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Modelling of radionuclide interception and loss processes in vegetation and of transfer in semi-natural ecosystems

Second report of the VAMP Terrestrial Working Group

Part of the IAEA/CEC Co-ordinated Research Programme on the Validation of Environmental Model Predictions (VAMP)

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FOREWORD

Following the Chernobyl accident and on the recommendation of the International Nuclear Safety Advisory Group (INSAG) in its Summary Report on the Post-Accident Review Meeting on the Chernobyl Accident (Safety Series No. 75-INSAG-1, IAEA, Vienna, 1986), the IAEA established a Co-ordinated Research Programme on "The Validation of Models for the Transfer of Radionuclides in Terrestrial, Urban and Aquatic Environments and the Acquisition of Data for that Purpose". The programme seeks to use the information on the environmental behaviour of radionuclides which became available as a result of the measurement programmes instituted in the countries of the former USSR and in many European countries after April 1986 for the purpose of testing the reliability of assessment models. Such models find application in assessing the radiological impact of all parts of the nuclear fuel cycle. They are used at the planning and design stage to predict the radiological impact of planned nuclear facilities, in assessing the possible consequences of accidents involving releases of radioactive material to the environment and in establishing criteria for the implementation of countermeasures. In the operational phase they are used together with the results of environmental monitoring to demonstrate compliance with regulatory requirements regarding release limitation.

The programme, which has the short title "***Va***lidation of Environmental ***M***odel ***P***redictions" (VAMP), was started in 1988; it is jointly sponsored by the Division of Nuclear Fuel Cycle and Waste Management and the Division of Nuclear Safety and is also supported by the European Commission. There are four working groups within the programme: the Terrestrial Working Group, the Urban Working Group, the Aquatic Working Group and the Multiple Pathways Working Group. This is the second report of the Terrestrial Working Group.

Other reports issued under the VAMP programme are:

Modelling of Resuspension, Seasonality and Losses during Food Processing. First Report of the VAMP Terrestrial Working Group, IAEA-TECDOC-647 (1992).

Assessing the Radiological Impact of Past Nuclear Activities and Events, IAEA-TECDOC-755 (1994).

Modelling the Deposition of Airborne Radionuclides into the Urban Environment. First Report of the VAMP Urban Working Group, IAEA-TECDOC-760 (1994).

Validation of Models using Chernobyl Fallout Data from the Central Bohemia Region of the Czech Republic. Scenario CB. First Report of the VAMP Multiple Pathways Assessment Working Group, IAEA-TECDOC-795 (1995).

EDITORIAL NOTE

CONTENTS

Chapter 1

INTRODUCTION

The Terrestrial Working Group of VAMP aims to examine, by means of expert review, the state-of-the-art in modelling the transfer of radionuclides from the terrestrial environment to man. Topics for review are decided upon by the working group taking into account the relative importance of a given process as a potential contributor to radiation dose to humans, the degree of uncertainty which exists in the area because of lack of knowledge and the possibilities presented by the Chernobyl accident for improvement of understanding of the process.

The first report of the working group considered the modelling of the resuspension of surface deposits into the air, the modelling of the variation of the contamination of crops with the season when the release occurs (seasonality), and the estimation of radionuclide loss from food products as a result of food processing (IAEA-TECDOC-647 (1992)).

The present report reviews (i) the modelling of radionuclide interception and retention by vegetation and the loss processes from vegetation and (ii) the modelling of radionuclide transfer to foodstuffs derived from semi-natural ecosystems.

(i) Radionuclide interception and loss processes in vegetation

Data available since the Chernobyl accident have strengthened the view that the transfer of radionuclides from air to vegetation is a primary area of uncertainty in the estimation of the contamination of food chains leading to human exposure. The processes affecting the overall transfer from air to vegetation involve wet and dry deposition, interception and initial retention, and post-deposition retention of radioactive substances by vegetation. During the growing season, the time-integrated concentrations of radionuclides on vegetation in the first few months after initial deposition are dominated by the direct foliar interception of deposited material. Chapter 2 contains a review of data for modelling the direct foliar interception and initial retention of radioactivity deposited by dry and wet processes, together with data on the factors affecting post-deposition retention of radioactivity on the vegetation.

(ii) Transfer of radionuclides by terrestrial food products from semi-natural ecosystems to humans

The potential radiological significance of radionuclide transfer to humans via foodstuffs derived from semi-natural ecosystems has become apparent since the Chernobyl accident. Foodchain models developed before this time usually did not take such transfers into account. The processes leading to contamination of food in these environments are complex and current understanding of the transfer mechanisms is incomplete. For these reasons the approach adopted in Chapter 3 is to represent, by means of aggregated parameters, the empirical relationships between ground deposits and concentration in the food product.

The two reviews presented in Chapters 2 and 3 were prepared by named authors but have had the benefit of a review process which involved the members of the Terrestrial Working Group of VAMP. The papers presented in this publication have been revised to take into account the comments received from the working group.

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Chapter 2

RADIONUCLIDE INTERCEPTION AND LOSS PROCESSES IN VEGETATION

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2.1. INTERCEPTION AND INITIAL RETENTION

2.1.1. Introduction

When materials are deposited from the atmosphere, whether by wet or dry deposition processes, some fraction of the materials will be intercepted by vegetation, with the remainder reaching the ground. Of the materials intercepted by the vegetation, some portion may bounce, roll, or be blown or washed off the vegetation to the ground while the rest is retained on the surface of the vegetation. The fraction of deposited materials intercepted and initially retained (i.e. not immediately blown or washed off) by vegetation is referred to as the interception fraction f. Chamberlain [1] provided a relationship between the interception fraction f and the standing biomass B.

$$f = 1 - e^{-(\mu B)} \tag{1}$$

where

B = above ground biomass (dry weight) of vegetation per unit area (kg m^{-2})
μ = absorption coefficient (m^2 kg^{-1}).

To account for the variation of f with different values of B, some authors analyse results in terms of μ, others in terms of a mass interception factor f/B. If f is less than about 0.3, there is little practical difference between f/B and μ (e.g., f = 0.3, f/B = 0.3/B, $e^{-\mu B}$ = 0.7, μ = 0.36/B). According to Chamberlain, when f approaches unity, as may happen when B is large, it is more appropriate to use μ.

Miller [2] showed that the spread of published values of f/B (or equivalently, μ) is smaller than that of f. He also concluded that the use of Equation (1) for pasture grass was well established, but that there were insufficient data for its validation for other crops. There may be some reason to doubt the applicability of Equation (1) for crops such as cabbage where the exposed leaf area does not increase in proportion to biomass as the crop matures. Pinder et al. [3, 4], however, have shown that the model that best described observations for corn, wheat, soybeans and cabbage crops was the one model in which f was adjusted for changing B during the growing season, according to Equation (1):

$$f/B = \frac{1 - e^{-\mu B}}{B} \tag{2}$$

where f/B = mass interception factor.

Experimental results for the mass interception factors are described in the following sections. The measurements of the total deposition as well as the fraction of activity retained on plants are used to derive mass interception factors. Such dry or wet deposits were applied artificially or released during nuclear bomb tests. Many data in the following compilation and discussion are taken from a recent review by Chamberlain and Garland [5].

2.1.2. Interception of dry-deposited materials

Interception of nuclear bomb debris

Some of the earliest measurements of interception were made during nuclear weapons trials. At Maralinga, Russell and Possingham [6] exposed trays of vegetation at various distances from the site of an explosion. After the test, the activity per kg dry weight in the vegetation was compared with the activity found in fallout collectors. The interception fraction f was found to vary from 0.03 to 0.15, and the mass interception factor f/B from 0.13 to 0.66 m^2 kg^{-1}. Both f and f/B were inversely correlated with the total deposited activity. This was attributed to a decrease in particle size with distance from the test centre. The particle size was not measured but, for the most part, was probably greater than 50 μm and may have been of the order of millimetres at close range.

Following ground-level explosions at the Nevada Test Site in 1955 and 1957, Romney et al. [7] measured fallout and contamination of herbage at distances varying from 11 to 420 km. The same effect of distance was seen, with f/B increasing from 0.01 close in to 0.4 m^2 kg^{-1} at a distance of 420 km; again this was attributed to reduction of particle size with distance. Comparisons of different plant species (wheat, clover, alfalfa) showed differences in f/B of not more than a factor of 2. In all the measurements of Russell and Possingham and of Romney et al., f was less than 0.2, and so f/B can be taken as equivalent to μ.

Based on an analysis of the literature, Simon [8] developed a simple model describing the relationship between the mass interception factor and the distance of the site of measurement to the site of the detonation. He also found increasing values for f/B for increasing distances, reflecting the influence of the particle size on the initial retention.

Martin [9] used autoradiographs and micrographs to study the fallout particles on leaves of desert shrubs after the Propit Sedan test at Nevada. Most of the retained activity was associated with particles of less than 5 μm in diameter.

In a review paper, Russell [10] concluded from investigations at nuclear weapons trials that particles which exceed 45 μm in diameter were seldom retained on leaves. These particles might rebound from surfaces and reach the basal parts of plants or the soil surface. If the particles are smaller, though sufficiently large to fall freely under gravity, they will lodge more readily on plant surfaces. The form of the vegetation seems to have a large effect on the extent to which it retains particles. In pastures, the greatest accumulation often occurs in the basal tissues which are below the levels to which most herbivores graze. This is also confirmed by the experiment of Eriksson [11]. However, the investigation of Peters and Witherspoon [12] gives no indication for a dependence of μm on the particle size. The reasons for this obvious discrepancy are not known.

TABLE I. ABSORPTION COEFFICIENTS (μ) OBTAINED FROM FIELD EXPERIMENTS

Deposited Material	Diam. μm	Crop	μ ($m^2\ kg^{-1}$ dry mass) Mean	SE	References
Lycopodium	32	Grass	3.08	0.15	[1]
Lycopodium	32	Wheat, dry	3.2	0.5	[13]
Lycopodium	32	Wheat, moist	9.6	3.7	[13]
Quartz	44-88	Grass	2.7	0.3	[12]
Silica sand	40-63	Grass, dry	0.44	0.15	[11]
Silica sand	40-63	Grass, wet[a]	0.88	0.13	[11]
Silica sand	63-100	Grass, dry	0.23	0.07	[11]
Silica sand	63-100	Grass, wet[a]	0.69	0.16	[11]
Silica sand	100-200	Grass, dry	0.24	0.07	[11]
Silica sand	100-200	Grass, wet[a]	0.46	0.11	[11]
^{238}Pu	~1	Corn	3.6	0.05	[3]
^{131}I	Vapour	Grass	2.8	0.14	[1]
^{212}Pb	Vapour	Artificial leaves	13	-	[14]

[a]Vegetation sprayed with water before experiment.

Field experiments with dry-deposited particles

Chamberlain [1] analysed the results of field experiments in which *Lycopodium* spores (32 μm), labelled with radioiodine, were dispersed over grassland at Harwell. At sampling points downwind of the release, the activities on grass and on the underlying mat and soil were measured and the fractional interception related to the herbage density. The results in terms of μ are shown in Table I.

Subsequently, Chadwick and Chamberlain [13] did further field experiments at Harwell and at Rothamsted in which labelled *Lycopodium* spores were dispersed over wheat crops. In 4 experiments, the crop in the sampling area was moistened with a water spray just before the spores were released. It was known from previous wind tunnel experiments that capture of particles by leaves is enhanced when the leaf surface is made wet or sticky. Only the wet weight of the crop was recorded. Values of μ have been calculated assuming a wet/dry weight ratio of 4. The values of μ for the dry wheat crop were similar to those previously obtained for grass (Table I). With the wetted crop, μ was variable, but was apparently enhanced by a factor of about 3 relative to the values obtained for the dry crop.

In the Harwell experiments, *Lycopodium* particles were dispersed by the wind, and the sampling areas were about 20 m downwind of the point of release. Witherspoon and Taylor [15] and Peters and Witherspoon [12] at Oak Ridge National Laboratory used a fertilizer spreader running on an overhead gantry to apply particles to crops. Witherspoon and Taylor used quartz particles labelled with ^{86}Rb, in two size ranges (48-88 μm and 88-175 μm), and applied them to crops of squash, soybeans, sorghum, lespedeza (clover), and peanuts. The results were somewhat variable, with f as measured exceeding unity in some cases. In all crops except soybeans, the average value of f was greater for the smaller particles.

Peters and Witherspoon [12] applied 48-88 μm quartz particles to four species of pasture grass. Values of the absorption coefficient, μ, ranging from 2.0 to 3.3 $m^2\ kg^{-1}$, with a mean of 2.7 $m^2\ kg^{-1}$, can be deduced from their results (Table I).

Eriksson [11] used a rotary device to apply silica sand particles to grass. The particles were in three size ranges (43-63, 63-100 and 100-200 μm) and were labelled with adsorbed fission products. They were applied to pasture grass with biomass varying from 0.05 to 0.3 kg m^{-2} dry weight. In some experiments the grass was lightly wetted by spraying an unspecified amount of water. Values of μ deduced from Eriksson's results are shown in Table I. Interception on wet grass was 2 to 3 times more efficient than on dry grass. Furthermore, μ declined as particle size increased. In one additional experiment, the grass was more thoroughly wetted by rain, and the interception of particles of all diameters was increased further.

Miller [16] measured the interception of particles emitted from a volcano on the foliage of several plant species. He associated higher interception with relative humidities greater than 90%, presumably due to films of water condensing on either the particles or the leaves.

Interception of plutonium-bearing particles

Pinder et al. [3] measured the interception of ^{238}Pu by corn plants at the Savannah River Ecology Laboratory. The plutonium came from a stack at the nearby nuclear separation facility; it was reported to be in particles, about 1 μm Average Mass Aerodynamic Diameter (AMAD), consisting of agglomerates of soil with Pu metal. Corn plants grown in pots, surrounded by a guard ring of similar plants, were exposed to the fallout for a week. Plants in varying stages of growth were used to give a variation in biomass per unit area. The ^{238}Pu activity per kg dry weight for the plants after exposure was compared with the activity on horizontal tacky papers exposed nearby.

Pinder et al. [3] found that the results fit the exponential model of interception, with a mean μ equal to 3.60 $m^2\ kg^{-1}$ (standard error, SE = 0.05), which is similar to that derived from the British experiments with *Lycopodium* spores (Table I).

Pinder et al. [17] assessed the plutonium inventory of different crops due to a mixture of dry and wet Pu deposits. In this work alternative estimates for the absorption coefficient μ and the weathering removal rate λ_w are applied, ranging from μ = 2.2 $m^2\ kg^{-1}$ to μ = 3.6 $m^2\ kg^{-1}$ and from λ_w = 0.025 d^{-1} to λ_w = 0.065 d^{-1}. Despite the range in μ and λ_w, the predicted inventories were in general similar to the observed inventories.

Pinder et al. [18] investigated the interception and retention of Pu-particles on orange trees and suggested an influence of the leaf characteristics on the absorption coefficient μ. He concluded that the interception of Pu particles on plant species with waxy surfaces might be lower than for other plant species. However, Pinder determined the interception during continuous depositions of plutonium. Therefore this observation could also be caused by a fast initial weathering of Pu on the orange leaves due to the pronounced hydrophobic character of these leaf surfaces.

Interception of elemental vapours

Chamberlain [1] analysed the results of experiments in which ^{131}I vapour was released over grassland at Harwell. The interception fraction was found to be consistent with μ = 2.78 $\pm$ (SE) 0.14 $m^2\ kg^{-1}$.

Chamberlain [14] studied deposition in a wind tunnel of ^{212}Pb vapour, for which all surfaces act as perfect sinks. An artificial grass surface made of paper-covered spills was used. This had a leaf area index of 2.1. (The leaf area index is the ratio of the area of one side of the leaves to the soil area under the plant canopy). Taking the mass of a real grass or cereal leaf as 35 g m^{-2} dry weight, the corresponding value of B would be 0.075 kg m^{-2}. At moderate wind speeds (friction velocity = 0.3 m s^{-1}), about 96% of the deposited ^{212}Pb was on the artificial leaves and only 4% reached the substrate; this would correspond to a value of about 13 m^2 kg^{-1} for μ.

2.1.3. Interception of wet-deposited materials

Experiments employing fine sprays and low amounts of precipitation

Milbourn and Taylor [19] used a modified agricultural spraying machine to apply ^{89}Sr in solution to pasture at 8 sites (Table II). Herbage densities varied from 0.058 to 0.23 kg m^{-2} dry weight. Two low-volume applications were made, separated by a 10 minute interval, and each gave a precipitation of 0.085 mm water. The applied amount of ^{89}Sr was known from the strength of the solution and the volume applied per unit area. Herbage was sampled after spraying, and the fractional interception was calculated. In the 8 experiments, f varied from 0.17 to 0.30. Values of μ calculated from the results averaged 3.33 $\pm$ 0.56 (SE) m^2 kg^{-1} dry mass, similar to some of the values obtained with the dry deposition of particles (Table I).

Aarkrog [20] sprayed a mixed solution containing isotopes of Sr, Cs, Mn and Ce onto small plots of rye, barley, oats and two varieties of wheat, at four stages of crop development. One of the wheats carried awns (Table II). Expressed in terms of Equation (1), the values for μ decrease as the crops mature. This decrease of the mass interception factor is probably due to the reduction of the leaf area index during the maturing process, while the above ground biomass still increases. Aarkrog and Lippert [21] applied a different range of elements (Cr, Fe, Co, Zn, Hg and Pb) to barley. Plants were sprayed at different stages of growth. Interception was not much affected when awns were removed from rye, barley and wheat before spraying. For mercury, f was lower than for the other elements in the second group, possibly because of evaporation, but otherwise differences between elements were small. Changes in morphology of the plants prevent the fitting of Equation (1) to the sequence of data for each crop, but for barley, Aarkrog and Lippert found that the data fitted

$$f = 1 - e^{-0.31\,B/h} \qquad (3)$$

where B was the dry biomass in grams per 40 cm diameter plot and h the height of the crop.

Eriksson [11] applied ^{59}Fe, ^{85}Sr, and ^{137}Cs in solution, and silica sand particles (40-63 μm and 100-200 μm) in suspension, as a spray with simulated precipitation of 0.1 mm. For ions in solution, μ was very similar to the results of Milbourn and Taylor [19]. Eriksson suggested that the results showed an effect of valency, with interception increasing in the order $^{137}Cs^{+} < ^{85}Sr^{++} < ^{59}Fe^{+++}$, but it is doubtful if the differences were significant. With particles in suspension μ was 1.45 $\pm$ 0.12 (SE) (40-63 μm particles) and 1.01 $\pm$ 0.31 (SE) m^2 kg^{-1} (100-200 μm particles). However, it should be kept in mind that the amounts of water applied in the experiments of Aarkrog and Eriksson are too low to be considered as typical for real situations with wet deposition.

TABLE II. ABSORPTION COEFFICIENT μ OBTAINED FROM EXPERIMENTS APPLYING FINE SPRAYS AND LOW AMOUNTS OF WATER

Experiment	μ ($m^2 . kg^{-1}$)	Biomass ($kg . m^{-2}$)	References
Spray on pasture containing ^{85}Sr (0.17 mm)	3.3 ± 0.56	0.06–0.23	[19]
Spray on rye with a mixture of isotopes (0.08 mm)	1.2 (71 d to harvest) 0.73 (62 d to harvest) 0.3 (35 d to harvest)	1.4 (71 d to harvest) 1.2 (62 d to harvest) 2.4 (35 d to harvest)	[20]
Spray on oats with a mixture of isotopes (0.08 mm)	1.7 (71 d to harvest) 1.6 (62 d to harvest) 0.32 (35 d to harvest)	0.57 (71 d to harvest) 0.6 (62 d to harvest) 1.8 (35 d to harvest)	[20]
Spray on barley with a mixture of isotopes (0.08 mm)	2.5 (55 d to harvest) 2.0 (42 d to harvest) 0.12 (15 d to harvest)	1.1 (51 d to harvest) 1.1 (42 d to harvest) 2.4 (15 d to harvest)	[20]
Spray on wheat without awns with a mixture of isotopes (0.08 mm)	2.1 (71 d to harvest) 1.7 (62 d to harvest) 0.57 (35 d to harvest)	0.32 (71 d to harvest) 0.39 (62 d to harvest) 0.89 (35 d to harvest)	[20]
Spray on wheat with awns with a mixture of isotopes (0.08 mm)	3.7 (71 d to harvest) 1.0 (62 d to harvest) 0.8 (35 d to harvest)	0.42 (71 d to harvest) 0.42 (62 d to harvest) 0.98 (35 d to harvest)	[20]
Spray with ^{59}Fe, ^{89}Sr, ^{137}Cs in solution on grass (0.1 mm)	1.9–3.4	0.05–0.19	[11]
Spray with suspended ^{59}Fe, ^{89}Sr, ^{137}Cs particles (40–63 μm)	1.45 ± 0.12	0.04–0.19	[11]
Spray with suspended ^{59}Fe, ^{89}Sr, ^{137}Cs particles (100–200 μm)	1.01 ± 0.31		[11]
Spray of TcO_4^{2-} on pasture (1 mm)	1.1 ± 0.05[a]		[22]

[a]In this experiment f/B was calculated instead of μ. According to Equation (1), the values for f/B and μ are very similar for $B < 0.3$ kg m^{-2} (dry mass).

Hoffman et al. [22] reported data for the interception of technetium, applied as pertechnetate during a simulated rain of 1 mm to a canopy of fescue. Values of f varied with crop density and were in the range of 0.079 to 0.166; f/B was 1.07 ± 0.05 (SE) m^2 kg^{-1}.

Experiments using simulated rain

Hoffman et al. [23, 24] carried out an extensive series of measurements at Oak Ridge National Laboratory with activity applied in simulated rain. The amounts of rain varied from 1 to 30 mm — much greater than the amounts used by Milbourn and Taylor [19]. The intensity at which the rain was applied was high, varying from 14 to 120 mm h^{-1}. The following materials were used:

(a) Polystyrene microspheres of 3, 9 or 25 μm diameter labelled respectively with ^{141}Ce, ^{95}Nb and ^{85}Sr.
(b) ^{7}Be in solution as $BeCl_2$.
(c) ^{131}I in solution as IO_4^- or I^-.

The activity was applied to plots of clover, fescue and mixed old field vegetation. Only slight differences were found in the results for the different herbage species. The field measurements of f/B carried out by Hoffman et al. [23, 24] are summarized in Figs 1–3. The measured values show strong differences according to chemical form (anions vs. cations). The f/B values for iodine (applied as I^- or IO_4^- are inversely correlated to the total amount of rainfall. The f/B values for ^{7}Be and the insoluble particles also decreased with increasing rainfall, but the dependence was less pronounced than for iodine. The rainfall intensity had very little effect on the f/B values.

Hoffman et al. [23, 24] gave results in terms of both f and f/B, and statistical analyses were performed on the log-transformed results to evaluate the variation with total applied rainfall, rainfall intensity and herbage biomass. The reduction in f/B as rainfall increased was statistically significant in all cases, but was most marked for ^{131}I (Fig. 1). Hoffman et al. [23, 24] also reported laboratory experiments showing that fescue leaves absorbed ^{7}Be and ^{137}Cs from solution much more strongly than ^{131}I.

Recalculated in terms of μ, the results for 1 mm rainfall in Figs 1, 2 and 3 range from about 2.8 for ^{131}I to 4.5 for ^{7}Be, assuming an average biomass of 0.15 kg m^{-2}. It appears, therefore, that the results of Hoffman et al. [23, 24] for ions in solution deposited in 1 mm rainfall are not greatly dissimilar from those of Milbourn and Taylor, who applied about 5 times less water. For particles in suspension, the values of μ obtained by Hoffman et al. [23, 24] are greater than those reported by Eriksson, but the particle size was substantially smaller.

In an experiment in which intermittent rain was applied in amounts ranging from 15 to 75 mm, values of f/B were relatively constant with respect to rain amount, although they were significantly lower for ^{131}I than for ^{7}Be and the three size classes of insoluble particles (Fig. 4). This observation implies that once initial retention occurs, subsequent rain is ineffective in removing previously deposited materials.

In a subsequent study (Hoffman et al., [25]), a wider range of plants was exposed to wet-deposited activity (Fig. 5). Again, inter-species differences were small (around a factor of 4 in range) in comparison to an order of magnitude variation observed among different chemical forms. Inter-species differences were further reduced when the data were normalized for leaf surface area (Fig. 6). The normalization of the interception fraction is obtained by dividing the deposition per unit leaf area surface by the total amount applied per unit ground area (the leaf area interception fraction, LAIF). Values of f/B for 8.5 mm simulated rain ranged from 0.15 to 0.6 for I^- and $SO_4^=$ and 0.6 to 3 for Cr^{+++}, Sr^{++}, Cd^{++}, Be^{++}, and 9-μm particles. The lower values for I^- and $SO_4^=$ are presumed to be due to the predominantly negative charge of leaf surfaces. [24, 26, 27]. These results also confirm the observations made by Eriksson [11].

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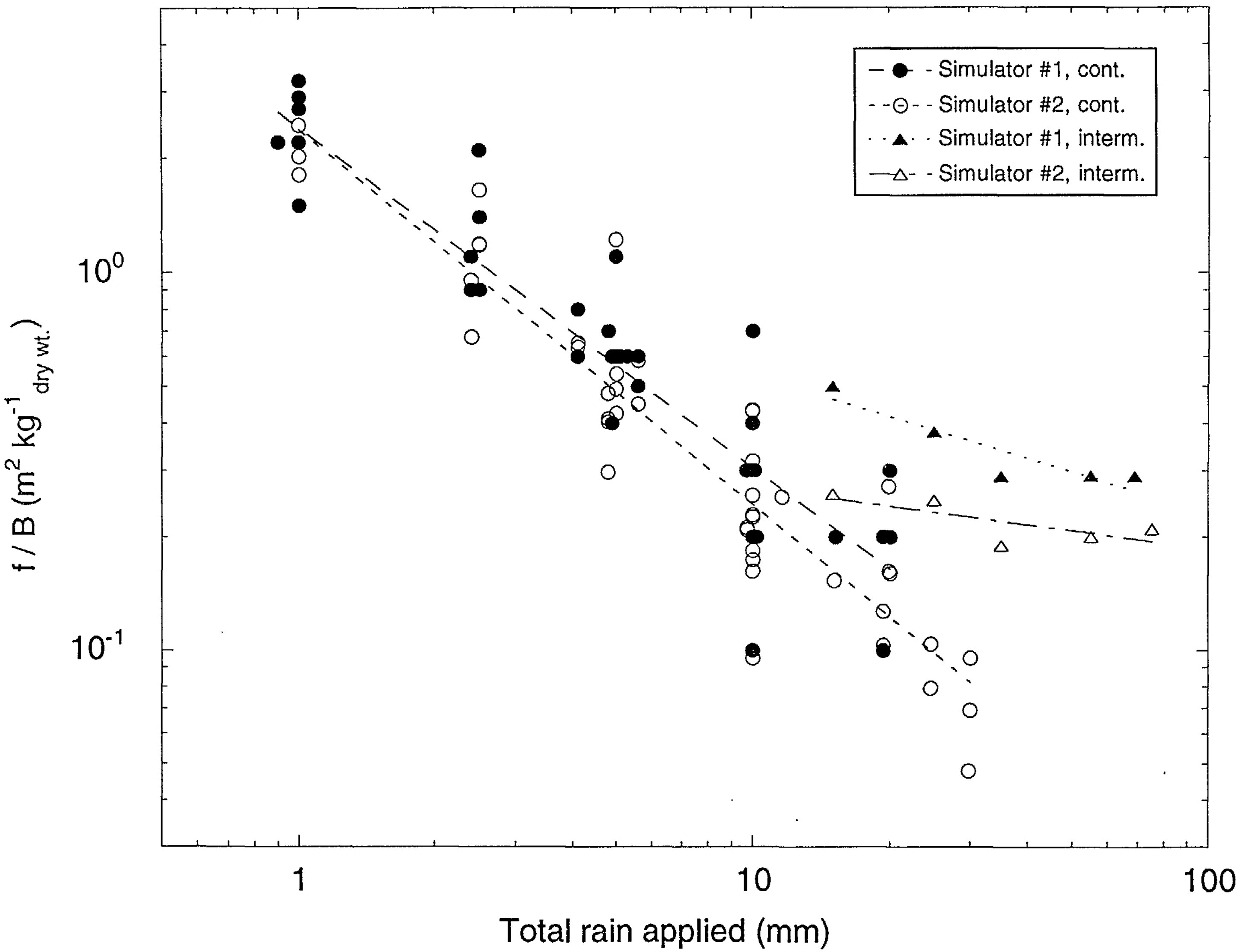

FIG. 1. Experimental values of the mass interception factor f/B (m^2 kg^{-1} dry) for ^{131}I from continuous and intermittent applications of simulated rain. The dashed lines represent the best fit to the data using a power function, $y = ax^b$ [24].

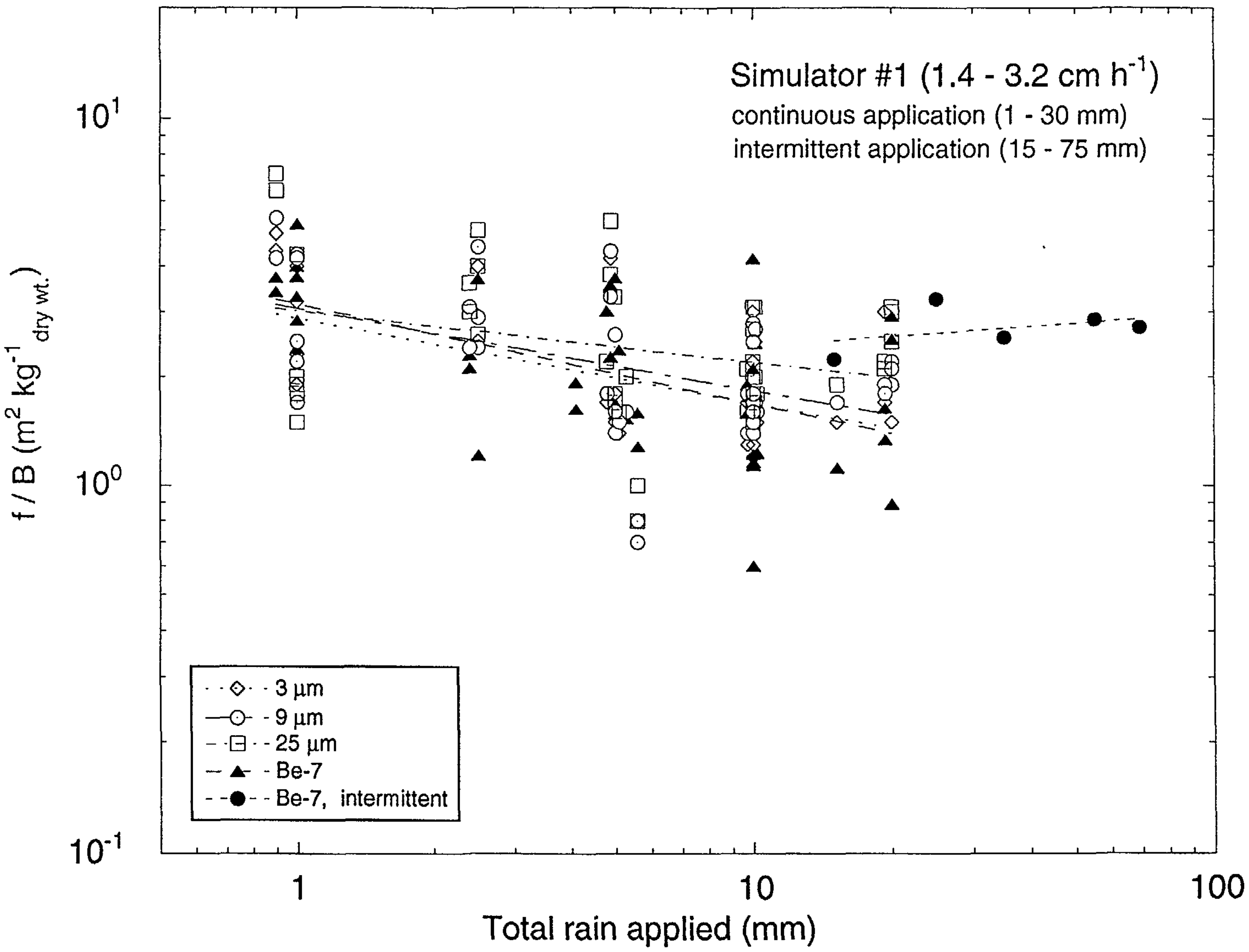

FIG. 2. Experimental values of the mass interception factor f/B (m^2 kg^{-1} dry) for ^7Be and the insoluble microspheres from continuous and intermittent (^7Be only) applications of moderate-intensity simulated rain. The dashed lines represent the best fit to the data using a power function, $y = ax^b$ [24].

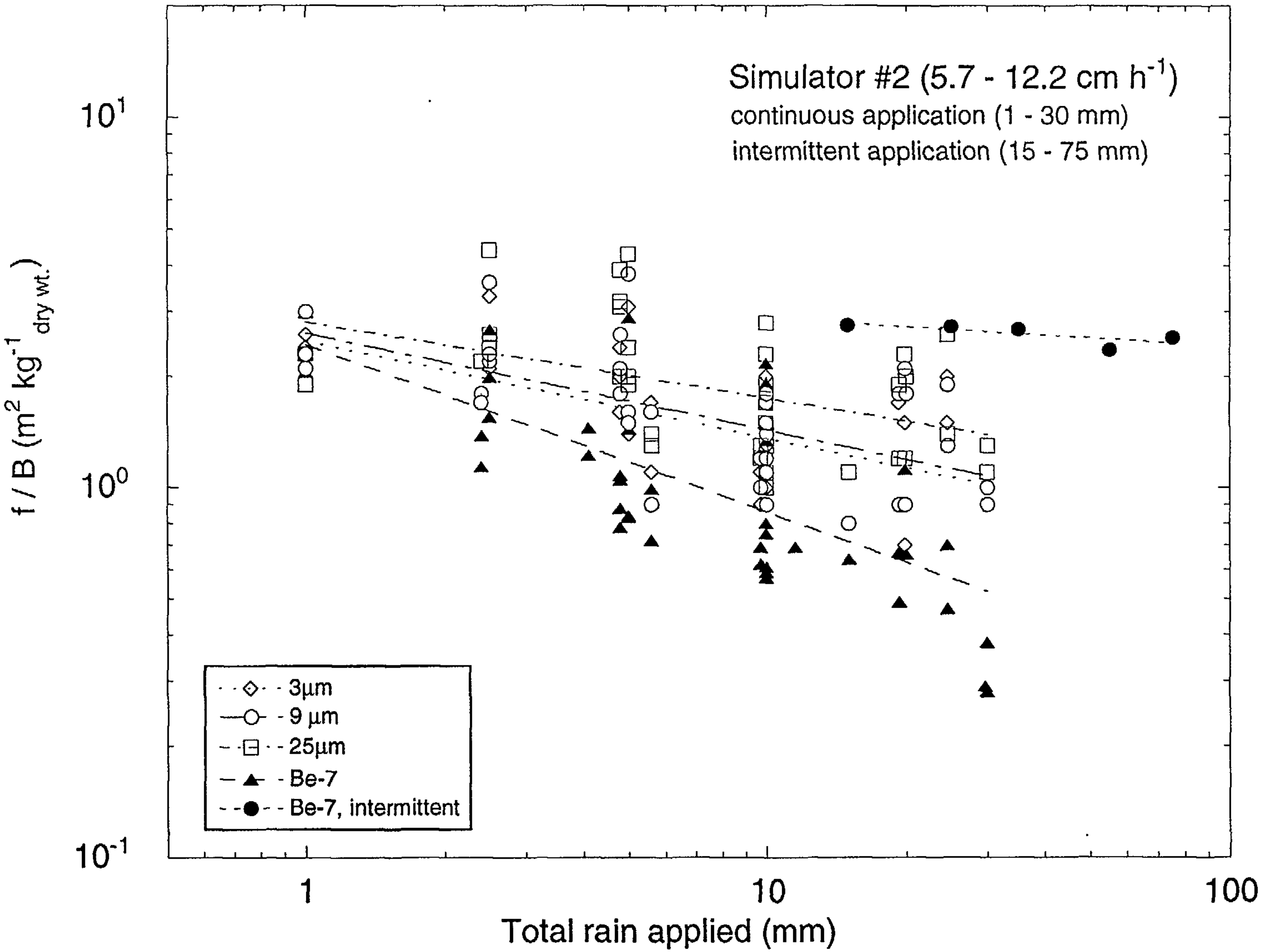

FIG. 3. Experimental values of the mass interception factor f/B (m^2 kg^{-1} dry) for ^7Be and the microspheres from continuous and intermittent (^7Be only) applications of high-intensity simulated rain. The dashed lines represent the best fit to the data using a power function, $y = ax^b$ [24].

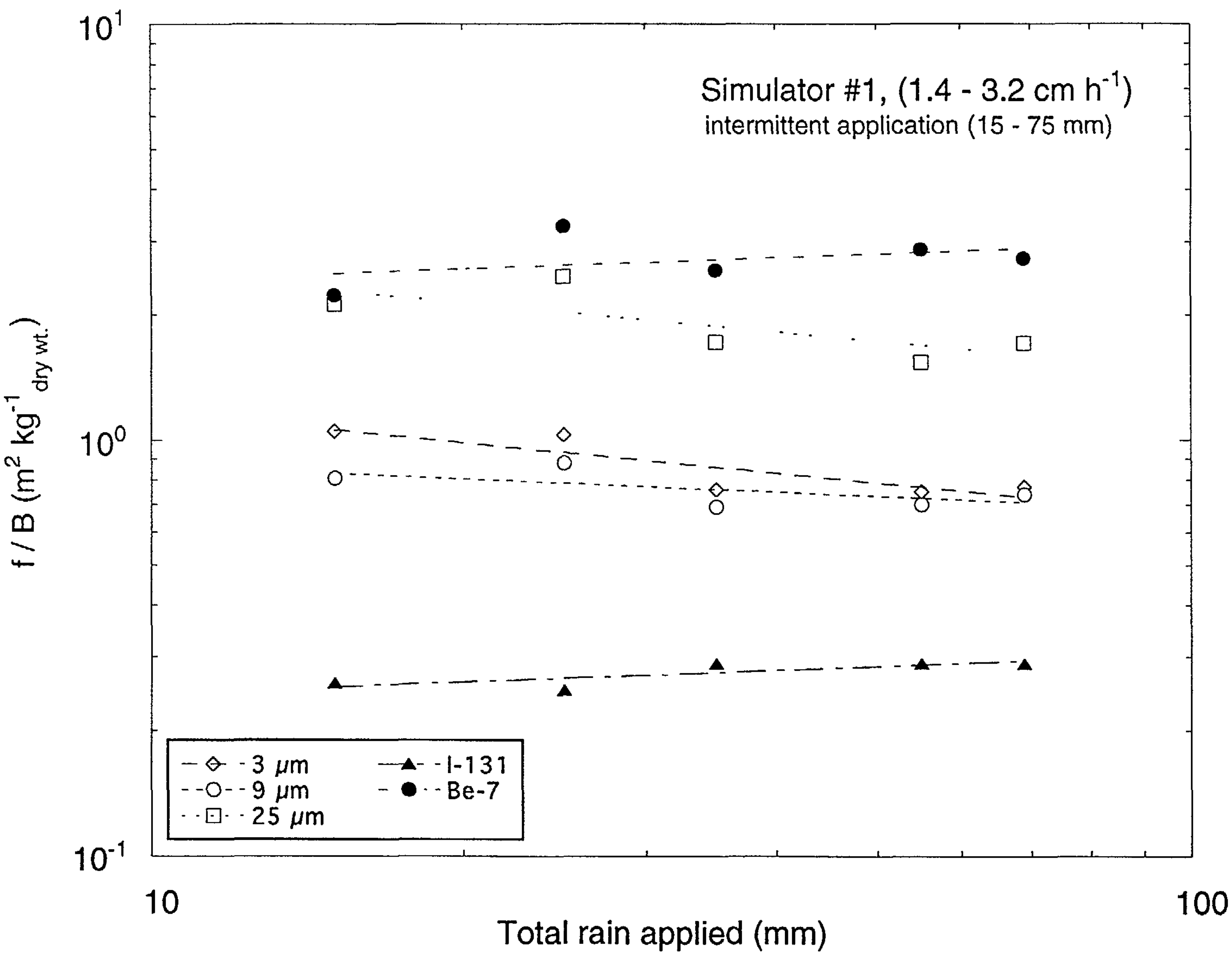

FIG. 4. Individual values of f/B for composite samples of fescue receiving intermittent rain applications [23].

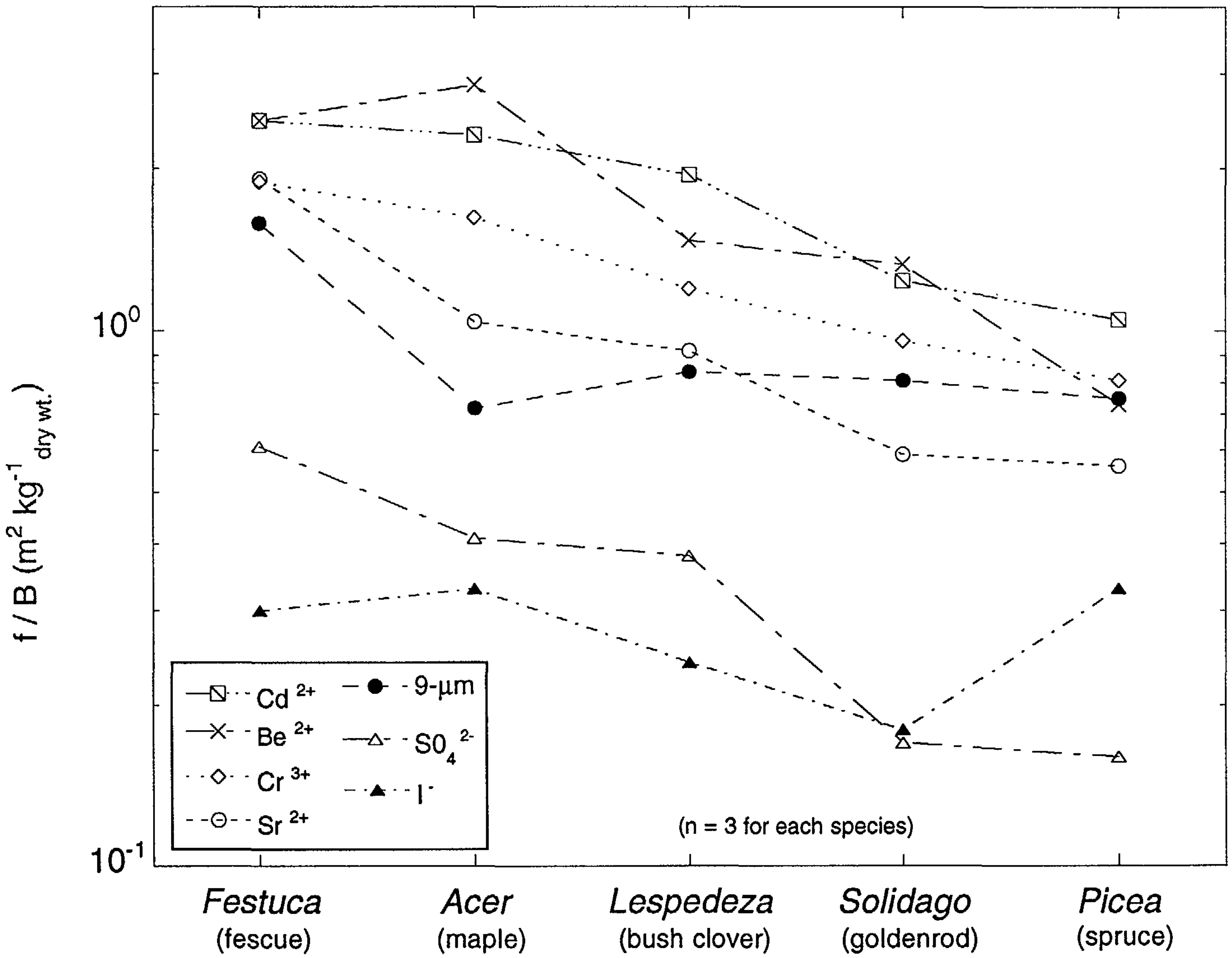

FIG. 5. Mean values of mass interception factors for five different plant types for radioactively labelled cations, anions, and one size-class of insoluble microsphere under controlled laboratory conditions. The amount of rainfall applied was 8.5 mm, the mean droplet size 1.7 mm in diameter, and the overall uncertainty less than 30% [25].

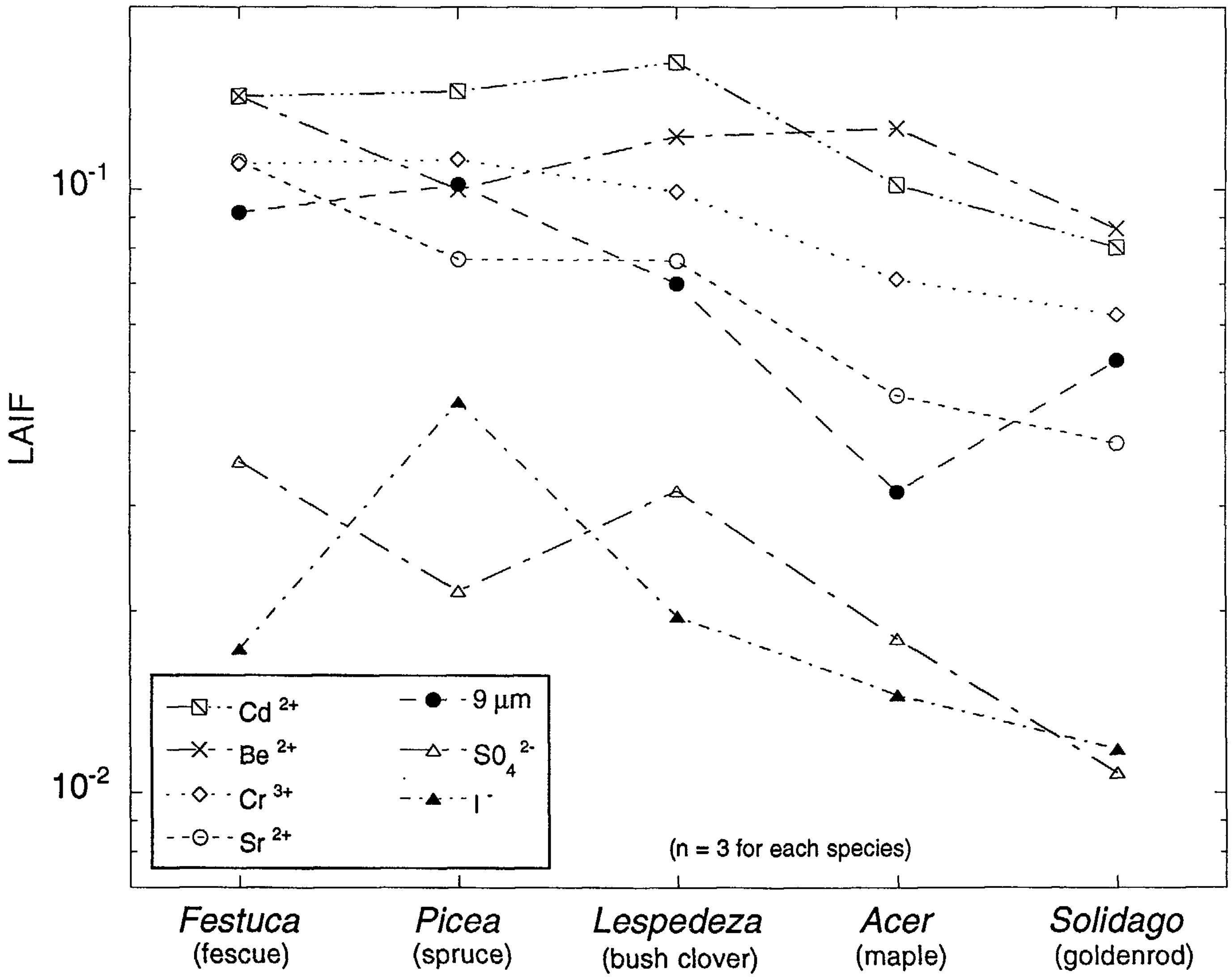

FIG. 6. Mean values of the Leaf Area Interception Fraction (LAIF interception fraction normalized to the leaf area) for five different plant types for radioactively labelled cations, anions, and one size-class of insoluble microsphere [25].

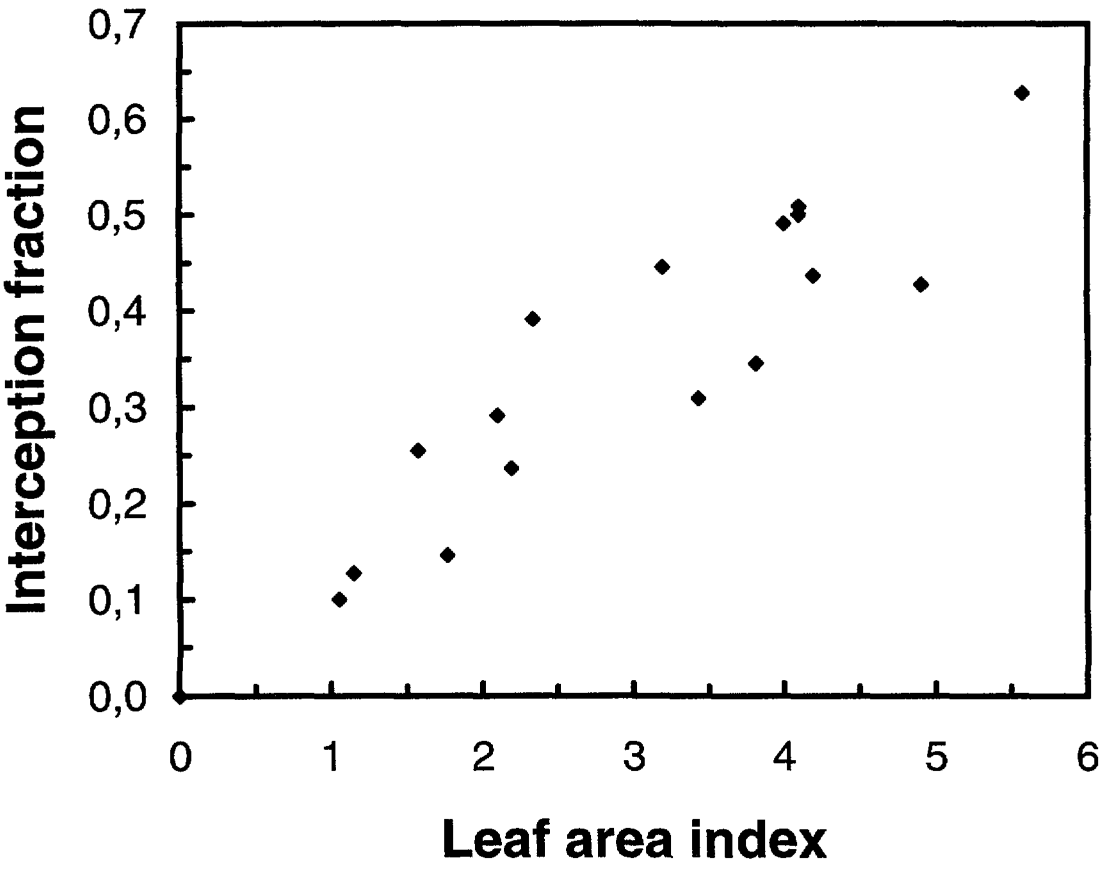

FIG. 7. Interception fraction for caesium deposited during an artificial rainfall of 1 mm as a function of the leaf area per unit area of ground [31].

Values of f/B measured by Angeletti and Levi [28] and Angeletti [29] are summarized in Table III. The values for iodine are in very good agreement with those measured by Hoffman et al. [23, 24]. The f/B values for strontium, however, are in the upper range of the values for ^{7}Be measured by Hoffman et al. Again, it is obvious that the mass interception factor f/B decreases with increasing amounts of precipitation during a rain (or precipitation) event. It is interesting to note that the mass interception factors of I^{-} are much smaller than those of Sr^{2+}. It is also obvious that the decrease of the mass interception factor for strontium with increasing rainfall is much less pronounced than for iodine; this might be due to active absorption of strontium by the leaves during the contact of the contaminated water with the leaves of the plants. These observations agree with results reported by Hoffman et al. [23, 24]; they can be explained by the fact that the plant cuticle acts as a cation-exchanger at pH-values above 3 [26, 30], which results in a strong absorption of cations and an inhibited absorption of anions. The data of Angeletti and Levi [28] seem to indicate higher mass interception factors for clover than for rye-grass. This could be due to a lower specific mass per leaf-area (expressed in terms kg m^{-2} leaves) or to the more horizontal orientation of the leaves of clover.

TABLE III. SUMMARY OF MASS INTERCEPTION FACTORS FOR I AND Sr FOR GRASS AND CLOVER MEASURED BY ANGELETTI AND LEVI [28] AND ANGELETTI [29]

Precipitation (mm)	f/B $m^2\ kg^{-1}$ dry mass					
	Grass			Clover		
	I^-	H_2O^a	Sr^{2+}	I^-	H_2O^a	Sr^{2+}
1	4.3	6.2	7.6	8.7	11.1	8.2
2	1.6	4.3	5.1	4.1	5.9	8.0
4	1.1	1.8	4.8	2.5	4.0	8.2
8-12	0.6	1.2	4.2	1.0	2.2	8.6
16-22	0.27	0.45	1.3	0.44	0.9	4.3

[a]The interception of water was measured indirectly as the difference between the total amount of irrigation water and the water content of the soil subsequent to the irrigation.

In the experiments of Voigt et al. [31], the interception of ^{137}Cs on cereals was determined. The Cs was applied with artificial rain of 1 mm. In Fig. 7 the measured interception fractions of the experiments are plotted against the leaf area index of the plants. As with the results of Hoffman et al. [24] (Fig. 6), this figure underlines the importance of the leaf area in the interception of wet-deposited radionuclides. A similar relationship to the leaf area index was observed by Pinder et al. [3] for dry-deposited plutonium particles on some plants.

2.1.4. Interception of Chernobyl fallout

Direct estimates of wet and dry interception of Chernobyl fallout by vegetation were difficult to make because of several complications. First, the total flux to the ground surface due to dry deposition is difficult to measure accurately. Although wet deposition occurred intermittently at most sites, dry deposition was continuous at all sites and was affected by the surface roughness created by the presence of vegetation. Dry deposition to ground covered with vegetation may be substantially higher than deposition collected by flat plates or funnel collectors (e.g. Jonas, [32]). The total amount of deposition to the ground surface could be estimated for ^{137}Cs during periods of rain as wet deposition typically dominated over dry deposition and is effectively sampled using funnel collectors. However, for ^{131}I, the amount of dry deposition to vegetation may have been substantial, and even enhanced, during periods of wet deposition. In central parts of Europe, the presence of elemental iodine vapour made up a fraction of 15 to 35% of the total amount of ^{131}I in air. Iodine vapour is readily deposited to the wet surfaces of leaves [33]. Furthermore, the measurements of vegetation were seldom made immediately after the period of initial deposition, and it is not easy to determine the amount of material intercepted that is due specifically to wet versus dry processes.

A dependence on chemical species of the interception of wet-deposited activity is reported by Jacob et al. [34] in the results of measurements collected after the first rainfall event (on 1st May 1986) following the beginning of the Chernobyl cloud in Munich. Taking into account the contamination of grass due to dry deposition, the mass interception factor f/B for ^{131}I was about 0.7, for ^{137}Cs about 1.0, and for ^{140}Ba (which should behave similarly to Sr) about 1.6. The estimated biomass of the grass was about 0.1 kg m^2 dry weight. The rainfall for this event was about 5-6 mm.

Winkelmann et al. [35] measured various radioisotopes of Cs, I, Ru and Ba in grass near Munich on 5th May 1986. The activities were given as Bq kg^{-1} fresh weight; they are converted to Bq kg^{-1} dry weight in Table IV assuming a dry matter content of 20%. The activity in soil was measured on 1st June 1986. With corrections for radioactive decay, the values of f/B obtained are in the range 0.28 to 0.58 m^2 kg^{-1}. However, in the Munich area, the main deposition from the passage of the Chernobyl cloud occurred on the 30th of April and 1st of May. Therefore, a part of the activity initially intercepted by the grass was already weathered off on the 5th of May, which results in lower mass interception factors than reported by Jacob et al. [34]. This weathering period implies that the f/B values given in Table IV are lower than would have been found immediately after deposition occurred. About 6 mm of rain fell in the area during the passage of the cloud.

Köhler et al. [36] estimated average values of f/B for data obtained from Neuherberg, Germany; Petten, Netherlands; Tokai, Japan; Roskilde, Denmark; Budapest, Hungary; and Transvik, Sweden (Table V). These average values were obtained by correcting estimated time-integrated vegetation concentrations at each location for radioactive decay and field loss to estimate a theoretical concentration, and dividing by the amount of total deposition reported for these locations.

For all locations, f/B ranged from 0.94 to 1.89 m^2 kg^{-1} for ^{131}I and from 0.46 to 0.97 m^2 kg^{-1} for ^{137}Cs (Table V). The values observed at the Neuherberg site were slightly more than two times the values calculated by Chamberlain [37] from the data reported for the same location by Winkelmann et. al [35], but the relative difference between ^{131}I and ^{137}Cs was approximately the same. A portion of this difference is due to the assumption of the ratio of wet to dry weight, but most of this discrepancy is attributed to the methods used to derive f/B from the measured data.

Martin et al. [38] measured the activity of ^{137}Cs in dried grass gathered at weekly intervals from 16th May 1986 onwards from a field near Aberdeen. Extrapolating backwards, it can be estimated that the activity on 3rd May, when the Chernobyl activity arrived in the area, was about 0.7 kBq kg^{-1}. The fallout of ^{137}Cs, measured at a site about 1 km from the field, was 2.7 kBq m^{-2}, most of it falling during a rainstorm of 5.8 mm on 3rd May. Hence, from these data, f/B = 0.7/2.7 = 0.26 m^2 kg^{-1}.

TABLE IV. INTERCEPTION OF FALLOUT FROM CHERNOBYL AT MUNICH [35]

Nuclide	Activity in grass on 5 May 1986 Bq kg^{-1} dry weight[a]	Activity in soil on 1 June 1986 (kBq m^{-2})	Activity in soil on 5 May 1986 (kBq m^{-2})	f/B m^2 kg^{-1}
^{103}Ru	5 000	11	18	0.28
^{131}I	21 000	5.6	57	0.37
^{134}Cs	3 400	8.9	9.1	0.37
^{137}Cs	7 000	16	16	0.44
^{140}Ba	4 300	1.7	7.4	0.58

[a]Assuming a dry matter content of 20%.

TABLE V. DERIVED AVERAGE MASS INTERCEPTION FACTORS (m^2 kg^{-1}) FROM CHERNOBYL FALLOUT DATA

Site	Iodine f/B	Caesium f/B
Neuherberg [36]	0.94	0.97
Petten [36]	0.97	0.60
Tokai [36]	1.05	0.46
Roskilde [36]	1.77	0.90
Budapest [36]	1.89	-
Transvik [36]	-	0.81
Gomel region [41]	-	1.2
St. Petersburg [42]	1.6	1.0

Fulker [39] measured the activities of ^{131}I and ^{134}Cs in grass and soil from 4 farms near Sellafield, Cumberland, on 3rd May 1986. Only fresh weight was recorded, but, assuming a dry matter content of 20%, the mean f/B ratios were about 0.4 m^2 kg^{-1} for ^{131}I and 1.1 m^2 kg^{-1} for ^{137}Cs. About 9 mm of rain fell in the area during the passage of the activity from Chernobyl.

Mass interception factors of 3.0 m^2 kg^{-1} dry weight for iodine and 2.6 m^2 kg^{-1} dry weight for caesium (relative error 40% for both) have been estimated for Chernobyl fallout on pasture grass in Russia and Belarus [40]. These estimates are based on data of 28-30 April for the following regions: Bryansk (Novozybkov district), Gomel (Bragin, Khoiniki, and Narovlya districts), Cherkassk (Kanev district), Leningrad (Lomonosov district), Moscow (in the vicinity of Moscow). Derived average mass interception factors (m^2 kg^{-1} dry weight; Table V) by May 1st were 1.2 for caesium for the Gomel region of Belarus [41] and 1.6 for iodine and 1.0 for caesium for St. Petersburg [42]. Other estimates for Russia range from 0.18 to 0.46 m^2 kg^{-1} wet weight for ^{131}I and from 0.11 to 0.28 m^2 kg^{-1} wet weight for ^{137}Cs.

2.1.5. Review of modelling approaches for interception

The approaches for the modelling of the interception of dry deposits as well as of radionuclides deposited with precipitation have been analyzed for a number of models recently applied during the BIOMOVS I and the VAMP study [36, 44].

The underlying assumptions in many of the models are very similar. Starting from the total deposition, for the estimation of the initial contamination, an interception fraction f is applied. In many models the interception fraction f is constant, often set at 0.2 to 0.25; in other models, it is dependent on the biomass of the standing crop, the plant species considered, the specific chemical, or the season. The approach of Chamberlain [1] is applied frequently in order to estimate the interception fraction f as a function of the standing biomass.

Most models do not take into account wet deposition explicitly. In one case wet deposition is considered simply by applying a higher deposition velocity on days with precipitation, with the interception of the total deposition estimated using Chamberlain's equation. Another model assumes that the dry-deposited activity is totally intercepted by the plant canopy, whereas for the wet-deposited activity an interception factor of 0.1 is applied.

In the German model ECOSYS [45, 46], dry deposition on the plant leaves is distinguished from deposition to the soil under the plants. The interception of the wet-deposited activity is modelled applying a "water-film-leaf-area approach." The basic assumption is that the interception fraction is dependent on the water film on the foliage of the plants after a rainfall event. Additionally, an element-dependent factor is applied accounting for the different ability of the elements to be fixed or absorbed on the leaves. The water film on the leaves is assumed to be dependent on the plant species; due to the more horizontally exposed leaves for non-Gramineae, slightly higher values are applied compared to Gramineae, the leaves of which are more vertically oriented. The leaf area is assumed to be dependent on the specific growing cycle of the plant.

Interception of dry-deposited activity

In many experiments the deposition velocity is derived from the activity deposited on the surface of the plants and the time-integrated activity concentration in air during that period of deposition. This means that the activity deposited onto the soil under the plant canopy is not taken into account. However, interception fractions sometimes are applied to such deposition velocities. Such approaches are not consistent with the experiments, unless an increased deposition velocity is used to account for the additional deposition under the plant canopy. The application of such an approach may lead to a significant underestimation of the contamination due to dry deposition, especially if relatively low interception fractions are applied.

The successful application of interception fractions for dry deposition requires a very careful choice of parameters in order to compensate for the inherent errors in this approach. This method might be appropriate in models for routine releases, where long-term average parameter values can be applied, but it is potentially inaccurate for models dealing with the consequences after nuclear accidents.

In the model ECOSYS [45, 46] a constant deposition velocity is applied for the dry deposition to the soil under the plant canopy, in addition to the leaf-area-dependent deposition velocity used for the plant surface. Especially for high biomass densities, this might lead to a slight overestimation of the deposition to soil, because the efficiency of the protection of the soil from deposition increases with increasing crop densities.

Interception of wet-deposited radionuclides

In many models the wet-interception fraction is modelled using the total wet deposition and applying the same interception fractions as for dry deposition. This is a rough simplification of the ongoing processes, since factors such as the actual biomass or leaf area, the plant species and the element are not taken into consideration. It might be possible to use such approaches in models for routine releases. In such cases, it could be possible to apply generic interception fractions which are adequate for the precipitation pattern in the region considered.

However, in emergencies in which specific situations have to be evaluated quickly, such generic approaches might lead to underestimations of the plant contamination in the case of rainfall events with little rainfall, and to overestimations if heavy rainfalls occur.

Interception of total deposition

In many models dry and wet deposition are added, and a common interception fraction is applied for both contamination pathways. For this calculation method the same disadvantages have to be mentioned as in the previous sections. However, due to the lumping of both processes, the disadvantages are masked and it is probable that they compensate each other in certain situations. For example, in the data reported by Winkelmann et al. [35] (see Table IV), the f/B-values for caesium and iodine are very similar, although the deposition velocities as well as the interception fractions for wet deposition differ considerably. However, compared to caesium, the higher dry deposition of iodine compensates for the lower wet-interception fraction, resulting in similar values for f/B. It should be expected that the f/B-value for iodine would decrease with increasing rainfall, but the effect may be masked because the high dry deposition for elemental I_2 dominates the contamination.

2.1.6. Recommendations for model improvement

From the analysis of the processes of wet and dry deposition and interception, it is obvious that these are two separate processes which also should be simulated separately in radioecological models. However, in reality, the processes overlap and a clear separation is often not possible. This is especially true for long-term emissions such as the weapons fallout of the 1960s or the routine releases from nuclear facilities. In such cases, it should be possible, as is done in common practice, to lump dry and wet deposition and to apply a generic interception fraction which is empirically derived from observations. These generic factors may be sufficient because the interception fraction of wet-deposited activity is nearly unity for well developed canopies and low amounts of rainfall. Furthermore, in reality, precipitation falls intermittently even on days with high total amounts of rainfall. As the results of Hoffman et al. [23, 24] indicate, values of f/B remain nearly constant, if the plants can dry between the rainfall events. However, such simplified approaches are not appropriate for the modelling of deposition over short time periods. In such cases the contamination due to dry and wet deposition must be modelled separately, taking into account the specific amounts of rainfall.

For modelling of the contamination of plants, the following aspects should be taken into consideration:

Contamination of plants due to dry deposition

The experimental determination of dry deposition velocities often is based on the activity deposited on the surface of the plant canopies. Therefore, the application of an interception fraction or a mass interception factor for dry deposition is not consistent with the underlying experiments. Either this practice should be avoided, or the value of the deposition velocity should be increased to account for the total deposition to vegetation and underlying soil.

In some investigations the variability of the dry deposition was reduced by normalising Vg to the standing biomass [32, 33, 47]. In this case the correlation between the deposition velocity and the biomass is implicitly taken into consideration. If other approaches are applied, one has to be sure that the correlation between the deposition velocity and the biomass is adequately considered.

The normalization of the dry deposition to the biomass leads to reasonable results for pasture grass, leafy crops and other crops in the vegetative growing period. In these cases a good correlation between the biomass and the leaf area index can also be observed. However, as the development of crops such as grain, vegetables, and fruit continues, especially during the generative phase of the growth, the correlation between the leaf area index and the biomass is no longer valid (Fig. 8) [45]. In these cases, there is some doubt whether the normalization of the deposition velocity to the biomass is still justified.

However, there are few data concerning dry deposition in relation to the leaf area. Further investigations in this field, taking into account the particle size, the wind speed and the plant species, would be desirable for an improvement of the knowledge and understanding of dry deposition processes.

Interception of wet-deposited activity

As the experiments of Hoffman et al. [23, 24, 25] and the experiences after the Chernobyl accident have clearly demonstrated, the interception of wet-deposited activity is mainly controlled by the biomass or leaf area, the amount of rainfall, and by the chemical form. Aarkrog [20] observed that aged plant leaves absorb less activity than green plants. In Fig. 9 the mass interception factor is plotted against the rainfall for ^{131}I, ^{7}Be, and insoluble microspheres applied with artificial rain. The dependence of the f/B-value on the amount of rainfall is strong for iodine, but it is much less pronounced for ^{7}Be and the insoluble microspheres. However, for particles over about 40 μm diameter, the interception fraction will probably be lower than for the microspheres as investigated by Hoffman et al.

For models predicting the consequences of routine releases, a constant, element-dependent interception factor might be appropriate if the rainfall pattern of the site considered is known.

In contrast, for models dealing with single releases, the interception has to be modelled by distinguishing between anions, cations and insoluble particles. For continuous rainstorms, the interception (f/B) for anions such as I^- or SO_4^{2-} can simply be related to the amount of rainfall.

2.1.7. Summary and conclusions

From the review of interception fractions and mass interception factors reported in the literature, the following conclusions can be drawn:

(1) The variations of the interception factor f are larger than for the mass interception factor f/B.

(2) The variation of the mass interception factors f/B observed for dry deposits is relatively low for small particles. For those particles a mass interception factor of about $3 m^2 kg^{-1}$ might be appropriate for a rough estimation of the initial contamination of vegetation due to dry deposition. However, this value should not be used without critical analysis of the situation to be considered. For particles of more than 40 μm the reported f/B-values are significantly lower. For large particles, wet plant surfaces favour the interception of dry-deposited particles by a factor of approximately 3 over that for dry surfaces.

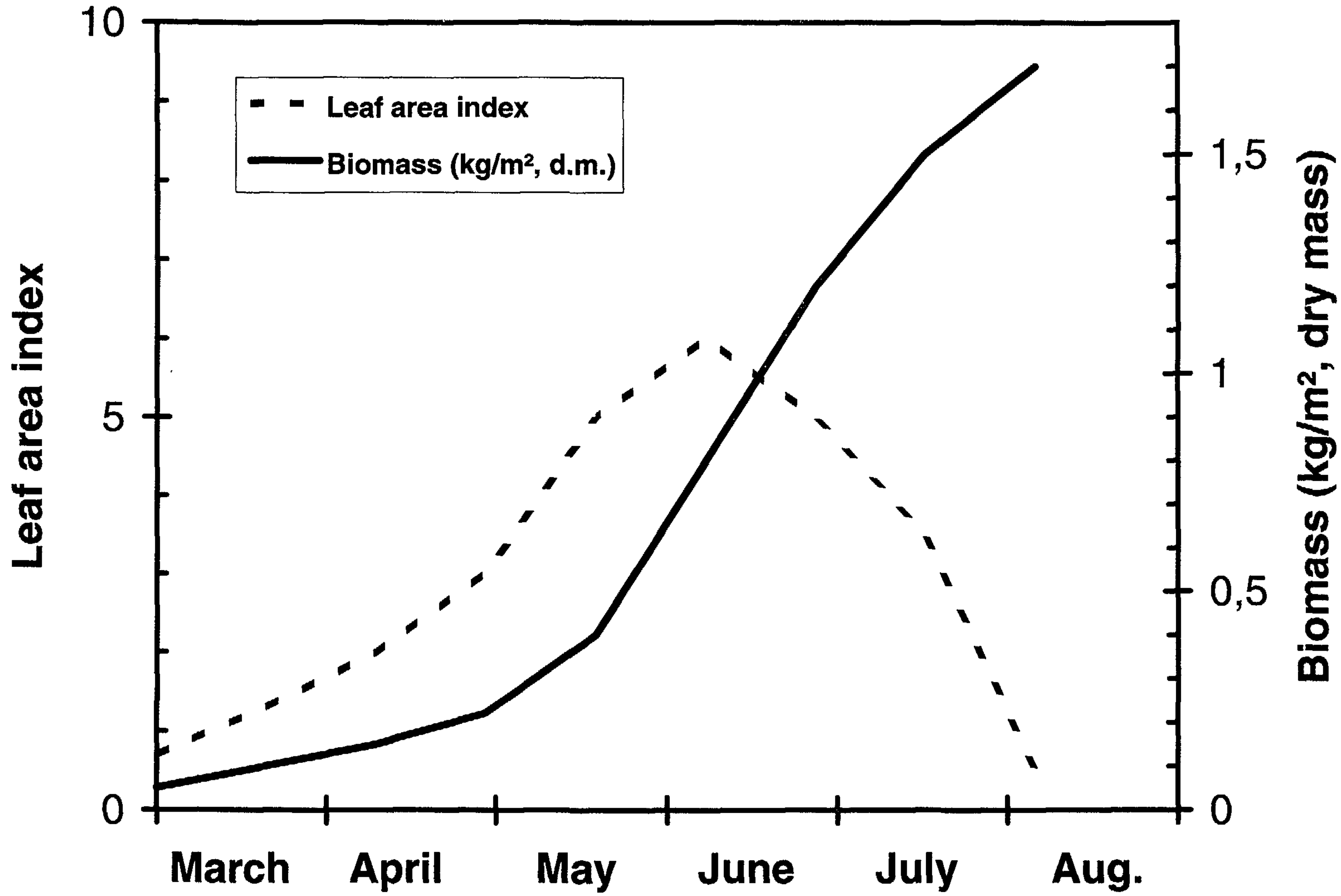

FIG. 8. The development of the leaf area index and the biomass during the growth period of winter wheat [45].

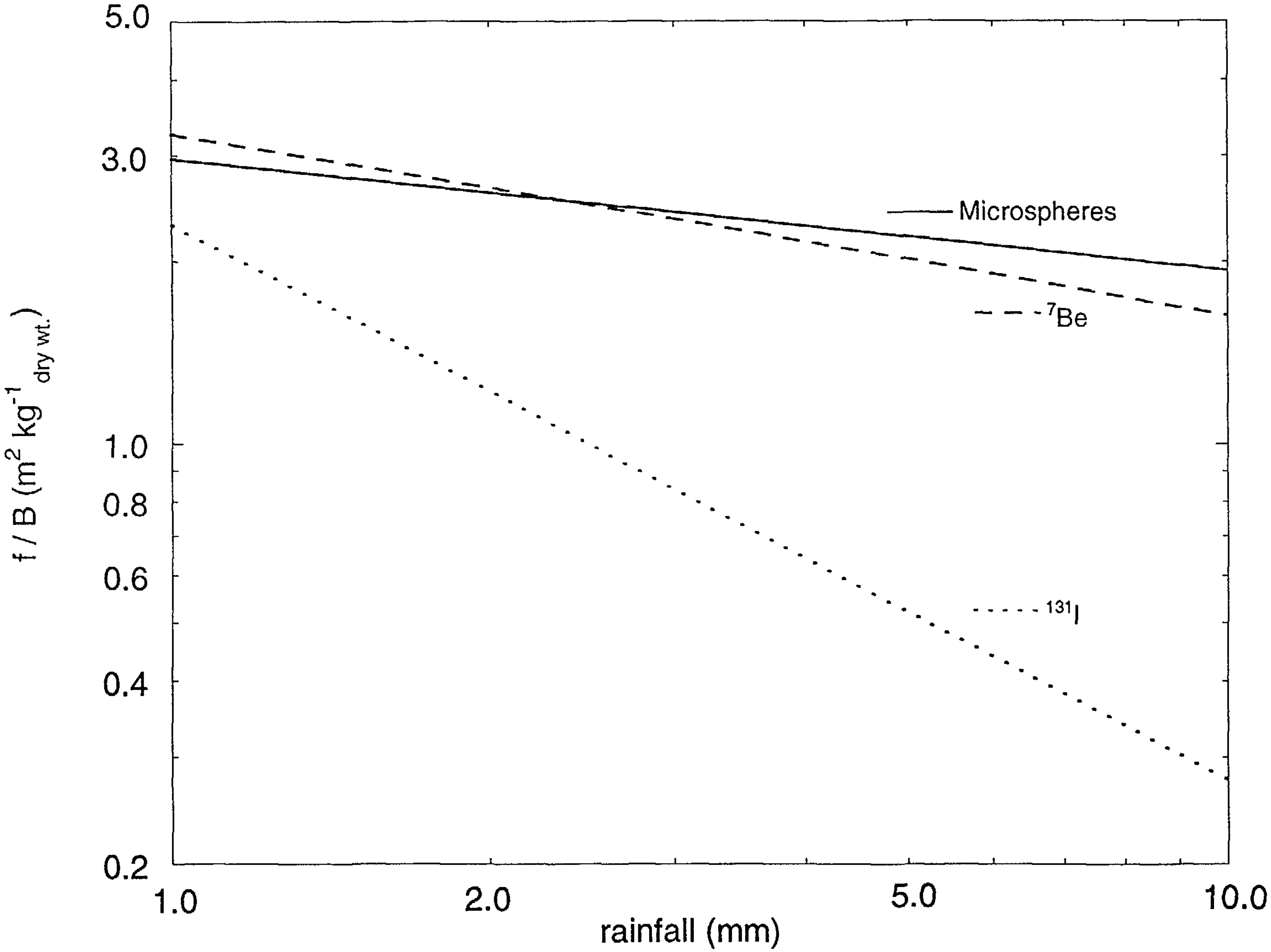

FIG. 9. Mass interception factor of activity in artificial rain. Regressions of Hoffman et. al. [23] (biomass 0.15 kg m^{-2} dry mass; rainfall intensity 17 mm h^{-1}).

(3) The mass interception factors f/B of wet-deposited activity are highly chemical-dependent; values for cations are significantly higher than for anions. Fig. 9 gives an impression of the dependence of f/B on the chemical form and the amount of precipitation for intermediate precipitation events. Furthermore, the f/B-values for cations decrease only slightly with increasing amounts of rainfall, whereas for anions such as iodine the f/B-values decrease in proportion to increasing precipitation. From Fig. 9, a concentration ratio vegetation/rainwater of 2.5 to 3.0 can be derived for iodine. For insoluble microspheres and reactive cations such a simple concentration ratio cannot be given, because the washoff effects during the precipitation are less pronounced than for iodine. The variations of f/B for different plant species are of minor importance.

(4) The correlation of the interception fraction to the biomass is strongly supported for pasture grass and other leafy crops. However, for other crops such as grain, fruits and vegetables, in the second half of the growing period, a normalization to the leaf area seems to be more appropriate. Unfortunately, the data available for such crops are very limited. Further investigations for such crops could fill some of these gaps of knowledge.

(5) In many models, interception fractions or mass interception factors are applied to the dry-deposited activity calculated by the use of deposition velocities. However, deposition velocities are often determined by relating the activity on the plant's surface to the time-integrated activity in air during the period of deposition. Therefore such approaches are inconsistent with the underlying experiments. This effect is often not obvious due to the considerable uncertainty associated with quantification of dry deposition; thus, this might be an area for model improvement.

(6) Wet and dry deposition to plant surfaces and the corresponding interception of wet- and dry-deposited activity are completely different processes which depend on different factors. However, in models for the prediction of the radiological consequences of routine releases, the use of generic, empirically derived interception fractions might be justified for both dry and wet deposition. For the estimation of plant contamination by single releases of activity, these processes should be modelled separately in order to take into account the situation-specific contribution of dry and wet deposition in a realistic way.

2.2. LOSS OF RADIONUCLIDES FROM VEGETATION

2.2.1. Introduction

After radionuclides are deposited on vegetation, environmental removal processes will combine with radioactive decay to reduce the quantity of initial contamination. Assuming strictly single-component exponential mechanisms, the time necessary for one-half of the radioactivity to be removed by environmental processes is referred to as the environmental half-time, T_w. This parameter is related to the environmental loss constant λ_w as follows:

$$\lambda_w = \frac{\ln 2}{T_w} \tag{5}$$

Considering radioactive decay with environmental loss processes, an effective half-life T_{eff} or loss constant λ_{eff} can be calculated as follows:

$$T_{eff} = \frac{T_w \cdot T_r}{T_w + T_r} \tag{6}$$

where T_r = radiological half-life. There are two possible methods to quantify the environmental half-time T_w:

- T_w may be determined by the loss of radionuclides from vegetation growing on a unit ground area.

- T_w may be determined by the decrease of the activity per unit mass of vegetation.

In the latter case, T_w includes the diluting effect of the increase in biomass during the plant's growth.

For radionuclides with small values of T_r, T_w will have relatively little effect on the resulting value of T_{eff}. For long-lived radionuclides however, T_w will directly control the value of T_{eff}. The evaluation of the potential variability in T_w is of importance for estimating the potential impact on man of long-lived radionuclides initially deposited on vegetation, as T_{eff} (or λ_{eff}) is frequently used in radiological models. For long-lived radionuclides, T_w has a direct influence on the calculated level of vegetation contamination when uptake from the soil is relatively low and the time between initial deposition and harvest is sufficiently long. It also controls the equilibrium concentration under chronic deposition. The process of loss of deposited materials from vegetated surfaces was termed "field loss" by Russell [10].

2.2.2. Factors influencing the environmental half-time

The environmental half-time T_w may be influenced by a number of processes. The exact contribution of specific processes to the removal of deposited radionuclides from vegetation is difficult to quantify. Therefore, the parameter T_w is determined empirically via regression of the concentration in vegetation with time. Nevertheless, several processes that affect the concentration of deposited radionuclides on vegetation with time and that ultimately will influence the variability of values reported for T_w.

Wind removal

Radionuclides associated with large particles may be shaken off of vegetation by wind action. This process may lead to a rapid initial loss of material immediately after deposition. There may be little direct effect by the wind on radioactivity removal from external plant surfaces after the first few days following the deposition [48]. However, it has been found that submicron particles can be detached from the surfaces of plants, especially during periods of rapid growth and high transpiration rates. Surface abrasion and leaf bending resulting from wind action may serve to dislodge salt particles, wax rodlets, and cuticular and other surface fragments from vegetation [49]. Thus, any radioactivity that might be associated with these particles may be removed from vegetation.

Water removal

Precipitation, fog, dew and mist may remove radionuclides from vegetation via two processes. Wash-off removes material from the surface of the plant, and leaching removes material incorporated into the interior of the plant. Like direct wind removal, wash-off seems to be most effective immediately following the deposition event [50]. However, the long-term retention of radionuclides on vegetation does not seem to be correlated to the amount of rainfall during the experimental periods. Nevertheless, in some experiments, in which the environmental half-time was determined simultaneously on experimental plots protected against rain, significantly longer values for T_w were observed [51, 52]. Leaching may occur throughout the plant's growing season, but it is the greatest just before maturity and death of the foliage [53]. Also, rain falling as a light continuous drizzle is more efficient as a leaching agent than is a large quantity of rain falling during a short period. Leaching also may be enhanced by high temperatures [54].

Growth

Up to maturity, growth of herbaceous vegetation can be approximately regarded as an exponential process [55–57]. As a result, the mass concentration of any material associated with the vegetation will decrease at about the same rate as the plant is growing. This process of growth dilution is implicitly included when values of T_w are based on measurements of radioactivity per unit mass of vegetation over time. This process is excluded, however, when values of T_w are based on measurements of vegetation radioactivity per unit area of ground. Such measurements, when obtained from an appropriate experimental design, provide an estimate of the total inventory of radionuclides in above-ground vegetation at the time of sampling. The effect of growth dilution depends on the farm management and the weather conditions during the growing period, as well as on the stage of the growth.

In intensive farming systems, different kinds of fertilizers are frequently applied to stimulate the plant growth and to maximize the yield; this practice principally increases the importance of growth dilution. Another important factor is the water supply of plants; water deficits inhibit the increase of biomass and therefore decrease the effect of growth dilution.

The increase of biomass is also subject to seasonal variations. For example, for pasture, the highest growth rates can be observed in spring; from the beginning of summer a more or less continuous decrease occurs until the vegetation senescence in autumn. The main factors for this decrease may change slightly from region to region. However, these factors are the inhibition of growth due to high temperatures, which might be accompanied temporarily by water supply deficits in summer, and low temperatures and the decreasing solar radiation in autumn.

In some crops senescence of individual leaves is a mechanism for radionuclide removal, for example, in grass where leaves die back and are continuously replaced. The removal of activity by grazing animals is another removal mechanism.

As stated above, the stage of a plant's growth plays an important role in determining the removal rate of radionuclides from the plant. Leaching of materials from plants increases with age, possibly in part due to the fact that young leaves are wetted with more difficulty than older leaves [54]. At the same time, growth dilution processes slow down as a plant ages. Some studies indicate that processes of radionuclide removal from the surfaces of herbaceous vegetation (mostly grasses) may be substantially reduced after the plant dies or becomes dormant [13, 21, 22, 33, 58, 59, 60].

2.2.3. Measured values for the environmental half-time

Miller and Hoffman [61] have given a comprehensive compilation of values for T_w obtained or derived from the literature (Table VI). The range of all values for T_w for herbaceous vegetation derived per unit mass is 2.8 to 19 d with a geometric mean of 8.6 d. From the experiments excluding the effect of growth dilution, a geometric mean of 11 d with a range of 4.5 to 34 d can be obtained. In general, T_w is lower for iodine (vapour as well as particle-bound iodine) than for the other particles.

The effect of growth dilution seems to be more pronounced for particulates than for iodine. However, it is very likely that this observation is an artifact, because the rapid physical decay as well as the shorter environmental half-life of iodine can mask the effect of growth dilution.

Values of the T_w for woody vegetation are generally larger than for herbaceous plants. This might be due to the effect of leaching. Radionuclides incorporated into woody plant parts will probably be much less subject to leaching by rain and fog than radionuclides incorporated into leafy plant parts.

The longest environmental half-lives were obtained for dormant vegetation, with a geometric mean value of 25 and a range of 5 to 49 d.

In Table VII, measurements of the environmental half-time T_w are summarized which are not included in the compilation of Miller and Hoffman [61]. The results are in good agreement to the observations summarized in Miller and Hoffman. The investigations of Aarkrog and Lippert [21] underline the influence of the stage of growth of plants on loss of radionuclides subsequent to initial retention. In these experiments, in the first two-thirds of the growing period of barley, a mean value for T_w of 18 d (per unit ground area) was observed, whereas from the middle of the growing period to harvest a value for T_w of 46 d was determined; this is a factor of 2.5 longer. In the first case, T_w was measured during the vegetative growth period, characterized by a high metabolic activity. In the second period of the experiment, the plants were in the corn filling (translocation to the grain of carbohydrates being produced during photosynthesis) and maturing phase. In this stage the foliage dies off and the barley plants become more and more similar to dormant vegetation.

The value for T_w of 130 d determined by Bondietti et al. [72] for Be-7 on semidormant pasture is the highest value which has been determined. Also this result underlines the differences in T_w of dormant and growing vegetation.

In an experiment with the pesticide diflubenzuron, Wimmer et al. [73] found that the levels of pesticide on leaves of hardwood trees declined rapidly (20–80%) during the first two weeks following application; after that, the levels were generally stable until leaf fall. The initial loss was much greater for two species of trees (yellow poplar and black oak) than for others (red maple, sugar maple, chestnut oak, white oak, and northern red oak). Rainfall did not seem to be major factor in pesticide loss, with the exception of a very hard rain immediately after application of the pesticide, in which case 40–50% was lost. Growth dilution also was not an important factor, as growth of the leaves was essentially complete at the time of spraying.

TABLE VI. REPORTED ENVIRONMENTAL HALF-TIMES

Experimental conditions	Geometric mean (days)	Range (days)	Number of observations	References
All values for herbaceous vegetation				
– Derived per unit mass	8.6	2.8-19	24	[61]
– Derived per unit ground area	11	4.5-34	26	[61]
I_2 vapour on herbaceous vegetation				
– Derived per unit mass	6.8	4.8-7.9	6	[58]
– Derived per unit ground area	7.3	4.5-14	13	[33, 48, 60, 62, 63]
I particulates on herbaceous vegetation				
– Derived per unit mass	8.2	2.8-16	10	[58, 59, 64–68]
– Derived per unit ground area	8.5	6-12	2	
Other particulates on herbaceous vegetation				
– Derived per unit mass	12	9-19	5	[13, 19, 20, 22, 64, 65,
– Derived per unit ground area	20	19-24	11	69, 70, 71]
I particulates and vapour on woody vegetation				
– Derived per unit mass	13		1	[64]
– Derived per unit ground area	7.9		1	[63]
Other particulates on woody vegetation				
– Derived per unit mass	20	12-28	4	[64, 69]
Dormant vegetation[a]				
all values	25	8.1-∞[b]	18	
I_2 vapour	22	8.1-50	5	[58, 60]
I particulates	22	5.3-∞[b]	8	[59, 60]
Other particulates	32	22-49	5	[13, 69]

[a]Dormant vegetation includes standing herbaceous vegetation sampled in winter as well as woody tissues of deciduous vegetation sampled after leaf-fall.
[b]For purposes of this statistical summary, a value of $T_w = \infty$ was set equal to $T_w = 44$ d, the highest measured value of T_w for I particulates.

TABLE VII. ADDITIONAL MEASUREMENTS OF ENVIRONMENTAL HALF-TIMES

Experimental conditions	T_w (d)	Range (d)	Observations	References
Fine spray of ^{89}Sr on pasture				[74]
- per unit mass	9	-	1	
- per unit area	14	-	1	
Fine spray of ^{51}Cr, ^{59}Fe, ^{58}Co, ^{65}Zn, ^{203}Hg, ^{210}Pb on spring barley				[21]
- per unit area determined from week 2 to 11	18	14–22	5	
determined from week 8 to 16 (harvest)	46	32–54	6	
^{7}Be on growing herbaceous vegetation	17	-		[72]
^{7}Be on semi-dormant pasture	130	-		[72]
^{238}Pu on corn	12			[75]
^{131}I, ^{7}Be and insoluble microspheres				[23]
- per unit mass		7.2–15		
- per unit area		7.5–18		
^{137}Cs on lettuce	8		1	[31]
^{131}I on grass	13	-	1	[76]

Table VIII summarizes some environmental half-times which were determined after the deposition of Chernobyl fallout. It should be mentioned that the two exponential functions for T_w reported by Müller and Pröhl [77] also include the activity in the second cut after the deposition. In addition to the data in Table VIII, environmental half-times determined for Chernobyl fallout at a larger number of sites have been summarized by Kirchner [78]. The mean weathering half-times on plants were 9.1 $\pm$ 0.6 d (median = 8.5 d) for iodine and 11.1 $\pm$ 0.8 d (median = 10.4 d) for caesium. The values obtained from Chernobyl fallout are in good agreement with the pre-Chernobyl values [61]. The summary by Kirchner, however, does not differentiate between environmental half-times determined on the basis of mass vs. those determined on the basis of area.

TABLE VIII. SUMMARY OF ENVIRONMENTAL HALF-TIMES DETERMINED FROM THE CHERNOBYL FALLOUT

Experimental conditions	T_w (d)	Range (d)	Observations	References
^{131}I on pasture		7.5–8.9	3	[80]
^{131}I on pasture	9.8		1	[81]
^{137}Cs on pasture	14		1	[81]
^{137}Cs on pasture				
short term		8–14 (95%)		[77]
long term		40–60 (5%)		
^{131}I on pasture	12			[37]
^{137}Cs on pasture	14			[37]
^{131}I on pasture		5.9–14		[36]
^{137}Cs on pasture		11–15		[36]
^{131}I on pasture	11			[82]
^{137}Cs on pasture	11			[82]
^{131}I on grass	14.3		1	[83]
^{137}Cs on grass	14.4		1	[83]
^{137}Cs on broccoli	11 ± 1		1	[84]
^{103}Ru on broccoli	7.5 ± 2.5		1	[84]
St. Petersburg				[42]
point 1 (30 Apr–2 Jun)				
^{131}I on pasture	9.6		1	
^{137}Cs on pasture	13.4		1	
point 2 (30 Apr–27 Aug, without cutting)				[41]
^{137}Cs on pasture				
short term		9–14.2 (92%)	3	
long term		49–52.2 (8%)		
point 3 (30 May–22 Jul, without cutting)				[41]
^{137}Cs on pasture				
short term	3.1 (75%)		1	
long term	53.2 (25%)			
Gomel region, 30-km zone ChNPP				[41]
^{137}Cs on pasture				
point 1 (10 May–10 Jun)	10.2	9.7–10.6	3	
point 2 (11 May - 5 Aug)				
short term	13.1 (85%)		1	
long term	46.2 (15%)			

Values are included in Table VIII for two areas in the former Soviet Union for the Gomel region of Belarus, within the 30 km zone of the Chernobyl NPP (two points) and for St. Petersburg (three points) [41, 42]. Some parts of St. Petersburg, including point 3, had heavy rain after deposition (1-3 May), followed by good weather. In the 30 km zone of the Chernobyl NPP, the weather was good throughout the observation period.

TABLE IX. VALUES OF T_w FOR SIMULATED FALLOUT PARTICLES ON CROP PLANTS ILLUSTRATING MULTIPLE TIME COMPONENTS OF THE REMOVAL PROCESS [15]

Time period after deposition (days)	Weathering half-life, T_w (d)					
	Squash	Soybean	Sorghum	Peanuts	Lespedeza	Mean value (all plants)
			44–88 μm particles			
0–1.5	2.5	2.1	2.8	5.1	1.4	2.8 ± 0.71
1.5–14	7.2	7.3	6.6	8.4	8.4	7.6 ± 0.35
14–28	42	18	35	18	18	26 ± 4.7
28–56	39	45	270	32	33	84 ± 46
			88–176 μm particles			
0–1.5	1.6	1.5	4.1	1.3	2.9	2.3 ± 0.53
1.5–14	7.4	7.2	7.4	16	2.6	9.1 ± 1.6
14–28	15	16	19	16	14	16 ± 1.0
28–56	57	35	38	29	38	39 ± 5.2

TABLE X. ENVIRONMENTAL HALF-TIME FOR ^{137}Cs ON GRASS [51]

Experimental conditions	Mode of determination		T_w (d)	
Fine spray of Chernobyl rain-water containing ^{137}Cs on grass grown in pots *protected* from rain	per unit mass	short term	8	(92%)
		long term	>60	(8%)
	per unit area	short term	6.2	(20%)
		long term	>60	(80%)
Fine spray of Chernobyl rain-water containing ^{137}Cs on grass grown in pots *not protected* from rain	per unit mass	short term	3.4	(90%)
		long term	23	(10%)
	per unit area	short term	2.4	(50%)
		long term	30	(50%)

However, other investigations also indicate that the decline of activity after deposition can be described by a function of at least two exponentials. During a period of 56 days after the deposit, Witherspoon and Taylor [15] have measured T_w for different types of vegetation using simulated fallout particles with size spectra of 44 to 88 μm and 88 to 176 μm (Table IX). They could distinguish 4 different phases during which T_w was significantly different. T_w increased during this 8-week experiment from some days in the initial phase to some tens of days in the last phase. However, the values for T_w for the last period in particular are probably associated with a considerable uncertainty because the half-life is longer than the experimental period during which it has been measured.

Witherspoon and Taylor [79] measured the loss of simulated fallout particles with a size of 1 to 44 μm from sorghum and soybean plants during a period of 5 weeks. For the first week, a value for T_w of approximately 4 days is reported, whereas in the subsequent 4 weeks of the experiment the loss was equivalent to an environmental half-time of 23 days.

In the experiments of Witherspoon and Taylor [15, 79] the initial loss rate is relatively high. However, in these experiments large particles have been used. For those particles an enhanced loss due to wind action might occur, as Mason et al. [48] speculated.

Also Ertel et al. [51] found that environmental half-lives followed a function with two exponentials (Table X) over a period of 60 d. A fine spray of Chernobyl rainwater was applied to grass grown in pots. One half of the pots were protected against rain, but watered during the whole experimental period; the other pots were not protected against rainfall. For the uncovered plots, T_w is about a factor of 2 shorter than for the plots protected against rainfall.

In both cases, covered and uncovered, the initial loss is very rapid. The long-term components might have been influenced by the very low growth rate in the second part of the experiment.

Such information about the time dependence of the environmental half-time is very useful, especially for application in dynamic radioecological models being used for the assessment of the radiological consequence of single radionuclide depositions. Nevertheless, effective values of T_w may be used to estimate long-term average concentrations in plants from routine releases or time-integrated concentrations resulting from acute releases.

2.2.4. Summary and conclusions

From the review of the environmental half-times of radionuclides deposited on vegetation the following conclusions can be drawn:

(1) The loss of radionuclides from vegetation is due to removal by wind and removal by water, including wash-off as well as leaching by rain and fog. Also the abrasion between leaves and shedding of cuticular wax can affect the post-deposition removal rate.

(2) The growth of vegetation causes a diluting effect for the retention measured in terms of activity per unit mass of vegetation. The growth dilution is most pronounced for growing herbaceous vegetation. It is less obvious for woody plants and negligible for dormant or dead standing vegetation.

(3) The removal rate seems to be highest immediately after deposition and then is reduced with time. Although removal by wind and water are factors that cause the post-deposition loss of activity, the long-term retention seldom appears to be correlated with weather conditions. However, in one experiment, the removal of activity from plants protected against rain has been a factor of 2 less compared to those exposed to normal rain.

(4) For iodine (elemental and particulate), the environmental half-time including growth dilution is about 7 days, with a range of 5 to 16 days. There is no significant difference if growth dilution is excluded. The rapid physical decay of ^{131}I possibly masks the effect of growth dilution.

(5) For particulate-bound radionuclides, the enviromental half-time is longer. Including growth dilution, a mean value of 12 d with a range of 9-19 days can be obtained. If the environmental half-time is determined per unit ground area, the mean is about 20 d.

(6) Some experiments indicate that the decline of activity can be described by a function of at least two exponentials, a short-term exponential with a half-life of several days and a long-term exponential with a half-life of some weeks.

(7) According to Chamberlain [37], the volatilization of iodine retained by plants is not a process which can explain the faster removal of iodine compared to particulates.

(8) For dormant vegetation the removal rates from vegetation are much lower, with environmental half-times ranging from about 20-130 d for particulates and from 5.3 to more than 50 d for iodine.

(9) The stage of development of the plants seems to influence the post-deposition retention considerably. The environmental half-times increase during the maturing process, when the foliage becomes more and more similar to dormant or dead standing plant canopies.

(10) The radiological models compared in the framework of the BIOMOVS A4 test scenario [36] used values for the environmental half-time which are within the range of values reported here. Most models use a single half-life to describe weathering loss. Bearing in mind the observed time-dependence of weathering, this could lead to an underestimation of the long-term contamination of plants after single deposits. However, the use of single exponential functions is appropriate for estimating contamination due to routine releases or for estimating the time-integrated concentrations from acute releases.

Appendix

COMPARISON OF CONCENTRATION RATIOS VEGETATION/AIR DETERMINED FROM THE CHERNOBYL DEPOSITION WITH THEORETICALLY DERIVED CONCENTRATION RATIOS VEGETATION/AIR FOR DRY AND WET DEPOSITION

The measurements performed after the Chernobyl accident of integrated activity concentration in air and in grass, as well as the total deposition of ^{131} I and ^{137}Cs, can be used to test the derivation of the mass interception factor f/B and the environmental half-time T_w. For some locations reported in Köhler et al. [36], concentration ratios vegetation/air ($CR_{v/a}$) are calculated according to:

$$CR_{v/a} = \frac{(D_{tot})\ (f/B)\ (T_{eff})}{(\ln 2)\ (C_{air})}$$

where

$CR_{v/a}$	=	concentration ratio vegetation/air ($m^3\ kg^{-1}$)
D_{tot}	=	total deposition ($Bq\ m^{-2}$)
f/B	=	mass interception factor ($m^2\ kg^{-1}$)
T_{eff}	=	effective environmental half-time (d) (including physical decay)
C_{air}	=	time-integrated activity concentration in air ($Bq\ d\ m^{-3}$)

In all these locations, the Chernobyl fallout was a mixture of dry and wet deposition. Therefore, two theoretical values of $CR_{v/a}$ were calculated: in one case it is assumed that the total deposition was dry, in the other case it is assumed that all deposition was during rainfall and no dry deposition occurred. These theoretical values represent the extreme cases and should give the lower and upper bounds of the actual $CR_{v/a}$. The calculated values for $CR_{v/a}$ are then compared against the $CR_{v/a}$ determined from measurements of the activity in air and grass at the sites considered.

The following parameters have been assumed for calculation of the theoretical values:

- For dry deposition, a mass interception factor of 3 $m^2\ kg^{-1}$ was used for both ^{131}I and ^{137}Cs.

- For wet deposition, a mass interception factor of 0.6 $m^2\ kg^{-1}$ was used for ^{131}I, and a mass interception factor of 1 $m^2\ kg^{-1}$ was used for ^{137} Cs. The data on measured values for the mass interception factor for wet deposition of ^{137}Cs are very poor. However, according to Pröhl [41], the initial retention of Cs seems to be a factor of 2 lower than for ^{140}Ba, which should behave similarly to ^{90}Sr and ^{7}Be. So, the value for μ used here is about a factor of 2 lower than for ^{7}Be.

- For the environmental half-time (including growth dilution), values of 7 d for ^{131}I and 14 d for ^{137}Cs were used.

Calculated and observed values for $CR_{v/a}$ at seven locations are given in Table XI. It is obvious from the table that the observed $CR_{v/a}$ are with two exceptions between the theoretical derived values for wet and dry deposition. Taking into account some additional information about the deposition pattern at these sites, it can be concluded that for sites with a high contribution of wet deposition, the observed values for $CR_{v/a}$ are generally closer to the theoretical $CR_{v/a}$ for wet deposition (e.g., Neuherberg, Tranvik, Budapest, Petten).

Due to the high rainfall at Neuherberg, the mass interception factor for wet-deposited ^{137}Cs used for the calculations might be slightly too high. The deviation observed at Tokai is probably due to wash-off during very heavy showers after the deposition.

Although the results of this comparison should not be given undue weight, they nevertheless indicate that the values T_w and f/B reported in this paper are consistent with environmental measurements performed after the Chernobyl accident.

TABLE XI. COMPARISON OF OBSERVED CONCENTRATION RATIO VEGETATION/AIR WITH THEORETICAL VALUE CALCULATED FOR DRY AND WET DEPOSITION

Location	Time integrated air concentration (Bq d m^{-3})	Total deposition (Bq m^{-2})	Deposition velocity (m d^{-1})	Concentration ratio vegetation/air (m^3 kg^{-1})		
				Observation	Theoretical Wet deposition	Theoretical Dry deposition
			^{131}I			
					f/B = 0.6 m^2 kg^{-1}	f/B = 3 m^2 kg^{-1}
Neuherberg	102	80000	784	5900	2500	13000
Roskilde	6.1	4000		7800	2100	11000
Tranvik	74	50000		2700	2200	11000
Budapest	51	33000		5900	2100	11000
Petten	36	17000		2500	1500	7700
Tokai	2.4	930		2100	1300	6300
Loviisa	270	100000		1200	1200	6000
			^{137}Cs			
					f/B = 1.0 m^2 kg^{-1}	f/B = 3 m^2 kg^{-1}
Neuherberg	12	19000	1600	28000	32000	95000
Roskilde	0.53	800	1500	35000	30000	91000
Tranvik	5.3	2000	377	14000	7500	23000
Budapest	4.5	5600	1244	36000	25000	75000
Petten	3.8	2400	631	12000	13000	38000
Tokai	0.15	140	933	6700	19000	56000
Loviisa	15	20000	1333	41000	27000	60000

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Chapter 3

TRANSFER OF RADIONUCLIDES BY TERRESTRIAL FOOD PRODUCTS FROM SEMI-NATURAL ECOSYSTEMS TO HUMANS

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3.1. INTRODUCTION

The Chernobyl accident focussed attention on the transfer of radionuclides, and in particular radiocaesium transfer through natural and semi-natural ecosystems to man. Within the VAMP programme attention has been drawn to the need to consider the potential significance of foodstuffs from these ecosystems (hereafter termed "natural food products") in radiological assessments.

Natural and semi-natural ecosystems, as discussed in this review, include non-intensively managed areas such as forests, heathlands, uplands, mountain pastures, mediterranean dry-shrub areas, marshlands and tundra [1]. In such ecosystems high and, in some cases, prolonged, bioavailabilities of some important radionuclides have been noted. Major foodstuffs taken from these ecosystems include fungi, berries, honey, meat from game animals and meat and milk from domestic ruminants. In these ecosystems, species diversity is much more pronounced than in agricultural systems, and many species are known to accumulate comparatively high levels of radiocaesium. Even within the same species the variation in radiocaesium levels can be considerable.

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Models developed before the Chernobyl accident were usually limited to considering agricultural food production, and they predicted radionuclide levels in such foodstuffs reasonably well. However, when these models were used they failed to predict radionuclide levels in natural food products from semi-natural ecosystems with an adequate degree of precision. Semi-natural ecosystems were previously considered only to a limited extent (with the notable exception of reindeer), because the quantities of food consumed from these systems were assumed to be comparatively small and usually by only a minor proportion of most countries populations. They were therefore usually not considered in dose assessments.

Although semi-natural ecosystems provide comparatively small quantities of food, the long-term dose commitment from these ecosystems to humans can be significant, particularly for radiocaesium. This is primarily due to the high levels of ^{137}Cs found in many natural food products and the much longer effective half-lives of ^{137}Cs observed in some of these ecosystems compared with agricultural systems [2,3,4,5]. Intake of food from semi-natural ecosystems is not evenly distributed in the population, and certain critical groups, such as hunters, sheep and reindeer breeders and berry and fungi pickers can have particularly large intakes of natural food products [6,7,8]. Consumption rates and the size of the critical groups which eat natural food products from semi-natural ecosystems vary greatly both between and within countries. Whilst high radiocaesium levels in natural food products often persist, those from agricultural areas generally decline quickly so that with each year after the deposition of fallout the comparative importance of transfer of radiocaesium by natural food products from semi-natural ecosystems to man increases.

The aim of this report is to provide a short summary of the most important factors affecting radionuclide transfer in semi-natural ecosystems, and to attempt to derive empirical relationships which can be used for modelling purposes.

For agricultural systems models have been developed which describe the transfer of radionuclides to food products with a reasonable degree of accuracy. However, modelling such transfers to food and finally to man is much more difficult for semi-natural ecosystems due to a lack of understanding of the processes involved in controlling the fate of radionuclides in these complex ecosystems. There is a pronounced heterogeneity in soil properties in these ecosystems, and furthermore, a much higher variability in dietary selection of food producing animals compared with agriculturally improved areas.

The transfer of radionuclides between soil and plants is often expressed using a concentration ratio; it is the ratio of the radionuclide concentration in the plant and that in soil (Bv). For plant to animal transfer, a transfer coefficient is often used; it is the equilibrium ratio between the activity concentration in milk or meat divided by the daily intake (F_m and F_f respectively). However it is often difficult to get reliable estimates of these parameters to use to estimate transfer ratios or coefficients in semi-natural ecosystems. For instance, the bulk density of soils in semi-natural ecosystems can vary markedly giving rise to very different transfer values to plants. For animals, the estimation of daily herbage intake is particularly difficult in semi-natural ecosystems. Therefore, it is questionable if it is appropriate to use such transfer parameters in these circumstances.

Therefore in this report we have attempted to provide easily derived, empirical transfer coefficients, termed aggregated transfer coefficients (T_{ag}) which can be used in predictive models instead of the commonly used transfer parameters. T_{ag}s are calculated using the expression:

$$T_{ag} = \frac{\text{activity concentration in the food product (Bq kg}^{-1}\text{ or Bq l}^{-1})}{\text{activity of deposit per unit area (Bq m}^{-2})}$$

and therefore are expressed as $m^2 kg^{-1}$ or $m^2 L^{-1}$. Activity levels are commonly expressed as fresh weight (fw), and whilst this is most appropriate for animal products more consistent values will be obtained for plant products by using dry weights (dw).

T_{ag} values are routinely used for agricultural ecosystems in the CIS. They are relatively easy to calculate although, in common with other transfer coefficients, they require specific knowledge of the ecosystems for which they are being used [5]. Given the complexity of the ecosystems and the lack of detailed knowledge there seems, at present, to be few practical, generally applicable, alternatives to the use of T_{ag} values for making long-term predictions in many types of semi-natural ecosystem.

Any transfer parameter is of limited usefulness when trying to predict radiological impact, unless it is combined with estimates of time-dependent changes in transfer. This may be of particular importance in semi-natural ecosystems where high radionuclide levels can persist for many years. However, if such changes with time are very slow, time dependence is of less importance. Reductions in activity levels with time are usually expressed using ecological (t_{ec}) or effective (t_{ef}) half-lives.

The effective half-life describes the time required for the activity concentration of a radionuclide in a food product to be reduced to one half of the original concentration in a specific system. Therefore the t_{ef} incorporates physical decay. In this report the t_{ef} takes into account all biological, chemical and physical processes within the ecosystem which influence the transfer of a radionuclide to a food product.

The ecological half-life does not take physical decay into account, and therefore can be adapted for different isotopes of the same element. For example, the t_{ef} of ^{134}Cs and ^{137}Cs will differ because of the differences in physical half-lives, while the t_{ec} would be identical. The relationship between t_{ef} and t_{ec} for a radionuclide with a physical half-life (t_{phys}) will be:

$$1/t_{ef} = 1/t_{phys} + 1/t_{ec}$$

The t_{ef} and t_{ec} values considered here describe only the long-term losses and may be comprised of more than one component. They do not include the short-term reductions in activity concentrations which often occur in the first few months after deposition. Similarly, T_{ag} values are not directly applicable in the period when directly deposited fallout could still be present on the vegetation. Therefore, they primarily refer to the relationship between ground deposit and contamination of the plant or animal, when the direct component is negligible. Furthermore, T_{ag}s cannot be used for prediction in conditions of continuous deposition for the same reasons.

The discussion will primarily consider radiocaesium; the few data which are available on the transfer of other radionuclides to foodstuffs are included where possible. Hopefully, further T_{ag} values may be derived in future from the more commonly used transfer ratios using information such as soil bulk density.

3.2. IMPORTANT PARAMETERS GOVERNING RADIONUCLIDE BEHAVIOUR IN SEMI-NATURAL ECOSYSTEMS

3.2.1. Interception and retention on plant surfaces

The rate of interception is highly variable in these ecosystems, and will depend on factors such as vegetation biomass, and the presence of coniferous or deciduous tree stands. Interception of radiocaesium can therefore range from below 5% of the total deposited on heavily grazed pastures in mountain areas with low vegetation biomass to >50% in semi-natural pasture [9] and 60-100% in dense coniferous forest [10]. Furthermore, part of the deposited radioactivity may be retained in the overstory of woodlands for some time. In thick lichen mats interception is nearly complete and the t_{ec} of retention of the original deposit is often in the order of years. Although it may take weeks or even months before all intercepted activity is transferred from the surface of many plant species to the soil, this intercepted component usually will not greatly influence the overall dose commitment to man from consumption of natural food products because of the high root uptake and long t_{ec} of radiocaesium in these ecosystems.

The physico-chemical form of the intercepted deposition may be important when plants which are externally contaminated are eaten by animals, as demonstrated by the relatively low bioavailabilty of Chernobyl fallout radiocaesium in 1986 compared with later years [11-13]. However, the effect will be limited to the period during which the initial deposit is retained on the plant surfaces. Furthermore, the physico-chemical form of the deposit, and how this changes with time, will also influence the rate of root uptake.

3.2.2. Soil properties

One of the most important factors responsible for the high radiocaesium levels in food products from semi-natural ecosystems is root (or hyphae) uptake from the soil. This was not extensively studied before the Chernobyl accident possibly because the fallout from atmospheric weapons testing was deposited continuously on to plant surfaces over a long period, thereby masking the importance of root uptake. Nevertheless the high uptake of ^{137}Cs from soils with a high organic matter content in the upper layers was noted [14]. The low capacity of semi-natural ecosystems to immobilize radiocaesium is one of the main factors responsible for the continuing high radiocaesium levels in plants and animals. A soil in which a high rate of root uptake of radiocaesium occurs has been characterized in N. Europe as typically having a low clay and potassium content, a low pH, and a high organic matter content [15]. Radiocaesium is present in a bioavailable state in these soils, not strongly bound to soil components, and consequently available for uptake from the soil solution [15] Nevertheless, radiocaesium usually migrates slowly down the soil profile and remains for long periods in the upper soil layers where root biomass is greatest.

In soils of many semi-natural ecosystems the fungal hyphae are usually confined to the upper organic horizons of the soil. Assessments of fungal biomass and measurements of radiocaesium uptake by hyphae suggest that a high proportion of deposited radiocaesium may be present in the fungal mycelium [16, 17].

Radiocaesium uptake by vegetation is also affected by the rate at which it moves down the soil profile. For example, Beresford et al. [18] found that T_{ag} values for Chernobyl ^{137}Cs in vegetation in upland ecosystems in the UK were higher than those for pre-Chernobyl ^{137}Cs ("aged deposits"). They attributed this to the greater depth of the "aged deposit", over 50%

of which was below the 0–4 cm layer down the soil profile and therefore out of the rooting zone of the vegetation. Such distribution in the soil profile will obviously also affect T_{ag} estimates (for comparison <25% of Chernobyl ^{137}Cs was below 4 cm). It should be noted that T_{ag}s do not take into account the distribution of activity in the soil.

3.2.3. Radiocaesium uptake into vegetation

The proportion of radiocaesium that resides in vegetation of semi-natural ecosystems is often much greater than that which occurs in agricultural ecosystems. In Norway 0.1–0.5% of the total deposit has been transferred yearly from soil to above ground vegetation in the period 1986-1990 in mountain pastures [19]. In comparison, for agricultural crops values would usually be in the range 0.01% per year for cereals and root crops.

The observed high radiocaesium activity concentrations in mosses and lichen are primarily due to efficient interception and retention of the original deposit. However, fungi have the highest radiocaesium activity concentrations among all the species in semi-natural ecosystems. Radiocaesium levels in vascular plants can be ranked in the following order: ericoid dwarf shrubs > herbs > grasses > shrubs and trees (e.g. Refs [20, 21]), however, there are many exceptions.

3.2.4. Animal feeding habits

Herbivores vary widely in their feeding habits when grazing in semi-natural ecosystems. In addition to differences in feed selection between animal species, seasonal differences in the available forage plants contribute markedly to variation in radiocaesium activity concentrations recorded in animal food products. High radiocaesium activity concentrations have been reported for animal products from many semi-natural ecosystems. This is due to the high radiocaesium levels in the vegetation species eaten by animals combined with the comparatively high transfer of radiocaesium to small ruminant species such as sheep, goats and roe-deer which predominate in these ecosystems [5].

3.3. TRANSFER OF RADIOCAESIUM TO FOOD PRODUCTS FROM SEMI-NATURAL ECOSYSTEMS

A brief description of the characteristics of radiocaesium accumulation in a range of natural food products is given below, together with T_{ag} values.

3.3.1. Fungi

High radiocaesium activities have been reported in the fruiting bodies (mushrooms) of a number of fungal species both before and after the Chernobyl accident (e.g. Refs [22–25]). However, radiocaesium levels are highly variable among fungal species (e.g. Ref. [26]) and within species variation can be considerable [27, 28]. Radiocaesium activity concentrations in the fruiting bodies of some species (e.g. *Cantharellus cibarius, Leccinum testaceoscabrum*) are comparable with higher plants growing in the same area. In contrast, in other species radiocaesium activity concentrations which are 10–100 times higher than those of higher plants can be found (e.g. *Rozites caperatus, Suillus variegatus, Lactarius rufus*) [23, 25, 29]. The various reasons suggested as causes of the variation include differences in soil properties, hyphal distribution in the soil profile, mycorhizzal physiology and biochemical transportation properties [26, 28, 30, 31]. Taking the depth of the mycelium and precipitation during the 10 days following the Chernobyl accident into consideration, Nimis et al. [32]

were able to subdivide radiocaesium contamination of macrofungi into several ecological categories (parasites, symbionts with deciduous plants, symbionts with coniferous trees and saprophytes). The most highly contaminated genera and species have been ranked into ecological categories by Guillite [33] and are summarised in Table I.

TABLE I. EXAMPLES OF FUNGAL SPECIES IN DIFFERENT ECOLOGICAL CATEGORIES WHICH ARE GENERALLY MOST HIGHLY CONTAMINED WITH RADIOCAESIUM

Ecological category	Genera	Other species
Mycorrhizal	*Cortinarius* *Dermocybe* *Inocybe* *Rozites* *Xerocomus* *Suillus* *Tylopilus* *Paxillus* *Hygrophorus* *Laccaria*	(thought to be facultative mycorrhiza species) *Lactarius rufus* *L. theiogalus* *L. camphoratus* *Cantharellus tubaeformis* *Tricholoma album* *Amanitopsis* (sub-genus of *Amanita*)
Saprophytic	*Clitocybe* *Lepista* *Cantharellula*	especially *C. clavipes*
Parasitic	*Armillaria*	*Pholiota squarrosa*

Changes in fungal radiocaesium levels with time since the Chernobyl accident generally depend on the depth of the mycelium. It has been suggested that the gradual migration of radiocaesium into deep soil layers will increase the uptake of radiocaesium by fungal species which have mycelium in deeper layers [24, 27, 28, 30, 34, 35]. Radiocaesium levels in fungi with deeper mycelium, such as *Boletus edulis*, had comparatively low levels soon after the Chernobyl accident [30, 36, 37] and also a low ratio of ^{134}Cs:^{137}Cs which indicates an uptake of nuclear weapons test fallout. In the first year after deposition of Chernobyl fallout, Wirth et al. [38] found that saprophytic species, which have a superficial mycelium in the upper litter horizon (L-horizon), were the most highly contaminated. After the first year following deposition the pattern changed and the mycorrhizal (symbionts) and parasitic fungi had higher radiocaesium activities than the saprophytes [38].

Few data in the literature are presented in an appropriate form, giving activity concentrations in fungi and ground deposition in soil which allow T_{ag} estimations to be made. Where possible T_{ag} for different species have been estimated and are presented in Table II. Dry weight conversions have been carried out where necessary assuming 8% dry matter in fungi.

Changes in mean activity concentrations with time in a few species from the most comprehensive data set available from Finland are also known in Fig. 1 [40, 41].

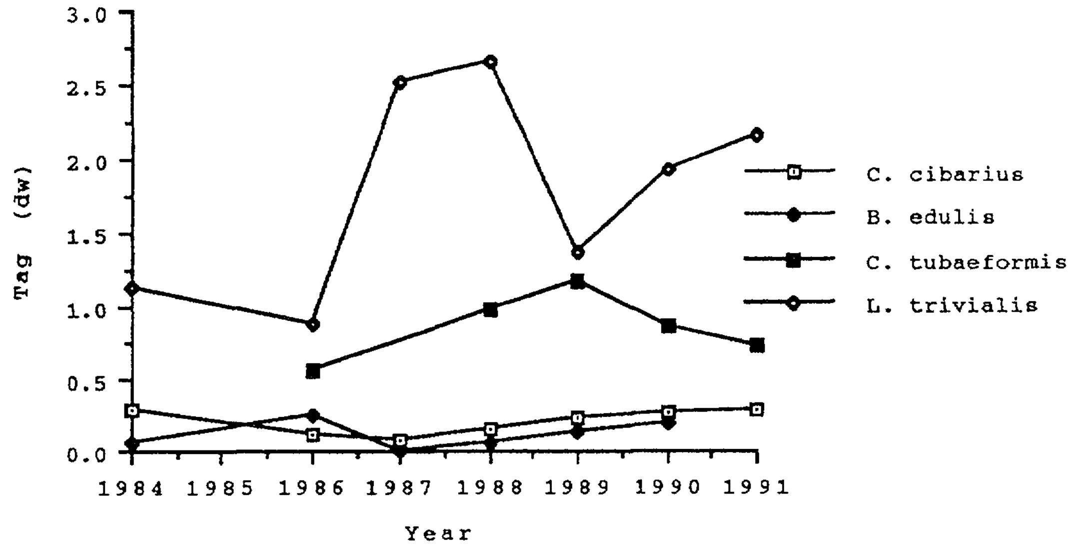

FIG. 1. Changes with time in mean T_{ag}s for Cs-137 in mushrooms of four fungal species [40, 41].

Even an intake of comparatively small quantities of fungi may transfer substantial activities of ^{137}Cs due to the high activity concentrations found in some fungi. In Sweden for example, about 24 million kg fw of mushrooms are picked each year, corresponding to about 3 kg fw per person (about 0.25 kg dw) [47]. A mean T_{ag} based on dry weights for all fungal species picked within one area in Sweden during 1988 to 1991 was 1.25 m^2 kg^{-1} [48]. This implies that the mean ^{137}Cs activity concentration for mushrooms in Sweden, where the mean deposition was 10 kBq m^{-2}, will be 12.5 kBq kg^{-1} dw, and the mean annual individual intake of ^{137}Cs from mushroom could be 3 kBq.

Similarly a small proportion of the population in Czechoslovakia, which were assumed to have an annual consumption of about 5 kg fw of mushrooms, were estimated to have a potential radiocaesium intake from fungi of 1.5 kBq y^{-1} [49]. In contrast, other foodstuffs from agricultural systems contributed 4-5 kBq during the first year after deposition, but only 0.5 kBq y^{-1} thereafter.

When using T_{ag}s for modelling transfer to humans it is important to note that we have not considered the reduction in ^{137}Cs activity concentrations that occur during food processing which can exceed 90% in mushrooms [50]. If people eat species with low transfers of radiocaesium and/or cook them using a method which reduces the radiocaesium content they will consume much lower amounts of radiocaesium.

Information given to the public after a nuclear accident may change food consumption habits markedly. In many countries affected by high deposition after the Chernobyl accident people were advised to reduce their consumption of species with particularly high radiocaesium content. In a household survey in Finland in 1990 the consumption of fungi was found to be reduced by 50% compared with a similar survey in 1985, while the consumption of wild berries was reduced by 30% [51].

TABLE II AGGREGATED TRANSFER COEFFICIENTS FOR RADIOCAESIUM IN VARIOUS FUNGAL SPECIES CALCULATED FOR DIFFERENT YEARS AFTER THE CHERNOBYL ACCIDENT

Species	References	Year	T_{ag} ($m^2\ kg^{-1}$ dw)			
			Mean	Median	Min	Max
Boletus edulis	[40,41]	1984	0 06		0 03	0 09
	[27]	1986	0 06			
		1986	0 25	0 02	<0 01	1 69
	[40, 41]	1987	0 01	0 01	<0 01	0 02
		1988	0 05	0 04	<0 01	0 19
		1989	0 13	0 05	0 02	0 32
		1990	0 20		0 01	0 38
		1991	0 06			
	[42]	1988-91	0 06	0 05	0 03	0 13
	[43]	1987-91	0 07	0 07	0 05	0 10
	[44,45]	1984	0 28	0 33	0 04	0 42
Cantharellus cibarius	[40,41]	1986	0 12	0 09	0 02	0 54
		1987	0 08	0 04	0 02	0 36
		1988	0 15	0 09	0 02	0 43
		1989	0 23	0 29	0 03	0 38
		1990	0 26	0 18	0 02	0 66
		1991	0 29	0 20	0 01	0 96
	[42]	1988-91	0 35	0 28	0 02	1 00
	[43]	1987-91	0 12	0 11	0 05	0 20
	[44,45]	1986	0 55	0 39	0 28	1 53
Cantharellus tubaeformis	[40,41]	1988	0 98	1 08	0 44	1 61
		1989	1 18	0 90	0 46	2 47
		1990	0 86	0 97	0 40	1 20
		1991	0 73	0 73	0 18	1 52
		1987-91	0 63	0 68	0 14	1 17
	[44,45]	1984	1 13	1 12	0 92	1 37
Lactarius trivialis	[40,41]	1986	0 89	1 03	0 02	3 00
		1987	2 51	2 01	1 65	4 50
		1988	2 66	2 63	0 85	5 15
		1989	1 37	1 02	0 07	3 47
		1990	1 92	1 91	0 69	4 14
		1991	2 15	2 21	1 17	3 07
		1984	0 10			
Leccinum versipelle	[40,41]	1986	0 05	0 03	0 01	0 14
		1987	0 14	0 06	0 01	0 57
		1988	0 11	0 05	0 02	0 74
		1989	0 06	0 05	0 02	0 11
		1990	0 06		0 02	0 11
		1988			1 88	3 13
Rozites caperatus	[2]	1990	2 25	2 15	1 89	2 70
	[40,41]	1991	1 54		0 78	2 30
		1987-91	0 08	0 08	0 01	0 14
Suillus granulatus	[44,45]	1986	1 70	1 44	0 29	4 80
Xerocomus badius	[40,41]	1987	1 25	0 84	0 58	3 90
	[46]	1988	3 22	2 30	0 44	7 10
		1988-91	2 19		0 43	6 10
	[43]	1987-91	0 55	0 45	0 19	1 37
	[44,45]	1987	1 29	0 76	0 23	5 20
Xerocomus chrysenteron	[2]	1988	1 63	1 38	0 64	2 50
		1989	0 03			
	[43,44,45]	1988-91	0 79	0 60	0 01	2 07

Since there are large variations both between and within different species, doses based on the mean values given in Table II are likely to be uncertain. Knowledge of T_{ag}s for fungi is, at present, fragmentary. Because of the high transfers of radiocaesium to fungi, and the potential importance for radiocaesium transfer to humans, it is clear that systematic collection of dry weight activity concentrations in fungi and deposition values for the soil in which they are growing are needed. An alternative approach to using T_{ag}s has been adopted by Wirth et al. [38] who have calculated transfer factors for soil to fungi based on the radiocaesium activity concentration in only the organic horizon of the soil. This method was suggested because fungal mycelium are preferentially found in organic horizons and are unlikely to absorb much radiocaesium from lower mineral horizons, where it will be strongly fixed. Such data, can, in some instances, be converted to calculate T_{ag}s (for example, Refs [44, 45], Table II).

Levels of ^{90}Sr in fungi are low compared with those of higher plants and correspond to the similarly low levels of its analogue calcium [26, 38]. Accumulation of ^{110m}Ag from the Chernobyl fallout in fungi was noted at activity concentrations of up to 500 Bq kg^{-1} dw in Slovenia [34].

3.3.2. Berries

Ericoid dwarf shrubs, such as bilberry (*Vaccinium myrtillus*) and lingonberry (also called cowberry) (*Vaccinium vitis-idaea*) accumulate comparatively high levels of radiocaesium [52, 53]. In northern parts of Europe cranberries (*Vaccinium oxycoccus*) and cloudberries (*Rubus chameomorus*), which grow on peaty, nutrient poor bogs, also have high levels of radiocaesium [53] Cloudberries growing in an area with a deposition of 35 kBq m^{-2} had radiocaesium activity concentrations of up to 8 kBq kg^{-1} dw.

Estimates of T_{ag}s for commonly consumed wild berries are listed in Table III. In comparison with many fungal species, transfer from soils to berries is low. Berries will therefore make only a small contribution to population dose, even when the average annual consumption of berries is high such as occurs in Nordic countries (e.g. 3–8 kg y^{-1} of fresh berries per person). However, as for fungi, there may be small critical groups who collect and eat large quantities of berries. It is possible that members of the critical group for fungi will also be in the critical group for berries.

TABLE III. AGGREGATED TRANSFER COEFFICIENTS FOR RADIOCAESIUM TRANSFER TO BERRIES USED FOR HUMAN CONSUMPTION

Species	T_{ag} (m^2 kg^{-1} dw)
Cloudberry	0.002–0.23^a; 0.13^b
Lingonberry (Cowberry)	0.04^a; 0.032^b
Bilberry (Blueberry)	0.017^c; 0.04^a; 0.041^b; 0.12^d

[a] Refs [40, 41].
[b] Ref.[53].
[c] Ref. [55].
[d] Ref. [56].

Data are also available from Ukraine which gives T_{ag} values for "fresh berries" of 0.001–0.004 m^2 kg^{-1}, which can be converted to dry weight using a dry matter content of 15% to 0.006–0.03 m^2 kg^{-1} [42]. Equivalent values for ^{90}Sr based on dry weight from this source are 0.0015–0.002 m^2 kg^{-1}.

3.3.3. Honey

The comparatively high levels of radiocaesium which are found in heather (*Calluna vulgaris*) result in high levels of radiocaesium in heather honey. Few data for transfer to heather honey are available, although in Germany heather honey was found to be nearly 100 times as highly contaminated as flower honey and levels of $^{239/240}Pu$ and ^{90}Sr were also higher in heather honey [57].

In Sweden a mean T_{ag} for ^{137}Cs for all types of honey was estimated to be about 0.005 m^2 kg^{-1} [58]. Since the mean ^{137}Cs deposition in Sweden is about 10 kBq m^{-2} the estimated mean activity concentration in honey will thus be 50 Bq kg^{-1}. The consumption of honey in Sweden is about 4 million kg or about 0.5 kg per person. Honey consumption by the critical group of bee keepers in Sweden is probable 10 times higher than the mean.

In Finland from 1977 and 1978 flower honey had a mean T_{ag} of 0.003 with a range from 0.00006–0.023 with an annual median of 0.0014. Pollen from ericaceous species dominated in honey samples with the highest T_{ag}s. From 1986 to 1990 the annual median T_{ag} value was 0.0010 with a range in T_{ag}s of 0.00004–0.01 [59].

Recent data for heather honey in Ireland in 1992 gave values of 0.08 (range 0.05–0.13) for honey from a lowland blanket bog, and 0.02 (range 0.01–0.02) from a mountain site with a shallow peat soil [60].

Few relevant data have been found on consumption of honey from semi-natural ecosystems. In Norway the average consumption of heather honey is 0.1–0.2 kg y^{-1} per person. However, the variation in intake is large, as it is for all foods from these ecosystems. The maximum concentration recorded in Norway is 2.6 kBq kg^{-1} per person [61]. Consumption by critical groups, estimated to consume 2–5 kg y^{-1} per person, would result in a maximum intake of 13 kBq y^{-1} per person.

3.3.4. Animals

(1) GAME ANIMALS

(i) Moose

Moose (*Alces alces*) graze mainly on herbs and leaves from shrubs in summer and increase their intake of more contaminated species such as bilberry and heather in the autumn [62]. However radiocaesium intake does not necessarily increase because there is a concommitant reduction in overall herbage intake [63].

Estimated T_{ag}s for moose are presented in Table IV. Mean T_{ag} values for calves have been found to be consistently higher than for adults [4]. The values quoted are usually mean values for T_{ag}s from within one area, but considerable temporal and spatial variation exists in some areas. For instance, in an area of Sweden where the mean value is 750 Bq kg^{-1} fw a variation from 100 Bq kg^{-1} to 3 kBq kg^{-1} was found [48]. Since the above T_{ag}s have been calculated from data from an area of Europe where most moose are found they would appear to be the most appropriate values to describe the transfer of radiocaesium for this particular foodchain from the forest ecosystem to man.

TABLE IV. AGGREGATED TRANSFER COEFFICIENTS FOR RADIOCAESIUM TRANSFER TO MOOSE

Year	T_{ag} (m^2 kg^{-1} fw)	References
1979	0.011-0.026 (all ages)	[64]
1985	0.015 (calves)	[4]
1985	0.010 (adults)	[4]
1986[a]-91	0.014 (calves)	[41]
1986[a]-91	0.010 (adults)	[41]
1986-90	0.009-0.032 (calves)	[4]
1986-90	0.006-0.017 (adults)	[4]
1986-88	0.02	[65]
1986-91	0.018-0.024	[8]
1989	0.009-0.019	[66]

[a]1986 data given [67].

Consumption of moose meat is only important in a few Nordic countries and in parts of the CIS and North America. In Sweden, for example, 135,000 moose were harvested during 1988 corresponding to 12 million kg of meat. This is equivalent to 1.5 kg per person, but the distribution of consumption is highly skewed and many of the critical group of hunters consume around 50 kg y^{-1} per person. In the Gävle district, the most contaminated area in Sweden, 938 moose were harvested in 1989 and the mean ^{137}Cs activity concentration was 1.3 kBq kg^{-1} fw. Each of the critical group of approximately 2000 hunters obtained meat containing 60 kBq (Fig. 2 [8]). During the period after the Chernobyl accident some yearly variation in ^{137}Cs levels has been noted, but no significant decreases in ^{137}Cs activity concentrations have been detected in moose meat [8]. Similarly both Bergman et al. [4] and Rantavaara [67] have found that T_{ag} values for ^{137}Cs were similar before and after the Chernobyl accident, indicating that the physical half-life of ^{137}Cs will determine the effective half-life.

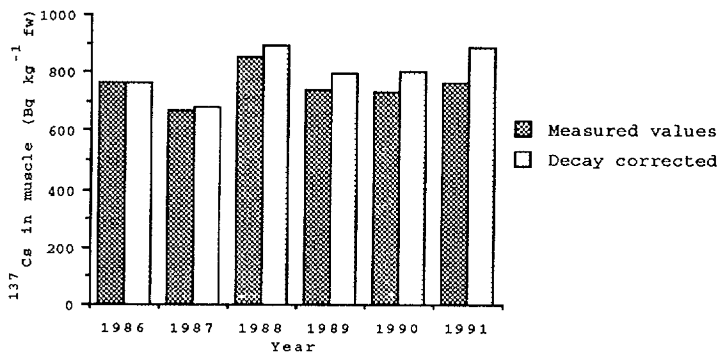

FIG. 2. Changes in muscle Cs activity concentration with time [8].

(ii) Roe-deer

Roe-deer (*Capreolus capreolus*) are the most common small wild deer in the areas which were most highly contaminated by Chernobyl deposition, and the small ruminant species for which we have the most comprehensive understanding of radiocaesium accumulation. It is therefore used as an example of the small deer for the present discussion. Unlike moose, roe-deer eat a wide variety of herbs, grasses and also fungi in large quantities when they are available.

Roe-deer have a pronounced annual variation in their ^{137}Cs activity concentrations with a peak in August-September when fungi are abundant. There is a considerable variation in the transfer of radiocaesium to roe-deer both between countries and even between areas within countries. Schönhofer & Tataruch [68] found high variation in small forested areas where roe-deer have access to agricultural land and suggested that this situation prevents the calculation of meaningful transfer coefficients.

The T_{ag} values presented in Table V are largely derived from, and therefore most relevant for, roe deer inhabiting Nordic forested areas with some access to farmland. In areas where farmland dominates less transfer of radiocaesium to roe-deer seems to occur.

TABLE V. AGGREGATED TRANSFER PARAMETERS FOR RADIOCAESIUM TRANSFER TO ROE DEER

Year	T_{ag} (m^2 kg^{-1} fw)	References
1988-89	0.15 (autumn)	[69]
1988-91	0.04 (rest of year)	[8]
1988-91	0.05 (annual mean)	[8]
1991	0.01-0.20	[70]

A roe-deer carcass provides about 10 kg of meat [8]. So, for example, 200 000 roe-deer harvested annually in Sweden provide a total of about 2 million kg of roe-deer meat, equivalent to 0.25 kg per person. However, this average figure is misleading since most of the meat is consumed within the critical group, the hunters and their families.

Reliable estimates of the T_{ef} for radiocaesium in roe deer have not, as yet, been possible in the Nordic countries due to the large seasonal variations in radiocaesium concentrations in roe deer. There is little evidence of a decline from 1988-1991 [8]. However, Lindner et al. [70] have calculated a T_{ef} of 2.6 y in Germany. In Scotland there has been a decrease from a typical value of 500 Bq kg^{-1} fw ^{137}Cs during 1986 to 170 in 1989 [71].

(iii) Other hunted animals

Other ruminants which are hunted such as red deer (*Cervus elaphus*), fallow deer (*Dama dama*) chamois (*Rubicapra rubicapra*), white-tailed deer (*Odecoileus virginianus*) have not been found to have particularly high levels of radiocaesium. Few data are available from which we can calculate T_{ag}s. Data from Scotland taken in 1989 gives approximate values for red deer of 0.02-0.07 m^2 kg^{-1} [72]. Considering the small volumes of meat from these animals consumed in most countries the contribution to collective dose is likely to be small.

In Norway, where levels of radiocaesium in 1986 were comparable to those in moose, about 10 000 red deer are harvested each year corresponding to about 700 000 kg meat.

T_{ag} values for white-tailed deer of 0.017 in 1986 and 0.03 in 1990 were estimated in Finland [41]. These values are slightly higher than those for moose in Finland. About 6000-8000 individual deer are hunted annually with an annual per capita meat consumption of 0.1 kg or less.

The few data available for Arctic hare (*Lepus timidus*) and Brown hare (*Lepus capensis*) from Finland and Sweden are shown in Table VI.

TABLE VI. AGGREGATED TRANSFER COEFFICIENTS FOR RADIOCAESIUM TRANSFER TO SOME SPECIES OF HARE

Species	n	Year	T_{ag} (m^2 kg^{-1} fw)			
			Mean	Median	Min	Max
Arctic hare	7	1967-68[a]	0.0009			
	75	1988-90[a]	0.038	0.027	0.006	0.104
	8	1986-89[b]	0.03		0.025	0.13
Brown hare	8	1967-68[a]	0.0021			
	11	1987-89[a]	0.008	-	0.0005	0.053
	11	1986-89[b]	0.003	-	0.00018	0.023

[a] Refs [40, 41].
[b] Ref. [73].

(iv) Wild boar

Wild boar (*Sus scrofa*) is hunted in many central and eastern European countries. Data from Austria [74] indicate that a wide range of values may be found even within the same forest area, possibly due to the large home range of wild boar. They reported typical ^{137}Cs activity concentrations of 37 Bq kg^{-1} fw for a forest contaminated with about 50 kBq m^{-2} in 1986, with levels apparently rising in 1988 to a maximum of 17.6 kBq kg^{-1} in February 1988. The calculation of T_{ag}s must await further data collection.

(v) Wildfowl and game birds

Some high levels of radiocaesium were reported in wildfowl during the first hunting season after the Chernobyl accident [75]. In Finland mean values of T_{ag} for both inland waterfowl and terrestrial game birds were 0.01 [41]. Lowe & Horrill [76] reported that ^{137}Cs levels in woodcock (*Scolopax rusticola*) muscle have been low when compared with ruminants. In willow grouse (*Lagopus mutus*) radiocaesium levels were much lower than in sheep grazing in the same area. Values for T_{ag} of 0.01-0.02 m^2 kg^{-1} can be calculated from the work of Pedersen [77]. In Norway there has been no noticeable decrease in ^{137}Cs levels in grouse. Radiocaesium levels in red grouse (*Lapogus lapogus*) and black grouse (*Tetrao tetrix*) are higher in Scotland than those recorded in Nordic countries, probably because these species of grouse eat large quantities of heather, whereas heather does not form an important part of the diet of willow grouse in the hunting season. Hence although the low levels in grouse meat from Nordic countries and the small quantities consumed imply that game birds are not contributing significantly to the doses from semi-natural ecosystems, the discrepancy between Scotland and Nordic Countries clearly shows that such conclusions would not necessarily be applicable for other ecosystems. It also demonstrates that species selected in the diet is important in determining radiocaesium levels in birds as well as in other animals inhabiting semi-natural ecosystems.

(vi) Reindeer

The transfer of ^{137}Cs through the food chain from lichens to reindeer to man was studied during the period of the above-ground nuclear tests. Effective half-lives of between 3 and 14 years were estimated from measurements of both reindeer meat and whole body monitoring of humans with intakes of large quantities of reindeer meat [78–83]. However, these studies did not take account of the continuing deposition of nuclear weapons fallout and, if they had, the calculated half-lives would have been more rapid. in some studies [82, 84] there seems to be a slower decrease in whole body contents after 1975 than before. During the period 1986 to 1992 effective half-lives have been recorded for ^{137}Cs in reindeer meat in Sweden of between 3.2 and 4.2, depending on the season [85].

Lichens form the major part of the diet of reindeer in the winter period and therefore contamination rates of lichen determine radiocaesium levels in reindeer in this period. Since lichens are directly contaminated, estimations of the bio-availability of radiocaesium in the lichen is more important than in green vegetation where contamination after the initial deposition occurs mainly by root uptake.

Values of T_{ag} for reindeer estimated during the first winter after the deposition of Chernobyl fallout were 0.6–1.1 $m^2\ kg^{-1}$. When radiocaesium levels in reindeer were at their lowest (late July and early August) the estimated T_{ag} in Sweden was about 0.025 [86]. A suitable T_{ag} for reindeer meat is difficult to define because lichens which are initially highly contaminated dilute the radiocaesium activity concentration by new growth. Nevertheless, the total deposition (Bq m^{-2}) remains the same and therefore it is particularly important to incorporate an estimate of t_{ef}. Due to grazing of contaminated biomass and dilution by new growth the radiocaesium content in lichen declined with a t_{ef} of 3–4 years in Norway [87] and around 7–10 years for both pre- and post-Chernobyl ^{137}Cs in reindeer lichen in Sweden [88]. Other post-Chernobyl studies in Sweden show a t_{ef} for ^{137}Cs in reindeer meat in the first few years after the accident (1986–1991) of between 3 and 4 years during the winter period [89]. Since lichen is the main feed source for reindeer (>75%) [90], use of the ratio between radiocaesium levels in lichen and reindeer meat during the winter season are probably a more appropriate and accurate measure than aggregated transfer coefficients. Data collected during field studies in Norway in 1987–1989 in November after the Chernobyl accident give a range of 1.0–1.2 kg^{-1} for meat per kg^{-1} dw in pooled samples of lichens collected from different locations within the grazing range of the studied herd. Therefore a meat to lichen ratio of 1.0 Bq kg^{-1} of meat per Bq kg^{-1} dw of lichen can be recommended.

The ^{137}Cs activity concentrations in reindeer during summer and early autumn have not been considered to be as important as those during the winter because they are generally about 10 to 20% of the latter. After the Chernobyl accident a recommendation to change the slaughter times in some Nordic countries was made in order to take advantage of the low summer values, thus decreasing the dose commitment to man. Reindeer generally graze the same pastures as sheep during summer, and the t_{ef} for lambs in these ecosystems has been much longer than for the lichen-reindeer foodchain. Differences between winter and summer activity concentrations in reindeer meat may therefore become less pronounced as time passes in areas where substantial root uptake is taking place. Finnish studies from 1983 of the ^{137}Cs levels in various fodder plants for reindeer found that lichens still had higher ^{137}Cs activity concentrations than most other fodder plants, but the differences were small [91]. During the period 1987–1992 in Norway both winter and summer radiocaesium activity concentrations in reindeer meat have decayed with a t_{eff} of 3.5 y. Radiocaesium activity concentrations in green vegetation six years after the Chernobyl accident were still below 10% of those in lichens. In this situation a lichen intake during the summer season of only 10–15% dw will dominate radiocaesium levels in meat to the extent that the t_{ef} will still approach that of lichens.

Radiocaesium levels in a substantial fraction of reindeer meat in Norway and Sweden have been above the intervention levels (6 and 1.5 kBq kg^{-1} fw respectively) at the normal time of slaughter since the Chernobyl accident. Considerable efforts have been made to reduce the radiocaesium levels in reindeer. As for game products, consumption of reindeer meat is not evenly distributed in the population. Average values for meat consumption of 0.3–0.5 kg y^{-1} in Scandinavian countries will contribute a maximum of 3 kBq y^{-1} of ^{137}Cs when eating meat with an activity concentration at the intervention level. In the critical group of hunters and reindeer herders much higher intakes have been noted; a maximum annual dose of 13 mSv was calculated based on whole body counting in Norway [7].

(2) DOMESTICATED RUMINANTS

(i) Sheep

In many countries sheep meat is the most important food product obtained from semi-natural ecosystems. Before the Chernobyl accident little emphasis was placed on measurement of radiocaesium levels in sheep from semi-natural compared with agricultural ecosystems, although there was some indication, based on limited data, that radiocaesium levels in upland sheep were elevated [92]. After the Chernobyl accident, high levels were observed in sheep from semi-natural ecosystems in many affected countries. The values observed exceeded intervention limits in the United Kingdom, Ireland, Norway, Sweden and Austria. For sheep grazing in semi-natural ecosystems the major factors responsible for the persistently high radiocaesium activities in meat are the comparatively high root uptake of grazed vegetation and, in certain areas, the selective intake by sheep of highly contaminated fungi and ericaceous species. Post-Chernobyl measurements of the transfer of radiocaesium to lamb has shown that the previously assumed transfer coefficient (F_f) for lamb was too low.

Major efforts have been undertaken to study the transfer of radiocaesium to sheep in semi-natural ecosystems because of the importance of sheep production in affected countries. It has become clear that there are considerable difficulties in determining accurate values for various transfer parameters for sheep grazing in semi-natural ecosystems. It is particularly difficult to estimate radiocaesium intake in freely grazing sheep. Therefore more effort has been devoted to trying to calculate T_{ag}s, particularly in the Nordic countries, where a collaborative study is in progress. Estimated pre- and post-Chernobyl values for T_{ag}s for sheep meat are given in Table VII.

TABLE VII. AGGREGATED TRANSFER COEFFICIENTS FOR RADIOCAESIUM TO SHEEP MEAT

Radiocaesium Source	Year	T_{ag} (m^2 kg^{-1} fw)		Comments, Site, References
		[a]Large area	[b]Pasture	
Nuclear weapons tests	1965	0.020-0.050		Norway [2]
	1991	0.011		ewes: Cumbria, UK [93]
Chernobyl	1988-90		0.020-0.030	Coastal, Norway [2]
	1988		0.05	Mountain pasture, Central Norway [2]
	1988		0.043	lambs: Cumbria, UK [93]
	1988		0.025	ewes: Cumbria, UK [93]
	1989		0.030-0.050[s]	ewes: Scotland
	1990		0.01-0.074	Nordic study [94]
	1991	0.036		ewes: Cumbria, UK [93]
		0.09[w]		ewes: Heather moor, Ireland [60]
		0.15[s] (0.03-0.29)		ewes: Heather moor, Ireland [60]

[a]Large area: Deposition calculated from a large area (several km^2).
[b]Pasture: Deposition measured in the specific pasture in which the sheep were grazing.
[w]Winter.
[s]Summer.

All T_{ag} values fall within the range 0.01 to 0.29. The highest values were obtained for ewes grazing on a montane blanket bog covered with shrub vegetation dominated by *C. vulgaris* in Ireland (calculated from data given in [95]. T_{ag} values of sheep in agricultural ecosystems are much lower. For instance ten-fold lower values have been found for agricultural ecosystems compared with semi-natural ecosystems in the same district in Norway.

In some intensively studied areas in Norway where estimates of T_{ag} are available for consecutive years after the Chernobyl accident, the maximum variation has been 50% prior to the autumn period [96]. In some of the contaminated pastures in Norway fungi occasionally produce a substantial crop of fruiting bodies. When this happened in 1988, consumption of highly contaminated mushrooms by sheep led to a 3-4 fold increase in T_{ag} values [97].

Although radiocaesium levels have declined in several areas, sheep from semi-natural ecosystems in Norway and Sweden, Ireland. In some closely monitored flocks in Ireland negligible declines in ^{137}Cs activity concentrations have taken place in the last few years [95].

This parallels the long t_{ef} values which have been calculated for above ground nuclear weapons test ^{137}Cs in Norway [2]. Similarly observations in the UK show that four years after deposition Chernobyl radiocaesium is being taken up by plants to a similar extent as aged deposits of ^{137}Cs, suggesting that future reductions in radiocaesium levels in vegetation, and therefore sheep, will be slow and predominantly due to physical decay and migration of radiocaesium down the soil profile [18]. T_{ag} values for ewe muscle for ^{137}Cs from "aged deposits" in this area in December 1991 of 0.011 were lower than those for Chernobyl ^{137}Cs of 0.036 [93]. Because conventional transfer coefficients (i.e. F_m and F_f) incorporate total daily radionuclide intake, they give higher values for radiocaesium transfer to animal products such as milk and meat from small ruminants compared with large ruminants, because food intake and also, therefore, radionuclide intake is proportional to metabolic body size. T_{ag}s remove this effect, nevertheless higher transfer of many radionuclides, including radiocaesium, occurs to sheep and goat milk compared with cow milk [98]. Consequently comparatively high levels of radiocaesium (and radioiodine) in sheep (and goat) milk will be found in contaminated areas and this is particularly important in those countries where milk is obtained from sheep grazing semi-natural ecosystems.

For many countries it is not possible to distinguish consumption of sheep meat from semi-natural ecosystems from that produced in agricultural ecosystems. However, in Norway the great majority of sheep are slaughtered when returning from summer grazing in semi-natural ecosystems. Care should be taken when t_{ef} is calculated in meat samples of unknown origin, as a random set of samples may contain meat from animals grazing in both semi-natural and improved pastures. An additional difficulty arises from the use of countermeasures during the grazing period, since reductions obtained by using prussian blue or feeding with clean feeds may cause reductions which vary both betwen districts and years.

(ii) Goats

Dairy goats graze in semi-natural ecosystems in many parts of the world, and as for sheep, transfers of radiocaesium to goat milk is high compared with cows milk [13]. Long effective half-lives of approximately 20 y have also been observed in semi-natural ecosystems for ^{137}Cs in goat milk products [2]. T_{ag}s for goat milk of 0.002-0.004 were calculated from Norwegian data for the period when ingestion of fungi was negligible, T_{ag} values increased 2-4 fold in years with a comparatively high fungal abundance [2, 97, 99]. It is common practice in Norway to feed 10-25% of the dry matter intake as concentrates, depending on pasture conditions. This feed usually has very low radiocaesium activity concentrations, which should be taken into account as a source of variation when estimating T_{ag}s for goat milk products.

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