also of Cambisol and Planosol, can be significantly separated from each other (t values 1.96 and 1.71).

The reason for the very great differences in some of the transfer factors within individual soil types can in part be attributed to differences in the soil parameters. Tables 25 and 26 list the individual soil parameters of the corresponding sampling points.

However, some of the differences in the transfer factors cannot be explained by the soil parameters listed above. In particular, the transfer factors of the soil with a perched water table with a standard deviation of $s = 0.0087$ fluctuate enormously about their average of 0.0067 without the fluctuations being attributable to the soil parameters covered.

The fluctuations of the transfer factors for the soil with a perched water table can be explained by the pedogenic inhomogeneities of this soil type in comparison to Cambisol and Fluvisol.

Sample	Clay(%)	Silt(%)	Sand(%)	N_{tot}(%)	C_{org}(%)	pH($CaCl_2$)
OS4a	11.7	61.4	27.0	0.17	1.97	5.6
OS4b	10.6	65.3	24.2	0.17	2.03	6.1
OS7a	18.5	36.6	45.0	0.13	1.45	5.6
OS12a	16.6	49.0	34.5	0.12	1.39	5.1
OS13a	15.7	48.8	35.6	0.14	1.39	5.6
OS16a	16.0	46.4	37.7	0.19	2.20	6.7
OS18a	15.2	43.1	41.8	0.16	1.94	6.3
OS21a	8.7	74.9	16.4	0.25	2.64	7.5
OS23	18.5	60.7	20.9	0.20	2.20	7.5
OS24	8.5	63.7	27.8	0.26	3.22	6.0
OS25	11.7	54.1	34.3	0.20	2.35	5.8
OS26	11.3	59.1	29.6	0.20	2.18	5.4
OS27	16.4	41.8	41.9	0.17	1.91	5.2
OS28	10.2	46.4	43.5	0.19	1.97	5.4
OS29	13.9	42.0	44.2	0.14	1.54	5.3
OS30	13.2	50.4	36.4	0.22	2.38	6.1
OS31	13.4	47.7	39.0	0.18	1.83	5.8
OS32	9.3	70.8	20.0	0.29	2.67	7.5
OS33	4.2	52.7	43.1	0.24	2.47	7.4
OS34	10.2	56.2	33.6	0.17	2.18	6.1
OS35	6.8	69.4	23.8	0.20	2.23	6.1
OS36	11.0	66.0	23.1	0.18	1.97	5.4
OS37	11.2	65.1	23.7	0.19	2.00	6.4
OS38	18.5	66.7	14.8	0.28	2.44	7.4

Table 25: Grain size, total nitrogen and also organic carbon and pH value of Upper Swabian soil samples.

Sample	Na^+	K^+	Ca^{++}	Mg^{++}	TEB	CEC	Base Saturation
OS4a	0.00	1.10	7.0	0.46	8.56	13.6	62.90
OS4b	0.025	1.08	9.7	0.69	11.49	14.8	77.64
OS7a	0.005	1.32	10.55	1.41	13.29	24.05	55.26
OS12a	0.14	1.06	4.00	0.97	6.17	6.17	100.00
OS13a	0.01	0.73	9.05	1.23	11.02	18.80	58.62
OS16a	0.03	1.24	12.95	2.47	16.69	17.20	97.03
OS18a	0.05	1.45	12.60	1.86	15.96	17.60	90.68
OS21a	0.09	0.17	21.40	2.54	24.20	24.90	97.19
OS23	0.10	1.15	17.01	1.95	20.20	20.20	100.00
OS24	0.10	0.10	11.70	1.21	13.11	17.80	73.65
OS25	0.20	0.13	11.20	1.26	12.79	23.20	55.13
OS26	0.15	0.15	9.65	1.13	11.08	23.60	46.95
OS27	0.00	0.35	10.0	1.14	11.49	31.1	36.93
OS28	0.01	0.45	7.90	1.46	9.81	15.70	62.98
OS29	0.01	1.15	6.90	0.91	8.97	14.80	60.61
OS30	0.01	0.93	12.15	2.16	15.24	18.35	83.05
OS31	0.01	0.43	11.50	1.61	13.54	16.95	79.88
OS32	0.01	0.43	21.71	2.31	24.45	24.45	100.00
OS33	0.01	0.60	15.08	1.38	17.07	17.07	100.00
OS34	0.07	0.88	5.80	0.55	7.30	7.3	100.00
OS35	0.15	0.83	6.60	0.74	8.30	8.3	100.00
OS36	0.01	1.63	6.85	0.53	9.01	15.9	56.67
OS37	0.01	0.70	12.70	0.62	14.03	16.15	80.87
OS38	0.01	0.38	20.29	2.76	23.43	23.80	98.45

Table 26: Interchangeable cations, Total Exchangeable Bases and CEC in meq/100 g, Base Saturation in %.

Whereas with these two soil types the uniform sedimentation of river gravels has formed a relatively homogeneous soil type, the soil with a perched water table is characterized by lively topography and the associated small-scale differences in soil type. Furthermore, this lively relief involves perceptible differences in the soil parameter characterizing this soil type. Depending on position, the influence of perched water is more or less greatly marked. However, precisely this parameter may greatly influence the availability of cesium ions in the soil. Nevertheless, the extent to which soil with a perched water table has been formed cannot be detected by laboratory analyses - in contrast to the present water content. Optical classification in the field is also made more difficult by amelioration measures as well as by the different patterning properties of the initial substrate.

It begins to become apparent that in particular the soils influenced by a perched water table display the highest transfer factors in this region of high precipitation in Upper Swabia. In contrast to the fluvially characterized soils, the variances within this soil type are so great that it is not meaningful to specify a "typical transfer factor" for the soil type. The averages for other soils, in contrast, tend to represent values which may be regarded as typical of the soil type.

2.3.1.2. Field Study in the Northeast of Italy

Radiocaesium in Soil and Plant Samples

^{137}Cs concentration in the soil collected in September 1986 range from 14 to 893 Bq/kg with an average value of 276±242 Bq/kg. This variation is due to the different deposition rates in the Friuli-Venezia Giulia plain after Chernobyl accident. ^{137}Cs deposition values infact, in this area range from 1 to 20 kBq/m^2. The classification of the sampling stations versus the deposition values shows a positive correlation between the different concentrations and the ^{137}Cs deposition. Infact in the area where deposition was above 15 kBq/m^2, the average value of ^{137}Cs concentration is 568±306 Bq/kg, while in the area where deposition was lower than 2 kBq/m^2 the average values of ^{137}Cs is 39±26 Bq/kg.

An analysis of ^{137}Cs concentrations in grass collected in September 1986 suggests the following considerations:

- all the examined samples show ^{137}Cs values between 2 and 113 Bq/kg d.m. with an average value of 33±30 Bq/kg d.m.;
- classification of ^{137}Cs in the fodder crops versus soil deposition shows a correlation between concentrations and deposition values. In fact in the area where deposition was highest the average value is 80±29 Bq/kg d.m. while in the area where deposition was lower than 2 kBq/m^2, the average value is 10±14 Bq/kg d.m..

In 1987 soils and fodder crops have been sampled at the same sampling stations. An analysis of the data bring up the following remarks:

- ^{137}Cs soil concentrations range from 10 to 800 Bq/kg with an average value of 158±164 Bq/kg. This variability is mainly due to the different values of deposition in the plain area;
- ^{134}Cs concentrations in fodder crops are mainly lower than detectable and ^{137}Cs concentrations range between 1 and 40 Bq/kg d.m. with an average value of 6±5 Bq/kg d.m.;
- ^{137}Cs concentrations (Bq/kg d.m.) in the first cut of forage are mainly higher than those measured in subsequent.

The ^{137}Cs high values measured in the first cut could be ascribed to the higher gramineae quantities present in the forage. These species have a larger absorbtion affinity versus the monocation and are characterized by a fasciculate root system which develops superficially. This root system allows them to absorb material on the superficial more contaminated layer of soil. Figure 4 shows the regression net which describes the development of ^{137}Cs concentrations in fodder crops versus those contained in soils and the confidence limits (95%). This development shows a linear correlation (r^2 = 76%) between concentration values in soils and in forage crops. The 2 values of ^{137}Cs concentrations exceeding confidence limits are related to the samples collected in natural meadows, which are generally composed of gramineae. This seems to confirm the higher absorption capacity of gramineae with respect to other kind of forage crops. As reported before, this behaviour is mainly due to the superficial root system of the gramineae.

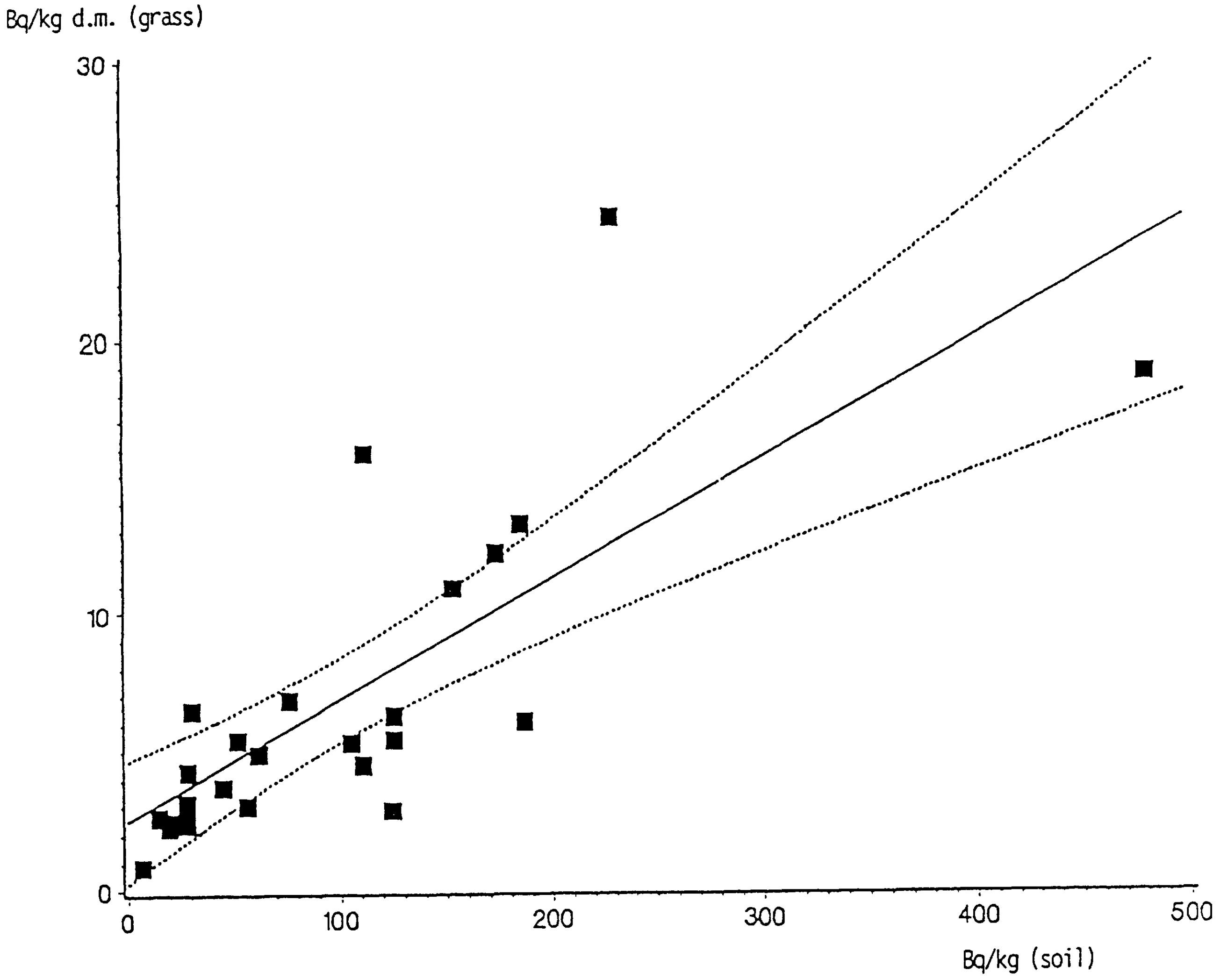


Figure 4: ^{137}Cs grass concentrations versus ^{137}Cs soil concentrations

Transfer factors

Table 27 shows ^{137}Cs transfer factors from soils to forage crops evaluated on samples collected in 1986 and 1987. These data have been obtained from soil samples collected to a depth of 5 cm (1986 and 1987) and to a depth of 10 cm in 1987.

^{137}Cs transfer factors from 1986 data, range from 0.02 to 0.66 with a mean value of 0.14±0.13. The transfer data obtained in 1987 from soils collected to a depth of 5 cm, range from 0.02 to 0.13, with a mean value of 0.06±0.03, while the data evaluated in soils collected up to 10 cm of depth range from 0.02 to 0.21 with a mean value of 0.09±0.04. These last data are, on average, 1.6 times higher than those obtained from soil samples collected to a depth of 5 cm.

Discussion

Figure 5 shows that transfer factors from 1986 data are generally higher than those from 1987 data (soil depth=0-5cm). This behaviour could be due to the different Caesium transfer processes like translocation, resuspension from soil and to the higher biological avalaibility of ^{137}Cs five months after Chernobyl deposition, in consequence of the higher presence of Caesium not yet bound to soil particles. The translocation process could have transfered, from the root system to the leaves, an aliquot of ^{137}Cs previously translocated (just after the Chernobyl deposition), from the leaves to the roots.

The chemico-physical characteristics of the soils, have been used to explain the variability of the transfer factors. The soil sampled have been classified using the simplified U.S.D.A. triangle. With this method the following 6 classes have been singled out: clayey soils (C), medium clayey soils (MC), medium silty soils (MS), medium-grained soils (MG) and sandy soils (S). Soils with a content >40% of particles with size greater than 2 mm, have been classified as CML soils, neglecting the distribution of particles with size lower than 2 mm.

FODDER CROPS	SAMPLING STATIONS	^{137}Cs SOIL-TO-FORAGE CROPS TRANSFER FACTORS		
		1986	1987	1987
		0 - 5 cm	0 - 5 cm	0 - 10 cm
alfalfa	02ab	0.15	0.02	0.04
alfalfa	02ac	0.15	0.03	0.05
alfalfa	03ab	0.07	0.03	0.04
alfalfa	04ab	0.06	0.02	0.04
alfalfa	08ab	0.16	0.06	0.09
alfalfa	09ab	0.12	0.10	0.14
alfalfa	11ab	0.10	0.02	0.03
alfalfa	12ab	0.14	0.04	0.07
alfalfa	13ab	0.05	0.05	0.08
alfalfa	14ab	0.23	0.06	0.08
alfalfa	15ab	0.15	0.08	0.11
alfalfa	19ab	0.11	0.04	0.07
alfalfa	24ab	0.24	0.04	0.07
meadow	27ab	0.13	0.06	0.11
alfalfa	29ab	0.16	0.07	0.12
alfalfa	32ab	0.24	0.13	0.21
alfalfa	33ab	0.03	0.06	0.05
alfalfa	34ab	0.66	0.07	0.11
alfalfa	37ab	0.02	0.03	0.05
alfalfa	38ab	0.02	0.06	0.08
meadow	39ab	-	0.10	0.14
alfalfa	44ab	0.15	0.09	0.11
alfalfa	48ab	0.21	0.12	0.17
alfalfa	51ab	0.03	0.07	0.11
alfalfa	57ab	0.02	0.01	0.02
alfalfa	58ab	-	0.06	0.10

Table 27: ^{137}Cs soil-to-forage crops transfer factors. (0 - 5 cm and 0 - 10 cm represent the soils sampling depth).

Tables 28 and 29 report the chemico-physical characteristics of each soil class and the related ^{137}Cs transfer factors from soil to forage crops. These tables show for each parameter its mean value, standard deviation, variation coefficient and the minimum and maximum value. In the description that follows, the classes C and MC have been considered as a single class because of the few data available.

pH values are homogeneous in the 6 different soilclasses (ranging from a neutral to sub-basic reaction).

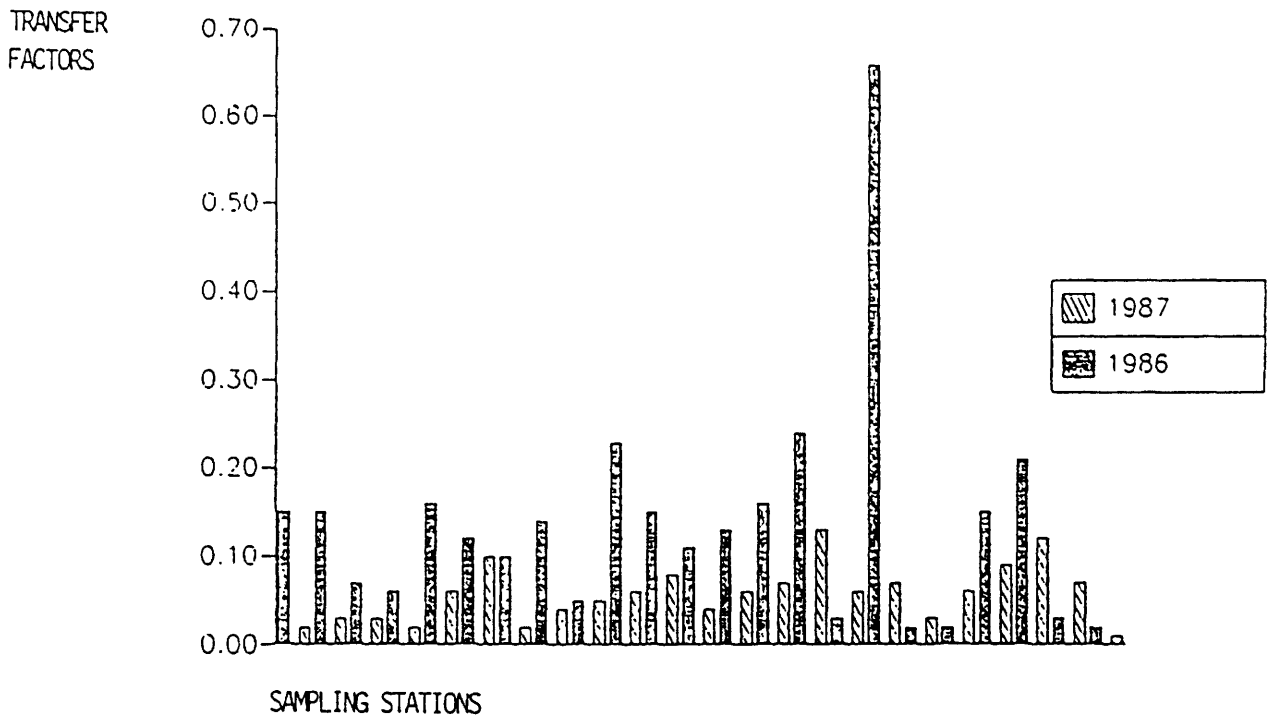


Figure 5: ^{137}Cs soil-to-forage crops transfer factors (1986 - 1987)

The highest values of organic matter (6%) have been found in CML, C and MC soil classes.

In the C and MC classes neverthless, we have found a high value of the variation coefficient (101%) that indicates a considerable variability of the organic matter content in these classes.

The extractable phosphorus and potassium are high in all soil classes. That is probably due to the mineral fertilization applied. The highest values (P=328±67 and K=146±103 mg/kg) are found in CML soil class.

CLASSES		PARTICLES Φ >2 mm (%)	SAND %	SILT %	CLAY %	pH KCl	HUMUS %
C M L	MEAN VALUES	59	29	57	14	7.2	6.2
	STANDARD DEV.	21	7	4	4	0.1	3.0
	VARIAT. COEFF.	35	24	8	30	1.6	47.8
	MINIMUM VALUE	41	20	51	8	7.0	2.9
	MAXIMUM VALUE	89	38	65	20	7.3	10.2
S	MEAN VALUES	13	53	35	11	7.4	2.3
	STANDARD DEV.	13	1	4	5	0.2	0.1
	VARIAT. COEFF.	100	2	10	41	2.7	4.3
	MINIMUM VALUE	0	52	32	7	7.2	2.2
	MAXIMUM VALUE	26	54	39	16	7.6	2.4
MG		35	32	50	18	6.9	3.8
MS	MEAN VALUES	8	21	61	17	7.2	3.5
	STANDARD DEV.	9	8	6	4	0.4	1.6
	VARIAT. COEFF.	117	37	10	24	5.3	44.8
	MINIMUM VALUE	0	9	50	9	6.2	1.7
	MAXIMUM VALUE	38	39	71	25	7.6	8.3
C and MC	MEAN VALUES	0	7	53	40	7.1	5.8
	STANDARD DEV.	0	7	6	4	0.2	5.9
	VARIAT. COEFF.	-	98	11	11	3.0	101.5
	MINIMUM VALUE	0	2	44	35	6.8	2.5
	MAXIMUM VALUE	0	21	61	46	7.5	17.5

Table 28: Chemico-physical characteristics of the following soil classes: clayey soils (C), medium clayey soils (MC), medium silty soils (MS), medium-grained soils (MG) and sandy soils (S). The soils with a content of particles with size greater than 2 mm above 40%, have been classified as CML soils, independently of the content of particles with size lower than 2 mm

CLASSES		EXTRACTABLE (mg/kg)					C.E.C. meq/ 100 g	^{137}Cs TF_{SP}
		P	K	Na	Mg	Ca		
C M L	MEAN VALUES	146	328	15	742	4475	38.6	0.04
	STANDARD DEV.	103	67	4	273	913	9.3	0.02
	VARIAT. COEFF.	71	20	25	37	20	24.1	46
	MINIMUM VALUE	64	244	10	481	3158	28.0	0.02
	MAXIMUM VALUE	346	432	21	1085	5589	54.0	0.07
S	MEAN VALUES	41	192	21	386	4816	19.7	0.07
	STANDARD DEV.	2	48	8	186	2290	1.1	-
	VARIAT. COEFF.	5	25	38	48	48	5.6	-
	MINIMUM VALUE	39	144	13	200	2526	18.6	-
	MAXIMUM VALUE	43	239	29	572	7105	20.8	-
MG		0	544	23	254	6684	28.0	0.08
MS	MEAN VALUES	49	268	28	526	5623	30.8	0.10
	STANDARD DEV.	36	217	8	316	1626	7.5	0.05
	VARIAT. COEFF.	73	81	28	60	29	24.5	51
	MINIMUM VALUE	0	88	17	213	3315	19.2	0.04
	MAXIMUM VALUE	152	810	40	1194	7894	47.5	0.21
C and MC	MEAN VALUES	61	257	36	614	7920	44.1	0.11
	STANDARD DEV.	37	41	19	427	612	23.8	0.02
	VARIAT. COEFF.	61	16	53	69	8	53.9	19
	MINIMUM VALUE	24	203	19	216	7210	21.9	0.08
	MAXIMUM VALUE	128	325	74	1443	8842	90.0	0.14

Table 29: ^{137}Cs transfer factors (TF_{SP}) from soil to forage crops and chemico-physical characteristics of the following soil classes: clayey soils (C), medium clayey soils (MC), medium silty soils (MS), medium-grained soils (MG) and sandy soils (S). The soils with a content of particles with size greater than 2 mm above 40%, have been classified as CML soils, independently of the content of particles with size lower than 2 mm

The lowest ^{137}Cs transfer factors from soil to forage crops (0.04±0.02) have been found in CML soil class.

In MS, MC and C classes, the transfer factor values are similar (0.10±0.05 and 0.11±0.02). In medium-grained soil class (MG) the transfer factor value found is 0.08 while in the sandy soil class (S) the value found is 0.07.

^{137}Cs transfer factor values from 1987 data, show an inverse correlation with the extractable phosphorus and potassium. Infact the lowest transfer factors have been found in the CML soil class where the mean values of the extractable phosphorus and potassium are the highest. Another process that can explain the lowest ^{137}Cs transfer values found in the CML soil class could be ascribed to the root system of forage crops. Infact in the presence of coarse soil and gravel, the roots need to explore deeper and therefore less contaminated soils.

2.3.1.3 Lysimeter Experiment

Radioactivity in Soil

All radioactivity data and transfer factors are decay-corrected to January 1, 1989.

The $^{134/137}Cs$ contents in the soil were determined before applying sewage sludge. On the pasture lysimeters, on average at a depth of 0-5 cm 22.75 Bq ^{137}Cs/kg was found, at 5-10 cm 5.96 Bq ^{137}Cs/kg, at 10-20 cm 3.13 Bq ^{137}Cs/kg and at a depth of 20-30 cm 0.28 Bq ^{137}Cs/kg. The individual values are minimally scattered about these averages. The corresponding ^{134}Cs contents were 5.70 Bq/kg (0-5 cm) and 0.70 Bq/kg (5-10 cm). No ^{134}Cs was detected at depths lower than 10 cm.

The radiocesium contents on the arable lysimeters fluctuated due to inhomogeneities during ploughing between 15.50 Bq ^{137}Cs/kg and 8.00 Bq ^{137}Cs/kg, and 2.22 Bq ^{134}Cs/kg and 0.93 Bq ^{134}Cs/kg respectively for a depth of 0-20 cm.

The radiocesium applied by sewage sludge fertilization, relative to 1.1.1989, was 10,120 Bq ^{137}Cs/m^2 and 2375 Bq ^{134}Cs/m^2. Relative to the ploughing depth of 20 cm, it was calculated that the radioactivity increased by 20.24 Bq ^{137}Cs/kg and 4.75 Bq ^{134}Cs/kg for the arable lysimeters, and by 40.48 Bq ^{137}Cs and 9.50 Bq ^{134}Cs/kg for the pasture respectively (depth 0-10 cm).

The increase actually measured after applying sewage sludge was 55 Bq ^{137}Cs/kg and 17 Bq ^{137}Cs/kg, and for ^{134}Cs in the region of 16 Bq/kg and 6 Bq/kg. The discrepancy between the calculated and measured values can be explained by the irregular distribution of sewage sludge in the soil.

Radioactivity in Plant Materials

The results of the first and second grass cut are available at present, as well as the potato analyses. At the first cut, the ^{137}Cs activity in the two control lysimeters with pasture vegetation amounted to 3.36 Bq/kg dry solid matter (DS) and < 2.8 Bq/kg DS. Relative to the plant fresh mass (20 % of the dry solid matter) this is 0.67 and < 0.56 Bq/kg. In the lysimeters treated with sewage sludge, the values are 42.51 and 33.33 Bq/kg DS (8.50 and 6.67 Bq/kg FM). ^{134}Cs was not detected on the control lysimeters, and on the areas treated with sewage sludge the values were 7.36 and 7.18 Bq/kg DS.

At the second cut, the ^{137}Cs contents on the control lysimeters were 0.47 and 2.90 Bq/kg DS, whereas on the sewage sludge lysimeters 7.43 and 5.19 Bq/kg DS were measured.

The ^{137}Cs contents in potatoes from the control lysimeters are 1.06 and 0.77 Bq/kg DS, whereas on the treated lysimeters they are 1.51 and 2.34 Bq/kg DS. It was not possible to detect ^{134}Cs in any of these cases (0.1 Bq/kg DS).

Transfer Factors

In the grass of the control lysimeters (pasture), the transfer factors relative to the plant fresh mass for ^{137}Cs amount to 0.0069 (lysimeter 1, second cut), 0.044 (lysimeter 3, first cut) and 0.038 (lysimeter 3, second cut) taking into consideration the average radioactivity content at a depth of 0-10 cm.

On the lysimeters treated with sewage sludge, the transfer factors for ^{137}Cs in the first cut are 0.15 and 0.12. The corresponding values for ^{134}Cs are 0.12 and 0.11.

In the second cut, the transfer factors for ^{137}Cs drop to 0.027 and 0.019, whereas the transfer factors for ^{134}Cs are 0.027 and 0.015.

For the soil-potato transfer factors, the soil activity is calculated relative to the average $^{134/137}$Cs content at a depth of 0-20 cm. Assuming an average dry solid matter fraction of 23 %, the radioactivity in the plant is related to the fresh mass. If the transfer factors are related to the measured ^{137}Cs contents then on the control lysimeters values of 0.025 and 0.016 result, whereas the potatoes from the sewage sludge lysimeters display transfer factors of 0.0056 and 0.019.

Based upon the computational increase in soil activity, the transfer factors are 0.015 and 0.020 for the untreated lysimeters and 0.012 and 0.017 for the treated lysimeters.

Discussion

A possible increase in the radiocesium activity in grass and potatoes was studied by applying sewage sludge contaminated by the reactor disaster in Chernobyl to lysimeters. The quantity of sewage sludge applied corresponds to the maximum permissible values pursuant to the German Sewage Sludge Ordinance.

In the first cut from the grass lysimeters, a clear increase in radiocesium activity by a factor of 10 was recorded. In the second cut, the ^{137}Cs activity was slightly increased in the two sewage sludge lysimeters at 7.43 and 5.19 Bq/kg DS, whereas ^{134}Cs was only detected in one lysimeter.

Only a very slight increase of radiocesium activity in potatoes was recorded due to sewage sludge application. The increase to 1.51 and 2.34 Bq/kg DS (in comparison to 1.06 and 0.77 Bq/kg DS in the control lysimeters) must be assessed in view of the natural ^{40}K activity in potatoes, which in this case is 820 Bq/kg DS.

The data of the transfer factors from the control lysimeters (pasture) are transfer factors of the ^{137}Cs released by the Chernobyl accident. The similarity between the transfer factors of the first and second cut in lysimeter 3 is interesting, whereas the transfer factor for lysimeter 1 is lower.

In specifying the transfer factors for the surfaces fertilized with sewage sludge it must be noted that particularly during the first cut sewage sludge particles adhering to the grass may increase the transfer factors. This is not so much root transfer as rather contamination by material which was not rinsed off after applying the sewage sludge. Care was taken in selecting the lysimeter soil to ensure that a pasture with an intact root network was found. Thus an uptake by plant material lying above the soil is also probable.

At the first cut, the transfer factors of both cesium isotopes on the sewage sludge lysimeters are surprisingly close at 0.077, 0.061, 0.058 and 0.057.

In the second cut, the transfer factors of the regrowth of considerably less contaminated grass drop to 0.014, 0.0095, 0.014 and 0.0076. In this case, the $^{134/137}$Cs contents in the grass are only insignificantly higher than the control lysimeters (see 2.3.1.3).

The transfer factors actually measured for the arable lysimeter are similarly close together at 0.025, 0.016, 0.0056 and 0.019. No differences in the transfer factors between treated and untreated lysimeters were established. The minimally higher ^{137}Cs values in the potatoes from the sewage sludge lysimeters were superimposed by the relatively large fluctuations in the ^{137}Cs content of the soil.

Even if the computational increase in the soil radioactivity of 20.24 Bq ^{137}Cs/kg is assumed, no differences in the transfer factors of the two lysimeter groups could be established. The overall fluctuation of the transfer factors is merely increased by this procedure. The values are very close together at 0.015, 0.020, 0.012 and 0.017. This type of calculation with the expected ^{137}Cs values in the soil is undoubtedly more realistic since the sewage sludge present in the soil in the form of aggregates makes measurements of the average ^{137}Cs activity in the soil more difficult.

By way of summary, it may be said that the increase of radioactivity in arable crops due to the application of highly contaminated sewage sludge is negligible.

As far as the pasture lysimeters are concerned, the first cut indicates a clear increase in the radiocesium content, but from the second cut onwards this also seems negligible.

2.3.2 Statistical studies

2.3.2.1 Evaluation of non-Chernobyl data

Soil organic matter

In order to assess the effect of soil OM on the TF_{SP} data were clustered according to:

- investigator
- type of experiment
- type of contamination
- plant species
- plant part
- soil type
- year of contamination
- time lag.

Results from lysimeter experiments by Stoutjesdijk and Lembrechts were not considered because in these experiments addition of OM coincided with other actions influencing the TF_{SP} of Cs (Lembrechts et al., 1989). Results from the few available pot experiments were not included either.

Peaty soil was regarded as sandy soil with an OM content of more than 10%. In total 271 data were used, distributed over nine classes of OM. Data on soils with an OM content of more than 22% were not equally distributed. Therefore only two classes, 28-35% and 70-80%, were created in this range.

TF_{SP} values appear to be significantly different between classes of OM (Kruskal Wallis, $P<0.001$) (Table 30). The effect of OM is maximal a factor 14. The TF_{SP} increases gradually with increasing OM content (Fig. 6) (Spearman, $P<0.005$). In fact, processes related to a high organic matter content of the soil may cause the observed increase in transfer of Cs. For example, under the prevailing climatological conditions microbial decomposition of soil organic matter can lead to the formation of NH_4^+ ions which may prevent Cs ions from adsorption on the solid phase.

From these results the following corrections are derived. These can be used when comparing TF_{SP} between soils with different OM content, as in the comparison of Chernobyl and non-Chernobyl data.

- OM <5%: no correction;
- OM 5-20%: division of TF_{SP} by a factor 2;
- OM 21-50%: division of TF_{SP} by a factor 4;
- OM >50%: division of TF_{SP} by a factor 10.

	CLASS OF ORGANIC MATTER CONTENT (%)								
		*	*						
	0.5-1	1.1-2	2.1-3	3.1-5	5.1-7	7.1-10	10.1-21	28-35	70-80
N	16	24	24	19	24	11	12	5	8
MEDIAN	0.9	1.0	1.0	1.7	1.3	2.7	2.4	4.3	12.6
10% POINT	0.3	0.7	0.9	0.4	0.4	1.0	0.6	0.4^{**}	6.8^{**}
90% POINT	2.0	1.0	1.3	3.6	2.1	4.4	6.1	10.9^{**}	26.8^{**}

**min. and max.; median values show an upward trend (Spearman, $P<0.005$).

Table 30: Median relative TF_{SP} value (relative cell value) of Cs, with 10%-90% points, in relation to OM content. N=number of cell values, *=reference class

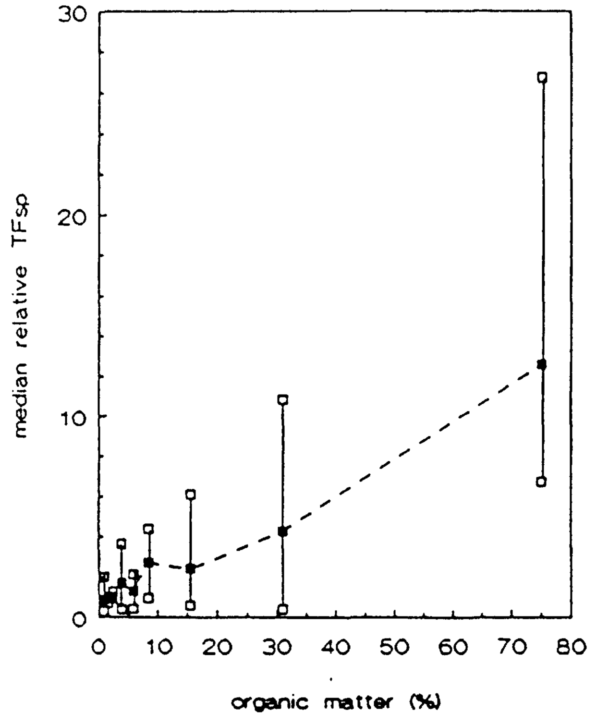


Figure 6: Median relative TF_{SP} of ^{134}Cs and ^{137}Cs (with 10% to 90% interval) as a function of the soil organic matter content.

Type of soil

To study the influence of type of soil on TF_{SP} of Cs data were clustered according to type of experiment and contamination, investigator, plant species, plant part, year of contamination and time lag. The class with data on sand was used as reference. A correction was applied for differences in OM content using the recommended correction factors mentioned in the evaluation of effects of OM on TF_{SP}. In total 767 values were used for the analysis. Peat soil is not considered in this analysis because peat soil was regarded as a sandy soil with a high OM content in the evaluation of effects of OM.

Transfer factors from both clay and loam are significantly lower than from sandy soil (Table 31) (see also Fredriksson et al., 1966). This implies that comparisons between TF_{SP} values should preferably be based on measurements on the same type of soil. If such is not possible, use of the following correction factors is recommended:

-a factor 5 in the comparison clay-sand;
-a factor 3 in the comparison loam-sand.

	CLAY/SAND	LOAM/SAND
N	69	149
MEDIAN	0.2	0.3
10%-90%	0.04-0.6	0.05-1.2

Table 31: Median relative TF_{SP} values (relative cell values) of ^{134}Cs and ^{137}Cs, with 10% to 90% points, resulting from the evaluation of the influence of soil type. N=number of cell values.
TF_{SP} values in the comparisons clay-sand and loam-sand are significantly different (sign test, $P<0.001$).

Soil pH

Before analyzing the effect of soil pH all pH values measured in water were transformed to pH values measured in KCl by subtraction of a factor 0.6. Data were clustered according to the same parameters as for the evaluation of OM. Within each cluster observations were distributed over ten continuous pH classes between pH 3.9 to 8.4. Four classes, ranging from pH 5.3 to 6.9, were selected as reference classes. The analysis included 494 TF_{SP} data.

The overall effect of pH on the TF_{SP} is less than a factor 2. Results of most of the individual investigators reveal a similar pattern: relative TF_{SP} values do not differ between pH classes (Kruskal Wallis, $P>0.10$) and median values do not show any trend (Spearman, $P>0.10$) (Fig. 7). However, results of one investigator show an exceptionally large pH effect of about a factor 10 in the range of pH 4.6-5.2. This large effect is probably due to other parameters than the pH.

In order to check whether an effect of pH could be observed in specific situations analyses were done for a selection of clusters, per type of experiment (field, lysimeter) or per plant species (barley, wheat, maize, grass, potato). These analyses lead to the same conclusions although for wheat relatively high TF_{SP} values (relative TF_{SP}=about 2.5) are found in the range pH 4.6 to 4.9. In this range of pH crop yields are already suboptimal so that other effects than the direct impact of pH may play a role. The pH effect on loam seems similar to that on sand. For other soils insufficient cell values were available for an evaluation.

In conclusion, corrections for differences in soil pH when comparing TF_{SP} values do not seem necessary.

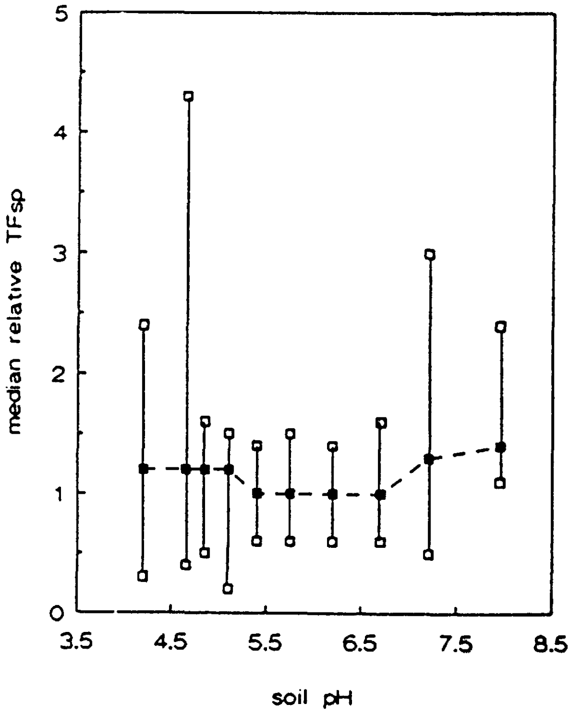


Figure 7: Median relative TF_{SP} of ^{134}Cs or ^{137}Cs (with 10% to 90% interval) as a function of soil pH. Medians do not show any trend (Spearman, P>0.10).

Time lag

For the analysis of effects of time lag data were again clustered in the same way as for the evaluation of effects of OM. Time lag is defined as the period elapsed between contamination of the soil and collection of samples (usually at time of harvest). In total 785 data were distributed over ten classes of time lag, ranging from 0 to >8 years. Classes 1.0-1.9 and 2.0-2.9 years were used as reference classes. No correction for OM was needed because this parameter hardly varied within each cluster.

In both the lysimeter (all data together) and the field experiments (with artificial contamination) relative TF_{SP} values are different between time lag classes (Kruskal Wallis, $P<0.001$). Furthermore, median relative TF_{SP} values for artificially contaminated fields show a significant decrease in TF_{SP} with increasing time lag (Spearman, $P<0.005$), especially after the first year (Fig. 8).

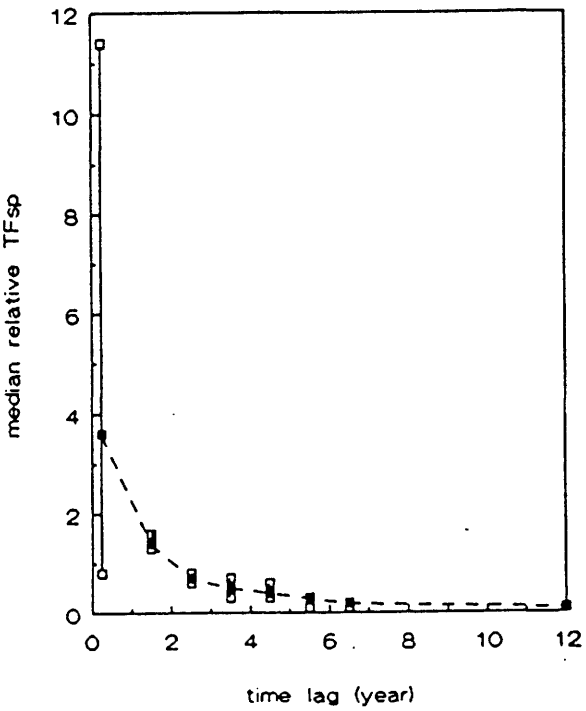


Figure 8: Median relative TF_{SP} (with minimum to maximum interval) as a function of time lag for artificially contaminated fields, showing a significant downward trend (Spearman, $P<0.005$).

On the contrary, data for all lysimeter experiments together do not show any correlation with time lag (Spearman, $P>0.10$) (Fig. 9). No interaction was found between type of soil and time lag, neither in field nor in lysimeter experiments.

Results of this analysis might have been influenced by changes in weather conditions over the different harvest years. Time lag is in this case related to the year of harvest; effects of both parameters cannot be separated. Nevertheless, the gradual decrease in TF_{SP} without large fluctuations for field experiments (Fig. 8) suggests that in this situation the influence of time lag is much larger than a possible influence of weather conditions. In the analysis of data from all lysimeter experiments together (Fig. 9) possible effects of time lag may have been masked by fluctuations in weather conditions over the different years.

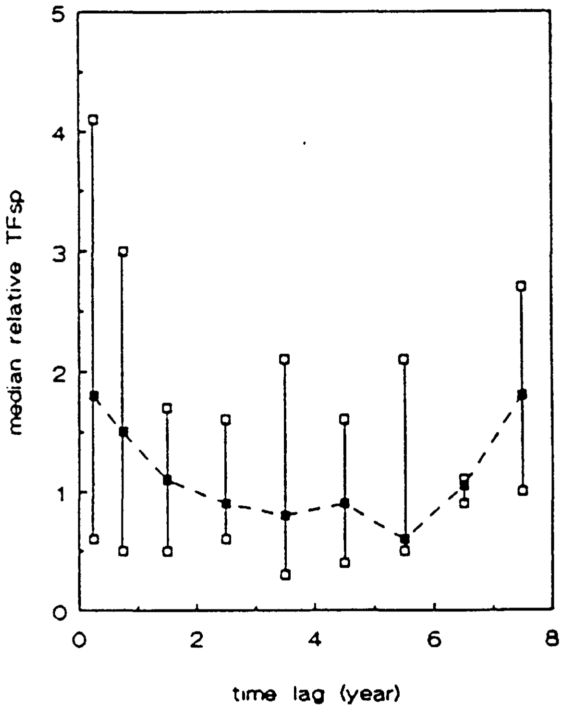


Figure 9: Median relative TF_{SP} (with 10% to 90% interval) as a function of time lag for all lysimeter experiments. Medians do not show any trend (Spearman, $P>0.10$).

For one specific location (ITAL lysimeters Wageningen, the Netherlands) additional information on climatological conditions and the experimental set-up allowed a more detailed evaluation of the influences of time lag. In each lysimeter ^{134}Cs and ^{137}Cs had been applied once but application of ^{134}Cs took place one or several years after that of ^{137}Cs. The concentrations of both isotopes were analyzed separately in each soil or plant sample. This approach supplied TF_{SP} values of the same element (Cs) after different time lags but measured in the same plant, under the same environmental conditions and the same weather situation. Therefore, this analysis (of 334 observations) renders an accurate estimate of the effect of time lag, although it refers to only one specific location, one experimental design, etc.

Relative TF_{SP} values for the Ital lysimeters appear to be significantly different between the time lag classes (Kruskal Wallis, $P<0.001$). Median TF_{SP} values reveal a small but gradual decrease with increasing time lag (Spearman, $P<0.005$) (Fig. 10).

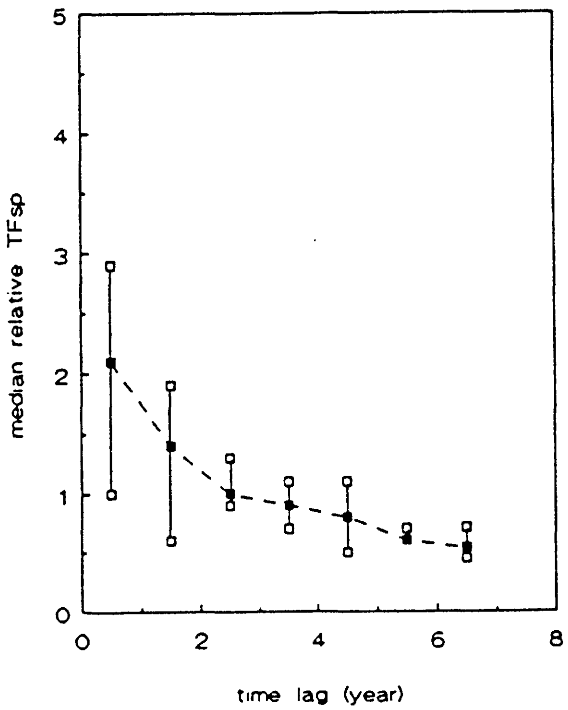


Figure 10: Median relative TF_{SP} (with 10% to 90% interval) as a function of time lag in lysimeter experiments in the Netherlands only (ITAL Wageningen). Weather effects are eliminated. TF_{SP} values show a downward trend (Spearman, $P<0.005$).

after the Chernobyl accident were not considered because effects of direct deposition on the plant might disturb the analysis on transfer from the soil. A total number of 289 observations from Belli & Sansone, Eriksson et al. and MacNeill could be used. Soils with more than 20% OM were regarded as sandy soil with a high OM content.

In the classes 20-60% and >60% OM there were too few comparable data to draw any conclusions. In the range 1-20% OM median relative TF_{SP} values seem hardly influenced by the OM content (Table 36). This finding does not agree with the result of the evaluation of non-Chernobyl data, where a significant effect of OM on TF_{SP} was observed (par. 2.3.2.1 'Soil organic matter').

Time lag

The effect of time lag could be analyzed with 220 data on alfalfa, barley and grass, collected by Belli & Sansone and Eriksson et al. over a period of two years. Soil-to-grass data collected shortly after the Chernobyl accident were neglected again. After clustering data, according to plant species, plant part, investigator, soil type and OM, relative TF_{SP} values were calculated as a function of time lag. The TF_{SP} determined in October 1986 was used as reference.

About one year after contamination transfer was less than half of its value in October 1986 (Table 37). In fact, this effect cannot be distinguished from influences of weather conditions which differ each growth season. Assumed that such influences in 1987 have been small, the observed effect of time lag on TF_{SP} is comparable to time-lag effects found on non-Chernobyl measurements (see par. 2.3.2.1 'Time lag').

PLANT	SOIL	OM%	TIME LAG 6 APRIL'87	 8 JUNE'87	 10 AUGUST'87
ALFALFA	LOAM	1-3	0.91	0.50	0.55
		3-7	0.61	0.32	0.48
		7-20	0.52	0.26	0.47
	SAND	1-3	0.51	0.24	0.29
		3-7	0.27	0.07	0.27
BARLEY	CLAY	1-3			0.67
		3-7			0.55
	LOAM	1-3			0.31
		3-7			0.23
		7-20			0.42
	PEAT	20-60			0.14
		>60			0.06
	SAND	3-7			0.20
		7-20			0.15
		20-60			0.18
GRASS	CLAY	1-3			0.30
	LOAM	1-3			0.34
		3-7			0.52
	PEAT	20-60			1.21
MEDIAN RELATIVE TF_{SP}			0.5[a]	0.3[a]	0.3[a]
	MIN TO MAX		0.3-0.9	0.1-0.5	0.1-1.2

[a] TF_{SP} values are significantly lower than in October 1986 (Wilcoxon's signed-ranks test, $P<0.05$).

Table 37: Relative TF_{SP} values (relative cell values) of Cs for different soil-plant systems in relation to time lag (month). Values are relative to those measured in October 1986 (time lag= 0).

2.3.3 Experimental studies in Denmark

The experiment was carried out at Risø National Laboratory in the summer of 1988 and 1989 as part of an experiment concerning root uptake of ^{137}Cs and ^{134}Cs. The experiment was made in order to identify crops with a relatively low or high root uptake of radiocaesium with regard to countermeasures after an accidental release of radionuclides (Øhlenschlæger M. and G. Gissel-Nielsen, 1989).

A part of the project was carried out in soil which originates from Gävle in Sweden, north-west of Stockholm, site of the largest deposition of radiocaesium in Sweden following the Chernobyl accident. Characteristics of the soil are given in table 38.

Soil type	organic
Content of organic matter (%)	66
Particle size in mm	
<0.002 mm	4.4
0.002-0.02 mm	7.7
0.02-0.2 mm	15.9
0.2-2.0 mm	6.0
pH	5.2
mg K/100 g soil	8.0

Table 38.

The soil, which was contaminated with ^{137}Cs from the Chernobyl accident was experimentally supplied with ^{134}Cs, in the summer 1988, in order to investigate differences between experimentally added caesium and Chernobyl caesium. The experimentally added caesium was added as $CsNO_3$ from a 0.1 m HNO_3 solution.

The results for barley grain given as concentration ratios, CR, (Bq/g plant per Bq/g soil, dry wight) are shown in Table 39. The results demonstrate differences between experimentally added Cs and Chernobyl Cs. In 1988 the uptake from experimentally added Cs is about a factor 4 higher than for Chernobyl Cs. In 1989 this difference is reduced, the root uptakes are of similar magnitude.

Soil and isotope	Variety	Year	Grain	Straw
Organic soil experimentally added Cs-134	Golf	1988	0.476±10 %	0.679±12 %
		1989	0.147±7 %	0.113±14 %
	Apex	1988	0.471±7 %	0.764±9 %
		1989	0.140±8 %	0.118±5 %
	Anker	1988	0.602±7 %	0.700±9 %
		1989	0.210±13 %	0.146±11 %
	Sila	1988	0.640±7 %	0.921±9 %
		1989	0.171±19 %	0.158±9 %
Organic soil Chernobyl Cs-137	Golf	1988	0.119±10 %	0.239±12 %
		1989	0.107±7 %	0.134±11 %
	Apex	1988	0.117±7 %	0.219±11 %
		1989	0.128±13 %	0.196±26 %
	Anker	1988	0.154±6 %	0.240±8 %
		1989	0.148±13 %	0.161±13 %
	Sila	1988	0.179±9 %	0.342±9 %
		1989	0.144±16 %	0.226±47 %

Table 39: Concentration ratios, Bq per g plant/Bq per g soil, dry weight, for Barley. Means of four replicates ±1SE.

It is well established that caesium is subject to consederable fixation in the soil as a result of its entrapment in the lattice structure of clay minerals (Nielsen B. and M. Strandberg, 1988; Squire H.M. and L.J. Middleton, 1966). Although the soil considered here is organic, and has a reduced absorption and subsequent fixation of caesium on clay minerals, the fixation is still of considerable importance. Squire and Middleton (1966) conclude that the fixation appears to be nearly complete after 3 years.

This agrees very well with our results where the fixation of the Chernobyl caesium is nearly complete after 3 years and very little further fixation can be seen. While the caesium experimentally added in 1988 still are exposed to fixation. An analysis of variance supports this showing a highly significant difference (>99.9 %) between the concentration ratios for the experimentally added caesium in 1988 und 1989, while there are no significant differences between the concentration ratios in 1988 and 1989 for Chernobyl caesium.

Part of the difference could be due to chemical differences between caesium from the Chernobyl accident and experimentally added caesium. In order to examine this further the CR values are plotted against the number of years since the contamination for the four varieties. The results are shown in Figure 15 and Figure 16 for grain and straw, respectively.

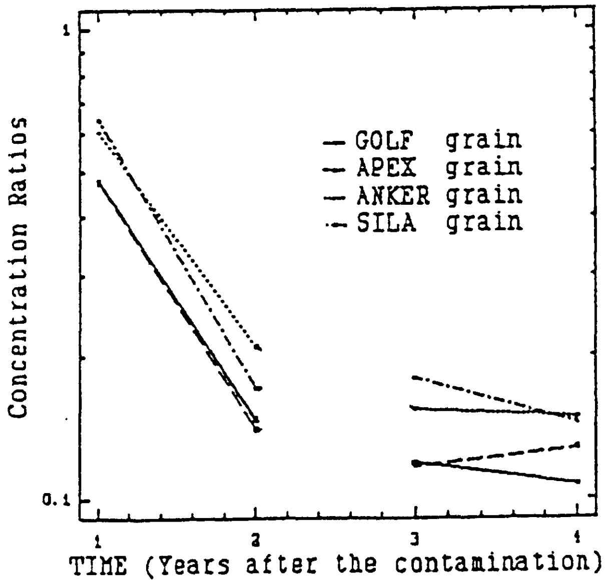


Figure 15: Concentration ratios. Showing the time trend for barley grain.

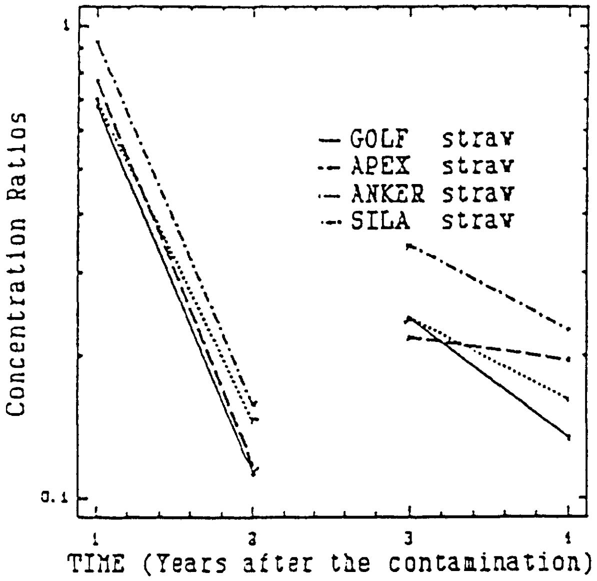


Figure 16: Concentration ratios. Showing the time trend for barley straw.

Assuming no difference between experimental added caesium and Chernobyl caesium, the CR values should decline in a similar way. With regard to the results for the grain this seems to be a plausible assumption but the results for the straw indicate some difference. In order to examine this further the experiment will continue and differences in the fixation trend for the two isotopes will be studied.

2.4 SUMMARY AND CONCLUSIONS

2.4.1 Objectives

Soil-to-plant transfer of Cs originating from Chernobyl was in some parts of Europe higher than expected on the basis of earlier observations. Main aim was to investigate whether Cs from Chernobyl is indeed more easily transferred from soil to plant than Cs from non-Chernobyl sources, i.e. remnants of nuclear weapon tests in the sixties, discharge of nuclear industry or artificial additions to soil. Because Chernobyl and non-Chernobyl data were generally sampled under different circumstances (region, year, crop, etc.) the second aim was to evaluate the effects of these parameters on the transfer of Cs. This analysis allowed corrections for environmental and experimental differences which have been used to compare Chernobyl and non-Chernobyl transfer measurements. These correction factors may also be valid for other comparisons of large amounts of soil-plant transfer data.

2.4.2 Experimental studies

Chernobyl transfer in South-Germany

Transfer factors (TF_{SP}) for radiocesium ($^{134/137}Cs$) in cereals grown on three different soil types - Cambisol (Terrassenbraunerde), calcaric Fluvisol (Allochthone Vega) and Planosol (Pseudogley) - in one of the highest contaminated areas in Germany (Upper Swabia) were determined. The variations of TF_{SP} within the group of Cambisols and Fluvisols were very small. Only in the group of soils with a perched water table (Pseudogley), variations were higher. This might be explained by local differences in the water regime of that soil type. In contrast to brown soils and alluvial soils, soils with a perched water table have much more differences in topography in that region. Therefore, water regime is changing drastically in small distances as well as other soil parameters.

The medium TF_{SP} of Cambisol and Fluvisol can be distinguished to the medium TF_{SP} of the soil with a perched water table, which is significantly higher. Differences in TF_{SP} of Cambisol and Fluvisol are not significant.

Lysimeter experiment with contaminated sewage sludge

In lysimeters filled with undisturbed soil monoliths of a podzolic gleysol (FAO classification: Dystric Gleysol) with an extremly high content of sand and a very low pH the transfer of $^{134/137}$Cs into potatoes, lettuce and grass due to the fertilization with sewage sludge contaminated after the Chernobyl accident was investigated. As compared to the untreated control the uptake of radiocesium into potatoes and lettuce showed a slight increase after fertilization of the soil with sewage sludge (1 kg D.M./m^2, 13,000 Bq $^{134/137}$Cs/kg D.M.). Nevertheless the content of radiocesium in potatoes and lettuce did not reach 1 Bq/kg of fresh plant weight. Even the radiocesium content in grass of the first cut increased significantly after fertilization with contaminated sewage sludge. In the grass from the second cut the radiocesium content decreased due to the lower contamination of the plants.

Conclusions from these results are that the use of sewage sludge as fertilizer even high contaminated with $^{134/137}$Cs does not lead to a serious increase of radiocesium in plants grown on arable land. Also in pasture grass the increase of radioactivity seems to be neglectable after fertilization with sewage sludge contaminated with radiocesium.

Chernobyl transfer in Northeast-Italy

^{137}Cs transfer factors from soil to forage crops have been evaluated from data collected in 1986 and 1987 in the Friuli-Venezia Giulia region. Sampling stations have been selected on the basis of a regional soils study and ^{137}Cs Chernobyl depositions.

^{137}Cs transfer factors from 1987 data have been evaluated in soils collected up to 10 cm of depth because all sampling stations were unploughed after Chernobyl and therefore the most part of ^{137}Cs contamination has remained in the superficial layers.

Soil to forage crops transfer factors from 1986 data are generally higher than those from 1987 data. This behaviour could be due to translocation from roots, stolons etc. to leaves, of ^{137}Cs previously translocated during the deposition period, from leaves to other parts of plants.

The 1987 data show that the lowest ^{137}Cs transfer factors from soil to forage crops have been found in soils with a content >40% of particles with size greater the 2 mm. In medium clayey, clayey and medium silty soils the transfer factor values are similar. This behaviour could be due to the highest mean values of extractable phosphorus and potassium in the CML class and to the deeper root system of forage crops in presence of coarse soils and gravel.

2.4.3 Statistical studies

Chernobyl transfer versus non-Chernobyl transfer

Results show that the soil-to-plant transfer factor of Cs originating from Chernobyl was about a factor 2 lower than the transfer factor for Cs from other contamination sources under comparable conditions. Three main factors (time lag, soil organic matter and translocation from older plant parts) may explain the difference between this result and the initially expected higher transfer of Cs:

1) Cs becomes less available in the course of time. After 5 to 10 years the availability has decreased with about a factor 10. Direct comparison, without correction for time lag, of Chernobyl data values with measurements on transfer of nuclear weapons fallout may thus overestimate the transfer up to about a factor 10.
2) Part of the Chernobyl data refers to upland soils in north and north-western Europe. The greater part of these soils is characterized by a high soil organic matter content, extremely low pH values, a low clay mineral content and a poor nutrient status, usually under a humid, cool climate. Transfer on soils with high organic matter contents were shown to be about ten times larger than on soils with less than 10% organic matter.

In fact, processes related to a high organic matter content of the soil may cause an increased transfer of Cs. Under the prevailing climatological conditions microbial decomposition of soil organic matter can lead to the formation of NH_4^+ ions which prevent Cs ions from adsorption on the solid phase.

3) High transfer factor values measured in spring and summer after the Chernobyl accident may partly refer to direct uptake by above ground plant parts in stead of uptake by the roots from the soil, even when the harvested plant parts were growing after the deposition. Translocation from plant parts (roots, stolons, etc.) present during deposition to parts which developed after deposition may be the cause of unexpectedly high Cs contents in the harvest.

Region and season

Effects of region and season can be traced back to differences in soil type, variation in crops, differences in agricultural practices and above all varying weather conditions (temperature, rainfall). Dutch observations collected over a period of 4 to 7 years show that climatological factors cause fluctuations in transfer per season of maximal a factor 10, depending on the type of crop. Increases in transfer seem to correspond with decreases in rainfall or temperature.

The transfer factor of Cs isotopes on sandy soils appears 3 to 5 times higher than on loam and clay respectively. Furthermore, transfer increases with increasing soil organic matter content. The impact of pH in the range pH 4.5 to 7.5 on soil-plant transfer is negligible. Experimental factors were also found to have impact on transfer of Cs. In lysimeters transfer appeared to be roughly 3 to 4 times lower than in the field, which might be caused by differences in soil and crop management as well as in the way soil was contaminated.

The impact of above-mentioned factors suggests that these factors must have caused regional and seasonal differences in the transfer of Cs originating from Chernobyl.

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after the Chernobyl accident were not considered because effects of direct deposition on the plant might disturb the analysis on transfer from the soil. A total number of 289 observations from Belli & Sansone, Eriksson et al. and MacNeill could be used. Soils with more than 20% OM were regarded as sandy soil with a high OM content.

In the classes 20-60% and >60% OM there were too few comparable data to draw any conclusions. In the range 1-20% OM median relative TF_{SP} values seem hardly influenced by the OM content (Table 36). This finding does not agree with the result of the evaluation of non-Chernobyl data, where a significant effect of OM on TF_{SP} was observed (par. 2.3.2.1 'Soil organic matter').

Time lag

The effect of time lag could be analyzed with 220 data on alfalfa, barley and grass, collected by Belli & Sansone and Eriksson et al. over a period of two years. Soil-to-grass data collected shortly after the Chernobyl accident were neglected again. After clustering data, according to plant species, plant part, investigator, soil type and OM, relative TF_{SP} values were calculated as a function of time lag. The TF_{SP} determined in October 1986 was used as reference.

About one year after contamination transfer was less than half of its value in October 1986 (Table 37). In fact, this effect cannot be distinguished from influences of weather conditions which differ each growth season. Assumed that such influences in 1987 have been small, the observed effect of time lag on TF_{SP} is comparable to time-lag effects found on non-Chernobyl measurements (see par. 2.3.2.1 'Time lag').

PLANT	SOIL	OM%	TIME LAG		
			6	8	10
			APRIL’87	JUNE’87	AUGUST’87
ALFALFA	LOAM	1-3	0.91	0.50	0.55
		3-7	0.61	0.32	0.48
		7-20	0.52	0.26	0.47
	SAND	1-3	0.51	0.24	0.29
		3-7	0.27	0.07	0.27
BARLEY	CLAY	1-3			0.67
		3-7			0.55
	LOAM	1-3			0.31
		3-7			0.23
		7-20			0.42
	PEAT	20-60			0.14
		>60			0.06
	SAND	3-7			0.20
		7-20			0.15
		20-60			0.18
GRASS	CLAY	1-3			0.30
	LOAM	1-3			0.34
		3-7			0.52
	PEAT	20-60			1.21
MEDIAN RELATIVE TF_{SP}			0.5[a]	0.3[a]	0.3[a]
	MIN TO MAX		0.3-0.9	0.1-0.5	0.1-1.2

[a] TF_{SP} values are significantly lower than in October 1986 (Wilcoxon’s signed-ranks test, $P<0.05$).

Table 37: Relative TF_{SP} values (relative cell values) of Cs for different soil-plant systems in relation to time lag (month). Values are relative to those measured in October 1986 (time lag= 0).

2.3.3 Experimental studies in Denmark

The experiment was carried out at Risø National Laboratory in the summer of 1988 and 1989 as part of an experiment concerning root uptake of ^{137}Cs and ^{134}Cs. The experiment was made in order to identify crops with a relatively low or high root uptake of radiocaesium with regard to countermeasures after an accidental release of radionuclides (Øhlenschlæger M. and G. Gissel-Nielsen, 1989).

A part of the project was carried out in soil which originates from Gävle in Sweden, north-west of Stockholm, site of the largest deposition of radiocaesium in Sweden following the Chernobyl accident. Characteristics of the soil are given in table 38.

Soil type	organic
Content of organic matter (%)	66
Particle size in mm	
<0.002 mm	4.4
0.002-0.02 mm	7.7
0.02-0.2 mm	15.9
0.2-2.0 mm	6.0
pH	5.2
mg K/100 g soil	8.0

Table 38.

The soil, which was contaminated with ^{137}Cs from the Chernobyl accident was experimentally supplied with ^{134}Cs, in the summer 1988, in order to investigate differences between experimentally added caesium and Chernobyl caesium. The experimentally added caesium was added as $CsNO_3$ from a 0.1 m HNO_3 solution.

The results for barley grain given as concentration ratios, CR, (Bq/g plant per Bq/g soil, dry wight) are shown in Table 39. The results demonstrate differences between experimentally added Cs and Chernobyl Cs. In 1988 the uptake from experimentally added Cs is about a factor 4 higher than for Chernobyl Cs. In 1989 this difference is reduced, the root uptakes are of similar magnitude.

Soil and isotope	Variety	Year	Grain	Straw
Organic soil experimentally added Cs-134	Golf	1988	0.476±10 %	0.679±12 %
		1989	0.147±7 %	0.113±14 %
	Apex	1988	0.471±7 %	0.764±9 %
		1989	0.140±8 %	0.118±5 %
	Anker	1988	0.602±7 %	0.700±9 %
		1989	0.210±13 %	0.146±11 %
	Sila	1988	0.640±7 %	0.921±9 %
		1989	0.171±19 %	0.158±9 %
Organic soil Chernobyl Cs-137	Golf	1988	0.119±10 %	0.239±12 %
		1989	0.107±7 %	0.134±11 %
	Apex	1988	0.117±7 %	0.219±11 %
		1989	0.128±13 %	0.196±26 %
	Anker	1988	0.154±6 %	0.240±8 %
		1989	0.148±13 %	0.161±13 %
	Sila	1988	0.179±9 %	0.342±9 %
		1989	0.144±16 %	0.226±47 %

Table 39: Concentration ratios, Bq per g plant/Bq per g soil, dry weight, for Barley. Means of four replicates ±1SE.

It is well established that caesium is subject to consederable fixation in the soil as a result of its entrapment in the lattice structure of clay minerals (Nielsen B. and M. Strandberg, 1988; Squire H.M. and L.J. Middleton, 1966). Although the soil considered here is organic, and has a reduced absorption and subsequent fixation of caesium on clay minerals, the fixation is still of considerable importance. Squire and Middleton (1966) conclude that the fixation appears to be nearly complete after 3 years.

This agrees very well with our results where the fixation of the Chernobyl caesium is nearly complete after 3 years and very little further fixation can be seen. While the caesium experimentally added in 1988 still are exposed to fixation. An analysis of variance supports this showing a highly significant difference (>99.9 %) between the concentration ratios for the experimentally added caesium in 1988 und 1989, while there are no significant differences between the concentration ratios in 1988 and 1989 for Chernobyl caesium.

Part of the difference could be due to chemical differences between caesium from the Chernobyl accident and experimentally added caesium. In order to examine this further the CR values are plotted against the number of years since the contamination for the four varieties. The results are shown in Figure 15 and Figure 16 for grain and straw, respectively.

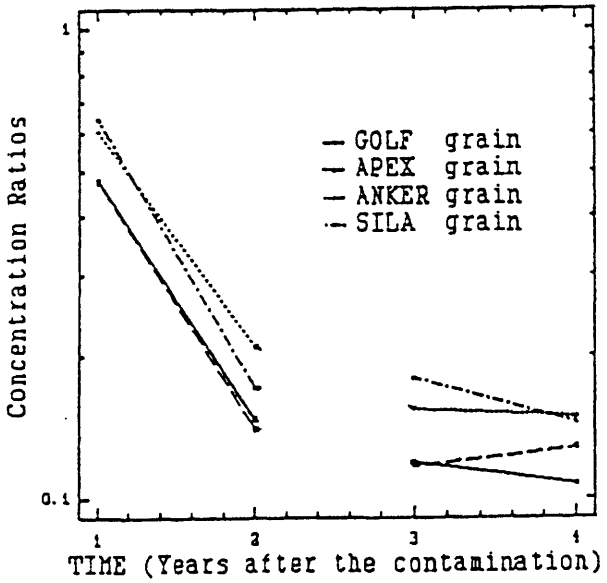

Figure 15: Concentration ratios. Showing the time trend for barley grain.

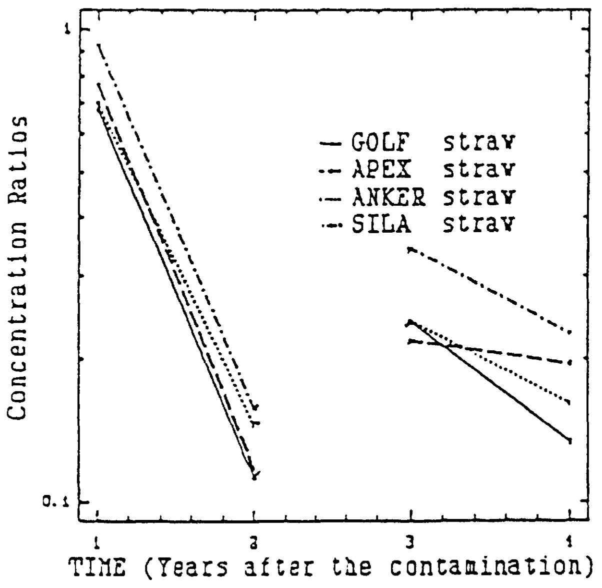

Figure 16: Concentration ratios. Showing the time trend for barley straw.

Assuming no difference between experimental added caesium and Chernobyl caesium, the CR values should decline in a similar way. With regard to the results for the grain this seems to be a plausible assumption but the results for the straw indicate some difference. In order to examine this further the experiment will continue and differences in the fixation trend for the two isotopes will be studied.

2.4 SUMMARY AND CONCLUSIONS

2.4.1 Objectives

Soil-to-plant transfer of Cs originating from Chernobyl was in some parts of Europe higher than expected on the basis of earlier observations. Main aim was to investigate whether Cs from Chernobyl is indeed more easily transferred from soil to plant than Cs from non-Chernobyl sources, i.e. remnants of nuclear weapon tests in the sixties, discharge of nuclear industry or artificial additions to soil. Because Chernobyl and non-Chernobyl data were generally sampled under different circumstances (region, year, crop, etc.) the second aim was to evaluate the effects of these parameters on the transfer of Cs. This analysis allowed corrections for environmental and experimental differences which have been used to compare Chernobyl and non-Chernobyl transfer measurements. These correction factors may also be valid for other comparisons of large amounts of soil-plant transfer data.

2.4.2 Experimental studies

Chernobyl transfer in South-Germany

Transfer factors (TF_{SP}) for radiocesium ($^{134/137}Cs$) in cereals grown on three different soil types - Cambisol (Terrassenbraunerde), calcaric Fluvisol (Allochthone Vega) and Planosol (Pseudogley) - in one of the highest contaminated areas in Germany (Upper Swabia) were determined. The variations of TF_{SP} within the group of Cambisols and Fluvisols were very small. Only in the group of soils with a perched water table (Pseudogley), variations were higher. This might be explained by local differences in the water regime of that soil type. In contrast to brown soils and alluvial soils, soils with a perched water table have much more differences in topography in that region. Therefore, water regime is changing drastically in small distances as well as other soil parameters.

The medium TF_{SP} of Cambisol and Fluvisol can be distinguished to the medium TF_{SP} of the soil with a perched water table, which is significantly higher. Differences in TF_{SP} of Cambisol and Fluvisol are not significant.

Lysimeter experiment with contaminated sewage sludge

In lysimeters filled with undisturbed soil monoliths of a podzolic gleysol (FAO classification: Dystric Gleysol) with an extremly high content of sand and a very low pH the transfer of $^{134/137}Cs$ into potatoes, lettuce and grass due to the fertilization with sewage sludge contaminated after the Chernobyl accident was investigated. As compared to the untreated control the uptake of radiocesium into potatoes and lettuce showed a slight increase after fertilization of the soil with sewage sludge (1 kg D.M./m^2, 13,000 Bq $^{134/137}Cs$/kg D.M.). Nevertheless the content of radiocesium in potatoes and lettuce did not reach 1 Bq/kg of fresh plant weight. Even the radiocesium content in grass of the first cut increased significantly after fertilization with contaminated sewage sludge. In the grass from the second cut the radiocesium content decreased due to the lower contamination of the plants.

Conclusions from these results are that the use of sewage sludge as fertilizer even high contaminated with $^{134/137}Cs$ does not lead to a serious increase of radiocesium in plants grown on arable land. Also in pasture grass the increase of radioactivity seems to be neglectable after fertilization with sewage sludge contaminated with radiocesium.

Chernobyl transfer in Northeast-Italy

^{137}Cs transfer factors from soil to forage crops have been evaluated from data collected in 1986 and 1987 in the Friuli-Venezia Giulia region. Sampling stations have been selected on the basis of a regional soils study and ^{137}Cs Chernobyl depositions.

^{137}Cs transfer factors from 1987 data have been evaluated in soils collected up to 10 cm of depth because all sampling stations were unploughed after Chernobyl and therefore the most part of ^{137}Cs contamination has remained in the superficial layers.

Soil to forage crops transfer factors from 1986 data are generally higher than those from 1987 data. This behaviour could be due to translocation from roots, stolons etc. to leaves, of ^{137}Cs previously translocated during the deposition period, from leaves to other parts of plants.

The 1987 data show that the lowest ^{137}Cs transfer factors from soil to forage crops have been found in soils with a content >40% of particles with size greater the 2 mm. In medium clayey, clayey and medium silty soils the transfer factor values are similar. This behaviour could be due to the highest mean values of extractable phosphorus and potassium in the CML class and to the deeper root system of forage crops in presence of coarse soils and gravel.

2.4.3 Statistical studies

Chernobyl transfer versus non-Chernobyl transfer

Results show that the soil-to-plant transfer factor of Cs originating from Chernobyl was about a factor 2 lower than the transfer factor for Cs from other contamination sources under comparable conditions. Three main factors (time lag, soil organic matter and translocation from older plant parts) may explain the difference between this result and the initially expected higher transfer of Cs:

1) Cs becomes less available in the course of time. After 5 to 10 years the availability has decreased with about a factor 10. Direct comparison, without correction for time lag, of Chernobyl data values with measurements on transfer of nuclear weapons fallout may thus overestimate the transfer up to about a factor 10.
2) Part of the Chernobyl data refers to upland soils in north and north-western Europe. The greater part of these soils is characterized by a high soil organic matter content, extremely low pH values, a low clay mineral content and a poor nutrient status, usually under a humid, cool climate. Transfer on soils with high organic matter contents were shown to be about ten times larger than on soils with less than 10% organic matter.

In fact, processes related to a high organic matter content of the soil may cause an increased transfer of Cs. Under the prevailing climatological conditions microbial decomposition of soil organic matter can lead to the formation of NH_4^+ ions which prevent Cs ions from adsorption on the solid phase.

3) High transfer factor values measured in spring and summer after the Chernobyl accident may partly refer to direct uptake by above ground plant parts in stead of uptake by the roots from the soil, even when the harvested plant parts were growing after the deposition. Translocation from plant parts (roots, stolons, etc.) present during deposition to parts which developed after deposition may be the cause of unexpectedly high Cs contents in the harvest.

Region and season

Effects of region and season can be traced back to differences in soil type, variation in crops, differences in agricultural practices and above all varying weather conditions (temperature, rainfall). Dutch observations collected over a period of 4 to 7 years show that climatological factors cause fluctuations in transfer per season of maximal a factor 10, depending on the type of crop. Increases in transfer seem to correspond with decreases in rainfall or temperature.

The transfer factor of Cs isotopes on sandy soils appears 3 to 5 times higher than on loam and clay respectively. Furthermore, transfer increases with increasing soil organic matter content. The impact of pH in the range pH 4.5 to 7.5 on soil-plant transfer is negligible. Experimental factors were also found to have impact on transfer of Cs. In lysimeters transfer appeared to be roughly 3 to 4 times lower than in the field, which might be caused by differences in soil and crop management as well as in the way soil was contaminated.

The impact of above-mentioned factors suggests that these factors must have caused regional and seasonal differences in the transfer of Cs originating from Chernobyl.

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IV PLANT-ANIMAL TRANSFER

(CEN, ITE, Macauly, NRPB,ENEA)

1. SEMI-NATURAL ECOSYSTEMS

1.1 INTRODUCTION

The Chernobyl accident occurred at a time of year when few animal-based food products were being taken from semi-natural ecosystems in western Europe. The combination of this timing and the isotopic composition of the fallout meant that only the comparatively long-lived radiocaesium isotopes, ^{137}Cs and ^{134}Cs, were of concern as sources of radioactivity in animal products from these ecosystems.

The majority of information available in the scientific literature on the behaviour of radiocaesium in semi-natural ecosystems has come from studies conducted in Scandinavia, the USSR and the USA. Hence there was little site specific information available on the transfer of radiocaesium to animals grazing in semi-natural ecosystems in the majority of European countries. However, some conclusions drawn from this work were relevant to the problems subsequently encountered in Europe. The most pertinent data available prior to the Chernobyl accident are reviewed in section 1.2.

Western European semi-natural ecosystems often occur in upland regions. Consequently the high correlation between rainfall and the deposition of Chernobyl fallout over Europe meant that they often received high levels of radioactivity. This led to high radiocaesium activity concentrations in many vegetation species which was subsequently reflected in grazing animals.

Animals grazing semi-natural ecosystems are the most common, direct route by which radioactive contamination in these areas reaches the human food chain. Although there is some cropping of species such as wildfowl, hare and rabbit and products such as honey, ruminants form the most important pathway to man. Some ruminants are hunted, such as moose and deer. Other species, which are more actively managed (eg hill sheep and cattle or reindeer), are allowed to graze freely for some of the year and are collected at intervals for culling etc. Some mountainous areas are only commercially grazed in the summer months, for instance by dairy cattle, goats and sheep in Alpine areas, Greece and Scandinavia.

Semi-natural ecosytems provide a wide variety of habitats for ruminants. Different ruminants may occupy different ecological niches in the same ecosystem. This natural diversity makes comparative studies of the transfer of radiocaesium more complex than in agriculturally managed systems. Since the Chernobyl accident there have been many studies of the transfer of radiocaesium to ruminants in semi-natural ecosystems. A review of these investigations is presented in section 1.3. The difficulties of studying the transfer of radiocaesium to grazing animals in these systems are discussed and potential methods for doing so evaluated.

The final section of this report discusses advances in our understanding of the behaviour of radiocaesium in semi-natural ecosystems since the Chernobyl accident. Potential countermeasures that are appropriate to these ecosystems are also reviewed.

1.2. RADIOCAESIUM CONTAMINATION OF ANIMALS IN SEMI-NATURAL ECOSYSTEMS PRIOR TO THE CHERNOBYL ACCIDENT

There is a considerable amount of literature from studies in Scandinavia and North America during the nuclear weapons fallout period of the 1950's and 60's. Such studies were able to identify various characteristics of semi-natural ecosystems which caused high radiocaesium transfers to ruminants, particularily in arctic areas. Some work was also conducted in the USA on the transfer of nuclear weapons fallout radiocaesium into animals in mountainous areas and coastal plains. Other studies were conducted in semi-arid ecosystems in the USA around the nuclear test sites, but these are not relevant to the ecological conditions prevailing in much of Europe. An exception are the studies on semi-natural ecosystems in the FRG by Bunzl and Kracke (1984a; 1984b).

Year	Country	Animal species*	^{137}Cs concentration ($Bq\ kg^{-1}\ FW^{+}$)	Reference
1961-65	Finland	reindeer	580 - 2680	Miettinen & Hasanen 1967
1962-64	USA	wild mule deer	10 - 120	Whicker et al. 1967
1962-65	USA	wild mule deer	16 - 23	Pendleton et al. 1964, 1965
1963-65	Canada	reindeer/caribou	max 6400	Rad. Prot. Div. 1967
1963-65	Sweden	reindeer	70 - 3920	Lindell & Magi 1967
1964-65	Alaska	caribou	90 - 1110	Hanson et al. 1967
1965	USSR	reindeer	2180 - 4880	Nevstrueva et al. 1967
1965-66	UK	upland sheep	max 220	ARCRL 1966
1966-70	Norway	sheep	104 - 250	Hove & Strand 1989
1963-66		goat cheese	max 800	
1966-67	FRG	red deer	max 410	Schonhofer & Tataruch 1988
1968-69	USA	whitetail deer	9 - 5660	Johnson & Nayfield 1970
1970	Austria	chamois	230	Schonhofer & Tataruch 1988
1978-79	Canada	moose	4	Zach et al. 1989
		whitetail deer	3	
1979	Canada	moose	230	Zach et al. 1989
1979	Finland	moose	30	Rantavaara 1982
1980-81	FRG	roe deer (liver)	4	Bunzl & Kracke 1984a
		moorland sheep (liver)	6 - 240	
		chamois (liver)	9	
1985	Finland	reindeer	300	Rissanen & Rahola 1989

* for muscle tissue unless otherwise stated

$^{+}$ FW = fresh weight

Table 1: Some pre Chernobyl radiocaesium activity concentrations in animals from semi-natural ecosystems contaminated by nuclear weapons fallout.

Indigenous and domesticated fauna in a wide variety of semi-natural ecosystems were found to readily accumulate radiocaesium (Table 1). Radiocaesium activity concentrations considerably higher than for domestic animals in agricultural areas were found in wild game (Whicker et al. 1968; Johnson & Nayfield 1970), moorland sheep (ARCRL 1966; Bunzl & Kracke 1984b) and in milk from cows grazing mountain pastures (Swedjemark et al. 1967) and natural vegetation on flooded riverbanks (Miettinen 1979). In muscle samples from red deer, roe deer and moose in the USSR ^{137}Cs concentrations were 3-20 fold higher than cattle in the same region (Sokolov et al. 1987).

Semi-natural ecosystems often experience high rates of annual precipitation and as a result radiocaesium deposition of nuclear weapons fallout was higher than average (Whicker & Schultz 1982). However, the high levels of radiocaesium in ruminants were not solely due to high deposition, but were also caused by ecological factors (Miettinen 1969).

A number of ecological characteristics, in different semi-natural ecosystems, which enhanced the transfer of radiocaesium into grazing animals were identified:

a. Effect of Soil Type

The soils were unmanaged (ie not fertilized etc), often shallow and of poor nutrient status. Soils with a high organic content which are present in some semi-natural ecosystems were shown to allow greater radiocaesium uptake than mineral soils (ARCRL 1963; Scott Russell 1967; Rantavaara 1982).

b. Contaminated Vegetation Species

Lichens, the dominant plant species in some of the areas studied, were shown to be efficient in trapping and retaining fallout radionuclides. Biological half-lives for radiocaesium contamination of lichen of up to 17 years were suggested (Liden and Gustafsson 1967). However some higher plants in temperate areas, such as ericaceous species, were also found to concentrate radiocaesium (Bunzl & Kracke 1984b).

c. Animal Diet Selection, Grazing Behaviour and Seasonal Variation

Often one or two highly contaminated vegetation species constituted an overwhelmingly important proportion of the diet at certain times of year. A classic example was the lichen - reindeer - man food chain in the Arctic (Aberg & Hungate 1967). Highly contaminated lichens formed practically the only food source for reindeer for approximately 6 months of the year (Miettinen 1969) resulting in adiocaesium activity concentrations exceeding 2500 Bq kg^{-1}FW in Scandinavia in the 1960's. However, during the summer, when the less contaminated grasses and forbs were being consumed, radiocaesium levels in reindeer fell to 13 - 52 % of those in the winter (Liden & Gustafsson 1967). The effect of diet was also demonstrated for deer in the USA. Whitetailed deer from coastal plains in Florida had high radiocaesium activity concentrations, apparently associated with the ingestion of highly contaminated mushrooms (Johnson & Nayfield 1970). Similarily sheep in West Germany, grazing mainly on contaminated Calluna vulgaris, were found to have comparatively high radiocaesium activities (Bunzl & Kracke 1984a;b).

In Colorado seasonal patterns in the ^{137}Cs activity of mule deer due to grazing behaviour were seen (Whicker & Shultz 1982). Between 1962 and 1965 the highest radiocaesium activity concentrations were found in the summer when the deer were grazing above 8500 ft. However, in Washington the opposite seasonal pattern was seen. The highest activities were found in winter and were thought to be due to deer grazing older plant parts, which had been exposed to fallout for longer than the succulent new growth grazed in the summer (Eberhardt et al. 1969).

Diet selection of ruminants grazing in the same area caused large differences in radiocaesium activities. Reindeer, which grazed highly contaminated lichen, were often found to have ten fold higher radiocaesium activity concentrations than moose, which do not graze large quantities of lichen (Whicker & Shultz 1982). In Wyoming, sheep were found to have a greater radiocaesium activity during the summer than antelope grazing the same pastures (Eberhardt et al. 1969). At other times of the year radiocaesium activities of the two ruminant species were similar. Unfortunately, although sheep were observed to graze mainly on grasses and antelope on shrubs, the vegetation species responsible for the differences were not identified.

d. High Transfer Coefficients

There was some indication that transfer coefficients for radiocaesium to wild ruminants were comparatively high when compared with cattle. Values recommended for white-tailed deer, black-tailed deer, reindeer and moose were 0.3, 0.07, 1.0 and 0.2 d kg^{-1} respectively (Zach et al. 1989; Ng et al. 1982) The transfer coefficient to beef was generally taken to be of the order of 0.03 d kg^{-1} (Ng et al. 1982). Similarily the transfer coefficient for radiocaesium to the flesh of sheep (an important domesticated animal in semi-natural ecosystems) was thought to be an order of magnitude higher than that for cattle (Ng et al. 1982).

e. Long Ecological Half-lives

The ecological half-life of radiocaesium contamination of reindeer meat was determined to be at least 5 y in Scandinavia (Lindell & Magi, 1967; Liden & Gustafsson, 1967). This was considerably longer than the half-lives of radiocaesium in animal products from agricultural areas which were generally in the order of a few weeks.

Contamination of animals in semi-natural ecosystems due to discharges from nuclear facilities

Apart from observations of weapons fallout ^{137}Cs in semi-natural ecosytems there are some reports of comparatively high radiocaesium activities in the tissues of animals in semi-natural ecosystems close to nuclear facilities. Examples of these are given in Table 2. However, they are site specific (eg studies on the environmental behaviour of liquid effluent from the Sellafield Reprocessing Plant) and of limited use in predicting the behaviour of radiocaesium in most semi-natural ecosystems.

Year	Country	Animal species	^{137}Cs concentration (Bq kg^{-1} FW)	Reference
1966	USA	whitetailed deer	80 - 230	Rabon 1968
<1968	USA	whitetailed deer	max 650	Brisbin & Smith 1975
1974	USA	pronghorn	max 60	Markham et al. 1982
1974-78	USA (Reactor Pond)	waterfowl	max 151000	Halford et al. 1981
1977-82	UK	saltmarsh sheep	8 - 64	Bradford & Curtis 1984
1979	UK	saltmarsh cattle	5 - 70	Sumerling 1981
1981	UK	saltmarsh wildfowl	60 - 160	Lowe & Horrill 1986
1982	UK	saltmarsh sheep	110 - 340	Howard & Lindley 1985

Table 2: Some pre Chernobyl radiocaesium activity concentrations in the muscle of animals from semi-natural ecosystems contaminated by nuclear facilities.

1.3. RADIOCAESIUM CONTAMINATION OF RUMINANTS IN SEMI-NATURAL ECOSYSTEMS AFTER THE CHERNOBYL ACCIDENT

High contamination of ruminants has been reported in a variety of semi-natural ecosystems following the deposition of fallout from the Chernobyl accident. In many countries contamination levels exceeded those during the weapons testing era (Table 3).

Results and conclusions that can be drawn from many of these radioecological studies are reviewed below.

Species	Country	Habitat	$^{137}Cs^{*}$ ($Bq\ kg^{-1}$ FW)	Reference
Sheep	UK	Upland pastures	5000	MAFF 1987
Red Deer	UK	Moorland	1600	Horrill et al. 1988
Sheep	Ireland	Upland pastures	3000+	Colgan & Scully 1989
Deer	FRG	Forest	4000	Kreuzer & Hecht 1988
Cow milk	Austria	Alpine valleys	>550	Henrich et al. in press
Roe Deer	Austria	#	11500	Schonhofer & Tataruch 1988
Red Deer	Austria	#	1000	Schonhofer & Tataruch 1988
Chamois	Austria	#	3800	Schonhofer & Tataruch 1988
Cattle	Norway	Mountain pastures	3000	Hove & Ekern 1988
Sheep	Norway	Mountain pastures	8000	Hove & Ekern 1988
Reindeer	Norway	Mountain pastures	90000	Hove & Ekern 1988
Goat milk	Norway	Mountain pastures	1000	Hove & Ekern 1988
Cow milk	Norway	Mountain pastures	500	Hove & Ekern 1988
Roe Deer	Sweden	#	20000	Jones 1989a
Moose	Sweden	Forest	5000	Karlen et al. pers comm.
Reindeer	Sweden	#	75000	Jones 1989b

* Results for muscle unless shown otherwise, results for milk are in Bq l^{-1}

+ ^{134}Cs + ^{137}Cs

Habitat not given

Table 3: Some examples of the high radiocaesium concentrations in the tissues of ruminants grazing natural and semi-natural ecosystems, following deposition from the Chernobyl accident.

a. Species Differences and Diet Selection

Considerable differences have been found in the radiocaesium activity concentrations of different animal species grazing in the same ecosystem. In Austria ruminants were found to be more contaminated than monogastric species (wild boar, hare, grouse) in the early months following deposition (Schonhofer & Tataruch 1988). Roe deer in both Austria and Sweden were found to have higher radiocaesium concentrations than other ruminants in the same area, ie moose in Sweden (Karlen et al. pers comm.) and chamois and red deer in Austria (Schonhofer & Tataruch 1988). In Austria this was thought to be due to the comparatively greater ingestion of more highly contaminated herb species by roe deer. However, recently the transfer of radiocaesium from contaminated hay to roe deer was found to be higher than that to sheep and cattle in feeding trials (Keszthelyi et al. 1989). The transfer coefficient was found to be 0.35 d kg^{-1} for roe deer compared with 0.08 d kg^{-1} for sheep and 0.007 d kg^{-1} for beef.

The selective grazing of highly contaminated vegetation species has been shown to cause high radiocaesium activity concentrations in ruminants in many areas of Europe. Studies using animals fitted with oesophageal fistulates in contaminated regions of Norway have been useful in demonstrating the importance of grazing selection (Garmo et al. 1988; 1989). Vegetation selected by sheep was generally found to have lower radiocaesium activity concentrations than that selected by goats. Vegetation selected by reindeer (often highly contaminated lichens and fungi) was more contaminated than that selected by either goats or sheep which grazed less contaminated forbs and grasses). Studies with the same animals in different semi-natural habitats which received similar levels of deposition have shown that the radiocaesium intake of goats and sheep is highest in areas of dry heather/semi-natural pastures compared with willow thickets, meadows or birch forest (Garmo et al. 1988).

Although the wild boar is not a ruminant, in Austria this species gives a good example of changes in radiocaesium concentration with time (Schonhofer & Tataruch 1988). In 1986 activity concentrations in wild boar were found generally to be below 40 Bq kg^{-1} FW (max 600 Bq kg^{-1}). However in 1988 values were usually greater than 2000 Bq kg^{-1} and a maximum concentration of 17600 Bq kg^{-1} FW was observed. This was the most contaminated meat sample found in Austria following the Chernobyl accident.

b. Seasonal Trends

Selective grazing of contaminated forage species at certain times of the year has led to seasonal patterns in the radiocaesium contamination of animals in semi-natural ecosytems.

As during the weapons testing era, highly contaminated lichens have been shown to be responsible for high levels of radiocaesium in reindeer during the winter (Jones et al. 1989; Garmo et al. 1989). Radiocaesium transfer coefficients for reindeer on winter (65 % lichen) and summer (>80 % vascular plants) feeds were found to be 0.65 d kg^{-1} and 0.3 d kg^{-1} respectively (Jones et al. 1989). It was suggested that the difference may be due to the low K^+ concentration of lichens. However, the authors stressed that the animals would not have been at equilibrium when these transfer coefficients were obtained.

The activity of moose in Sweden increased in the autumns of 1987 and 1988 when they were grazing higher quantities of *Calluna vulgaris* and *Vaccinium myrtillus* (von Bothmer et al. 1990). The activity of these two vegetation species in 1987/88 was found to be approximately 13000 Bq kg^{-1} and 4100 Bq kg^{-1} DW respectively compared with <2000 Bq kg^{-1} DW for herbaceous and grass species grazed by moose. These two ericaceous species are also thought to be responsible for maintaining comparatively high levels of radiocaesium in forest roe deer during the winter (Karlen et al. pers comm.).

The considerable effect of the seasonal ingestion of highly contaminated fungi species has been highlighted since the Chernobyl accident. In July 1988 contamination levels of goats' milk and reindeer from mountainous districts of Norway increased greatly although little change was observed in the activity concentration of green vegetation (Hove & Strand 1989). The increase coincided with an abundant crop of highly contaminated fungi (>500 kBq kg^{-1} DW, Bakken & Olson 1988) which ruminants were seen to graze. No such increase was observed in 1989 when fewer fungi appeared. The availability of radiocaesium from fungi for gut absorption has been shown to be high (Hove & Solheim Hansen 1989). The ingestion of fungi was also thought to be responsible for high radiocaesium concentrations in the tissues of forest deer in Sweden and Austria during late summer and early autumn (Karlen et al. pers comm.; Schonhofer & Tataruch 1988).

There are other factors which may influence the variation in radiocaesium activities of grazing animals throughout the year. For instance, Howard et al. (in press) have found that the radiocaesium activity concentration of sheep grazing upland pastures in the UK is higher in the summer than in the winter. Although grazing selection will contribute to seasonal differences, throughout the year grasses such as *Festuca* spp. and *Agrostis* spp. appeared to be the main component of the diet. They suggest that the combination of higher herbage intake and digestibility during the summer months (see Armstrong & Hudson 1986) will lead to more radiocaesium being absorbed in the gut.

c. Management Effects

Management practices may influence radiocaesium concentrations. For instance, animals which are removed to less contaminated agricultural land during the winter will initially have lower radiocaesium activity concentrations when they are returned to natural pastures the following spring. Variations in the radiocaesium levels of sheep due to management occur in upland areas of the UK (Howard & Beresford 1989). In these areas sheep are removed from open pastures to nearby enclosed fields for short periods throughout the year, their radiocaesium activity decreases whilst they are in enclosed grazing and increases again when they are returned to the open pastures. Wild ruminants with access to agricultural land have been found to have lower radiocaesium concentrations than those purely inhabiting natural ecosystems (Karlen et al. pers comm.; Kreuzer & Hecht 1988; Johanson & Bergstrom 1989).

d. Longterm Changes in the Radiocaesium Contamination of Ruminants

Sufficient time has elapsed since the deposition of Chernobyl fallout to enable long term comparisons of the behaviour of Chernobyl radiocaesium in a variety of ecosystems and to allow estimates of ecological half-lives to be made. Radiocaesium levels of ruminants in many semi-natural ecosystems are generally declining only slowly. In Sweden an average yearly reduction in the radiocaesium activity of moose and roe deer of 10-15 % has been reported (Jones 1989a). This is similar to the average reduction of 20% in radiocaesium activity of Norwegian sheep between 1986 and 1987 (Garmo et al. 1988). However, in some areas there has been no significant decrease and in others levels have increased. Some upland sheep in the UK had similar activities in 1987 and 1988 to those in autumn 1986 (Howard et al. in press). Similarily no decline in the radiocaesium activity of upland sheep was found at some farms between 1987 and 1988 in Ireland (Colgan & Scully 1989) and in some areas of Norway between 1986 and 1987 (Hove & Ekern 1988). In Sweden no change was observed in the milk of some of the cows grazing unimproved pastures between 1986 and 1988 (Johanson & Bergstrom 1989).

It seems that the availability of Chernobyl radiocaesium for absorption by grazing animals is higher now than immediately after it was deposited in 1986. This is thought to be due to a greater availability of radiocaesium when incorporated into plant tissues rather than when present as fallout on vegetation surfaces. Transfer coefficients derived for sheep on upland pastures in the UK were 2-3 times higher in the summer of 1987 than those derived immediately following deposition of fallout (Beresford et al. 1989). This observation has since been confirmed in other studies (Ward et al. 1989; Hove & Solheim Hansen 1989). It may help to explain why, although activity concentrations of vegetation were slowly declining, sheep grazing upland pastures (UK) had similar levels of ^{137}Cs in their muscle in the summer/autumn of 1987 and 1988 as in autumn 1986 (Howard & Beresford 1989). Similarily the activity concentration ratio of upland Norwegian sheep tissues to that of grazed vegetation was two fold higher in 1987 compared with 1986 (Garmo et al. 1988).

Many studies have noted that it is usually the low caesium immobilization capacity of the soil (often acid, with a high organic and low content of the clay minerals which bind radiocaesium) and subsequent high radiocaesium uptake by vegetation which is responsible for the continuingly high radiocaesium activities in the grazing animals of most semi-natural ecosystems (eg Howard et al. in press; Hove & Strand 1989; Henrich et al. in press).

Coniferous tree needles are efficient filters of radioactivity, but have a slow leaf turnover time. Therefore any direct deposit remaining on the needle may not reach the soil surface for 5-7 years where its decomposition will be slower than leaves in deciduous woodland. They may therefore act as a slow input source, helping to maintain high radiocaesium levels in grazing animals in conifer forests (Schonhofer & Tataruch 1988; Henrich et al. 1989).

e. Half-life Predictions

A number of predictions on the likely half-life of Chernobyl radiocaesium in ruminants of semi-natural ecosystems have now been made. These are heavily dependant on the characteristics of the ecosystem. An effective half-life for the lichen-reindeer foodchain of 10-15 y has been suggested in Sweden (Jones 1989a). In contrast, the ecological half-life of Chernobyl radiocaesium was estimated in May-July 1986 to be approximately 28 d for roe deer in a UK forest (Lowe & Horrill 1988). This value would obviously be too short for forest roe deer in other parts of Europe (Schonhofer & Tataruch 1988; Karlen et al. pers comm.) and may be a reflection of either the short term nature of the study or ecological characteristics. Following the Chernobyl accident scientists in Norway have calculated ecological half-lives for nuclear weapons fallout ^{137}Cs in sheep meat and goats' whey cheese (Hove & Strand 1989), to be approximately 20 and 15-25 y respectively. They also report similar transfer ratios from soil to lambs' tissues, goats milk and fungi for weapons fallout and Chernobyl derived radiocaesium implying that similar ecological half-lives can be expected for Chernobyl radiocaesium. Hence it appears that in the semi-natural systems of Norway physical decay of radiocaesium will be the major process in reducing levels in animal products from the affected areas. Similar results are now being found in some of the affected upland areas of the UK where the transfer of Chernobyl and pre-Chernobyl radiocaesium to vegetation appears to be similar (Howard unpublished data). It has been predicted that countermeasures to reduce the contamination of animals in Norwegian semi-natural systems will have to be used for 20 to 50 y (Hove & Strand 1989).

1.4. COUNTERMEASURES FOR REDUCING ANIMAL RADIOCAESIUM CONCENTRATIONS

Following the Chernobyl accident the high levels of radiocaesium contamination of animals and animal products from semi-natural ecosystems has necessitated the use of countermeasures in many European states. The radiocaesium activity above which meat cannot enter the human food chain is usually set between 300 and 1000 Bq kg^{-1} FW. There are some cases where it is much higher (eg 6000 Bq kg^{-1} FW for reindeer in Norway). However for goats milk in Norway it is lower at effectively 50 Bq L^{-1}, since the production of whey cheese increases the radiocaesium activity concentration approximately 10 fold (Hove et al. 1989).

The simplest countermeasures employed in semi-natural ecosystems have been to delay hunting seasons and to monitor animals before they are allowed to leave affected areas or enter the human food chain. Some countries have adopted a policy of feeding uncontaminated feeds before slaughter. In other countries (eg UK and Ireland) upland lambs are usually fattened on lowland pastures before slaughter. As the radiocaesium is lost from these lambs with a half-life of about 10 d, the radiocaesium content of their meat is reduced before entry into the food chain without the need for countermeasures. In Sweden the more contaminated pastures were used to graze horses, breeding animals or young animals not intended for slaughter (Jones 1989b).

Accepted methods, whereby caesium binders such as bentonite, vermiculites or hexacyanoferrate salts are administered daily to reduce absorption of radiocaesium in the digestive tract of animals receiving a contaminated diet are unsuitable for most animals in semi-natural ecosystems. Animals in these ecosystems may range over large areas and it would be impossible to administer caesium binders with their feed every day, although this is possible for some dairy animals. For instance, it is now common practice in Norway to incorporate ammonium-hexacyanoferrate

(ACFC) into feed pellets which dairy goats receive daily whilst they are being milked. In this way the radiocaesium levels in the milk of goats in some mountainous areas of Norway were reduced by 70-85% in 1989 (Garmo et al. 1989).

Potassium fertilizers have been shown to reduce radiocaesium uptake by vegetation of unimproved pastures (Karlsen et al. 1989). However, the size and inaccessibility of many areas which would have to be treated if this method were adopted make it impractical on a large scale. In the UK attempts to reduce the radiocaesium activity of free-ranging sheep included the treatment of pastures with bentonite (Beresford et al. 1989). There was a reduction in the radiocaesium concentration of sheep grazing pastures repeatedly treated with bentonite. However, ewes on the treated pastures experienced a loss in body weight combined with a reduced herbage intake.

The most successful countermeasure techniques used since the Chernobyl accident are those developed in Norway. Two methods of administering ACFC to free-ranging animals have been developed: incorporation into slow release boli and saltlicks.

The slow release boli contain 20% ACFC in a fatty acid matrix (Hove et al. 1988) and are now routinely used to reduce radiocaesium contamination of reindeer in affected areas of Norway. A 60% reduction in the radiocaesium activity of reindeer meat has been achieved with an effective operation time of 2 months (Hove & Solheim Hansen 1989). Trials with goats, sheep and lambs have also been conducted: the radiocaesium activity concentration of goats milk was reduced by 80% for more than a month (Hove et al. 1989); sheep administered 2 boli were found to have 30-70% lower radiocaesium concentrations than untreated sheep after 6 wks grazing on mountain pastures (Karlsen et al. 1989); whilst in a further experiment a 50% reduction was found in lambs' tissues (Hove & Solheim Hansen 1989). The prolonged effectiveness of the boli makes them suitable for use in semi-natural ecosystems where animals are infrequently handled. Evaluation of their use in contaminated areas of the UK will begin in 1990.

ACFC impregnated NaCl saltlicks are now used to reduce contamination levels of mountain sheep in Norway. The salt-licks are left in areas where ordinary salt-licks are traditionally left. In 1988 treated sheep were found to have 75% lower radiocaesium activities than neighbouring untreated animals (Hove et al. 1989). However, results within groups of animals were very variable. The reduction in activity of individual reindeer ranged from 0 - 70%, suggesting that some of the animals do not use the saltlicks (Hove & Ekern 1988). The effectiveness of salt-licks, if employed in other areas of Europe, would depend on the willingness of animals to use them, especially at times of year when they may not be used to visiting ordinary saltlicks.

1.5. MEASURING THE TRANSFER OF RADIOCAESIUM TO ANIMALS IN SEMI-NATURAL ECOSYSTEMS

Measuring the transfer of radiocaesium to animals in contaminated semi-natural ecosystems is considerably more difficult than in agricultural areas; some authors have expressed the view that it is not possible (Schonhofer & Tataruch 1989). Few estimates of this parameter are therefore available and those given are usually accompanied by cautions concerning their accuracy.

Radiocaesium activity concentration in animals grazing in semi-natural ecosystems will be influenced by a large number of habitat-specific variables. There are many problems in trying to make precise measurements of the radiocaesium concentration of different components of the ecosystem and the transfer between different trophic levels. The problems are discussed below, drawing on general information concerning ruminants (Mayes 1989; Susmel et al. in press) and studies on ruminant radioecology, including a CEC sponsored study designed to help in this assessment conducted in the Highland Region of Scotland and in central Norway (Full details and results from these studies will be presented elsewhere):

a. Estimating Total Radiocaesium Intake

It is obviously necessary to locate animals so that vegetation can be sampled from grazed areas, which often vary with time of day as well as season. It is seldom easy to find study animals in many semi-natural ecosystems, particularily forests or very large areas of open pasture. Even if the area being grazed can be identified it is often difficult to sample it in a representative manner. Most ruminants are selective grazers, preferentially grazing certain vegetation species at different times of the year. Species differences occur: goats and sheep are highly selective grazers compared with cattle (Mayes 1989), hence it may be necessary to sample the same sward in a different manner for different ruminants.

To estimate the daily intake of radiocaesium three parameters must be estimated: species composition of the diet, total daily herbage intake and radiocaesium content of the grazed herbage.

One of the simplest methods of estimating species composition of the diet is by direct grazing observations of the study ruminants. This is extremely difficult for many studies conducted in semi-natural ecosystems. Vegetation species ingested can be estimated by examining plant fragments in rumen contents or faeces. However the faeces technique is only qualitative, since many faecal particles are not identifiable and the cuticular remains of some plants are more readily identified than others. Examination of rumen contents is also inaccurate since different plant species may be degraded at different rates. Oesophageal fistulas can be used to give a more precise indication of species intake since less degradation of ingested vegetation occurs. It also has the advantage that repeated measurements can be made on the same animals. But the use of this method is limited since animals have to be surgically prepared and continually supervised. There is also some doubt as to how representative results from such experimental animals are compared with free-ranging animals. Faecal and rumen content examination and oesophageal fistulates all give relatively short term estimates of species intake (< 2 d). The n-alkanes of plant cuticular waxes can be used as faecal markers to estimate diet composition, averaged over a week, since different species contain odd-chain alkanes (C_{23}-C_{35}) in differing proportions (Mayes 1989). However, daily dosing with even chain alkanes is needed to enable faecal output to be calculated.

Furthermore it is unlikely that the method would work if more than three herbage species are grazed.

The parameter which is often not measured when trying to estimate radiocaesium intake is the total daily herbage intake of the animal. For some wild animals this is virtually impossible to measure and estimates have to be taken from the literature. However for other, more accessible animals this should be an integral part of the study. A few attempts have been made to measure intake by calculating faecal output and herbage digestibility or using oesophageally fistulated animals (Beresford et al. 1989; Garmo et al. 1988). Oesophageal fistulates have the problems discussed above although they do allow direct estimates of radiocaesium intake. The accuracy of faecal markers (eg chromic oxide) will be limited in semi-natural ecosystems since a range of herbage species with differing digestibilities and radiocaesium activity concentrations may be ingested. Previously the use of faecal markers needed daily administration, however, this problem has recently been overcome by the development of slow release chromic oxide capsules which allow measurements to be made over a month (Ellis et al. 1981).

Even if herbage intake and species composition of the diet can be estimated the radiocaesium activity of the diet also has to be determined. Considerable uncertainty can be associated with this measurement due to the high degree of variability in radiocaesium levels of vegetation collected from even a small area (Table 4).

b. Factors Affecting the Availability for Absorption in the Gut

Uptake of ingested radiocaesium may be affected by many factors such as the type of vegetation, extent of soil adhesion, time after deposition of the fallout and age of animal. Many of these factors are inherently more variable in semi-natural ecosystems and few have been investigated sufficiently to allow them to be taken into account. Further studies, under controlled conditions, are needed to determine the availability of radiocaesium from natural vegetation species and its transfer to important wild herbivores.

Vegetation species	Radiocaesium activity concentration ($Bq\ kg^{-1}$ DW)	
	^{134}Cs	^{137}Cs
Calluna vulgaris	70-1380	360-7790
Nardus stricta	<20-80	140-460
Molinia caerulea	50-280	140-1590
Carex spp.	10-120	120-730
Ranunculus spp.	<20-100	410-900
Tricophorum cespitosum	30-340	210-2140
Deschampsia cespitosum	<40-190	120-1180

Table 4: Radiocaesium activity concentrations of vegetation species collected from an enclosed area grazed by sheep in Scotland.

c. Changes in the Radiocaesium Activity Concentration of Animals with Time

Since most animals in semi-natural ecosystems are handled infrequently, access to study animals is often limited. Studies of game animals often rely on samples from hunters or road kills their samples; as few samples are obtained out of the hunting season, seasonal trends may be based on more than 100 animals for some months and less than 10 in others. Combining this problem with the variability in radiocaesium activities found within groups of animals from semi-natural ecosystems it is evident that the results of such studies must be treated with caution. Radiocaesium activities of animals in semi-natural ecosystems vary with time due to changes in radiocaesium intake and availability or the influences of management practices. Therefore tissue levels are unlikely ever to be at equilibrium with dietary radiocaesium. This casts doubt on the validity of transfer coefficents derived in such ecosystems.

1.6. GENERAL DISCUSSION

Some conclusions drawn from the work in the 1960's and 70's in Scandinavia, the USA and the USSR could be applied to semi-natural ecosytems after the Chernobyl accident. However, the data was ultimately of limited use in continental Europe, particularly with respect to the dominant vegetation species, climate, soil types, animals and management practices.

Since semi-natural ecosystems often receive higher than average precipitation they are more liable to receive high levels of deposition following a nuclear incident. Hence comparatively high activities of radiocaesium will be deposited into systems with ecological characteristics which promote high radiocaesium levels in grazing animals. Therefore animals in semi-natural ecosystem will often be a potentially important source of radiocaesium following a nuclear incident.

The deposition of fallout from weapons testing occurred over many years and plant uptake of radiocaesium was thought to be relatively unimportant by some investigators (Scott Russell 1967). In contrast, fallout from the Chernobyl accident was deposited over a few days, largely as a consequence of the coincidence of rainfall with the contaminated plume. The importance of the link between soils which allow high radiocaesium uptake by vegetation and high radiocaesium concentrations in ruminants from semi-natural ecosystems has therefore been more evident since the Chernobyl accident. An example of this was presented by Livens and Loveland (1988). They classified soils in Cumbria (UK) in terms of their ability to immobilize caesium, based on clay mineral content and type, organic content, pH, ammonium content, and potassium status of the soils. These data were then combined with results of a survey of Chernobyl radiocaesium deposition in the region to identify areas in which high levels of radiocaesium in flora and fauna would persist. The area identified coincided closely with the area in which the movement and slaughter of sheep is currently restricted because of the high radiocaesium levels in their tissues. It is evident that there was sufficent knowledge before the Chernobyl accident to enable us to predict that semi-natural ecosystems would experience problems of high radiocaesium activities in grazing animals following deposition from a nuclear incident. However, the existing data were probably not adequate to predict the magnitude of the resulting problem or its longevity.

There has been an improvement in transfer data describing the uptake of radiocaesium into ruminants since the Chernobyl accident. It is now generally accepted that the transfer coefficient Ff for sheep is higher, at about 0.3 d kg^{-1} for ewes and 0.49 d kg^{-1} for lambs (Coughtrey 1989), than the previously accepted value of 0.12 d kg^{-1} given by Ng et al. (1982). Although there are further indications that comparatively high transfer occurs in other, generally small, ruminants, including the smaller species of deer (Zach et al. 1989; Keszthelyi et al. 1989) the data is limited and further studies are needed in controlled laboratory conditions. Given the lack of good transfer data for ruminants in semi-natural ecosystems prior to and since the Chernobyl accident it is difficult to compare the transfer of pre-Chernobyl radiocaesium with that from the Chernobyl accident in these ecosystems. However it seems that the availability of Chernobyl radiocaesium increased from soon after deposition, when some of it would have been present as a surface deposit, compared with when it was incorporated into plant tissue (Beresford et al. 1989; Ward et al. 1989; Hove & Solheim Hansen 1989). Once this had occurred there is no evidence to suggest that it was absorbed to any greater or lesser extent than pre-Chernobyl radiocaesium, mainly because data prior to 1986 is too sparse to allow an adequate comparison.

There are obvious difficulties in determining a parameter such as the transfer coefficient for animals in semi-natural ecosystems. Even for studies of domesticated animals a great deal of time, effort and money has to be devoted to overcome the difficulties listed above. For some wild ruminants such studies are impossible. Indeed it is questionable whether transfer coefficients are of much value in these situations. We suggest that for comparative studies of these sorts of ecosystems it may be more useful to determine aggregated transfer parameters where the activity concentration in an animals tissue (Bq kg^{-1} FW) is compared with that in the soil (either Bq m-2 or Bq kg^{-1} DW). This would overcome many of the uncertainties of estimating radionuclide intake. However, it must be recognized that these would be highly ecosystem-specific and that their applicability in predictive models may be limited.

In agricultural systems it has long been recognized that the time of deposition will have a profound influence on the contamination levels of foodstuffs (Simmonds 1985). This would also be true of semi-natural ecosystems. If deposition occurs in winter immediate doses may be reduced if domesticated ruminants have already been removed to winter grazing. The presence of snow may also influence the distribution of fallout causing amplified levels in areas in which it accumulates and decreased deposition in sites exposed to the wind (Whicker & Schultz 1982). In the summer semi-natural ecosystems may be used for dairy animals, particularly in alpine areas, hence potential doses from milk products would be enhanced. There is an obvious influence of the time of deposition with regard to the hunting season.

There are considerable financial implications for governments in trying to limit radiocaesium dose to populations via this route. For instance since the Chernobyl accident many millions of pounds have been spent in compensating farmers and on countermeasures. However, countermeasures for animals in semi-natural ecosystems have been much improved and could be implemented in any future incident.

There are secondary ecological implications which may result from either governmental or individual responses to the high contamination levels in animals. Browsing damage to trees in forest ecosystems may occur if less hunting takes place. One of the potential measures to reduce radiocaesium levels in vegetation is to fertilize more accessible areas, however, this may cause changes in the vegetation species structure of the ecosystem.

The Chernobyl accident has reinvigorated research into the radioecology of natural and semi-natural ecosystems (Desmet & Myttenaere 1988) and it is apparent that these areas will have problems for many years to come. We still need more information to quantify the importance of these environments, relative to agricultural system, to predict eventual transfers to man.

1.7 SUMMARY

In the advent of deposition from a nuclear incident ruminants in semi-natural ecosystems are likely to have comparatively high radiocaesium activity concentrations in their meat and milk, compared with ruminants from agricultural areas. Ecological characteristics in these environments which make them vunerable to this form of pollution include the presence of: (i) soils lacking the specific clay minerals which immobilize radiocaesium and which therefore allow its uptake into vegetation; (ii) vegetation species with high uptake rates and/or retention of radiocaesium; (iii) ruminant species with a high transfer of radiocaesium from gut to tissues and (iv) climatic factors such as high precipitation rates.

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2. AGRICULTURAL ECOSYSTEMS

2.1 INTRODUCTION

Discharges of radioactive elements into the environment result in the external and internal exposure of populations to ionising radiations. Among the various exposure pathways, the main route of entry of fission products and most other artificial radionuclides into man has been identified as uptake from the diet (Van Den Hoek and Kirchmann, 1968; Stara et al., 1971; Paretzke et al., 1986; Simmonds, 1986). Since agricultural products constitute the basic diet of most populations, the fate and behaviour of radionuclides in agricultural ecosystems are of primary importance when assessing the exposure risk of man from environmental releases of radioactivity.

Prior to the Chernobyl accident, numerous investigations on the transfer of radionuclides into agricultural (plant and animal) products had already been carried out. Controlled experiments conducted under laboratory and field conditions with radioisotopes artificially added to the system as ionic forms had produced a wide set of radioecological data. Beside these experimental data, information was also available from studies carried out on the nuclear weapons test sites or considering the behaviour of the worldwide dispersed radionuclides originating from the weapons test fall-out. Finally, a last set of information had been provided by studies on the radioactive releases from the nuclear fuel cycle under routine and accidental conditions (eg Windscale 1957, Kyschtym (Oural) 1957).

The Chernobyl accident on April 26th, 1986 released over 12 days about 2.7 10^{17} Bq, or 20% of the inventory of the reactor core, into the atmosphere, providing a new source term. The characteristics of this source term, especially regarding the speciation of the deposited radionuclides, could differ substantially from other source terms already investigated. As a consequence of this large-scale release, radioactivity measurements were performed in many countries throughout the Northern hemisphere. From the time of the accident onwards, numerous and varied samples of agricultural products were collected and analysed, both for food contamination control and scientific studies. These measurements revealed, in particular ecosystems (uplands), an unpredicted behaviour and transfer to animal products of radiocaesium. Moreover, a posteriori calculations carried out in the months after the accident pointed out large discrepancies between models predictions and field observations. On another hand, one of the conclusions reported by the CEC Article 31 Working Group (Radionuclide transfer factors for animal feedingstuffs and animal products) was that some transfers were poorly documented (transfer to milk other than cow's milk, transfer to growing animals, transfer to poultry). These findings and conclusions reactivated the interest of the scientific community for radioelements like iodine and caesium which were, before Chernobyl, generally considered as sufficiently documented.

The aim of this research, in the framework of the action 2 of the CEC post-Chernobyl programme, was to investigate the transfer of the main radionuclides deposited by the Chernobyl fall-out on pasture to animals and their products. It also aimed to compare the transfer data for the Chernobyl radionuclides with data obtained for other source terms, and to generate new data in less documented fields.

Three animal species of major importance for human nutrition have been considered : cattle, sheep and chicken. The results presented below were obtained by four laboratories, CEN/SCK in Belgium, ITE/NERC (in collaboration with MLURI) and NRPB in United Kingdom and ENEA in Italy. CEN/SCK and ENEA were financially supported for this work by the Radiation Protection Programme of the CEC, the other laboratories were supported by contracts with MAFF, DOE and CEGB (United kingdom).

2.2 MATERIAL AND METHODS

2.2.1 Retention of radionuclides in pasture ecosystems

Two meadows located in the vicinity of the Belgian Nuclear Research Centre (CEN/SCK) at Mol (North-East of Belgium) were selected to investigate the change with time of the activity associated with grass. The two pastures were continuously grazed by cattle. The meadow of the CEN/SCK experimental farm (about 3 ha), hereafter denoted as the "Mol" meadow, was grazed by one cow from April 30th to May 5th, by two cows from May 6th to May 26th and by three cows thereafter. The meadow located in Geel (about 3.5 ha), the "Geel" meadow, was grazed by four cattle from May 3rd to May 25th and by fourteen cattle thereafter. Grass (from a 1 m» area) was cut with shears to within 2 cm of the ground. Samples were taken twice daily until May 11th, then once daily until May 23th and weekly thereafter. The vegetation biomass was recorded and the radiocontamination levels were measured by gamma spectrometry using NaI(Tl) detectors. In situ gamma-ray spectrometry measurements were also performed on these two meadows using a portable HPGe detector mounted on a tripod in a down-looking position at a height of 1 m above the ground level (Deworm, 1987).

The same kind of investigation were conducted in the United Kingdom by Wilkins et al. (1988) on pasture from two farms using different grazing patterns for their cattle. The first one is located in Berkshire ("Berkshire" meadows). Due to the dry weather conditions prevailing in this region in the beginning of May contamination by the Chernobyl fall-out was limited to dry deposit ; iodine and caesium were nevertheless measurable in both pasture grass and milk. This farm operates a block grazing pattern; each morning the cattle are given access to an area of pasture that has not been grazed recently. During early summer a block of 6 ha is allocated to 100 cattle. The second farm is situated in Cumbria ("Cumbria" meadow). Due to wet deposition, the contamination level on grass in this region was at least an order of magnitude higher than in Berkshire . In this farm, the herd, of about 50 cattle, was allowed to graze relatively freely and continuously over about

10 ha of pasture. The grass sampling procedure was the same at both farms : pasture was cut from the area available for grazing that day, with shears to within 2 cm of the ground from an area of 1 m». The foliage density was recorded and the activity concentrations were determined by gamma spectrometry using GeLi or NaI detectors.

Retention of radiocaesium was also estimated in ungrazed pastures located in the north-eastern part of Italy (Friuli-Venezia Giulia region). Grass was sampled weekly from 8 plots (two plots each week) and measured for radiocaesium content using HPGe detectors (Belli et al., 1989).

2.2.2 Transfer to cow's milk under field conditions

As the grass production in the beginning of May at the experimental farm of CEN/SCK was insufficient to allow cows to graze, the cattle was still stalled. However, one cow (cow n° 56.0) was turned out to grass during day time and brought in for the night from April 30th. She received 4 kg/d of concentrates, 3 kg/d of beet pulp and 3 kg/d of hay in order to sustain lactation. After May 25th, she remained on the pasture night and day receiving a supplementary feed of solely 4 kg/d of concentrates. After they had calved, two other cows were put on the pasture, the first (cow n° 50.1) from May 6th and the second (cow n° 56.2) from May 26th; they both stayed outside night and day and received a daily complement of 2 kg/d of concentrates. At the Geel farm, one cow (cow "Geel") was turned out to grass from May 3rd and fed exclusively on grass, receiving no dietary supplements until May 25th ; afterwards, she received 0.5 kg/d of concentrates and 5 kg/d of beet. The amount of grass eaten by cows has been estimated on the base of their body weight, milk production and quantity and quality of complementary feed they received. Milk was sampled twice daily until May 11th, then daily until May 23th and weekly afterwards. The samples were measured for their radioiodine and radiocaesium content by gamma spectrometry using NaI(Tl) detectors.

At the two English farms studied by Wilkins and coworkers (Wilkins et al., 1988), the cattle were already grazing outside during early May and were feeding on open pasture throughout the period of investigation, without dietary supplements. From both farms milk samples were taken daily from the bulk tank. Radioactive contamination levels were determined by gamma spectrometry using GeLi or NaI detectors. Direct measurements were possible when the activity concentration was higher than 5 Bq/l. This applies to all results presented here for the "Cumbria" farm. Otherwise a large sample was taken and I-131 abstracted onto an anion exchange resin (Wilkins and Steward, 1982) ; similarly caesium isotopes were concentrated by complexation on ammonium molybdophosphate (Green and Major, 1983). These methods are in regular operational use and lend themselves to this task because they are simple, rapid and invariably have a chemical yield of almost 100 %.

The radiocaesium transfer to milk was investigated in 13 Italian dairy farms (Friuli-Venezia Giulia), where livestock was fed in cowshed during all the year (Belli et al., 1989). These farms were classified into three groups depending on the main component of the cow's diet : alfalfa, grass or maize silage. The daily intake was calculated on the basis of the weight of the different components of the diet and their respective caesium content. Milk and samples of the diet components were measured for their radiocaesium content using HPGe detectors.

2.2.3 Transfer to sheep's milk under field conditions

When the radioactive cloud from Chernobyl arrived above Belgium on May 2nd 1986, the sheep flock (Suffolk strain) of the CEN/SCK experimental farm was already grazing on pasture and were not receiving any dietary supplements. The amount of grass eaten by sheep has been estimated on the base of their body weight and milk production. Weekly sheep were milked by hand, without oxytocin injection, and samples were taken for radioactivity analysis by gamma spectrometry using NaI(Tl) detectors.

In the months following the Chernobyl accident, numerous milk samples were collected by ITE from ewes grazing different types of pastures or fed with vegetation harvested from different ecosystems before or after the deposition of the Chernobyl radionuclides. Ewes were milked by hand after intravenous injection of oxytocin. The samples were measured by gamma spectrometry using GeLi detectors. The experimental design and procedure are detailed in reports and publications produced by the ITE, referred in the discussion.

2.2.4 Transfer to sheep's organs under field conditions

Sheep from the CEN/SCK experimental farm were sacrificed at various times and several organs : muscle (psoas), liver and kidney, were sampled and analysed for their radiocaesium content by gamma spectrometry (NaI detectors).

The radioactivity levels in upland sheep in the UK have been measured by ITE. Experiments were designed to understand the changes in contamination levels in sheep meat when they move from fells to improved pasture near the upland farmstead and returned to the fells, or when they are sold for fattening on lowland pasture. Tissues from slaughtered animals were analysed by gamma spectrometry using high resolution GeLi detectors, but also in vivo measurements were made using a portable hand held NaI(Tl) detector (4.4 x 5.1 cm), linked to a single channel analyser. For these in vivo measurements, the detector was placed on the rump of the animal and the mean of 3 times 10 second counts was recorded. The detector has been calibrated against muscles taken from slaughtered sheep at the end of the study. Separate calibration curves were obtained for ewes and lambs. The experimental designs and procedures are detailed in reports and publications produced by ITE and will be referred to in the discussion.

2.2.5 Transfer to cow's milk under experimental conditions

To obtain a more accurate estimation of the transfer coefficient of radiocaesium from grass to cow's milk than that obtained under field conditions, and to compare the bioavailability of the Chernobyl Cs with the one of a well-known Cs chemical form (CsCl), an experiment was performed under controlled conditions at CEN/SCK

(Vankerkom et al., 1988). Two cows received relatively uncontaminated hay for three weeks, harvested in 1985, in order to reduce as much as possible their radiocaesium body burden ; they then received "Chernobyl" hay (contaminated by the Chernobyl fall-out to a level of 197 ± 48 Bq Cs-134/kg dry weight and 464 ± 117 Bq Cs-137/kg dry weight (activities measured on September 7th, 1987)) harvested in mid-June 1986 for 30 days. Afterwards they received relatively uncontaminated hay harvested in 1985, for a 60 day decontamination period. Two other cows received hay, during the experimental period, collected in 1987, that was much less contaminated (17.5 ± 6.0 Bq Cs-137/kg DW) than the hay harvested in 1986. For 30 days, these two cows received 5 kBq of Cs-134 daily as CsCl, corresponding approximately to the daily intake expected for the cows fed with "Chernobyl" hay and were thereafter allowed to lose Cs-134 activity for 60 days. The quantity of hay ingested by the cows was recorded. Milk samples from the four cows were collected daily during the contamination period and for the first 26 days of the decontamination period, and at least every two days thereafter. The samples were measured for their radiocaesium content by gamma spectrometry (NaI).

2.2.6 Transfer to pregnant sheep and progeny under experimental conditions

Transfer data for sheep are limited compared to the extensive set of data available for cows. It seemed then worth documenting this topic, especially laying emphasis on the transfer to developing animals during foetal life and via the lactation. About two months after mating, pregnant ewes of the Suffolk strain were contaminated daily by oral administration of 74 kBq of Cs-134 as CsCl given in a small amount of concentrate (Vandecasteele et al., 1989). The daily ration consisted in approximately 200 g of concentrates, 500 g of beet pulp and 700 g of oats; hay and water were provided ad libitum. The animals were housed in metabolism cages during 76 days and thereafter in individual boxes on the floor for lambing. After birth the lambs stayed for a three days period with their mother to allow absorption of collostrum. The lambs were than distributed into two groups, lambs from the first group stayed with their dams and consequently received contaminated milk while lambs from the second group were fed non contaminated artificial milk. The lambs were sacrificed at selected intervals over a period of 60 days after birth. After weaning, the ewes were again housed in the metabolism cages and contamination was stopped. They were sacrificed at various intervals from this time over a 64 days period of decontamination. Urine and faeces were collected from ewes during the periods they stayed in the metabolism cages. Ewes' blood samples were taken at various times during contamination and decontamination periods. Milk samples were obtained by milking by hand without hormone injection at various times since day 3 after lambing. Selected organs of the sacrificed animals were sampled. A total of 6 ewes and 10 lambs were used in this investigation. The mean weight of the ewes at the start of the contamination was 73.7 ± 14.9 kg. The radiocaesium activity was measured by gamma spectrometry using GeLi or NaI(Tl) detectors. Compartmental analyses were performed by non-linear least square methods.

2.2.7 Influence of the incorporation of Cs into plants on its absorption by sheep

Availability of radionuclides for gastro-intestinal absorption depends on their own chemical form as well as on their accessibility in the matrix within which they are included. Radionuclides incorporated into plant material need to be solubilised and released from their cellulose matrix by the digestive processes prior to absorption. In order to estimate the relative availability of radiocaesium incorporated into plants to that of ionic forms, maize plants were grown in a greenhouse and injected at flowering with an aqueous solution of CsCl into the stem. The plants were harvested at maturity, dried and powdered. A small amount (90 g/d) of this highly contaminated material was then fed to four young male sheep while four others received CsCl. During the 4 weeks contamination period, blood samples were collected at regular interval for radioactivity measurements.

2.2.8 Transfer of radiocaesium to laying hens

Thirty-five laying hens (Sexalline) aged 155 days were put on battery. They were fed standard hen food ad libitum. During 56 d, the hens were given an aqueous solution of Cs-134 (5.57 kBq/d) as CsCl, injected with a syringe into their throat. Fifteen hens were sacrificed during the contamination period while 20 were allowed to decontaminate and were sacrificed at regular intervals, up to 77 d after the end of dosing. Eggs and faeces of 6 hens were sampled daily for radioactivity measurements. Eggs were boiled and separated into white, yolk and shell.

2.2.9 Transfer of radiocaesium to growing chickens

Sixty chicks (Hubbard) aged one day were put on battery and were randomly divided into three groups depending on the contamination scenario. The first group received contaminated feed until they reached the age at which they are normally slaughtered for consumption (8 weeks). Animals in the second group were fed contaminated flour during the 4 first weeks and uncontaminated feed thereafter. Chickens of the third group were given contaminated flour only during the 4 last weeks before slaughter. The chicken feed consisted in a standard flour, artificially contaminated when needed, with ^{134}Cs (61.6 Bq/g D.W.) as CsCl. Faeces were regularly collected and animals from each group were sacrificed at various times. Faeces and selected organs were analysed for their radiocontamination levels.

2.3 RESULTS AND DISCUSSION

2.3.1 Initial retention in pasture ecosystems

The initial retention of the deposited radionuclides by vegetation depends on several parameters peculiar to the physico-chemical characteristics of aerosols and particles, climatic conditions and plant morphology. It also varies with the herbage density at the time of the deposit (Kirchmann and Boulanger, 1965a ; Kirchmann et al., 1966). This clearly appears when comparing (Table 5) the radioactive content in grass of two meadows located in the vicinity of the CEN/SCK. At the two locations, the total deposit has been measured by in situ gamma spectrometry and measurement of grass and surface soil samples. The results obtained with both techniques were comparable at the two sites and in agreement with the measurements of the total fall-out performed using rain-water collectors (Deworm, 1987).

Station	Total fall-out		Herbage density (g/m^2)	radioactivity			
				per area unit (Bq/m^2)		per weight unit (Bq/g F.W.)	
	^{131}I	^{137}Cs		^{131}I	^{137}Cs	^{131}I	^{137}Cs
MOL 04/05	9364	1500	233	740	220	3176	944
GEEL 04/05			973	2100	360	2158	370

Table 5: Initial retention measured in pasture grass at two sampling stations in the vicinity of the CEN/SCK. The values reported for the total fall-out are those obtained using rain-water collectors and may be assumed to be representative of the deposit on the two meadows ; these values are corrected for decay at the time of grass sampling.

At the beginning of May, the meadow located in Geel had 4 times as much vegetation cover as the "Mol" meadow. The interception efficiency by the vegetation calculated per unit area was thus higher (by a factor of 1.5 for radiocaesium, and by a factor of 3 for radioiodine) in the "Geel" meadow than in "Mol" meadow. However, due to dilution by a much higher vegetation mass, the specific activity (in Bq/g) of the grass from the "Geel" meadow was approximately half that of the "Mol" meadow. This is an important consideration since it is obvious that grazing animals ingest kilogrammes of grass rather than square meters of pasture.

2.3.2 Evolution of the radioactive contamination on grass

In the weeks following the deposit, the contamination of grass is reduced due to physical decay, weathering processes (removal of particles by wind, leaching by rain, fog, dew and mist or irrigation water), senescence processes (shedding of cuticular wax, die-back of old leaves). Decrease of the radiocontamination of vegetation also results from dilution by plant growth and removal contaminated parts by harvesting and grazing, depending on farming practices and grazing patterns.

The changes in Cs-137 and I-131 content of the grass with time in the two meadows investigated near the CEN/SCK are given in figure 1. The results obtained for Cs at both sampling sites were fitted as the sum of two exponential functions. The retention half-time associated with the first one was shorter for the "Mol" meadow (2.6 d) than for the "Geel" meadow (6.5 d). This may be partly explained by the higher relative growth rate of the grass in the "Mol" meadow compared to that in the "Geel" meadow which had started to grow earlier in the season. The retention half-times for the second compartment in both meadows (about 60 d) were not statistically different, but the capacity of this second compartment was more than 10 times higher in the "Mol" meadow compared with the "Geel" meadow. This difference may be attributed to either a lower grazing pressure and/or to a higher availability of the Chernobyl radiocaesium for soil-plant transfer in the "Mol" meadows. For the short-lived I-131, only one compartment could be discerned. The retention half-time estimated expressed on the grass weight basis was about 2.6 d at the two sampling sites.

At the two English farms investigated by Wilkins et al. (1988), one in Cumbria and the other in Berkshire, the cattle were feeding exclusively on open pasture throughout the period of investigation (May and June, 1986), and were not receiving any dietary supplements. At the "Berkshire" farm (block grazing pattern), pasture samples were taken from the block grazed that day at the time the grazing began so that half-times estimated from these data essentially represent ungrazed conditions ; those derived from data from the "Cumbria" farm represent continuous grazing,

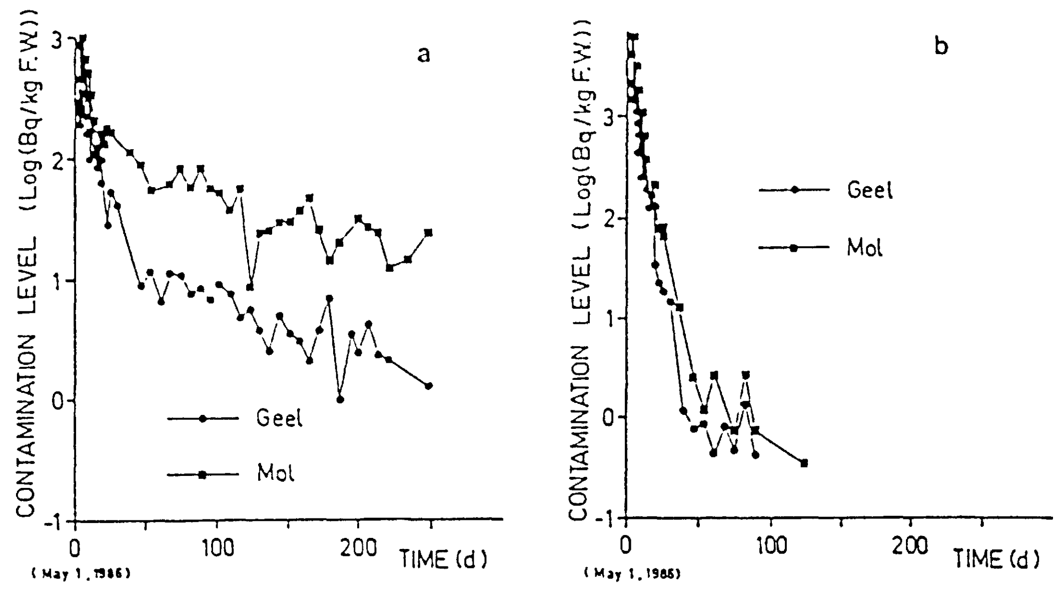

Figure 1: Changes in the Cs-137 (a) and I-131 (b) contamination level (Bq/kg fresh Weight) measured in the grass of the two Belgian meadows.

and should therefore be less than the values from the "Berkshire" farm (fig. 2). The estimated retention half-times for Cs-137 were 7 d at the "Cumbria" farm and 23 d at the "Berkshire" farm. The corresponding values for I-131 were 4.6 and 5.2 d. This values are estimated on an area basis, which do not take into account the dilution by growth. However, the density of the vegetation cover at the "Cumbria" farm remained rather constant (about 0.3 kg F.W./m^2) throughout the investigation period and that of the "Berkshire" farm, after an initial increase by almost one order of magnitude within a few days in the beginning of May, stayed in the range 1-2 kg F.W./m^2.

The retention half-time of radiocaesium in grass of pastures located in the Friuli-Venezia Giulia region was estimated to 7 d (Belli et al., 1989).

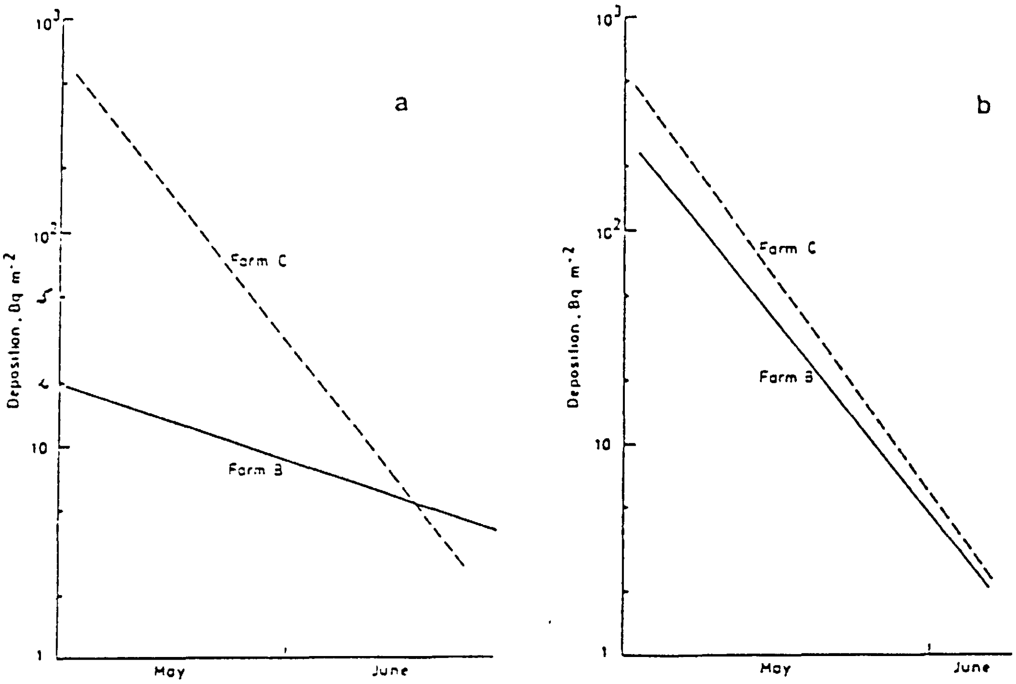

Figure 2: Changes in the Cs-137 (a) and I-131 (b) contamination level (Bq/kg fresh weight) measured in the grass of the two English meadows.

2.3.3 Transfer of radionuclides to cow's milk under field conditions

The changes in the Cs-137 contamination in milk from cows grazing on the "Mol" and "Geel" meadows measured during the months following the deposit until the cattle was stalled for winter is presented in figure 3.

The differences in the caesium excretion pattern observed between the cows at the CEN/SCK experimental farm at the beginning of the observation period can be explained by the fact that the cattle were turned out to grass at different times after the deposit. It must be also noted that, until May 25th, one of the cows (cow n° 56.0) grazed during the day period only and was stalled for the night. After this period the radiocaesium excretion in milk of the three cows behaved very similarly. The radiocaesium transfer into milk for the cow grazing on the "Geel" meadow declined more rapidly and reached lower levels than the cows grazing on the

"Mol" meadow. This discrepancy reflected the characteristics of caesium retention by grass at the two sites.

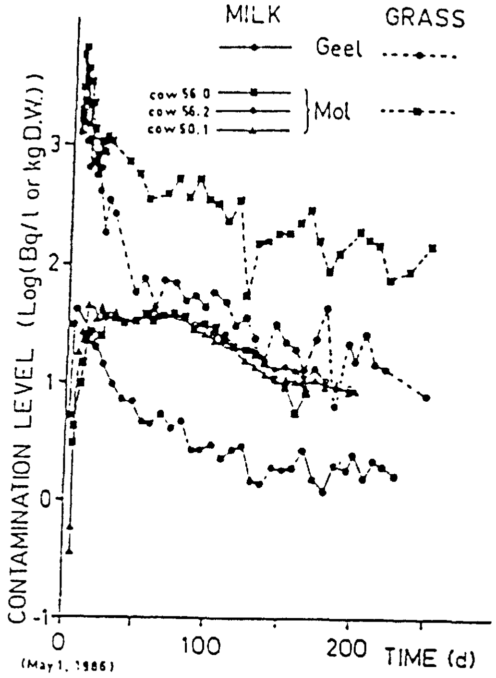

Figure 3: Changes in the Cs-137 contamination in pasture grass and milk from the cows grazing on these pastures at two Belgian farms.

One single compartment with a half-time of about 80 d (ranging from 68 to 89 d) was identified for the loss of radiocaesium in milk from the "Mol" cows. For the milk from the "Geel" cow , statistical analysis reveals two compartments characterised by half-times of about 11 and 175 d, respectively. In both cases the half-times estimated for milk were longer than the corresponding ones estimated in the grass consumed by the cows. At the time a pseudo-equilibrium was achieved, the activity ratio between milk and grass collected on the same day amounted to about 0.6 kg f.w./l at both locations. Assuming a daily intake of 50 kg grass, the transfer coefficient between grass and milk estimated as the quotient of the activity in milk by the total activity intake for that same day was 0.012 d/l. This value is in the range of the previously reported data.

The excretion patterns for the Chernobyl radioiodine obtained at the farms are presented in figure 4. The effective half-times for I-131 in milk do not differ statistically between the two farms and have been estimated to be about 4 d. The longer half-time for milk compared to grass can be explained by the incorporation of iodine into the animal tissues (Loutit, 1961).

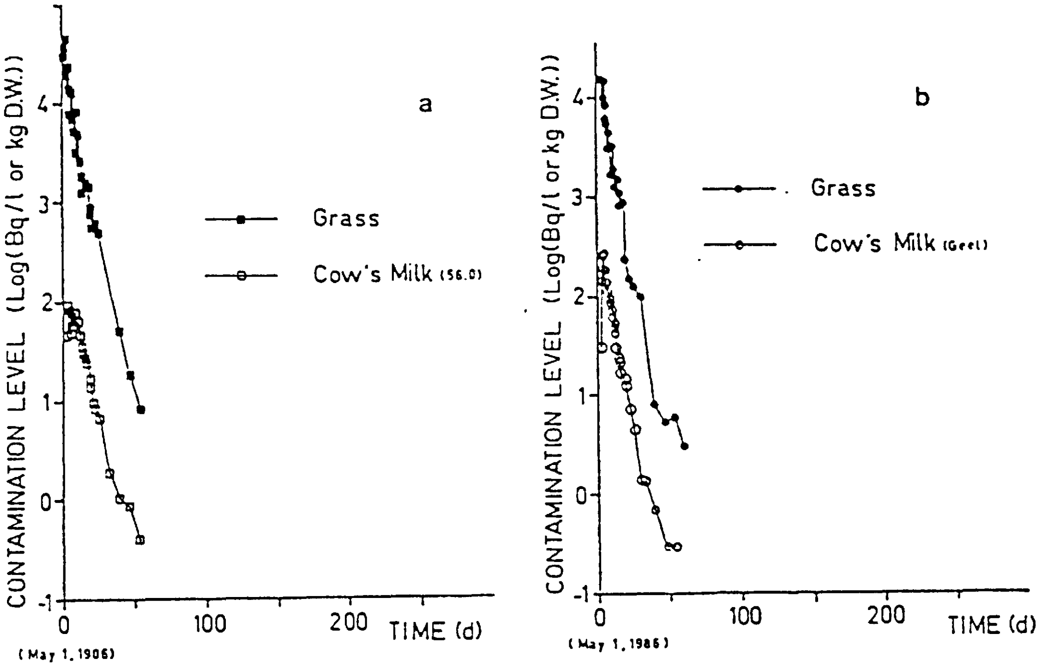

Figure 4: Changes in the I-131 contamination in grass (Bq/kg dry weight) and cow's milk (Bq/l) at the "Mol" farm (a) and "Geel" farm (b).

The activity ratio between milk and grass reached a pseudo-equilibrium value of 0.11 kg F.W./l at the CEN/SCK experimental farm and a value of 0.15 kg F.W./l at the Geel farm where the cow did not receive any complementary food in the first weeks following the deposit. Assuming a daily grass intake of 50 and 80 kg F.W. for the "CEN/SCK" and "Geel" cows, respectively, the transfer coefficient are estimated to 0.002 d/l at the two locations. This value corresponds to the lower limit of the range reported by Kirchmann and Boulanger (1965b) for the transfer to milk in cows grazing on pastures artificially contaminated by spraying with elementary iodine and is less than the consensus figure of 0.01 d/l given in a compendium of earlier data (Hoffman, 1977). Such a low figure may be attributed to the peculiar speciation of iodine from the Chernobyl fall-out leading to a lower availability of the Chernobyl iodine compared to elemental or ionic forms.

The changes during the two first months after the deposit in the activity intake by cattle and the excretion of radioiodine and radiocaesium into milk from cows at two English farms has been considered by Wilkins et al. (1988). At the "Berkshire" farm, the period immediately following deposition was one of a high rate of growth of pasture leading to an important dilution effect on the radiocontamination levels in grass and subsequently on the activity intake (Fig.5).

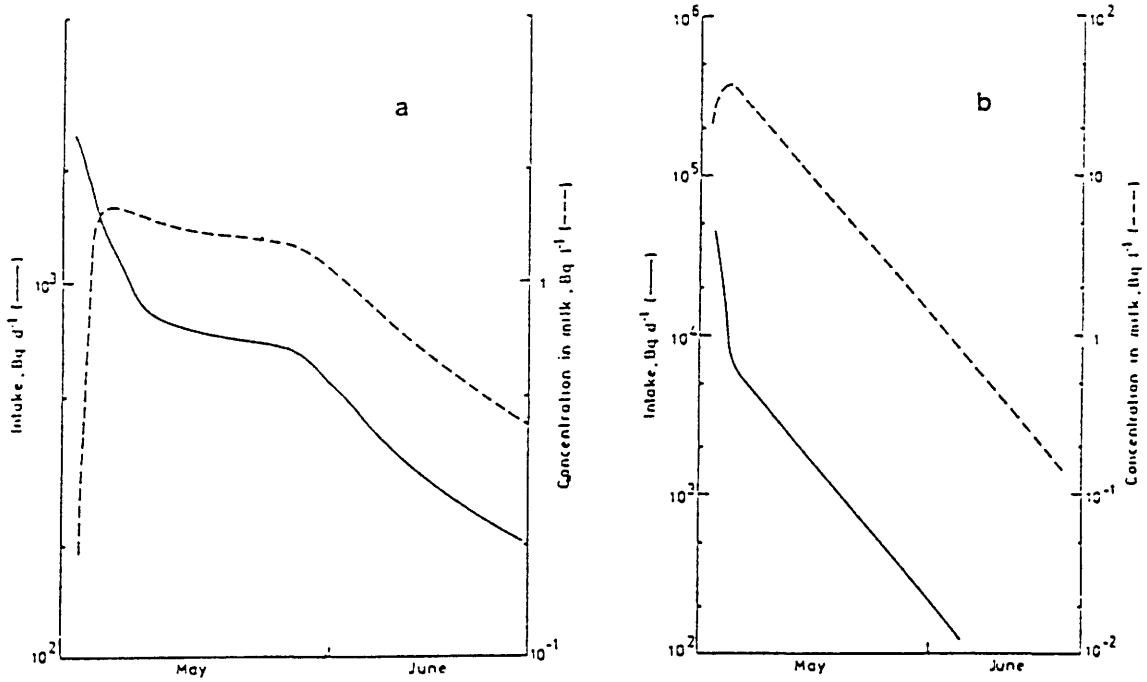

Figure 5: Intakes of Cs-137 (a) and I-131 (b) in Bq/d and associated milk concentrations in Bq/l at the "Berkshire" farm.

The growth then slowed down and the block grazing system became the controlling factor : intakes of caesium remained relatively steady for about 15 days, after which the cattle returned to previously grazed blocks that had since grown fresh grass. Intakes of I-131 were more influenced by the short half-life of this radionuclide than by the block grazing system. The ecological half-time estimated for iodine in milk was 5.6 d, slightly higher than the value obtained for the pastures in the Mol region.

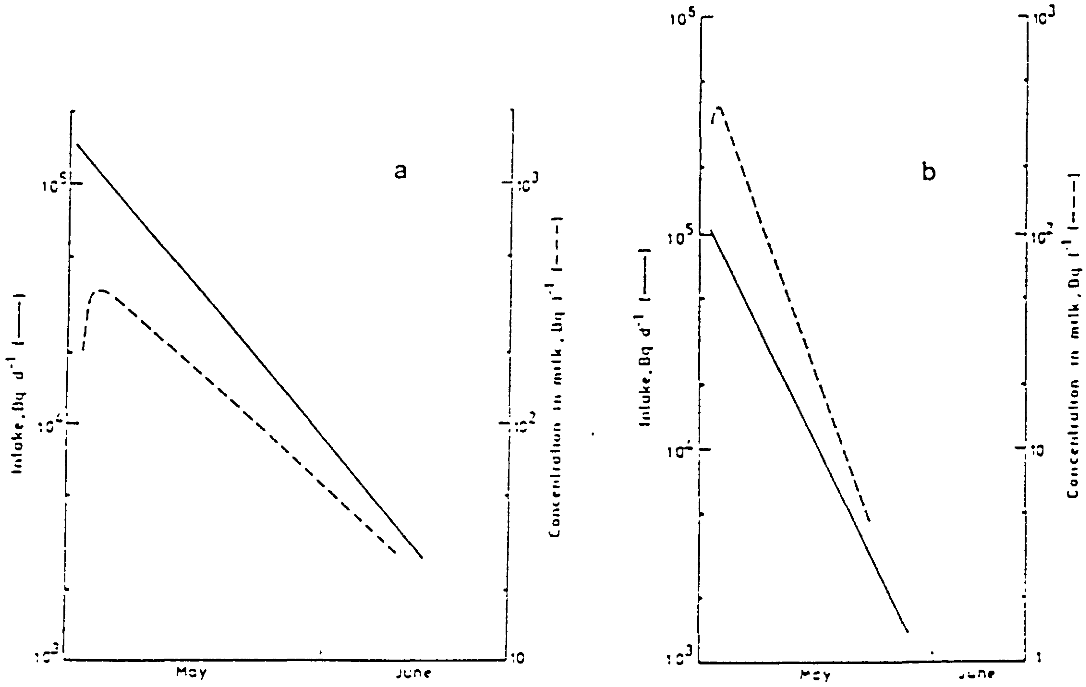

Figure 6: Intakes of Cs-137 (a) and I-131 (b) in Bq/d and associated milk concentrations in Bq/l at the "Cumbria" farm.

At the "Cumbria" farm, the vegetation densities remained relatively constant (0.3 kg f.w./m^2) throughout the 40-day measurement period, a reflection of the continuous grazing of the area and the latter onset of growth in northern England. Daily intakes of Cs-137 declined steadily with a half-time of 7.4 d (Fig. 6a) in contrast to observations at the "Berkshire" farm (Fig. 5a). Concentrations in milk followed changes in intake closely. Although only limited results for I-131 have been obtained before concentrations fell below the detection limit, the ecological half-time in milk has been estimated at 3 d (Fig. 6b), lower than the value obtained at the "Berkshire" farm but close to those determined for the cows studied in Belgium, which had a similar grazing pattern.

The ratios of the activity concentrations in milk and the previous days' intake of activity have been estimated. The quotient for I-

131 at the "Berkshire" farm rapidly approached an equilibrium value of around 0.002 d/l ; the figure extrapolated from the available data at the "Cumbria" farm was 0.003 d/l. These values are similar to the ones calculated for the "Geel" and "Mol" cows, and are comparable to that estimated for continuous deposition of I-129 close to the Sellafield reprocessing plant (Wilkins and Stewart, 1987). The quotients for radiocaesium had not attained an equilibrium value by the time concentrations in grass reached detection limits. By early June, the quotient at the "Cumbria" farm reached about 0.009 d/l. The quotient at the "Cumbria" farm is somewhat lower than the value obtained in the Belgian pastures near Mol but was still increasing towards an equilibrium value. These values obtained in England (Cumbria) and in Belgium can be compared with the transfer coefficient, of around 0.007 d/l, estimated for continuous deposition around Sellafield (Sumerling et al., 1984). Compared to the transfer coefficient estimated at the "Cumbria" farm for wet deposition, the value calculated at the "Berkshire" farm, where the pasture contamination was due to dry deposit only, is about 5 times lower implying a reduced availability of the dry-deposited caesium.

The transfer factors to milk estimated for radiocaesium in farms located in the Friuli-Venezia Giulia region (Italy) varied depending on the cow's diet (Belli et al.,1989). For cows fed the "alfalfa" and the "grass" diets the transfer factor amounted to 0.0026 and 0.0033 d/l, respectively. It was higher for cows fed the "maize silage" diet, 0.013 d/l. Although those differences were not statistically significant, they point out some influence of the regime on the absorption of radiocaesium by ruminants.

2.3.4 Transfer of radionuclides to sheep's milk under field conditions

Although the sheep from the flock of the CEN/SCK experimental farm are of the Suffolk strain and thus are not milk sheep, samples of milk were obtained from ewes with lambs. These sheep grazed on a pasture adjacent to the "Mol" meadow grazed by the "Mol" cows. The changes in the I-131 and Cs-137 contamination level in sheep's milk are presented in figure 7.

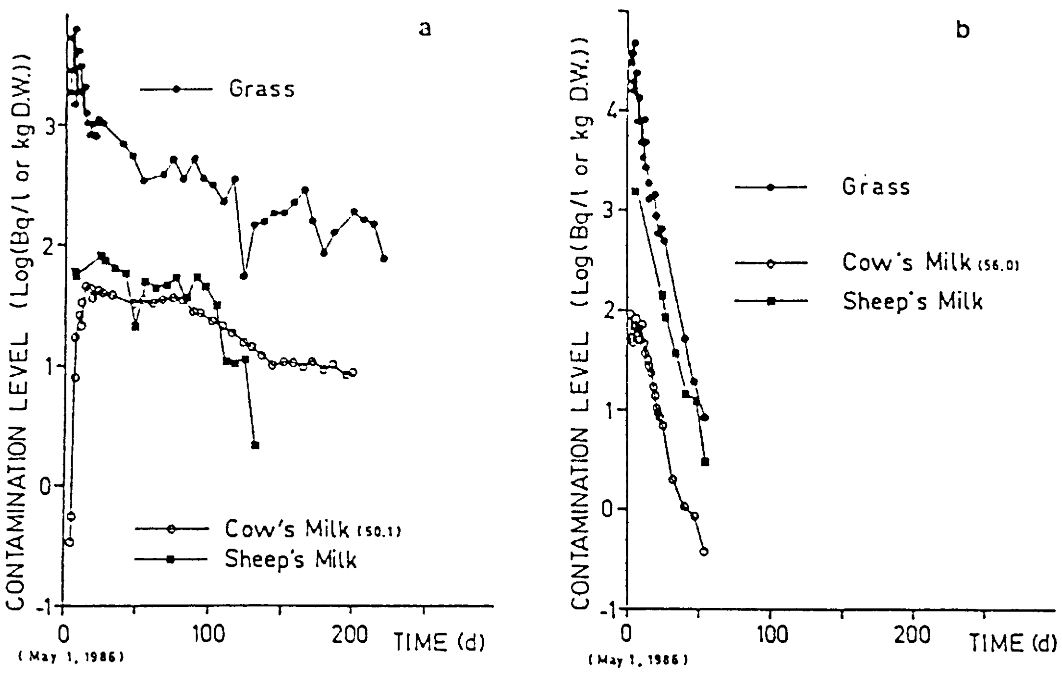

Figure 7: Changes in the Cs-137 (a) and I-131 (b) concentration in sheep's milk (Bq/l) at the CEN/SCK experimental farm. Contamination levels in grass (Bq/kg dry weight) and cow's milk (Bq/l) are reported for comparison.

The contamination levels in grass and cow's milk are also reported to facilitate comparison. The concentration of iodine in sheep's milk is higher by a factor 14 compared with that in cow's milk. Due to the limited data available, an accurate estimation of the transfer coefficient for iodine is not possible. However, based on an estimated daily grass consumption of 10 kg fresh weight (corresponding to 1.6 kg dry weight), the ratios between the radioactivity level in milk and the daily intake of activity were calculated and range, when an equilibrium seems to be reached, between 0.12 and 0.2 d/l. The effective half-time for I-131 in sheep's milk is 5.2 d, which is somewhat longer than for cows. The half-time calculated for Cs-137 was 87 d in sheep's milk. This estimation does not take into account the last data showing an important decrease of the contamination level, probably due to a reduction in milk production and modifications of lactation metabolism at weaning of lambs. The difference in milk concentration between cow and sheep is lower for caesium than for iodine and amounts to less than a factor 2. The activity ratio calculated between milk and grass amounts to about 0.7 kg F.W./l. The estimated transfer coefficients, based on the same assumption for grass intake as mentioned above, gives an equilibrium value 0.07 d/l. In the case of Cs-137 the transfer from grass to milk is higher by a factor 6 in sheep than in cows. The transfer coefficient estimated for sheep under field conditions is in good agreement with the value of 0.058 reported for sheep given grass contaminated by direct deposition of radiocaesium from Chernobyl under controlled condition in Greece (Assimakopoulos et al., 1987).

In the United Kingdom, the highest deposits from Chernobyl were observed in the uplands where large areas are mainly devoted to hill sheep farming. In these regions, the British Government restricted the movement and slaughter of sheep. These restrictions had to be maintained in several regions for longer than initially thought since the contamination level of the vegetation in some uplands areas remained persistently higher than anticipated. The reason for this persistence is that these areas are poor ecosystems of which the soil is characterised by low pH and organic content ; in these conditions, Cs remains more available for plants (Howard et al., 1988 ; Beresford et al., 1987a). In improved pastures close to hill farms, the activity of grass was less than on fells. Several mechanisms could contribute to the difference : higher rainfall and hence greater deposition on the fells ; the leaf surfaces of the fell vegetation may have retained more caesium because of their texture ; fresh growth that is lower on the fells than on improved pastures may not have diluted to a large extent the deposited activity ; the already mentioned soil characteristics on the fells are more favourable to the soil-plant transfer than that from the improved pastures (Beresford et al., 1987b). The transfer coefficients for I-131 and Cs-137 present in the Chernobyl fall-out have been estimated during an indoor experiment conducted in Edinburgh during May-July 1986. During a 32 day period three ewes received Chernobyl contaminated herbage (perennial rye-grass) harvested from a pasture near Edinburgh, Scotland on May 6th, 1986, approximately 3 days after the peak deposition. The Fm values obtained were 0.06 and 0.29 d/l for Cs-

137 and I-131, respectively. The estimation for Cs is close to the values reported in Belgium (CEN/SCK, Mol) and Greece (Assimakopoulos et al., 1987). For comparison, the grass-to-milk transfer coefficient for Cs-137 in sheep fed saltmarsh vegetation harvested before the Chernobyl accident and contaminated by marine discharges from Sellafield was of 0.006 d/l. This very low value is explained by the association of the caesium activity with silt adherent to the vegetation, a form that is poorly available for gut transfer (Howard, 1989). The transfer coefficient for Cs to milk of sheep grazing on fells in July-August 1987, was 0.12 d/l, higher than the values obtained for ewes fed rye-grass harvested 3 days after the peak deposit. This difference may be due to a greater availability of Cs-137 when biologically incorporated into the plant material rather than when present as a recent deposit (associated with insoluble particles?) on the leaf surface (Beresford and Howard, 1988).

2.3.5 Transfer of radionuclides to sheep's organ under field conditions

The changes with time in the radiocaesium content in muscles (psoas), liver and kidneys of sheep from the CEN/SCK flock is presented in figure 8. A distinction has been made between animals younger than 8 months (lambs) at the time of sacrifice and older adult animals whose growth was complete. During the first months after the accident, the contamination measured in lambs is nearly twice as high as that of ewes. This difference is however not observed during the second year probably due to the lower contamination levels in grass consumed by the animals,and to the subsequent lower contamination of organs and higher variability in counting.

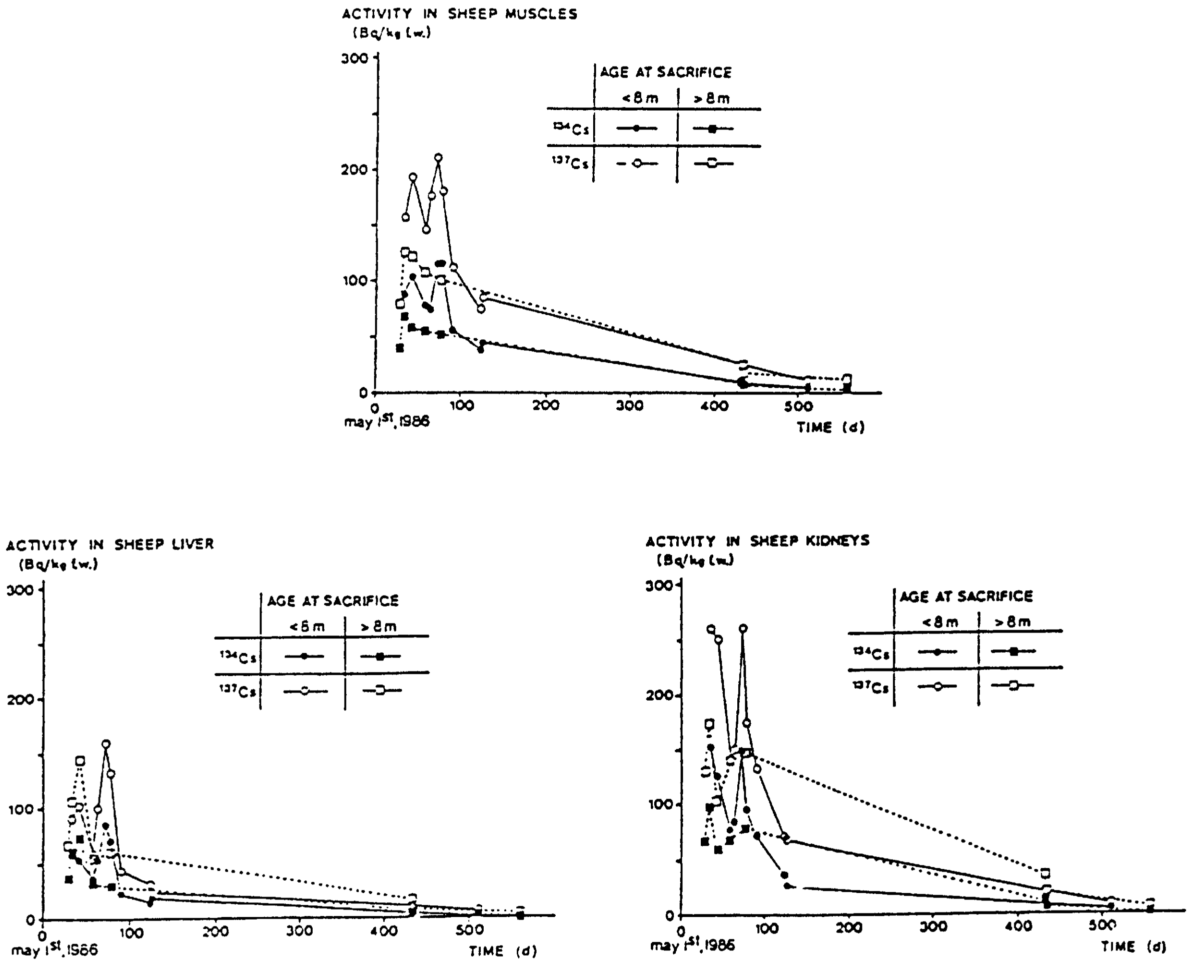

Figure 8: Changes in the Cs-137 contamination level (Bq/kg fresh weight) in sheep's muscle, liver and kidneys.

The half-times were estimated by a non-linear weighted least squares method. The statistical analysis revealed the existence of at least two compartments for the three organs considered. The half-times estimated for the two compartments range respectively from 8 to 12 d and from 143 to 240 in lambs ; in ewes, the estimated values range respectively from 15 to 24 d and from 134 to 216 d. The transfer coefficients (in d/kg) from grass to meat has been roughly estimated assuming a daily intake of 1.6 kg d.w. herbage for ewes and 0.6 kg d.w. herbage for lambs : they are highest for kidneys and being 0.73 ± 0.39 in lambs and 0.19 ± 0.08 in ewes ; they are lower for liver with values of 0.38 ± 0.22 in lambs and 0.12 ± 0.08 in ewes ; for muscles the transfer coefficients are 0.63 ± 0.24 in lambs and 0.14 ± 0.06 in ewes.

Contamination levels were measured by ITE/NERC for organs of sheep grazing fells and improved pastures around the farmstead of the uplands. The ewes graze for much of the year on the open fell, but are moved to improved pastures around the farms at various times of the year, for lambing, shearing, weaning of lambs, mating, worming and compulsory dipping.

The contamination levels measured in the muscles of these sheep varied with their movements from highly contaminated fells to less contaminated improved pastures (Fig. 9) : a decrease was observed when the flock was removed to improved pastures but the Cs body burden of the sheep increased again when the flock returned to the fells. An important decrease of the radiocaesium concentration in muscles was also observed for store lambs (lambs to be sold in early autumn for fattening on lowland pasture where the radiocaesium contamination level was much less -100 to 200 times less- than in upland areas). These lambs lost caesium activity rapidly, it took 10 days for them to eliminate half of their body burden of caesium (Howard et al., 1987). This half-time is comparable to that measured for the first compartment in the organs of young sheep from the CEN/SCK flock.

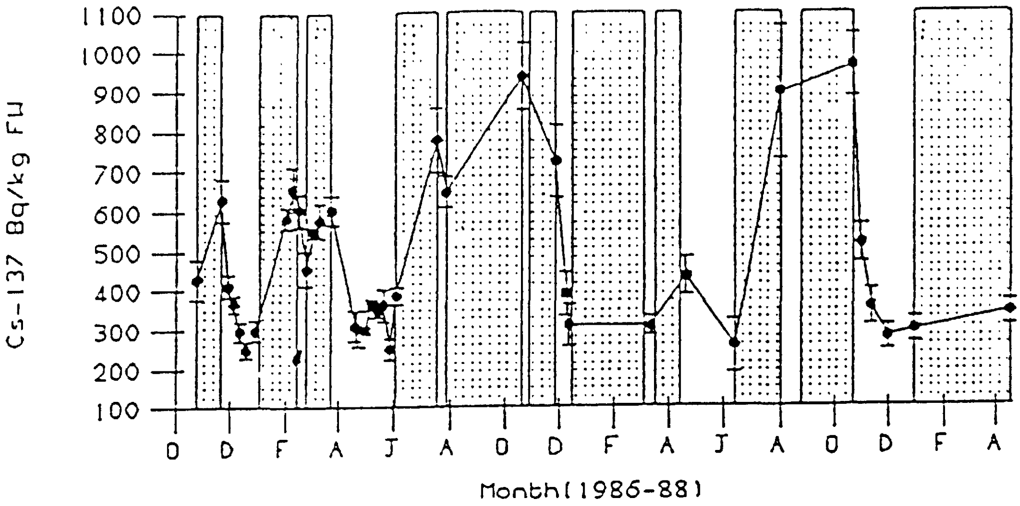

Figure 9: Changes in the Cs-137 activity (Bq/kg fresh weight) of muscles from ewes and lambs as they are moved between improved pastures and fells. (Shading = on fell).

The transfer coefficients calculated for the Chernobyl radiocaesium from grass to the sheep's organs (muscles, liver and kidney) for ewes and lambs fed rye-grass, harvested from a pasture near Edinburgh 3 days after the peak deposition of the Chernobyl fall-out, during an indoor experiment conducted in Edinbourgh in May-June 1986, or grazing on open fells are presented in table 6. Values obtained for sheep grazing on saltmarsh or pastures contaminated by releases from the Sellafield reprocessing plant (Howard and Lindley, 1985 ; Howard and Beresford, 1987) are also presented for comparison. Transfer factors for ewes and lambs grazing on open fells in 1987 are considerably higher than those obtained for ewes and lambs (of similar age) fed rye-grass harvested soon after the maximum deposit. This difference may be due, as already mentioned for milk, to a greater availability of caesium when bioincorporated into the plant material than when present as a recent deposit on the leaf surface. It is also obvious that the values calculated for the radiocaesium originating from Chernobyl are higher than those estimated for radiocaesium released by Sellafield.

TREATMENT	AGE	MUSCLE	KIDNEY	LIVER	REF.
Chernobyl					(48)
Rye-grass	ewes	0.12 ± 0.01	0.10 ± 0.01	0.07 ± 0.01	
indoor					
1986	lambs	0.50 ± 0.08	0.57 ± 0.06	0.27 ± 0.02	
Chernobyl					(10)
Open fells	ewes	0.33 ± 0.03	0.22 ± 0.02	0.14 ± 0.02	
field stud.					
1987	lambs	1.61 ± 0.21	1.37 ± 0.15	0.80 ± 0.09	
Sellafield					(41)
pasture	ewes	0.05 ± 0.02	0.15 ± 0.05	0.03 ± 0.01	
field stud.					
1984	lambs	0.10 ± 0.03	0.32 ± 0.18	0.06 ± 0.02	
Sellafield					(46)
saltmarsh	ewes	0.06 ± 0.03	0.15 ± 0.08	0.06 ± 0.03	
field stud.					
1986	lambs	0.18 ± 0.09	0.49 ± 0.26	0.20 ± 0.11	

Table 6: Transfer coefficients for Cs-137 (d/kg) measured for tissues from ewes and lambs fed contaminated rye-grass harvested 3 days after the peak deposition of the Chernobyl fall-out, grazing on open fells or grazing on saltmarsh or pastures close to the Sellafield reprocessing plant before the Chernobyl accident (mean ± standard deviation).

Higher transfer factors were obtained for lambs compared to their dams, although the radiocaesium concentrations measured in February 1987 in foetuses of ewes from upland farms were consistently lower than that of their dam's muscle : the ratio varying from 0.14 to 0.38 (Howard and Beresford, 1989). The markedly higher transfer factors for lambs' tissues could be explained by a number of metabolic parameters, including the rates of uptake, retention and loss of radiocaesium. One reason could be the supply of more readily available radiocaesium in ewes' milk.

It must also be noted that, in the case of the Chernobyl radiocaesium, the highest transfer coefficients were obtained for muscles ; however, the values obtained for the kidneys do not differ to a large extent from those for muscles. On the contrary, in the case of the Sellafield releases, the kidneys exhibit much greater transfer factors than muscles.

2.3.6 Transfer of radionuclides to cow's milk under controlled conditions

Estimation of grass-to-milk transfer coefficients under field conditions are generally affected by the lack of data on the daily intake. To verify the transfer coefficients from grass to cow's milk estimated from field data and compare the bioavailability of the Chernobyl Cs with the availability of a well-known chemical form, an experiment was performed under controlled conditions. The changes in the radiocaesium concentration in milk during the contamination and decontamination periods is presented in figure 10.

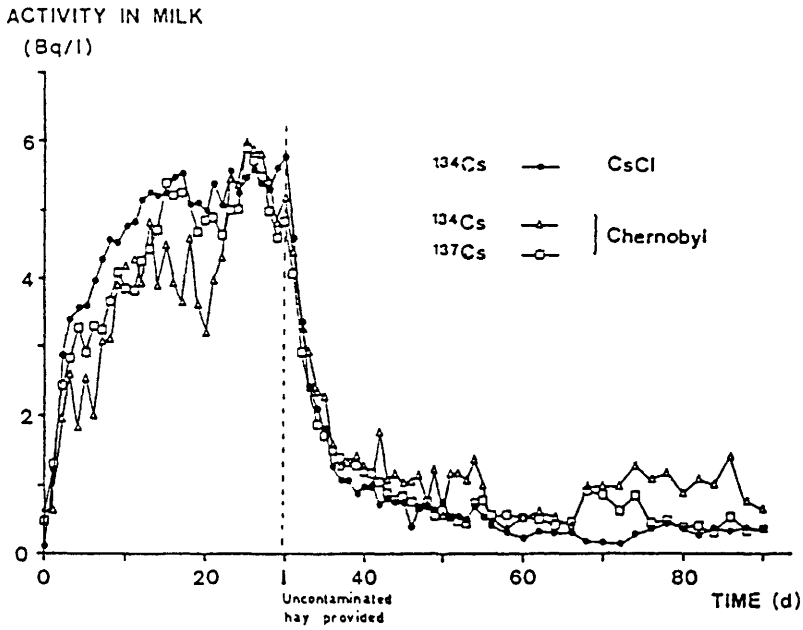

Figure 10: Excretion of radiocaesium (Bq/l) in milk from cows contaminated during the 30 first days with caesium as CsCl or present on hay harvested on mid-June 1986 and contaminated by the Chernobyl fall-out. The concentrations in milk are normalised to a daily intake of 1 kBq.

For Cs-134 administered to the cows as CsCl, the reported values represent the contribution of this form only (the contribution of the Cs-134 from Chernobyl was deducted from the total Cs-134 activity measured). In the case of the contamination by "Chernobyl" hay, a similar correction of the gross values was not possible so that the data are affected by a contribution of the Chernobyl radiocaesium present in the food during the decontamination period as well as of the radiocaesium body burden previous to the start of the experiment.

The results obtained for the transfer to milk in cows fed with "Chernobyl" hay are less homogeneous than the ones obtained for the Cs-134Cl treatment. This can be explained by the daily variations in the ingested quantity and in contamination level of hay given to the "Chernobyl" cows ; the amount of Cs-134Cl administrated to the two other cows was constant during all the contamination period. Both Chernobyl caesium isotopes behave similarly, but are apparently transferred at a slightly lower rate than CsCl. However, the transfer coefficients estimated near equilibrium for caesium as CsCl and for the chemical forms deposited by the Chernobyl fall-out are similar and amount to about 0.0055 d/l. This means that the Chernobyl caesium present on or in (foliar absorption) the forage at the time the hay has been harvested was as available as Cs as CsCl, but this does not imply that the whole caesium from the Chernobyl deposit was under soluble and available forms ; it is not improbable that the less available form has been washed from the plants by rain between the time of the deposit and the harvesting of hay. The transfer coefficient calculated for the Cs-137 present in the hay harvested in 1987 (fed to the cows receiving Cs-134Cl) amount to about 0.011, a greater value than for Cs as CsCl or from recent deposit of the Chernobyl fall-out. As mentioned for sheep grazing on fells one year after the deposit (Beresford and Howard, 1988), it seems that the availability of bioincorporated Cs is greater than the availability of the recently deposited Cs from the Chernobyl fall-out and even than the availability of Cs as CsCl. This needs to be confirmed. The biological half-times estimated during the decontamination phase by a non-linear weighted least squares method are about 2 d for the first compartment, regardless of the isotope and chemical form of Cs, and about 24 and 52 d for the second compartment for CsCl and Chernobyl Cs, respectively. The apparently longer half-time estimated for the second compartment in milk from cows fed with "Chernobyl" hay compared to milk from cows given CsCl is probably due to the unavoidable contamination of the feed given during the decontamination period. If the results obtained during the two last weeks for the cows fed "Chernobyl" hay are not taken into consideration for the calculation, then the half-times estimated for the second compartment are 17 and 25 d for Cs-137 and Cs-134, respectively.

2.3.7 Transfer of caesium to pregnant ewes and their progeny under controlled conditions

The evolution of the percentage of activity excreted by urine and faeces during the contamination period is presented in figure 11 for one of the ewes as an example.

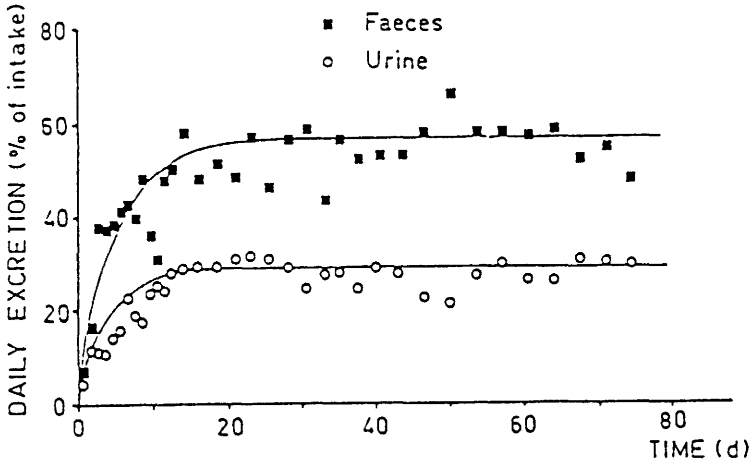

Figure 11: Evolution of the percentage of the daily administered Cs-134 excreted during the contamination period with faeces and urine.

For both excretion routes a plateau is estimated by eye to be reached after about 20 days, approximately the same time needed to reach the plateau for caesium secretion in cow's milk (Vankerkom et al., 1988). Twice more caesium was eliminated by faeces than by urine. Considering for all ewes the input-output balance after the first 76 days of contamination, a total of 74 ± 6 % of the administered activity is excreted : 24 ± 3 % by the urine and 49 ± 5 % with the faeces pathway. The fraction of the daily intake excreted via the faeces was reported to be 50 % by Beresford et al. (1989) for sheep grazing upland pastures contaminated by the Chernobyl deposit. By difference, the retention in the animal body was estimated to 26 ± 6 % of the total intake.

The evolution pattern of activity in blood is similar to those observed in urine and faeces with a plateau after some 20 days (Fig. 12).

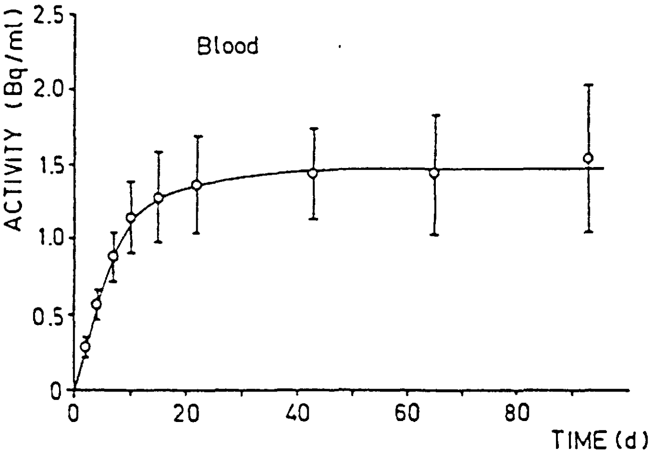

Figure 12: Evolution of the Cs-134 concentration in blood from pregnant ewes chronically contaminated by oral administration of 74 kBq/d as CsCl.

The radiocaesium contamination in wool collected from the back of the animals, increases steadily during the contamination phase, reaching rather high values (Table 7). Newly grown wool collected from an area previously shaved has a specific activity higher than old wool sampled at the same time ; nevertheless it is still not clear whether this contamination results from an internal or external contamination of the hair.

TIME (d)	ACTIVITY (Bq/g f.w.) OLD GROWTH	 NEW GROWTH
14	0.15 ± 0.09	
49	5.26 ± 3.02	
91	17.83 ± 4.91	64.50 ± 21.70
134	25.20 ± 4.24*	

* n = 2

Table 7: Contamination levels measured in wool collected from the back of the animals as a function of time after the beginning of the contamination. Mean ± standard deviation (n = 6).

ORGAN	ACTIVITY (Bq/g f.w.)	TRANSFER COEFFICIENT (d/kg f.w.)
MUSCLE	42.06 ± 0.24	0.57
KIDNEY	36.70 ± 0.21	0.50
LIVER	24.07 ± 0.16	0.33
LUNG	13.77 ± 0.12	0.17
SPLEEN	17.63 ± 0.11	0.24
PANCREAS	30.51 ± 0.12	0.41
RIB	3.05 ± 0.05	0.04
FEMUR	1.08 ± 0.01	0.01
SKIN	7.55 ± 0.10	0.10
SALIVARY GLAND	77.17 ± 0.44	1.04
BLOOD	1.77 ± 0.09	0.02
PLACENTA	3.11 ± 0.32	0.04

Table 8: Activities (± counting error) and transfer coefficients in selected organs of a pregnant ewe sacrificed at lambing, 76 days after the beginning of dosing at an administration rate of 74 kBq/d.

The transfer coefficient in milk samples collected from suckling ewes between day 5 to day 35 after lambing amounts to 0.09 ± 0.01 d/l. This value is in the range of values previously obtained under various experimental conditions (see Howard, 1989).

The evolution of the activity excreted by the urine and faecal routes after the administration had been stopped is presented in figure 13. The experimental results for faeces, urine and total of both excretion routes are best fitted by two-exponential models :

$13.5 \exp(-\ln 2 * t/1.2) + 14.0 \exp(-\ln 2 * t/16.7)$ for faeces,
$7.0 \exp(-\ln 2 * t/1.4) + 8.2 \exp(-\ln 2 * t/20.5)$ for urine and
$20.7 \exp(-\ln 2 * t/1.3) + 21.8 \exp(-\ln 2 * t/18.6)$ for the total of both routes.

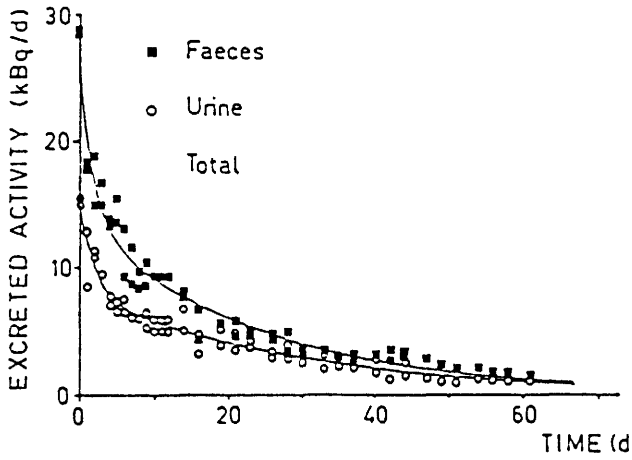

Figure 13: Excretion of radiocaesium by ewes via faeces and urine, during the decontamination period following chronic oral administration of 74 kBq Cs-134/d as CsCl during pregnancy and lactation.

In the three cases, the two compartments have about the same capacity and are characterised by half-times of about 1.3 d for the first compartment and 20 days for the second one. These parameters are similar to elimination parameters found in reindeer in which 50 % of the Cs-137 body burden had a biological half-time of 1.5 d and 50 % had a half-time of 20 d (Nevstrueva et al., 1967).

The decrease of the radiocaesium activity in blood (Fig. 14) proceeds with half-times for the two compartments of about 2.5 and 20 days respectively and is represented by the following equation:

$0.50 \exp(-\ln 2 * t/2.6) + 0.97 \exp(-\ln 2 * t/19.2)$

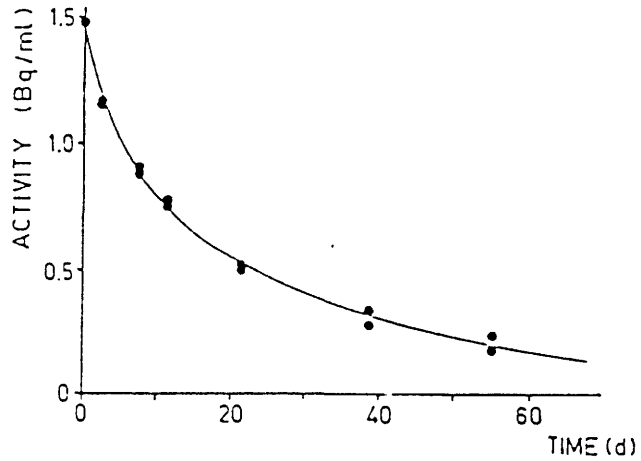

Figure 14: Evolution of the Cs-134 concentration in blood from ewes, during the decontamination period following chronic oral administration of 74 kBq Cs-134/d as CsCl during pregnancy and lactation.

The decrease of the activity from three important organs during the decontamination phase (fig. 15) is also best fitted by two-compartments models and may be expressed as :

$32.7 \exp(-\ln 2 * t/2.9) + 9.37 \exp(-\ln 2 * t/174)$ for muscles,
$31.9 \exp(-\ln 2 * t/2.8) + 4.90 \exp(-\ln 2 * t/123)$ for kidneys and
$22.0 \exp(-\ln 2 * t/2.3) + 2.16 \exp(-\ln 2 * t/157)$ for liver.

For these three organs the half-times estimated for the first compartment is similar to that of the blood but the one of the second compartment is 6 to 8 times longer, varying from 120 to 175 d. This is rather long compared to a value of 19 d derived from the formula proposed by Coughtrey and Thorne (1983) for sheep but comparable with the biological half-time of the long-term

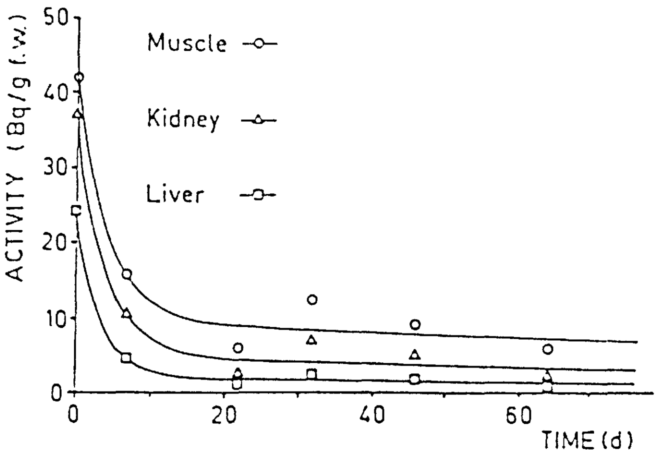

Figure 15: Evolution of the Cs-134 contamination levels measured in selected organs from ewes sacrificed at various times during the decontamination period following chronic oral administration of 74 kBq Cs-134/d as CsCl during pregnancy and lactation.

compartment in man with values ranging from 50 to 150 d (Coughtrey and Thorne, 1983) and the half-time of 158 d already reported in the late sixties for a long-term compartment in rats (Thomas and Thomas, 1968). The capacity of the second compartment is for the muscles (≒ 20 % of the body burden) twice that of the kidneys or liver (≒ 10 % of the body burden). The changes with time of the contamination levels measured in muscles, kidneys and liver of lambs from the two groups are presented in figure 16.

In lambs kept with their contaminated dams, the radioactivity levels increase very rapidly during the first days after birth and at progressively lower rates thereafter. In lambs fed non-contaminated artificial milk from day 3 after birth the activity concentrations in organs decreased with a global half-time estimated, despite of the limited numbers of experimental data, to 11 d in muscle, 10 d in kidneys and 8 d in liver.

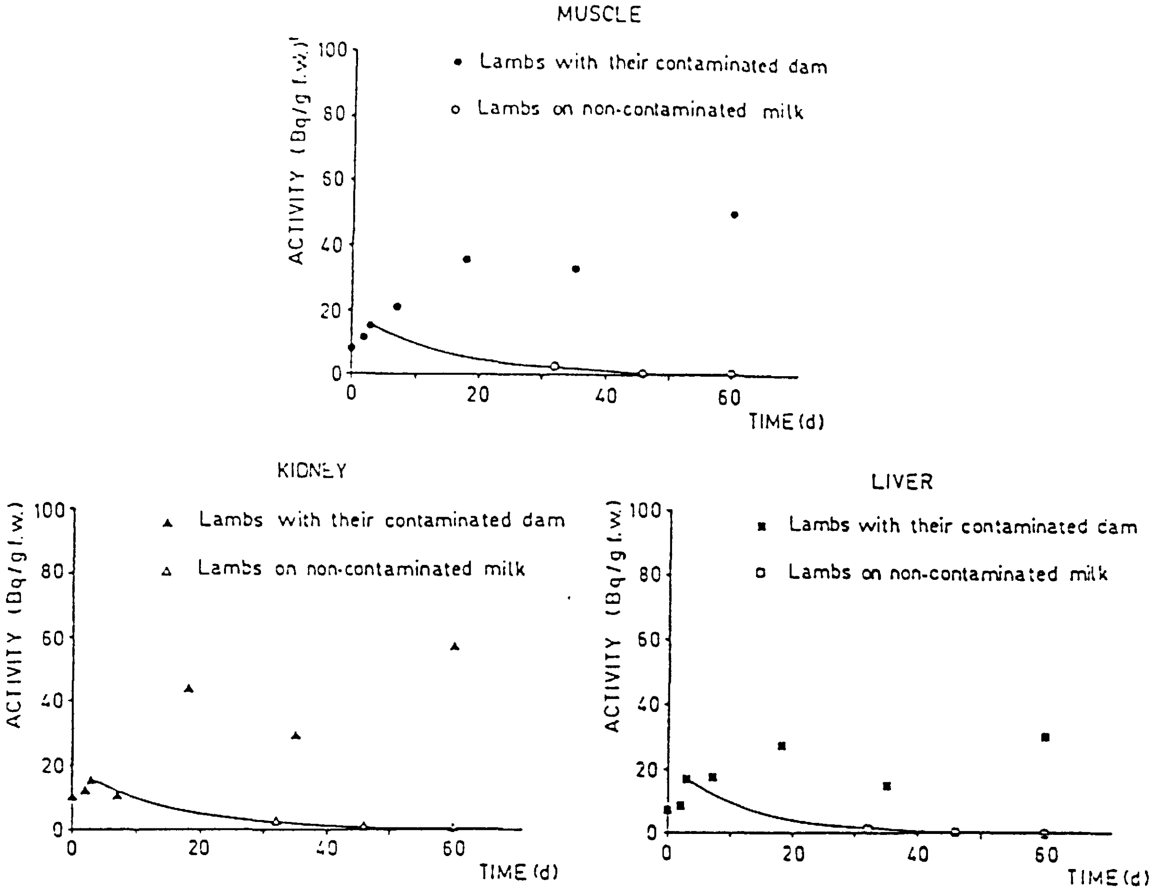

Figure 16: Evolution of the Cs-134 concentration in muscles, kidney and liver from lambs born from ewes contaminated during pregnancy and lactation. Lambs from one group were kept with their dam, receiving contaminated milk ; lambs from a second group were separated from their dam at the age of 3 d and fed uncontaminated artificial milk.

These values are in good agreement with the 10 d (Howard and Livens, 1987) and 10 to 12 d (Colgan, 1988) biological half-times determined after the Chernobyl accident for lambs moved from highly contaminated upland pastures to less contaminated lowland pastures. It is moreover interesting to notice (Table 9) that the contamination levels measured in the organs of a new-born lamb are about four times lower than those measured in the corresponding organs of a contaminated ewe, as already reported by Howard and Beresford (1989), but are higher than the contamination level in the placenta. Two months later the transfer of radiocaesium via the milk gives rise to contamination levels higher than in ewes.

ORGAN	ACTIVITY Bq/g f.w.) EWE	LAMB AT BIRTH	60 DAYS OLD LAMB
MUSCLE	42.06 ± 0.24	8.00 ± 0.03	49.72 ± 0.23
KIDNEY	36.70 ± 0.21	9.65 ± 0.05	56.93 ± 0.23
LIVER	24.07 ± 0.16	6.89 ± 0.03	30.45 ± 0.15
PLACENTA	3.11 ± 0.04		

Table 9: Comparison between the contamination levels (± counting error) measured in selected organs of an ewe at lambing, a lamb at birth and a 60 days old lamb.

Higher transfer coefficients in lambs compared to adult animals had also been observed after the Chernobyl accident for sheep and lambs of the same strain as that used for this experiment grazing lowland pasture at the CEN/SCK experimental farm (Table 10) or had been reported for several different experimental conditions (Howard, 1989).

The transfer coefficients in sheep for radiocaesium from Chernobyl are two to four times lower than those estimated for Cs administered as CsCl. This could be due to a reduced bioavailability of the radiocaesium deposited after the Chernobyl event but could also be partly explained by the physiological status of the animals, pregnant in the later case and not pregnant nor lactating in the former one.

2.3.8 Influence of the incorporation of Cs into plants on its absorption by sheep

The availability of radiocaesium bioincorporated into maize plant for gastro-intestinal uptake in sheep has been compared with that of ionic caesium given as CsCl.

ORGAN	TRANSFER COEFFICIENT (d/kg f.w.)		
	CHERNOBYL-Cs		CsCl
	ADULT	LAMB	ADULT
MUSCLE	0.14 ± 0.06	0.63 ± 0.24	0.57
KIDNEY	0.19 ± 0.08	0.73 ± 0.39	0.50
LIVER	0.12 ± 0.08	0.38 ± 0.22	0.33

Table 10: Comparison of transfer coefficients in selected organs of sheep contaminated with CsCl under laboratory conditions and sheep contaminated by the Cs from the Chernobyl fall-out (samples collected from June to July, 1986). Mean ± standard deviation.

Contamination levels measured in blood at equilibrium were twice higher for ionic caesium than for caesium incorporated in maize leaves. The transfer coefficient in blood were estimated to 0.019 d/l for CsCl and to 0.016 d/l for caesium present in maize. The reason for the lower availability of caesium bioincorporated into plant tissue compared to ionic caesium is most probably due to the "sheltering" of the radionuclide in a structure which is but partially destroyed by the digestive processes and retains part of the incorporated radioactivity. Only the fraction of caesium that can be solubilised is available for gastro-intestinal transfer. Similar observations were reported by Wilson et al. (1969, in Eisenbud, 1987). These authors noted that high crude fibre content diets (such as alfalfa or corn silage), compared to a low fibre content diet (mixed grain), reduced by a factor of 4 the transfer of caesium to cow's milk.

2.3.9 Transfer of caesium to poultry

Data on the transfer of radionuclides to poultry are scanty. Therefore, two controlled experiments were conducted : the first one considered the transfer of caesium to eggs and the second one dealt with the incorporation of radiocaesium in chicken meat.

Transfer to the eggs

The transfer coefficients to the various fractions of egg, estimated when equilibrium was reached (on data from day 19 to day 56), amounted to 0.48 ± 0.07 , 0.27 ± 0.03 and 0.11 ± 0.04 d/kg F.W. for white, yolk and shell respectively. At steady state, 2.4% of the daily intake was excreted with the whole egg.

The contamination pattern measured in eggs (fig. 17) produced during the decontamination phase could be fitted by exponential curves. For white and shell the contamination level decreased immediately after the end of dosing; the activity in yolk showed a kind of lag phase (about 6 days) This lag phase could be attributed to the time needed for this part to be formed in the egg factory. The parameters of the decay curves for the three fractions considered are :

white: A_t (Bq/g F.W.) = $2.29 \exp(-\ln 2 * t/1.23\,d) + 0.49 \exp(-\ln 2 * t/12.8\,d)$

yolk : $5.13 \exp(-\ln 2 * t/1.89\,d) + 0.20 \exp(-\ln 2 * t/17.3\,d)$

shell : $0.34 \exp(-\ln 2 * t/1.45\,d) + 0.12 \exp(-\ln 2 * t/31.1\,d)$

The high value for the capacity of the first compartment in yolk is due to the presence of a lag phase. The variation coefficients for all these parameter are lower than 15%. For white (for yolk and shell this was not possible, maybe due to the larger fluctuations between replicates) a three compartments model could be applied with a better fit of the curve to the point and variation coefficients of the estimated parameters lower than 11%. The equation obtained was :

$$A_t = 1.59 \exp(-\ln 2 * t/0.84\,d) + 0.97 \exp(-\ln 2 * t/3.07\,d) + 0.25 \exp(-\ln 2 * t/22.9\,d)$$

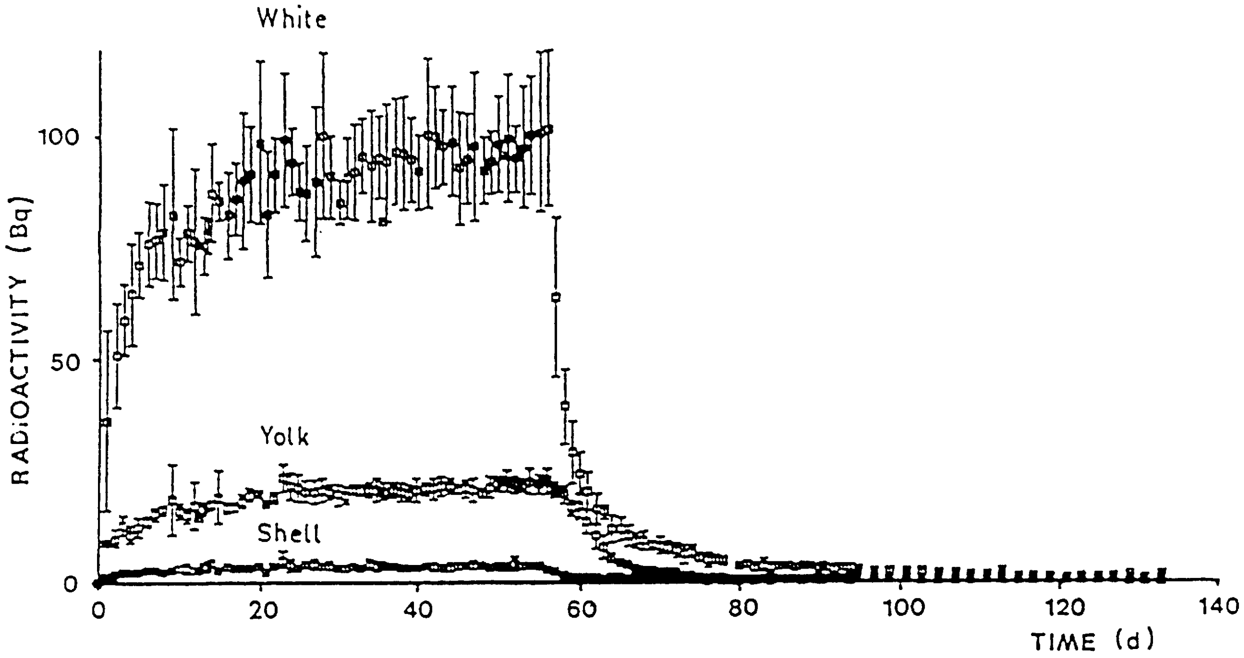

Figure 17: Evolution of the total Caesium content in the different fraction of hen's eggs during a 56 d contamination period followed by a decontamination phase.

<u>Transfer to the meat of laying hens</u>

The activity measured in four hens sacrificed at the end of the contamination period (two on day 42 and two others on day 56) were considered to calculate the transfer coefficients (d/kg F.W.) for various organs. Muscles (breast) did not reach equilibrium within the 56 days of the contamination period ; the ratio of the concentration in muscle to the daily intake amounted at this time to 3.53 ± 0.76. For other organs, equilibrium seemed to be reached.

The transfer coefficient for blood was 0.26 ± 0.04; for liver, 1.06 ± 0.19; for kidneys, 1.62 ± 0.34; for heart, 1.79 ± 0.40 and for gizzard 2.02 ± 0.54.

The retention of caesium in various organs during the decontamination period was best fitted as the sum of two exponential functions, except for muscles for which only one compartment could be identified (table 11).

Organ	1st Compartment		2nd Compartment	
	Cap. (Bq/g FW)	Half-time (d)	Cap. (Bq/g FW)	Half-time (d)
blood	1.4 ± 6%	2.0 ± 10%	0.3 ± 28%	16.0 ± 27%
muscle	20.6 ± 3%	27.7 ± 7%		
gizzard	8.5 ± 4%	0.8 ± 14%	2.3 ± 12%	14.5 ± 15%
heart	8.4 ± 9%	1.0 ± 27%	2.0 ± 32%	17.2 ± 39%
liver	5.2 ± 5%	1.1 ± 13%	1.1 ± 20%	18.5 ± 25%
kidneys	7.5 ± 5%	0.9 ± 17%	2.1 ± 16%	15.2 ± 19%

Table 11: Parameters of the retention of caesium in selected organs of laying hens (mean ± variation coefficient).

Faeces were also collected. Total excretion by faeces during the decontamination phase could be represented as a sum of two (possibly three) exponentials of which the parameters are :

$$2578 \exp(-\ln2*t/1.07d) +658 \exp(-\ln2*t/13.0d)$$

or

$$3095 \exp(-\ln2*t/0.49d) +1152 \exp(-\ln2*t/3.07d) +342 \exp(-\ln2*t/24.2d)$$

For none of these parameters the variation coefficient is higher than 25%. The biological half-times estimated for excretion of caesium by faeces are similar to those calculated for the white of eggs.

2.3.10 Transfer of caesium to growing chicken.

The caesium activity (Bq/g F.W.) measured in selected organs of the chicks continuously fed contaminated flour and sacrificed at various times from the start of the experiment are given in table 12.

Time (d)	Muscles	blood	liver	kidneys	heart	gizzard
3	19.8±0.9	3.2±0.5	16.0±0.4	21.3±0.4	18.4±2.3	
7	31.8±3.2	5.9±0.7	17.9±3.2	24.7±2.7	19.5±4.8	

15	36.6±4.7	5.6±0.7	20.1±2.1	23.2±0.2	21.3±0.8	
21	36.5±2.0	4.1±0.4	13.3±0.8	19.6±1.5	17.9±1.2	
28	31.2±0.6	3.6±0.3	11.8±0.9	18.0±2.7	17.6±2.0	
35	33.4±3.6	3.7±0.3	12.0±0.5	16.6±3.0	16.7±2.3	
42	31.2±8.5	2.5±0.5	12.2±1.5	15.8±3.7	17.4±4.4	
49	26.9±6.4	2.6±0.2	10.3±3.3	18.7±5.4	21.6±6.8	
56	25.5±4.2	2.2±0.5	7.2±2.0	10.9±2.3	13.5±3.5	12.4±2.3
56*	21.7±3.1	2.3±0.2	7.0±1.4	11.0±2.2	13.8±2.5	12.4±0.6

Table 12: Changes with time of the caesium content (Bq/g F.W.) in selected organs and tissues of growing chicken continuously fed a standard chicken flour contaminated with 61.6 Bq ^{134}Cs/g D.W. as CsCl (mean ± standard deviation).The last line of data (56*) corresponds to the activities measured in chicks that have received contaminated feed only during the four last weeks.

The retention in selected organs of chicks from the second group fed contaminated flour during the four first week and the excretion by faeces have been estimated. Activities have been corrected for dilution by growth roughly estimating that the growth of organs is proportional to the increase of the total body weight. A one compartment model has been tested but except for muscles, no good fit was obtained (the estimated curve decline too quickly). A two compartments model was then applied with a very good agreement between experimental points and estimated curve ; the uncertainty regarding the value of the parameter of the first compartment is however very large, due to the lack of points between d0 and d3. The estimated capacity of the compartment(s) and their (respective) half-time(s) are given in table 13.

Organ	1st Compartment		2nd Compartment	
	Cap. (Bq/g FW)	Half-time (d)	Cap. (Bq/g FW)	Half-time (d)
blood	3.5 ± 5%	3.4 ± 11%		
	1.0 ± 126%	0.5 ± 995%	2.6 ± 47%	4.8 ± 38%
muscle	30.0 ± 7%	15.7 ± 18%		

heart	17.3 ± 6%	1.9 ± 13%		
	11.9 ± 13%	0.4 ± 260%	5.7 ± 25%	8.2 ± 27%
liver	11.7 ± 5%	1.7 ± 11%		
	8.2 ± 8%	0.5 ± 80%	3.6 ± 18%	7.9 ± 19%
kidneys	17.6 ± 6%	1.9 ± 13%		
	11.7 ± 18%	0.4 ± 630%	6.3 ± 32%	7.1 ± 33%

Table 13: Parameters of the retention of caesium in selected organs of growing chicken fed radiocaesium from the age of 1 d for four weeks (mean ± variation coefficient).

A regression analysis has also been performed on the variation of the concentration in faeces (data were not corrected for the increase of feed consumption with time and subsequent dilution of the activity in faeces) of chicks from the second group and the following equations were obtained for models with one and two compartments :

$$157.0 \exp(-\ln 2 * t/2.5d)$$

or $$145.4 \exp(-\ln 2 * t/1.2d) + 48.4 \exp(-\ln 2 * t/6.3d)$$

The two compartments model fits the experimental points very well except for the few last ones. the variation coefficients associated with the estimated parameters are rather low (< 10%).

2.4 CONCLUSIONS

The amounts of activity (particularly radiocaesium) deposited on soil surfaces after the Chernobyl accident were dependant on the weather conditions prevailing at the time the radioactive cloud from Chernobyl passed over each region : higher deposition was observed in regions where the passage of the radioactive plumes coincided with rainy conditions.

Initial interception of fall-out radionuclides by pasture grass was function of the herbage density at the time of the deposit. The greater the herbage density, the greater the fraction of the deposit that is intercepted by the green cover. However, the increase of interception efficiency is less than proportional to the increase in herbage density (Kirchmann et al., 1966). Thus, due to a higher dilution in a larger vegetation mass, the specific radioactivity on herbage may be significantly lower in pasture with high vegetation density than in those with a low grass yield at the time of the deposit. As a consequence, animals grazing on high vegetation density meadows will ingest less radioactivity than those grazing on low vegetation density meadows.

Short-term retention of deposited radionuclides is governed by physical decay, weathering processes, dilution by plant growth and removal of contaminated parts by harvesting and grazing. Agricultural practices such as grazing pressure (number of animals per unit area) or grazing pattern (continuous or block grazing pattern), profoundly affect the retention of radioactivity on pastures and, in fine, the evolution of contamination levels in animal products, especially in milk. The short-term effective half-times for the Chernobyl radiocaesium on pasture grass calculated at the different sampling sites considered in this study ranged from 2.6 to 23 d ; those estimated for radioiodine were more comparable (4 to 5.2 d).

Long-term contamination of forage plants is mainly dependant on the soil properties (pH, clay and organic matter content, ...) which influence upon the availability of radionuclides deposited on or leached to the ground for uptake by roots. In this regard, the quality of the pasture (improved <u>versus</u> "natural" ecosystems like the fells in the uplands) plays an important role on the long term availability and transfer of the deposited radiocaesium. In lowland pastures, generally characterised by a rather high clay content in soil, caesium is adsorbed onto the exchange complex and its availability for plant uptake is reduced. Moreover, the potassium is generally present in large quantities, competing with Cs for uptake by roots. A long-term compartment has been identified for the retention of caesium in the two Belgian meadows investigated up to 300 d after the deposit ; the half-time associated with this compartment was 60 d. In the soils of uplands pastures, poor in clay minerals and potassium, caesium remains available for much longer period of times (see chapters on natural ecosystems). All these parameters affecting the retention in forage should be kept in mind when assessing the changes in the contamination of farm produce with general models that were not necessarily designed for the particular set of conditions, characteristic of the region or ecosystem considered. It is recommended that site specific data should be compared with models predictions in order to check the validity of the models assumptions.

There were some evidences that the availability of Chernobyl radiocaesium was less than that of the ionic forms. The grass-to-milk transfer coefficients were lower in sheep fed contaminated grass collected in Scotland 3 days after the deposit (large proportion of Cs present as particles ?) than in those grazing on upland fells during summer 1987 (caesium incorporated into the plant material from soil by root uptake). However, comparing under controlled conditions the transfer to cow's milk of radiocaesium associated with hay collected in mid-June 1986 and that of ionic forms, only a slight reduction, if any, in availability was observed for the Chernobyl caesium. The rainfalls during the 6 weeks between the time of deposit and the harvesting of hay may be suspected to have leached from the grass the contaminated particles in which caesium was poorly available.

Despite of the difficulty to measure or estimate herbage intake, and subsequently intake of radionuclides, transfer coefficients were calculated from the field data collected after the Chernobyl accident. Estimates of the transfer coefficients to milk and meat vary widely but some trends are evident :

- transfer coefficients for milk and meat vary as a function of the animal species (higher transfer in sheep compared to cows).
- the age of the animal is another parameter that influences the transfer of radionuclides (higher transfer in youngs than in adults, although concentration in the foetus is lower than the concentration measured in the muscles from its dam).
- transfer coefficients also vary depending on the physico-chemical form of the radioactive element considered and, by the way, with the source term (higher transfer for the

Chernobyl Cs than for the Cs on saltmarsh - associated with silt - originating from Sellafield) and with the time after the deposit (higher transfer for the Chernobyl Cs one year after the accident than in the months following the deposit).

These aspects have been extensively discussed for sheep by Howard (1989). A better description of the gastro-intestinal absorption depending on animal species, age and physiological status (pregnancy, lactation) as well as on feed characteristics and chemical form administered is needed to explain the wide variation observed under different conditions. Such studies will allow to built more reliable predictive models.

The field and experimental data obtained in the framework of this study, regarding the transfer of radiocaesium to animal products and its retention in animals, have been used by Coughtrey (1989) for the preparation of an extensive review of the "Radioactivity transfer to animal products". The reader will find in this report an updated information of the present knowledge in this field.

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V. TRANSFER IN AQUATIC SYSTEMS
(IFE, LNETI, MAFF, ENEA)

(Improvements in the understanding of radionuclide transport processes in freshwater systems after Chernobyl)

1. INTRODUCTION

Considerable quantities of data were collected from lakes and rivers in the immediate aftermath of the Chernobyl accident. Often it was obtained with no thought to its consequent scientific use and had little supporting data. Hence, interpretation of unusual results was often very difficult if not impossible. Foulquier and Baudin-Jaulent (1990) have already reviewed this data. The present report will concentrate on limitations in our understanding which have been exposed in the wake of Chernobyl and will make recommendations on both the collection of data in the future and the most important directions for future work. Much of the work relates to Cs, as this was the most abundant and easily measured element in Chernobyl fallout, but the general principals are common to all nuclides although they may differ in subtle details.

2. COLLABORATIVE RESEARCH IN FRESH WATERS UNDER THE CEC POST-CHERNOBYL ACTION

Four collaborators were involved: Institute of Freshwater Ecology (IFE), Windermere, UK (coordinator); Laboratório Nacional de Engenharia e Technologia Industrial (LNETI), Lisbon, Portugal; Ministry of Agriculture, Fisheries and Food (MAFF), Lowestoft, UK; Comitato Nazionale per la Ricerca e per lo sviluppo dell'Energia Nucleare e delle Energie Alternative (ENEA), Rome, Italy (no financial contribution from the CEC). Each laboratory encountered very different conditions but work was focused towards elucidating specific sub sections of a conceptual model of radioactive transfer from catchments to the aquatic system and from there into aquatic fauna (fig. 1). The main aim being to use the opportunities offered by accidental releases, particularly Chernobyl, to identify areas where understanding was poor and further, more detailed, research was required.

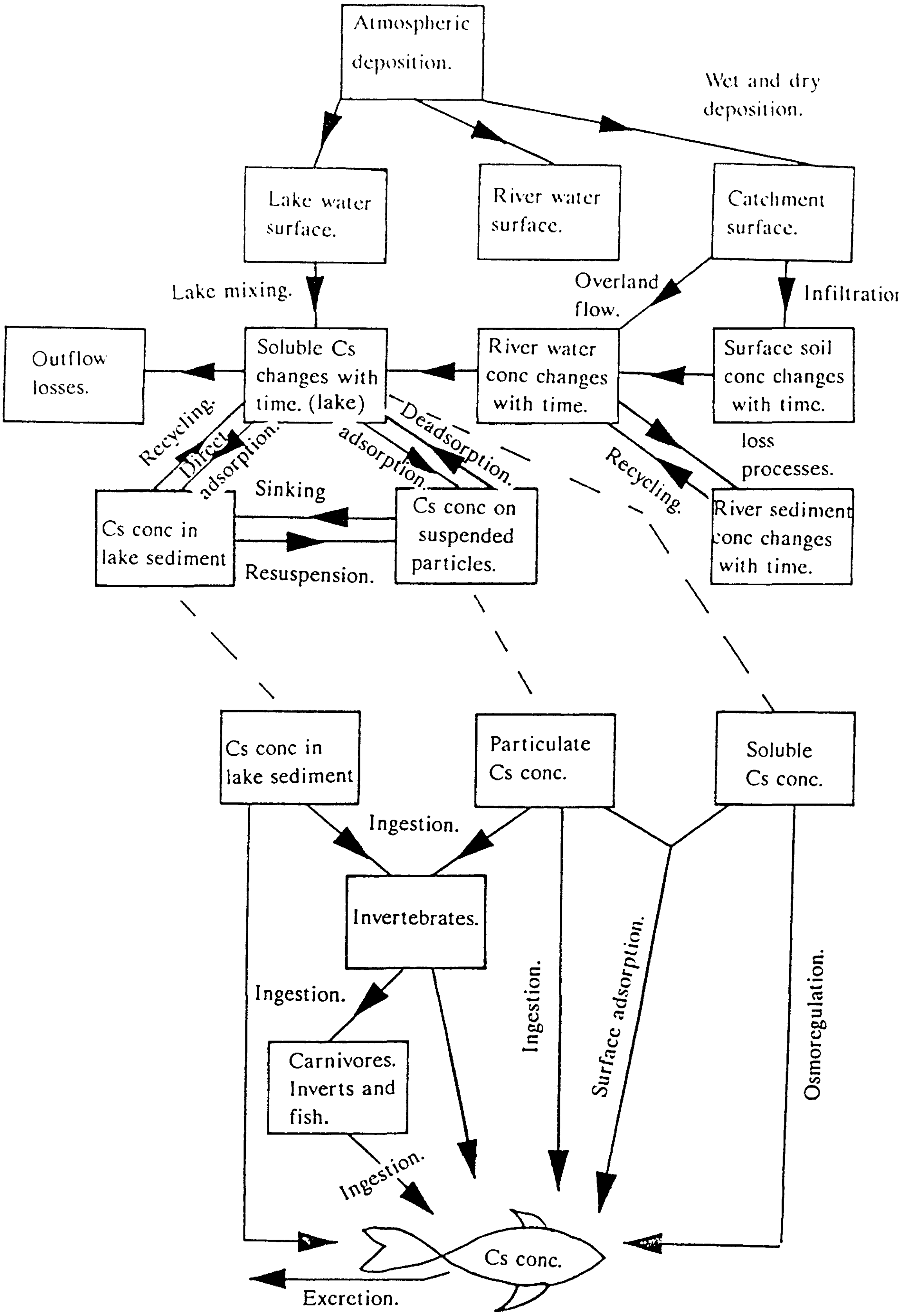

Figure 1: A conceptual model of the transfer of radioactive Cs from atmospheric deposition into fish.

MAFF studied three aspects: speciation of Chernobyl Cs in several water bodies using size differentiation ie ultra-filtration, (Leonard et al., 1989); relationships between K_d and particle/ sediment chemistry (Leonard et al., 1989); and (in conjunction with IFE) the effect of food type on the development with time of the radioactive body burden of fish in field situations (Elliott et al., 1990).

As part of a much larger study of the pollution dynamics of the Marano-Grado lagoon in the Friuli-Venezia Giulia region of Italy, ENEA studied the transport of radioactivity from land to sea in an actively eroding, sediment laden river and some of the variables which affected the final distribution of Cs in the sediments of the lagoon (Ventura et al., 1988).

As part of an on-going study of the transfer of radionuclides in the Fratel dam lake system on the Tejo river (Carreiro and Sequeira, 1987; Freitas et al., 1988; Carreiro, 1988; de Oliveira et al., 1989; Ferreira, 1989), LNETI have studied two areas in detail: the effect of several variables on the K_d of Cs-134 (Madruga et al., 1988) and Co-60 (Madruga and Carreiro,1990); and the dynamics of the transfer of radiocaesium both directly from water and through a trophic chain from water to algae, to daphnia and finally to fish (Corisco and Carreiro, 1990a,b,c).

In conjunction with the National Radiological Protection Board and the Atomic Energy Research Establishment, Harwell the IFE carried out an investigation into the chemical form of Chernobyl Cs in atmospheric dusts (Hilton, Cambray and Green,1990). In addition two, adjacent lakes with very different physical properties and productivity levels were intensively monitored to provide information on the chemical transport processes operating within lakes (Hilton, Hamilton-Taylor et al., 1990; Hilton, Davison et al., 1990). A survey was made of several lakes along lines of constant deposition in order to ascertain the importance of catchment runoff in maintaining high radioactivity levels in water long after the Chernobyl event (Hilton, Lishman and Carrick, 1990). In conjunction with the Institute of Hydrology as much as possible of the understanding gained in the study was incorporated into a mathematical model which predicts the change with time of the concentration of radionuclides dissolved in the water, attached to suspended particles, transported to the sediment and into fish (McDougall et al., 1990).

In order to exchange ideas and cross fertilise research aims Dr J Hilton (IFE) visited ENEA staff in Rome and Venice; Dr Hilton and Dr CA Mills twice visited staff at LNETI, Lisbon for discussions and Ms Madruga (LNETI) visited IFE, Windermere.

3. DIFFICULTIES IN OUR UNDERSTANDING OF RADIONUCLIDE TRANSPORT EXPOSED BY CHERNOBYL

3.1 ESTIMATION OF INPUT FLUXES FROM ATMOSPHERIC DEPOSITION

Radioactive material can be deposited in one of two ways: dry deposition or wet deposition. Both can be fairly complex processes but dry deposition tends to produce much more constant concentration within a given area, although differences in land surface roughness, including vegetation type, within the deposition area will greatly influence the final surface concentration. Wet deposition fluxes, on the other hand, are dictated by the quantity of rain. Fallout from atmospheric weapons testing was well dispersed in the upper atmosphere over a long period of time, so that deposition maps in a single region could easily be obtained from long term rainfall maps (Cawse and Horrill, 1986).

However, much of the Chernobyl material in the atmosphere remained in very localised clouds, so that Clark and Smith (1988) required a knowledge of both rainfall patterns and the trajectory and time of passage of the radioactive cloud in order to model the general distribution of Chernobyl deposition observed in a 1000 square mile area of north western Britain (Cumbria). The major variable was found to be altitude, as rainfall is much higher at higher altitudes in this region. However, Hilton et al. (1988) were unable to estimate the deposition flux in a much smaller, 30 square mile, area within better than a factor of 2 because of differences in the measurement of rainfall at four sites, at the same altitude in the catchment. Since measurements of radionuclide concentrations in rainfall were also scarce (Dept of Agriculture for Northern Ireland et al., 1986), errors reported in the direct estimation of deposition are likely to be considerably lower than the true errors.

In the event of a future accident, arrangements (both practical and financial) should be made beforehand to measure radionuclides in integrated rainfall samples from as many different sites as possible covering the area of interest in order to obtain a reasonable measure of the total deposition in a region. Daily samples from only a few sites would be required to obtain a picture of the change in deposition with time.

3.2 BEHAVIOUR OF DEPOSITED RADIOCAESIUM POST DEPOSITION

Experience from weapons testing fallout suggested that the majority of deposited radiocasium (in particular) was immobilised immediately on making contact with the ground or, when it fell directly onto water, immediately on making contact with the sediments. This resulted in very clear records of the change in Cs deposition with time in many lake sediments (Pennington et al., 1973). However measurements made within a few months of the Chernobyl deposition have shown many examples of Cs-134 being found at depths in sediments well below those expected from the accumulation rates calculated from clear Cs-137 profiles from weapons fallout (Hilton et al., 1989; Figure 1). Similar effects have been observed in ombrotrophic peat bogs which have good Pb-210 dates retained in their profile (Appleby et al., 1988) but, in which Cs-134 moved rapidly to depths of several cm (up to 15 cm in one case) below the surface (Appleby et al., in prep.). Since transport to these depths by mixing processes such as bioturbation by sediment living animals would have also destroyed the fallout or lead profiles, the data suggest that Chernobyl material was more mobile than material from weapons fallout.

This conclusion is consistent with analyses of atmospheric particulate material (Hilton Cambray and Green, 1990) which showed that only about 8% of the bomb fallout was in a water soluble form, whereas at least 80% of the Chernobyl material was water soluble. Although these measurements were not completely definitive as there may have been a change in availability in the Cs derived from weapons fallout during the 20 years storage, they show that the Chernobyl fallout, at least, was mobile for some time immediately after deposition. Similar measurements made on lake sediments only nine months after Chernobyl showed that the Cs had rapidly become unavailable (Hilton Cambray and Green, 1990).

Santschi et al. (1988) cited calculations from which the likely chemical form of radionuclides can be estimated knowing the properties of the source, particularly the fuel and temperature of the source reactor, but these calculations can only be made some time after the event and it makes more sense at this state of our knowledge to use simple analytical measurements such as absorption by cation or anion exchange resins or by activated charcoal for non ionic forms to delimit the possible chemical forms present and hence to have a better idea of the likelyhood of chemical and biological uptake after any event. Details of an apparatus for obtaining large volume samples from field situations is given by Spezzano et al. (1988). Leonard et al. (1989) gave details of an alternative approach using ultra filtration which showed that, on occasion, up to 17% of the Cs in some lakes was associated with molecules greater than 100K Daltons. In any future accidental release more work should be directed at the chemical form and availability of the radionuclides introduced to the environment.

3.3 RETENTION BY CATCHMENTS

Following the work of Evans et al. (1983) and others, it was established that, although Cs could be absorbed onto special sites on clay minerals from which it could be released under certain circumstances, with time it became locked in the crystal lattice and unavailable. On this premise it was assumed that Cs deposited onto lake and river catchments would rapidly become immobilised and only be transported in the solid form as natural erosion took place. Data from Chernobyl confirmed this supposition in catchments with clay soils (Hilton, Hamilton-taylor et al., 1990). However, water concentrations monitored at some sites remained very high for several years after the Chernobyl deposition (Camplin et al., 1989). These high concentrations were much greater, and lasted much longer than would be expected if only deposition directly onto lake surfaces had been the sole contributor and normal loss processes were operating (see later).

Transport of material from the catchment was suspected and a survey of several lakes along a line of constant deposition (Hilton et al., 1990c) showed that lakes within a few kilometres of each other could have very different radiocaesium concentrations even though the original deposition was the same. Data suggested that this may be the result of deposition onto organic/ peat catchments where Cs was only weakly bound and that chemical processes in the catchment could bring it back into solution to be transported into lakes or rivers. Although the data implicate peat catchments, and Cremers et al. (1988) have used their knowledge of the adsorption properties of clay minerals to model the remobilisation of Cs by ammonia in rain falling on such catchments, the survey data suggest that the presence of peat in a catchment is not sufficient on its own to produced enhanced runoff. More work is required to clarify this anomaly, particularly as the high quality water found in these upland catchments is often transported to distant urban populations for use in potable supply. It is interesting to note that evidence of this phenomenon has been observed and recorded in the literature of weapons fallout inventories of lake sediments in the Adirondacks (Heit et al., 1987) but it was not recognised as constituting transport of Cs from the catchment.

3.4 MODELING THE TEMPORAL CHANGE IN RADIONUCLIDE CONCENTRATION IN WATER AFTER A NUCLEAR RELEASE

Mathematical models can be used both to test our understanding of an ecological system and as a management tool in the control of discharges. In this report we will concentrate on those models which are based on the underlying processes of chemical transport in waters and associated material. By comparing the output of these models with actual measurements made in the field after Chernobyl, flaws in our understanding were highlighted. Although models have been written to predict concentration changes in rivers (eg CEC/NRPB, 1979), they are complicated, compared to lake studies, by the large changes in chemical, and particularly suspended solids concentrations which are induced by changes in flow. Since the physical properties of lakes can be approximated very easily by assuming instantaneous, complete-mixing in all or part of the water body, individual processes can be measured more easily than in rivers. Lake models can also be transferred to rivers very easily by approximating the river to a chain of completely stirred reactor tanks, hence most of the major developments have been made in lake studies.

3.4.1 Transfer from the catchment

Carlsson (1978) proposed an early model which is still one of the best models of transfer from a catchment to a water body. In this study he assumed that some of the deposited (in his case atmospheric) radioactivity would be washed off immediately while a second fraction accumulated in the catchment soils to be eroded at a very slow rate. He suggested that the fraction of the long term accumulated Cs was a function of the runoff of surface water, however his removal rates for fresh and accumulated radionuclide on the catchment could only be obtained from a least squares fit to the data. As with all models an inability to define constants in terms of underlying physical and chemical properties reduces the general application of the model as they must be calibrated at every site before use, and require a good set of deposition - transport data in order to do so. Although Smith et al. (1987) showed how fast and slow erosional components may be estimated from sediment core data in ideal circumstances. There has been little attempt to relate radionuclide removal rates with physical transport models developed in geomorphology. Similarly, until recently, there has been little attempt to look at the binding properties of catchment soils.

However, recently Cremers and co workers (1988) have looked more closely at the binding of Cs by clay minerals, particularly illite. In this mineral three binding sites can be identified: (i) large numbers of non selective sites on the top and bottom of the plate-like crystals; (ii) a few sites on the edge of the plates which are very selective for Cs; and (iii) sites inside the crystal which take time to fill but from which release is very slow. They were able to measure the Cs on the different sites and have used these data to show that the edge sites control the release of Cs and that only very small molecules can displace the Cs. Hence, they suggest that ammonium concentrations in rainfall can displace Cs from these sites and have modeled Cs release from upland catchments using this approach (Cremers, pers. comm.). Further work is required to link the chemical and biological processes on the catchment with the geomorpological models of solid transport in catchments before Carlsson's model will be super ceded.

3.4.2 Processes of radionuclide loss from the water column

In the recent past a number of new models have appeared (eg Snodgrass et al., 1986). Most have been based on the original proposals of Santschi et al. (1986). Who defined three physical pathways (excluding radioactive decay) for removal of radionuclides from the water column: hydraulic washout (or dilution); absorption onto particles followed by settlement; direct adsorption onto surface sediments.

Hydraulic washout occurs when polluted water enters a water body followed by more clean water. Assuming instantaneous mixing then the concentration of radionuclide in the lake is the same as in the outflow and decreases exponentially with time. The higher the flow, compared to the volume, the faster the rate of dilution.

For lakes this introduces a seasonal cycle onto the rapidity of hydraulic dilution. In the winter (for a monomictic lake, see later) or in spring and autumn (for dimictic lakes. see later) the lake is completely mixed and the volume of interest in modeling is the total volume. However, in the summer (monomictic lakes) or in summer and winter (dimictic lakes) the lake separates into two distinct parts which do not mix together, although each mixes within itself to form isolated, homogeneous water bodies. Under these conditions the upper mixed volume (epilimnion) is considerably smaller than the total lake volume and dilution of radionuclides is much more rapid. The seasonal effect can range from very small, in conditions where heavy winter rainfall into the whole lake is more than balanced by much reduced rainfall runoff in summer into a smaller epilimnion, to a very large effect in some deep alpine lakes where the onset of stratification coincides with very high flows due to snow melt. In this case dilution can be extremely rapid.

[The surface water of monomictic lakes is heated by the sun in summer and becomes less dense. Eventually the density difference becomes so great that wind energy cannot mix the warmer, less dense water into the body of the lake and stratification occurs. Stratification is maintained until, in the autumn, the heat input decreases and wind strength increases until eventually the lake mixes completely again. In dimictic lakes summer stratification occurs due to the same process but winter stratification also occurs because ice on the lake surface removes the mixing effect of the wind. Complete mixing occurs in dimictic lakes in autumn and spring.]

Adsorption of radioactive substances in solution onto particles is governed by the K_d value where:

$$K_d = \frac{\text{Concentration of radionuclide in the solid phase (Bq/kg)}}{\text{Concentration of radionuclide in solution (Bq/l)}}$$

Once a radioactive atom, ion or molecule has adsorbed onto a particle then radioactivity can be lost from the water column by settlement of the particle. Typically settlement rates vary from 0.1 to about 3m/d (Hilton et al., 1989) depending on the size and constituents of the particle, for example, a 6 μm particle consisting solely of quartz would have a settlement velocity of about 2m/d, a 70μm diatom with a silica skeleton would have a settlement velocity of about 1.3m/d and a 70 μm green alga has a velocity of about 0.2 m/d.. Settlement times are a function of both the settlement velocity and the mean depth of the mixed layer, ie the whole lake in winter and the epilimnion in summer (monomictic lake). In deep lakes this will introduce a delay between the appearance of radioactive material in the water column and in the sediment.

The third process incorporated into recent models is the direct adsorption of radioactivity onto the sediment from the water. This process was originally identified when small, plastic bag enclosures in lakes were used as test systems (Hesslein et al., 1980). Since the test lakes were simple plastic bags floating below rubber collars with the open end level with the lake water surface and sediment on the closed bottom end, there were no hydraulic losses. Similarly, as the experiments only lasted for a few weeks, particulate material could not be replaced by inflows or generated internally by algal growth so that settlement losses were minimal. However, even under these conditions radionuclides disappeared at considerable rates. Simple calculations using Fick's laws of diffusion showed that direct transport of radionuclides across the boundary layer of static water separating the sediment from the mass of moving water in the body of the lake could be sufficient to remove radioactivity to the sediments at the correct rate.

The boundary layer is a function of the velocity/ turbulence of the water above a solid surface, but is typically of the order of 100-150 μm. The rate determining step is the diffusion of radioactivity across this boundary, driven by the concentration gradient across it. Hence the rate of loss of radioactivity from the water column depends on the turbulence at the water -sediment boundary and the concentration gradient. The latter is initially dependent on the concentration in the water, but as time progresses the transport rate will slowly reduce as the concentration in the interstitial water increases, reducing the concentration gradient. In order for diffusion to continue across the boundary there must be a mechanism for removing radioactive material from the interstitial water in the sediments, such as adsorption onto the solid sediment. The adsorption coefficient need not be particularly large for this process to be very efficient as the concentration of solid surfaces in the sediment is very high.

By statistically fitting data to the model equation, Hesslein (1987) found values of the settlement velocity and boundary layer which were consistent with expected values. However, the model consists of several components which relate concentration to time in an exponential form and, since a curve fitting approach was used to fit data to a model which was practically reasonable, constants of the expected order were found. No independent measurement of the proportion of material being transported by each process was made. Hilton et al. (1989), using similar numbers of data points in the fitting process, found that, within the limits of the field measurements, their data could be explained by models incorporating hydraulic losses combined with just one of either particle transport or direct adsorption to the sediments. In order to prove that all the mechanisms suggested by the Santschi- Hesslein model are equally important in a normal lake, studies must be carried out which measure either the particle flux to the sediments using sediment traps and /or the diffusional flux across the sediment water boundary (no methodology is available at present) as well as the loss of activity in solution in the water column. Studies to date have used either traps (Sanschi et al., 1989) or a mass balance (Hilton, Davison et al. 1990) not the more useful combination of techniques.

A further problem with the particle transport process is that there is no agreement as to the most important particles which carry radionuclides out of the system. Normally total particle concentrations would be measured in the form of suspended solids, but, if only one type of particle is important for a specific radionuclide then the total suspended solids concentration could change without changing the concentration of the important particles. For example, if the arguments of Evans- Cremers (1983,1989 respectively) are taken to their logical conclusion then the only particles which are important in the transport of Cs are clay mineral particles, especially illite. Any changes in the concentration of other particles such as algal cells would not affect the transport rate even though the solids concentration could change by an order of magnitude over a period of a few weeks in spring. Leonard et al. (1989) have made initial studies to address this problem and have shown that the K_d varies inversely with organic content and directly with Al and Si concentration in the sediments. Similarly Ventura et al. (1989) have shown that K_d varies with particle size and redox status.

Algal cells also pose another question. Do they act simply like inert particles so that the only process transferring radioactivity to the cell is adsorption on the surface or, conversely, is the radionuclide taken into the cell by active uptake mechanisms. Pally and Foulquier (1981) reviewed the evidence for caesium and concluded that adsorption is the major uptake mechanism and that uptake was usually very small. However, using desorption experiments, Corisco and Vaz Carreirro (1990a) showed that at least 25% of the uptake over seven days was due to incorporation into the cell.

3.5 SEDIMENTS

Sediments play a major role in the removal and storage of radionuclides from the water column. They can act as short term stores, long term sinks or sources of recycled radionuclides.

Radionuclides reach the sediments either by direct adsorption across the boundary layer or by particle transport. In the sediment they can undergo physical or chemical changes which strengthen the hold of the sediments, such as the change observed for Chernobyl Cs (Hilton, Cambray and Green, 1990) which became completely unavailable to chemical leachates within nine months of deposition. As time passes the sediment containing deposited radionuclide becomes overlain by freshly deposited, uncontaminated sediment. In lakes burial rates can vary from a mm to a few cm a year (eg Pennington et al., 1973). However during the time it takes to bury the material several cm below the surface and into an, essentially permanent store, wave action may resuspend the sediment into the water column of shallow lakes or, chemical conditions may change and release the nuclide from the sediments into the interstitial water and hence, by diffusion, into the main body of water. The latter process can be enhanced by the presence of animals living in the sediment whose burrows increase the rate of release of radioactivity because of the greater surface area they create and because they irrigate the burrows with an induced flow of water which actively pumps out the remobilised radionuclides.

Sholkovitz (1985) discussed the mechanisms which may be operating to remobilise Cs in sediments. He concluded, at that time, that the evidence was inconclusive. However more recent work by Cremers et al. (1990) has shown that certain sites on illite are very selective for Cs and that the same sites are also very selective for NH_4^+. Production of ammonium ions by microbial reduction processes in the sediments release bound Cs whereas excess K or Na, which are not absorbed as selectively, will not. Using very low background counting facilities, Comans et al. (1989) have now measured coincident peaks of ammonia and Cs in lake sediments proving the mechanism. However, it is interesting to note that Cs release has only been observed in anoxic hypolimnia (bottom waters in stratified lakes) whereas the ammonium displacement mechanism only requires that sediments become sufficiently anoxic that nitrate is reduced to ammonium. Hence there is no reason why Cs release should not be observed in oxic hypolimnia. Is this a case of finding only what we want to find?

Hilton et al. have shown that Cs release into the anoxic hypolimnion of a small lake only mobilized 1% of the Chernobyl material stored in the sediment in the year following deposition. This constituted a very small contribution to the total load to the lake. It is interesting to speculate that, even though it has not been observed, remobilisation in anaerobic sediments and release into oxic waters for long periods of time each year could generate much higher fluxes than in the anaerobic hypolimnia where Cs release has been observed.

3.6 TRANSFER INTO FISH

Because quasi steady state conditions were induced during the deposition of weapons testing fallout it was possible to use simple factors to estimate the concentration of Cs in, say fish, given the concentration in another phase, usually water. However the dynamic nature of the Chernobyl event did not allow a steady state to develop so that concentrations in fish predicted by this method bore no resemblance to reality (fig 2).

In terms of modeling the transfer of radionuclides into fish, considerable difficulties arise as to the most important mode of ingestion, either through the water by respiration and osmoregulation or by food. There is a considerable amount of evidence, which has been reviewed by Foulquier (1979), which suggests that food is the main source of ingestion in the long term, but direct transfer from the water may be more important in the short term. The effect of increasing monovalent cation concentrations, particularly potassium, is reasonably well understood and adequate relationships have been developed to predict the lower concentrations in fish flesh which result as potassium concentrations increase. However the majority of models still assume that the fish in question feeds in the water column, so that at steady state the concentration in fish flesh is a simple multiple of the water concentration. There appears to have been little appreciation that many fish, particularly those most often used as food by man, do not feed in the water column but on invertebrate animals living in the sediments. Data from a field survey was unable to distinguish differences between fish feeding on different food sources (Elliott et al., 1990). However one model has been found which has addressed the problem (Lambrechts, 1984). In this case the concentration of radionuclides in the sediment was used as the basis of the prediction of concentrations. A test in the Rhône river (Foulquier and Lambrechts, 1983) showed that the model assumptions were reasonable. By combining models of the transport of radionuclides to the sediment with the model approach of Lambrechts, Mcdougall et al. (1990) were able to reproduce concentration changes in fish flesh after Chernobyl (fig 2).

4. DIFFERENT AQUATIC ECOSYSTEMS

For atmospherically deposited Cs,at least, the soil type in the catchment controls the change of concentration change with time in water. Lakes and rivers with organic or sandy (probably) catchments are likely to experience higher inputs of

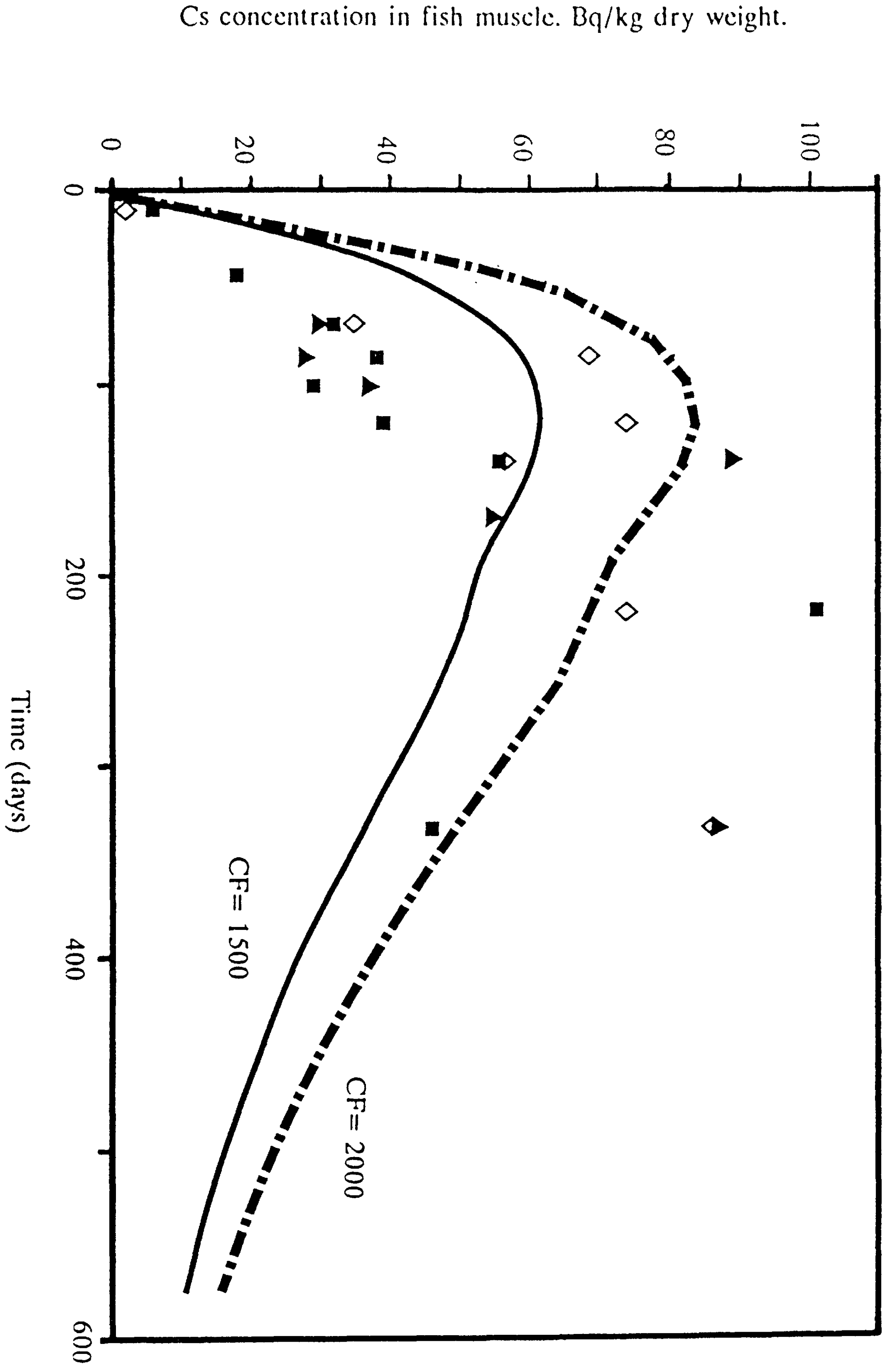

Figure 2: Observed Cs concentrations in Windermere fish: brown trout (■), pike (Δ) and perch (◇). Simulated concentrations shown by lines.

radionuclides, for longer time periods than catchments with well developed clay soils. No run off will be experienced in the latter case and lake concentrations will be dictated simply by the depositional flux directly onto the lake surface and the mean depth. Hence, the greater the mean depth the lower the concentration in the water. Once the radioactivity has reached the water it reacts in exactly the same way, assuming it is present in the same chemical form, whether it is derived from atmospheric deposition or from a liquid discharge.

The two obviously different aquatic environments are rivers and lakes. Although the same loss processes operate in both environments, rivers can be considered as plug flow reactors, with water passing at an initial time t_0 not mixing with water passing at a later time $t_0 + t$. Lakes, however, approximate to completely mixed reactors and water entering at time t mixes with water entering at time t + t,smoothing out the effects of a spike input of radioactivity. In rivers the dominant property is the flow. Conversely, in lakes the equivalent property is the retention time, which is the average length of time water spends in the lake and is given by the lake volume divided by the flow. Although rivers can be subdivided in terms of the "flashyness" of their hydrograph (the speed at which the river responds to an increase in flow in its upper reaches) and lakes can be subdivided in terms of the ratio of their depth to area ("shallow" lakes being more likely to resuspend bottom material than "deep" lakes) or in terms of their trophic status (productivity), in general all rivers behave in the same way as do all lakes. This is a slight oversimplification but differences between lakes are usually explainable in terms of their physical and chemical properties and, as such do not constitute "different ecosystems". The same is true of rivers.

The only obvious exceptions to this generalisation are lakes created on large rivers by damming the flow. In this case the water body can act as a river in periods of flood when the gates are open, but at other times, when flow is greatly reduced, it will respond like a lake. However the approximation to complete mixing is much less valid as the lakes tend to be long and thin reducing the ease of mixing between the ends. Although unrelated work (Hilton, 1985) has shown that these systems can follow the same rules as true lakes for much of the year, it is not entirely clear yet whether these lakes should constitute a separate group or whether they should simply be considered as lakes at one time and as rivers at another.

5. REMEDIAL ACTION IN THE EVENT OF ACCIDENTAL RELEASE

Using the increased understanding obtained from models and from measurements made in the aftermath of the Chernobyl accident it is now possible to make some informed comments about the treatment of aquatic systems to aid their recovery after a pulse input of radionuclides from the atmosphere, such as occurred after Chernobyl. A pulse of radioactivity will pass through a river system relatively quickly, although the original pulse will spread over a longer length of river in the extra time it takes to pass down a long river, compared to a short river. Hence the first line of defense is simply to monitor and shut water treatment inputs appropriately to avoid contaminating water supplies. Assuming the pulse is short; that only relatively little runoff occurs from the contaminated catchment and that flows in the river are reasonably high, then the river water will clear itself fairly rapidly.

If the initial contamination was high then the sediments may have been contaminated, in which case one of two options is open. In a regulated river it may be possible to open control gates to create surges of water of sufficient turbulence to keep contaminated solids in suspension until the system has flushed itself. Alternatively, the river bed can be mud pumped in the worst affected areas to remove polluted mud. This approach should be attempted as soon as possible after the clearance of the water column so that the minimum of diffusion into the sediments and the minimum of dilution by uncontaminated fresh sediment has occurred so that the minimum of sediment has to be transported and disposed of. In lakes and reservoirs there will never be a short event. The events after Chernobyl confirmed that the mixing in lakes extends the time for which a lake is contaminated, compared to a point on a river system and it is seldom that problems will last less than a few months. From the models which have been developed it would appear that there are two options: if considerable quantities of uncontaminated water are available, then it may be possible to increase the flow rate through the system, increasing the hydraulic washout losses from the lake. This option is seldom likely to be viable unless the pulse input is very localised. The second option is to treat the lake with a relatively fine (a few tens of microns diameter) solid material which has a high, and preferably irreversible, affinity for the radionuclide(s) in question. For Cs, illite would be ideal. The radionuclide would absorb to the solid and settle to the lake bed within a few days. This approach would be less applicable to shallow lakes where resuspension regularly occurs. If chemical changes in the sediment could bring about a remobilisation of the contaminant then the introduced material would need to mud pumped from the surface of the sediment within a short time of the treatment.

If the radioactive input were long lasting then there is little that can be done directly for the aquatic environment until the input ceases, apart from issuing warnings about the dangers of eating fish caught in the water and making extra provision for treatment at water supply treatment works.

6. FIELD MEASUREMENTS AND THE REPORTING OF RESULTS

In order to make the best use of radioecological data which is collected in the future (for whatever purpose) every effort should be made to measure the following parameters with every sample taken for radioactivity measurements and these data should always be reported with the activity data:

1) A major ion balance (Ca, Mg, Na, K, Cl, SO_4 , NO_3 , Alkalinity/ carbonate, pH and conductivity). These parameters define the water. They are generally present in mg/l quantities and hence usually compete with low concentration radionuclides in absorption and ingestion processes. Particularly important examples are the effect of K on Cs uptake in fish and the competition between Cs and K and Na for adsorption sites on particles.

2) Suspended solids concentrations. Both the total and organic component (ignition loss at 550° C) should be reported. These measurements are a controlling variable in defining the solid liquid distribution of radionuclides. It would also be very helpful to have the cation exchange capacity reported and XRD data for the inorganic fraction, as the K_d is very dependent on the minerals which are present.

3) Total phosphorus is a major parameter defining the productivity of lakes, and larger, slow flowing rivers.

4) Ammonia concentrations (for Cs). Concentrations of ammonia of the order of a few hundred μg/l are sufficient to compete effectively for Cs specific adsorption sites and release radioactivity into solution.

5) Weight and length of fish (as well as species identification) and, ideally, age. These data can be used to assess the growth rate of the fish. In addition, for a given concentration of radioactivity in the water, larger fish will contain higher concentrations compared to smaller fish of the same species. Comparisons should only be made between fish of the same size.

6) Concentrations of major algal species suspended in the water. In productive lakes algal cell can provide an enormous surface for adsorption. Information on the species numbers (and size from the literature) allows some estimate of the surface area and biomass volume to be made.

8) Concentration of radioactivity in the top few specified) cm of sediment with % organic matter, a major cation (at least) analysis and, ideally, an XRD analysis of the major mineral content.

9) For rooted aquatic plants the same data as for 8 above should be reported, but for a homogenised sample covering the whole of the root zone depth.

10) Physical information on the lake or river should always be given such as: river cross section or lake bathymetry (mean and maximum depth, mean thermocline depth, total surface area and surface area above the thermocline at least); instantaneous flow rate and annual mean flow rate (for rivers); mean and summer retention time for lakes.

11) Whenever possible concentrations of as many radionuclides as possible should be measured. Data from Carreiro et al. (pers. comm.), after an accidental release on the Tejo river, showed that different nuclides followed different pathways and, Hilton et al. (1990) showed that such data, when used in conjunction with models of radionuclide transport, can give more information on the transport mechanisms of all nuclides than could be obtained from one nuclide alone.

7. REQUIREMENTS FOR FUTURE WORK

Considerable advances have been made in the past few years in the production of models based on fundamental properties. A major advantage of this approach to modeling for management use is that models are usually portable, ie they can easily be applied at any site of choice with the minimum of calibration because, in the best circumstances, all the constants involved in the formulae are known, and do not vary between sites. It is possible now to build a fundamental model of radionuclide transport within a given lake (McDougal et al., 1990) allowing predictions of the change in concentration with time in different compartments such as dissolved in the water, fish flesh, etc based the models of Carlsson (1978), Sanschi at al (1986), Aoyama (1978) and Lambrechts (1984). However, in order to calculate the distribution of radionuclide between solid and solution in the water column, a K_d value must be used. Unfortunately K_d values in the literature vary over at least four orders of magnitude, with no guidelines as to the most appropriate value to use in any specific situation. Hence it is not possible to estimate water and sediment concentrations without a considerable amount of work at any new site.

The K_d is theoretically the ratio of free ion in solution to the reversibly absorbed ion on a particle. Field or laboratory measurements of this parameter can be biased in many ways (fig 3). It is also dependent on the properties of the solid such as particle size, mineral or substance type, the ease of release of the absorbed material,etc. Unfortunately no systematic study has been carried out to try and either lay down any guidelines to aid in the choice of K_d values or to evaluate the most important parameters influencing the variability of K_d so that it can be modeled from easily measurable parameters.

A similar problem occurs with the incorporation of concentration factors (CF) in models of radionuclide transfer into living material. In the same way as K_d, CF can vary over several orders of magnitude. Although Foulquier (1979) has listed some of the parameters in his review, there appears to be little systematic work to use as a guide in choosing CF values at different sites. Both of these parameters require more work to define the underlying variables.

8. CONCLUSIONS AND RECOMMENDATIONS

The Chernobyl accident provided a, scientifically, very useful test of our understanding of the transfer of radionuclides through aquatic environments. It highlighted several flaws in our understanding of the dynamics of transfer, particularly the contribution made by certain types of catchment to elevated water concentrations and the importance of using correct food chain pathways for predicting fish flesh concentrations. An initial outline of the ideal requirements for information which should be collected during any repeat of the Chernobyl incident have been made, along with recommendations for the reinstatement of polluted water. Future work should concentrate on areas which limit the portability of predictive models, particularly sources of variability in K_d values and CF values, but also including the effect of catchment type, transfer processes from water to sediment and sediment release mechanisms.

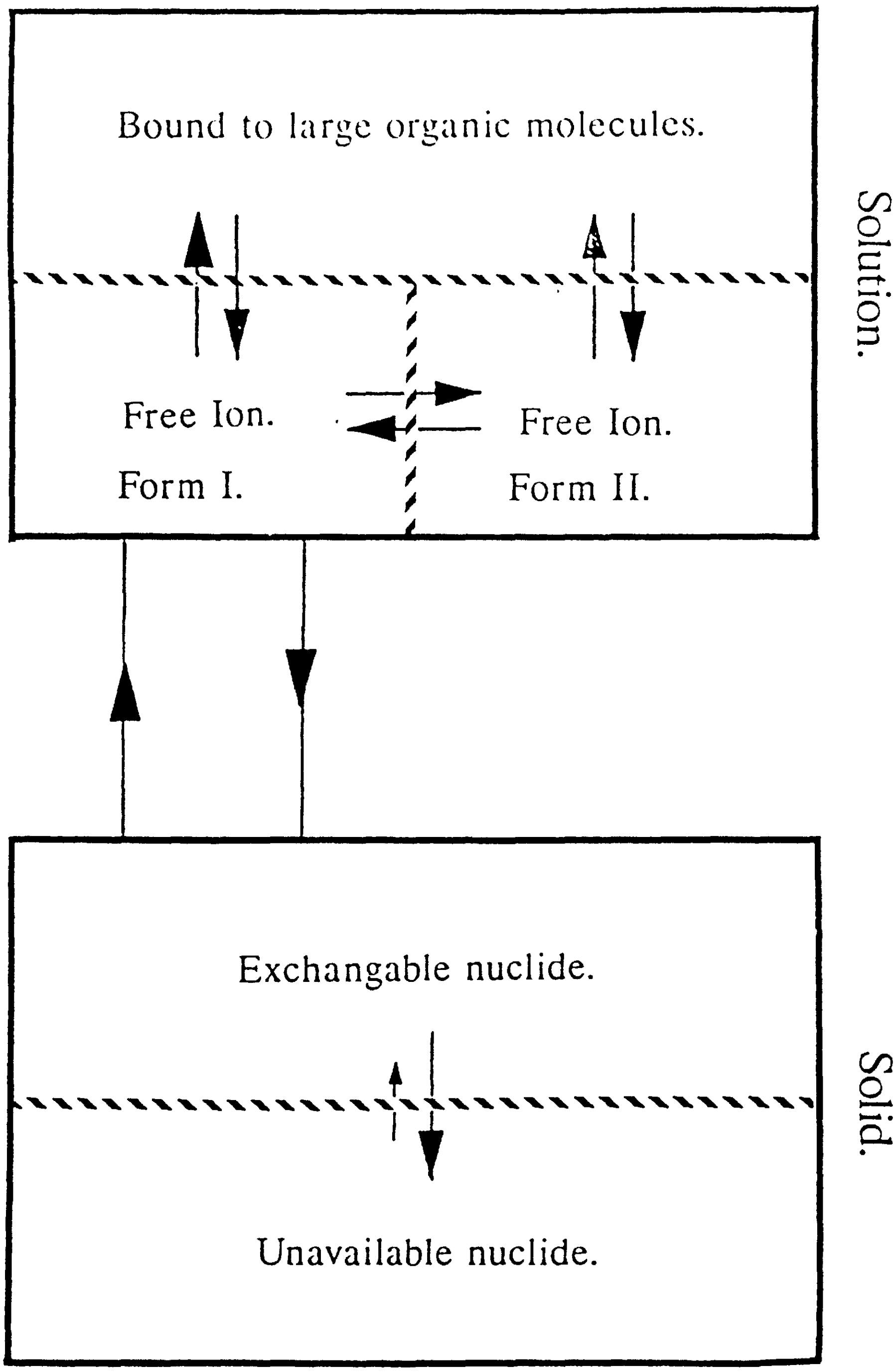

Figure 3: A model of the adsorption of a radionuclide to a solid phase. K_d(theory) = exchangeable nuclide on solid / free ion in solution. K_d(actual) = radionuclide in solid form / radionuclide in solution.

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VI. CONCLUSIONS

Action 2 of the CEC Post-Chernobyl Programme has considered four main research areas; chemical speciation, soil-plant transfer, plant-animal transfer and transfer in aquatic systems. Aspects of the behaviour of radionuclides in these different systems have been studied by a large number of laboratories from many member countries. The main conclusions drawn from these extensive investigations are reported below.

Sixteen radionuclides were detected in the aerosols from the Chernobyl accident. The aerosols had a median diameter of 1μm. Iodine was found in only small quantities in the aerosols. Theoretical predictions of the chemical forms of many of the radionuclides present in the aerosols from the Chernobyl accident were found to be reasonably accurate, and laboratory aerosol generation experiments satisfactorily simulated the accident source term. Caesium iodide (CsI), caesium hydroxide (CsOH), metallic ruthenium and silver were present. However some unexpected compounds were also present, such as cadmium-caesium nitrite. The solubility of the radioecologically important radionuclides appeared to be similar to that of the relevant stable elements, thereby depending on their chemical properties. Strontium exhibited a low to moderate solubility. Caesium was found to be moderately soluble when it is generated from uranium-rich mixtures, as occurred in the Chernobyl fallout. Indeed a fraction of the radiocaesium was probably associated with colloids, suggesting that the transfer of radiocaesium from Chernobyl in the environment might initially be expected to be lower than previously anticipated in predictive models.

Statistical evaluation of a large data set on soil to plant transfer from many countries seemed to confirm this view. In agricultural ecosystems soil-to-plant transfer ratios for radiocaesium originating from the Chernobyl accident were about two times lower than that of radiocaesium from other contamination sources. Discrepancies in the data set were apparently attributable to three main factors; (i) changes in availability with time (ii) large data sets from areas with high soil organic matter contents and (iii) foliar absorption and translocation. Apparent effects of region and season could be explained by differences in soil type, variation in crops, differences in agricultural practices and varying weather conditions. The importance of soil type in determining soil-to-plant transfer of radiocaesium was emphasized. For example cereals grown on a soil with a perched water table gave significantly higher radiocaesium transfer factors than cereals grown on cambisol and fluvisol. The use of lightly contaminated sewage sludge had little influence. On soils with high content of coarse sand and gravel transfer factors were lower, presumably due to the presence of high levels of extractable potassium in these soils, and to deeper penetration of plant roots.

In natural and semi-natural ecosystems soil-plant transfer factors were consistently higher for radiocaesium from Chernobyl compared with that on the aged deposit from weapons fallout. This appears to be because the aged deposit has migrated into sites in these soils from which it is less likely to migrate in the soil solution. In contrast Chernobyl radiocaesium initially had a greater mobility in these soils and moved more readily from soil components into the soil solution. In many of the contaminated soils, particularly podzols, radiocaesium from Chernobyl has been retained in the top few cms of the soil profile and is hence still within the rooting zone of many plant species. The aged deposits of radiocaesium have migrated further down the soil profile, particularly in highly organic peats. Unfortunately the paucity of data in these ecosystems prior to Chernobyl combined with the different characteristics of the source terms (a single pulse dose compared with continual deposition over a long period) make it difficult to compare the availability of the different sources of radiocaesium immediately after deposition.

After an environmental release of radiocaesium ruminants inhabiting natural and semi-natural ecosystems are likely to have comparatively high radiocaesium activity concentrations in their meat and milk, compared with ruminants from agricultural areas. Ecological characteristics in these environments which make them vulnerable to this form of pollution include the presence of :

- soils lacking the specific clay minerals which immobilize radiocaesium and which therefore allow its uptake into vegetation;
- vegetation species with high uptake rates and/or retention of radiocaesium;
- ruminant species with a high transfer of radiocaesium from gut to tissues.

Valuable information has been obtained on the transfer of radiocaesium into animal products from both unimproved areas and agricultural areas. From these studies a number of general points can be highlighted. Although estimates of transfer coefficients to milk and meat vary widely some trends are evident :

- transfer coefficients for milk and meat vary with animal species. Generally higher transfers occur in sheep and goats compared with cows;
- age is an important factor influencing transfer of radionuclides. Higher transfer occurs in younger animals. However radionuclide concentrations in foetuses are low compared with the mother;
- transfer coefficients are very dependent on physico-chemical form of radionuclides.

The Chernobyl accident highlighted several flaws in our knowledge of the dynamics of transfer, in aquatic ecosystems. In particular the importance of (i) the contribution made by certain types of catchment to elevated water concentrations of radionuclides and (ii) of using appropriate food chain pathways for predicting fish flesh concentrations was emphasized. Recommendations for information collection from aquatic ecosystems, should such an incident as the Chernobyl accident reoccur, have been made. Similarly recommendations for the reinstatement of polluted water have been suggested.

VII. RECOMMENDATIONS

1. After the Chernobyl accident concentrations of radiocaesium in plants and animals in natural and semi-natural ecosystems were higher than those from agriculturally improved areas and the high levels persisted for a comparatively long time. Predictive models based on agricultural areas were inadequate to cope with these ecosystems. The mechanisms determining the mobility of radiocaesium in such ecosystems, were insufficiently well understood, particularly for a pulse deposition of radiocaesium. Further studies are needed on the behaviour of many radionuclides in these ecosystems, particularly strontium and the actinides.

Important areas for future research in the natural and semi-natural of ecosystems of Europe could include;

- The effects of climatic factors on radiocaesium movement;
- Further studies of the Chernobyl material to document and explain long term changes in behaviour;
- The development of models, particularly concerned with such ecosystems;
- Study of the binding sites of radiocaesium in upland soils and of factors which influence the migration of radiocaesium into soil solution;
- Investigations into the reasons for differential uptake of radiocaesium by species in the same habitat.

2. It has become apparent that the bioavailability of radiocaesium changed with time. Initially the particulate radiocaesium appeared to be less available for uptake. However once radiocaesium was incorporated into plant tissue its uptake increased. In contaminated soils with a high organic content the radiocaesium seems to have gradually migrated to stronger binding sites so that its availability for plant uptake has declined. The time dependent nature of bioavailability and the effect of chemical form is a neglected area of radioecology which should be addressed. The consequences of changing transfer rates between environmental compartments are considerable for predictive models.

3. One factor which appears to influence transfer of caesium from soils to plants is prevailing climatological conditions. Currently few relevant studies have been carried on this aspect and therefore the importance is difficult to assess.

4. Studies after the Chernobyl deposition were focused on contamination of agricultural systems and their food products. Some of these studies were poorly structured due to the need for quick assessments of the doses to the population. Well designed studies using internationally standardised methodology would enable the implications of any future event to be assessed quickly and suitable remedial measures applied. It is recommended that a wide range of ecosystems should be studied and appropriate models developed.

5. In aquatic ecosystems more detailed supporting data should be collected to supplement radiochemical data and suggestions of minimum requirements have been made. Future work should concentrate on areas which limit the portability of predictive models, particularly sources of variability in K_d values and CF values, but should also include studies of the effect of catchment type, transfer processes from water to sediment and sediment release mechanisms.

VIII. GLOSSARY

Activated charcoal

particles of carbon with a small particle size which have a very large surface area for a given size due to the creation of large numbers of holes on the surface by heat treatment.

Adsorption

adherence of an atom, ion, or molecule to the surface of another substance. In soil adsorption of cations present in soil water to organic matter or mineral particles often occurs (see also cation exchange and and exchange sites).

Adsorption coefficient

a number defining the strength of adsorption of a chemical to a defined solid surface (see K_d).

Aerosols

a suspension of liquid or solid particles in a gas or mixture of gases such as air. Fog and smoke are common examples of natural aerosols.

Agricultural ecosystem

ecosystem in which soil and plants are influenced by agricultural techniques as ploughing, fertilizing, irrigation, plant protection, etc.

Algae

microscopic plants which live in water.

Angiosperms

a large, diverse group of plants characterised by the possession of flowers although they may be relatively inconspicuous. They have two major subgroups - monocotyledons and dicotyledons

Anion exchange resin

a small particle size resin which removes negatively charged ions from solution by exchanging them for another anion with which the resin was originally impregnated.

Artificial contamination

Addition of radionuclides to soils as part of an experiment.

(Bio-)Availability

Quantification of the extent to which an element or isotope present in a particular form or association can be absorbed by a living organism.

Bioincorporation

association with tissues as the result of biological processes

Bioturbation

disturbance of bottom sediments by the movement, particulary tunnelling, of invertebrate animals in their search for food.

Boundary layer

a thin layer of static water which acts as a boundary between moving water and a static object. The thickness of the boundary is inversely related to the velocity of the water

Bryophytes

a group of simple plant forms that lack vascular tissues and roots. They are usually small terrestrial plants that grow in damp places and attach themselves to their surroundings by rhyzoids.

Catchment

the land surrounding a lake or a river which contributes water, which originally fell as rain, to the water body.

Catchment runoff

water which runs off a catchment into a lake or a river.

Cation exchange

a mechanism by which cations in solution change places with cations adsorbed to adjacent substrates such as mineral particles or organic matter (see also adsorption and exchange sites)

Cation exchange resin

a small particle size resin which removes positively charged ions from solution by exchanging them for another cation whith which the resin was originally impregnated.

Chernobyl TF_{SP} data

all soil-to-plant transfer measurements on Cs isotopes which are deposited in the environment as a consequence of the accident in the nuclear reactor of Chernobyl in April 1986.

Clay minerals

alumino silicate minerals with plate-like crystals which are present in manu soils.

Climate

- **Arctic** - typically has a growing season of 50-60 days, the summer temperature is low, soil moisture levels are low tree growth is absent and there is snow cover for much of the year.

- **Atlantic** - typically produces cool, moist summers and and mild wet winters on the western side of European land masses adjacent to the Atlantic, with dryer, hotter summers and colder winters on the eastern side.

- **Boreal** - characterised by a short growing season with relatively low temperatures; rainfall is spread fairly evenly throughout the year. Low evaporation rates often lead to wet soils and slow decomposition of organic matter Mainly occurs in Scandinavian mountains and mountains of northern Scotland.

Compartment

Distinct entity with respect with kinetic parameters; each compartment is characterised by capacity and input-output rates.

Compartmental analysis

Statistical estimation of the parameters (half-time and capacity) characteristic of a retention curve modelled as a sum of decreasing exponential functions, each of them being assumed to represent a compartment.

Concentration ratio

Ratio of the concentration of an element or isotope in one material (sink) to the concentration of this element or isotope in a different material (source).

Correlation

a statistical relationship between otherwise unrelated variables for a specific range of values

Correlation coefficient

a dimensionless quantity using values in the range -1 to 1 measuring the degree of linear association between two variables. A value of -1 indicates a perfect negative linear relationship, a value of 1 a perfect positive relationship.

Dalton

unit of size for large molecules

Daphnia

small (1 mm) free swimming invertebrate animal which lives in the water column and feeds on algae.

Decomposers

an organism such as bacteria and fungi that breaks down dead organic matter into simpler compounds.

Deposition processes

- wet: mechanism by which particulate matter and aerosols that have become suspended in the atmosphere can be returned to the ground in rainfall, snow, sleet and hail.

- dry: mechanism by which particulate matter and aerosols that have become suspended in the atmosphere slowly fall to the ground due to gravity. Usually occurs during still conditions.

Detritivores

soil animals that feed on dead organic matter in upper soil layers

Diffusion

the movement of material, at the molecular level, from a region of high concentration to one of low concentration due to random movements of molecules.

Dimictic

lakes which stratify twice a year.

Dispersion efficiency

the ability to spread particulate matter evenly throughout a solution.

Ecosystem

a relatively self-contained natural environment that is defined by the types of animals, plants and insects that it contains. eg forest, grassland, soil etc...

Environmental contamination

radionuclides present in the soil as an undesired consequence of washout and fallout from nuclear weapon tests and from releases of nuclear facilities.

Epilimnion

the upper part of a lake during stratification.

Ericaceous

plants that belong to the heather family. It includes heaths, heathers and rhododendrons.

Exchange sites

very small areas on substrates such as mineral particles and organic matter that have a negative or positive charge associated with them to which ions from adjacent solutions can adsorb (see also adsorption).

Ff (transfer factor or transfer coefficient for meat)

ratio of the concentration of an element or isotope in an organ or tissue to the total amount of this element or isotope ingested daily, at steady state equilibrium (d/kg).

Fission products

- gaseous: gaseous radioactive elements that are released when the nucleous of an atom is split. For example, ^{131}I.

- particulate: solid radioactive particles that may be released when a nuclear reaction takes place. For example, ^{137}Cs.

Fm (transfer factor or transfer coefficient for milk)

ratio of the concentration of an element or isotope in milk to the total amount of this element or isotope ingested daily, at steady state equilibrium (d/l).

Geomorphology

study of the physical features of the earth and its geological structures.

Ground water

water that leaches through soils until it reaches the water table. Ultimately it permeates into streams and rivers and drains to the sea

Growth strategy

mode of living adopted by plants and animals using resources in their environment. For example, development of large root systems in soils lacking specific nutrients required by particular plants.

Half-time (biological)

half-time associated with the biological elimination processes and possibly dilution by growth.

Half-time (ecological)

half-time associated with the biological elimination processes, possibly dilution by growth and weathering processes.

Half-time (effective)

half-time associated with the biological elimination processes, possibly dilution by growth, weathering processes (if relevant) and physical decay.

Half-time (retention half-time)

time needed to decrease the total amount of an element or isotope or concentration of an element or isotope in a compartment by a factor 2.

Humic material

black organic material of complex composition which is the end product of microbial breakdown of plant and animal residues in soil

Hydraulic washout

the removal of material in a water body by replacement with water which containes less of the material of interest.

Illite

a hydrous potassium alumino silicate clay mineral.

Interface

the boundary between two different substrates. For example:

Ions

an atom or radical that has lost or gained one or more electrons so that it has a positive or negative charge.

- anion: an ion with a negative charge. For example, NO^{3-}

- cation: an ion with a positive charge. For example, K^+.

K_d

the ratio between the amount of a radionuclide adsorbed onto solid material and the amount of radionuclide in solution.

Leachate

a chemical solution which removes parts of a solid material by dissolution.

Leaching processes

washing of ions and nutrients out of soils that occurs as soil water drains through soils.

Lichens

a composite organism formed from the symbiotic association of some fungi and a green algae or cyano-bacterium. They are usually found encrusting rocks or tree trunks and can often survive in extreme environmental conditions.

Lysimeter experiment

experiment executed on plants grown in containers filled with at least 0.125 m^3 of soil and characterized by the presence of a drainage system.

Major ion balance

summation of all the major chemical components of a water to show that the sum of cations equals the sum of anions.

Monomictic

a lake which stratifies only once a year.

Non-Chernobyl TF_{SP} data

all soil-to-plant transfer measurements on Cs isotopes with exclusion of the Chernobyl TF_{SP} data.

Nutrient elements

any element used or required by plants and animals as food. For example, carbon, nitrogen, oxygen etc...

Ombrotrophic peat bog

a peat bog which obtains all its nutrients etc. from the atmosphere.

Organic matter

plant or animal material. Organic matter in soils comprises of dead plant and animal matter that is partially decomposed.

pH

a value used to represent the acidity or alkalinity of^AA an aqueous solution. Values are expressed on a scale between 1 and 14. pH 7 denotes a neutral solution, less than pH 7 an acidic solution and more than pH 7 an alkaline solution. These values are also applied to soils.

Productivity levels

the amount of productivity, or biological growth, a water body can maintain.

Retention time

the average length of time that water spends in a lake or stretch of a river.

Semi-natural ecosystem

ecosystem in which impact of men is limited to actions like removal of part of the vegetation by extensive grazing, wood cutting etc.

Soil characteristics

descriptive properties of soils that can be used to differentiate between soil types. For example, pH, particle size, organic matter content etc...

Soil density

weight per unit volume (W/V) of soils usually expressed as kilograms per cubic metre.

Soil type

- brown earths - loamy soils with a brown or reddish, friable, non-calcaereous subsurface horizon.

- peat - a soil that is almost entirely composed of organic matter, is often waterlogged and in which decomposition processes are slow.

- podzols - sandy or coarse loamy soils, normally well drained with a bleached horizon and/or dark brown or black compact subsurface horizon enriched in humus.

- rankers - non-calcareous soils, normally well drained and usually shallow over non-calcareous rock or massive limestone.

- rendzinas - calcareous soils, normally well drained and usually shallow, over shattered limestone, chalk or soft extremely calcareous material.

Soil water

water present in the soil due to rain or flooding. It leaches nutrients, ions and/or pollutants out of the soil as it drains down the soil profile.

Speciation

identification of the chemical form of an element.

Supernatant solution

a liquid or fluid forming a layer on the surface of another solution.

Stratification

the separation of a water body into two or more parts due to density differences, usually brought about by differential heating of the top of the water column.

TF_{SP}

the transfer factor of radionuclides from soils to plants. It is defined as the concentration of a radionuclide in plant material, divided by its concentration in the soil on which the plant was grown.

Thermocline

a region of rapid temperature change with depth in a stratified lake.

Time lag

the period elapsed between contamination of the soil with radionuclides and harvesting of the crop.

Tropic chain

the chain of plants and animals from the simple plants, which are eaten by herbivores, which in turn are eaten by carnivores, which in turn are eaten by larger carnivores.

Ultra-filtration

concentration of large molecules in solution using a membrane containing small holes which allow the solvent to pass through but retain the large molecules.

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