



# Detection limits for $^{90}\text{Sr}$ , Pu, Am and Cm in soil and pasture vegetation shortly after a nuclear accident

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## Abstract

This work estimates the critical activity concentrations of  $^{90}\text{Sr}$  and the  $\alpha$ -emitting isotopes of Pu, Am and Cm in soil and pasture vegetation that would be required to exceed the action levels for foodstuffs recommended by the IAEA. The results show that the common detection limits for environmental analysis of these nuclides may be increased by orders of magnitude if the aim of the analysis is to determine whether or not the action levels will be exceeded. This information is useful in the development of more simple and rapid analytical methods to be used shortly after a nuclear accident. In addition to activity concentrations, the critical deposition densities on soil and grazing areas are estimated. Critical limits are also derived for  $^{137}\text{Cs}$  and  $^{131}\text{I}$ . © 1999 Elsevier Science Ltd. All rights reserved.

## 1. Introduction

Environmental analysis of  $\gamma$ -emitting radionuclides, such as  $^{137}\text{Cs}$  and  $^{131}\text{I}$ , is relatively easily carried out, whereas the analysis of the pure  $\beta$ -emitter  $^{90}\text{Sr}$  and the  $\alpha$ -emitting isotopes of Pu, Am and Cm is more complicated, involving sample pretreatment (e.g. ashing and leaching) and a radiochemical separation procedure. Many of the currently employed methods for determining  $^{90}\text{Sr}$ , Pu, Am and Cm in environmental materials are designed to reach very low detection limits. These methods are well adapted for investigations of the environmental behavior of different radionuclides and in thorough analyses of a radioactive fallout. However, the analytical procedures are generally time-consuming and labor intensive and there is a lack of simple and

rapid radioanalytic methods suitable in urgent situations and/or when large amounts of samples have to be processed, such as shortly after a nuclear accident.

The lowest activity concentrations detectable with most methods are well below the limits at which any counter-measures would be required. The detection limits shortly after an accident, however, do not necessarily have to be lower than to clarify if any actions have to be taken. It is therefore possible that more simple and rapid methods can be developed by allowing increased detection limits. This would shorten the counting times and/or justify the use of smaller samples. The latter would simplify the chemical separation process and decrease the need of time-consuming evaporations for reducing the sample volume.

The required detection limits can be derived from the critical activity concentrations in different environmental materials that would require actions to be taken. However, no such critical limits for  $^{90}\text{Sr}$ , Pu, Am and Cm, applicable to the case of a nuclear acci-

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dent, appear to have been recommended in the literature. The IAEA (1994) recommends generic action levels for foodstuffs to be used after nuclear accidents, but no limits for environmental materials are given.

In this work, we relate the action levels of  $^{90}\text{Sr}$ , Pu, Am and Cm given by the IAEA to activity concentrations in soil and pasture vegetation and regard these concentrations as the critical limits which would require actions to be taken. In addition, the action levels are related to critical deposition densities on soil and grazing areas. The study is divided into three parts: (1) transfer from soil to plant products (e.g. fruit and vegetables) via root uptake, (2) transfer from well-mixed soil to animal products (e.g. milk and meat) via root uptake by pasture vegetation and soil ingestion by grazing animals and (3) transfer to animal products from direct deposition on pasture vegetation. The values of the transfer factors used in the calculations are, chiefly, the default values used by the National Radiological Protection Board (NRPB) in food-chain modelling (Brown and Simmonds, 1995; NRPB, 1998). Some transfer factors derived from bomb-fallout data are also used.

The transfer to plant products from direct deposition on plants is not considered in the estimations of the critical deposition densities. This transfer depends on several factors, including the agricultural conditions at the time of deposition and the weather conditions. Worst-case limits for different plant products can be estimated from the crop yield ( $\text{kg}/\text{m}^2$ ) and the action levels for foodstuffs ( $\text{Bq}/\text{kg}$ ).

For comparison, the corresponding values (action levels, deposition densities, activity concentrations) for  $^{137}\text{Cs}$  are included throughout this work. In the case of direct deposition on pasture vegetation, also the short-lived  $^{131}\text{I}$  ( $t_{1/2} \sim 8$  d) is included. The critical activity levels of these nuclides are interesting, but increased detection limits would not significantly simplify the analytical procedure.

The action levels for the radionuclides of interest in this work are given in Table 1. These levels agree with

the international standards for food trading after nuclear accidents (FAO/WHO, 1991). The nuclides are grouped depending on their dose per unit intake factor. In the group of  $\alpha$ -emitters, one may include  $^{238-240,242}\text{Pu}$ ,  $^{241,243}\text{Am}$  and  $^{242-244}\text{Cm}$  (although the action level for  $^{242}\text{Cm}$  may be set higher due to its lower radiotoxicity). It is, however, highly unlikely that all these nuclides would be of similar importance in a release/fallout. To estimate the critical limits for the individual nuclides, we will assume that five, or less, of them will contribute significantly to the total  $\alpha$ -activity, as would be the case in a fallout from a nuclear reactor accident (Friberg, 1998).

## 2. Results and discussion

### 2.1. Transfer from soil to plant products via root uptake

The common soil-to-plant transfer factors (TFs) refer to radionuclides that are well-mixed in the soil. The critical activity concentrations in well-mixed soil can therefore be obtained directly from the TFs and the action levels for foodstuffs. Radionuclides deposited on top of the soil will, however, essentially stay in the top soil layer and not be mixed with the deeper layers until the soil is ploughed. For analyses of undisturbed soils, we assume that the radionuclides will be retained in the upper 2-cm layer until ploughing and, thus, that the required sample depth is 2 cm. The vertical distribution of radionuclides in this layer will be somewhat heterogenous, but we assume that the average activity concentration of this layer can be compared with the critical concentrations obtained for well-mixed soil. This assumption should not underestimate the transfer from undisturbed soil to plants, e.g. when the deposition occurs shortly after sowing, as the root-systems usually reach below the penetration depth of the deposited radionuclides. We consider ploughed soil to be well-mixed, as far as our calculations are concerned.

Table 1  
Generic action levels for foodstuffs recommended by the IAEA<sup>a</sup>

Radionuclide(s)	Foods destined for general consumption (Bq/kg)	Milk, infant foods and drinking water (Bq/kg)
Sr-90	100	100
Pu, Am, Cm ( $\alpha$ -emitters)	10	1
Cs-137	1000	1000
I-131	1000	100

<sup>a</sup> Modified excerpt from IAEA (1994). The levels refer to food prepared for consumption and to the summed activity of the radionuclides in each group. Several fission products (other than  $^{131}\text{I}$ ) were originally included in the same group as  $^{137}\text{Cs}$ .

Table 2

Activity concentrations in well-mixed soil required to exceed the generic action levels for plant products

Nuclide(s)	Transfer factor <sup>a</sup> (Bq/kg food per Bq/kg soil)	Soil concentration (kBq/kg)
Sr-90	$3 \cdot 10^{-1}$ (green vegetable)	0.3
Pu	$3 \cdot 10^{-4}$ (fruit)	33
Am, Cm	$8 \cdot 10^{-4}$ (fruit)	12
Cs-137	$2 \cdot 10^{-2}$ (fruit)	50

<sup>a</sup> Taken from the NRPB (Brown and Simmonds, 1995; NRPB, 1998). The highest TFs were selected among those presented (domestic fruit, potatoes and root vegetables, green vegetables and cereals).

#### 2.1.1. TFs obtained from the NRPB

Table 2 displays a selection of soil-to-plant TFs obtained from the NRPB, along with the resulting critical activity concentrations in well-mixed soil.

The values used by the NRPB appear to be restrictive since experiments with different types of soils have indicated that the true TFs are more than a factor of two lower for <sup>90</sup>Sr and more than a factor of 10 lower for Pu/Am, compared with the values listed in Table 2 (Green et al., 1997). Consequently, there should be a safety margin included in the critical soil concentrations presented.

#### 2.1.2. TFs obtained from bomb-fallout data

To check for indications of any major deviations in the case of a real deposition, TFs are also derived from bomb-fallout data. These data refer to a continuous deposition but some information applicable to the case of short-term release after an accident can be extracted. The transfer of <sup>90</sup>Sr and <sup>137</sup>Cs from continuous deposition to diet has been modelled with the following formula (UNSCEAR, 1993):

Table 3

Activity concentrations in well-mixed soil required to exceed the generic action levels for plant products. Calculated from bomb-fallout data

Nuclide(s)	Transfer factor <sup>a</sup> (Bq/kg food per Bq/m <sup>2</sup> )	Soil concentration <sup>b</sup> (kBq/kg)
Sr-90	$0.3 \cdot 10^{-3}$ (vegetables)	0.7
Pu	$6.5 \cdot 10^{-7}$	34
Am	$1.5 \cdot 10^{-6}$	15
Cs-137	$1.1 \cdot 10^{-3}$ (fruit)	2

<sup>a</sup> Modified from UNSCEAR (1982, 1993). The highest TFs for <sup>90</sup>Sr and <sup>137</sup>Cs were selected among those presented for different foodstuffs. The values for Pu and Am are calculated from average concentrations in food.

<sup>b</sup> Assuming the deposition to be well-mixed in a 30-cm ploughing layer (soil density = 1.5 kg/dm<sup>3</sup>).

$$C_i = b_1 \cdot F_i + b_2 \cdot F_{i-1} + b_3 \cdot \sum_{n=1}^{\infty} e^{-\lambda n} \cdot F_{i-n},$$

where  $C_i$  (Bq/kg) is the concentration of the nuclide in a certain food component in the year  $i$  due to: the deposition density rate in the same year,  $F_i$  (Bq m<sup>-2</sup> y<sup>-1</sup>); the deposition density rate in the previous year,  $F_{i-1}$ ; and the deposition density rate in all previous years,  $F_{i-n}$ , reduced by exponential decay (both radioactive decay and environmental loss). The transfer coefficients  $b_x$  (Bq y kg<sup>-1</sup> per Bq m<sup>-2</sup>) reflect the transfer per unit annual deposition:  $b_1$  is the transfer in the first year, primarily from direct deposition;  $b_2$  is the transfer in the second year from lagged use of stored foods and uptake from the surface deposit; and  $b_3$  is the transfer via root uptake from the accumulated deposit. The transfer coefficients for the different nuclides have been calculated from experimental data on  $C$  and  $F$  during a number of years. The transfer of Pu and Am (but not Cm) from deposition to diet has been modelled in a similar way (UNSCEAR, 1982).

Table 4

Transfer from animal feed to foodstuff. Transfer factors

Nuclide(s)	Transfer factor <sup>a</sup> (Bq/kg food per Bq/day of animal intake)				
	Milk	Meat		Liver	
		Cattle	Sheep	Cattle	Sheep
Sr-90	$2 \cdot 10^{-3}$	$3 \cdot 10^{-4}$	$3 \cdot 10^{-3}$	$3 \cdot 10^{-4}$	$3 \cdot 10^{-3}$
Pu, Am, Cm	$1 \cdot 10^{-6}$	$1 \cdot 10^{-4}$	$4 \cdot 10^{-4}$	$2 \cdot 10^{-2}$	$3 \cdot 10^{-2}$
Cs-137	$5 \cdot 10^{-3}$	$3 \cdot 10^{-2}$	$5 \cdot 10^{-1}$	$3 \cdot 10^{-2}$	$5 \cdot 10^{-1}$
I-131	$5 \cdot 10^{-3}$	$2 \cdot 10^{-3}$	$5 \cdot 10^{-2}$	$2 \cdot 10^{-3}$	$5 \cdot 10^{-2}$

<sup>a</sup> Taken from the NRPB (Brown and Simmonds, 1995; NRPB, 1998).

Table 5

Transfer from well-mixed soil to animal products. Activity concentrations in the soil required to exceed the generic action levels for foodstuffs

Nuclide(s)	Activity concentration in well-mixed soil* (kBq/kg)				
	milk	meat		liver	
		cattle	sheep	cattle	sheep
Sr-90	12	83	49	83	49
Pu	$2.0 \cdot 10^3$	200	83	1.0	1.1
Am, Cm	$1.8 \cdot 10^3$	175	81	0.9	1.1
Cs-137	77	13	3.8	13	3.8

\* Calculated from the action levels, the feed-to-food TFs given in Table 4 and the soil-to-pasture TFs given in the text. The daily intakes by grazing animals used in the calculations were: 14 kg feed (dry weight) and 0.5 kg soil for cattle, and 1.5 kg feed (dry weight) and 0.3 kg soil for sheep. The dry weight of the pasture vegetation was set to 20% of the fresh weight.

Table 6

Transfer to animal products from direct deposition on pasture vegetation under assumptions described in the text. Animal intakes, activity concentrations in feed and ashed pasture vegetation and deposition densities, required to exceed the generic action levels for foodstuffs

Food type	Nuclide(s)	Animal intake (kBq/day)	Act. conc in dry pasture vegetation (kBq/kg)	Act. conc. in ashed vegetation (kBq/kg)	Deposition density (kBq/m <sup>2</sup> )
Milk	Sr-90	50	3.6	18	0.71
	Pu, Am, Cm	1000	71	360	14
	Cs-137	200	14	71	2.9
	I-131	20	1.4	7.1	0.29
<i>Meat</i>					
Cattle	Sr-90	330	24	119	4.8
	Pu, Am, Cm	100	7.1	36	1.4
	Cs-137	33	2.4	12	0.48
	I-131	500	36	179	7.1
Sheep	Sr-90	33	22	111	4.4
	Pu, Am, Cm	25	17	83	3.3
	Cs-137	2	1.3	6.7	0.27
	I-131	20	13	67	2.7
<i>Liver</i>					
Cattle	Sr-90	330	24	119	4.8
	Pu, Am, Cm	0.5	0.036	0.18	0.007
	Cs-137	33	2.4	12	0.48
	I-131	500	36	179	7.1
Sheep	Sr-90	33	22	111	4.4
	Pu, Am, Cm	0.33	0.22	1.1	0.044
	Cs-137	2	1.3	6.7	0.27
	I-131	20	13	67	2.7

The animal intakes are obtained from Tables 1 and 4. The values used in the calculations are: a daily feed intake of 14 and 1.5 kg/d (dry weight) for cattle and sheep, respectively; a pasture vegetation ash content of 20% of the dry weight; a pasture veg. density of 0.2 kg/m<sup>2</sup> (d.w.).

The two first coefficients,  $b_1$  and  $b_2$ , include the transfer from continuous direct deposition during a whole year and they are therefore difficult to apply to the case of a short-term release. However, the value of  $b_3$  should reflect, or at least indicate the magnitude of, the long-term transfer from soil to food after an accident. Table 3 shows the (unit-modified)  $b_3$  transfer factors for different nuclides along with the corresponding soil concentrations assuming a 30 cm deep ploughing layer. For  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ , the highest  $b_3$ s were selected among those given for grain products, vegetables and fruit in either Argentina, Denmark or the US. The  $b_3$ s for Pu and Am have been given as an average food concentration and we assume an annual food intake of 540 kg. Although the uncertainties in the transfer factors derived this way might be large, at least for Pu and Am, it is useful to compare the soil concentrations in Tables 2 and 3 as they have been obtained in different ways.

As seen by comparing Tables 2 and 3, the critical soil concentrations for the different nuclides agree rather well, except for  $^{137}\text{Cs}$  (the very good agreement for the  $\alpha$ -emitters is likely to be a coincidence). It appears reasonable to select the lowest values as the critical activity concentrations in well-mixed soil, i.e. 0.3 kBq/kg for  $^{90}\text{Sr}$ , 12 kBq/kg for the group of  $\alpha$ -emitters and 2 kBq/kg for  $^{137}\text{Cs}$ . Since only five or less of the  $\alpha$ -emitters will be of importance, the critical concentration for each of them can be set to 2 kBq/kg.

### 2.1.3. Deposition densities

For a 2 cm deep sample of undisturbed soil ( $\rho = 1.5 \text{ kg dm}^{-3}$ ), the deposition densities corresponding to the critical concentrations are 9 kBq/m<sup>2</sup> for  $^{90}\text{Sr}$  and

60 kBq/m<sup>2</sup> for each of the  $\alpha$ -emitters. If only root uptake has to be considered (perhaps shortly after sowing), these values are the approximate critical deposition densities on soil below which no actions have to be taken. For a sample of soil mixed by ploughing and assuming a plough layer of 30 cm, the corresponding deposition densities are 135 and 900 kBq/m<sup>2</sup>. These values are the approximate critical deposition densities on soil below which ploughing (i.e. dispersing the radionuclides more evenly through the soil profile) would be sufficient for growing edible plants without exceeding the action levels. Higher deposition densities would require further actions to be taken (e.g. restrictions in the use of plants with high uptake or removal of topsoil layer).

Similarly, the critical deposition densities for  $^{137}\text{Cs}$  become 60 and 900 kBq/m<sup>2</sup>. This nuclide requires, however, a more thorough consideration of the external exposure.

### 2.2. Transfer from well-mixed soil to animal products

We assume that the animals get their full feed intake by grazing on contaminated land and that the grazing continues for a sufficient time period to reach up to the equilibrium TFs listed in Table 4 (the TFs for iodine are used in the next section). The critical activity concentrations in well-mixed soil for the transfer to animal products via root uptake and soil ingestion are shown in Table 5. The soil-to-pasture TFs (Bq/kg fresh vegetation per Bq/kg dry soil) used in the calculations are:  $5 \cdot 10^{-2}$  (Sr),  $1 \cdot 10^{-4}$  (Pu),  $1 \cdot 10^{-3}$  (Am, Cm) and  $3 \cdot 10^{-2}$  (Cs), as given by the NRPB (Brown and Simmonds, 1995). The values used for the daily intakes

Table 7  
Summary of critical activity concentrations required to exceed the generic action levels for foodstuffs

Nuclide(s)	Activity concentrations (kBq/kg)					
	Well-mixed soil		Direct deposition on grazing area			
	Transfer to plant products	Transfer to animal products	Pasture vegetation			Soil <sup>c</sup>
			Ashed	Dry	Fresh	
Sr-90	0.3	12	9			6
Pu, Am, Cm <sup>a</sup>	2	16	3.5			8
		0.2 <sup>b</sup>	0.02 <sup>b</sup>			0.1 <sup>b</sup>
Cs-137	2	4	3.5	0.7	0.14	2
I-131				0.7	0.14	20

<sup>a</sup> The values refer to the activity level of each of the  $\alpha$ -emitting isotopes.

<sup>b</sup> For the transfer of Pu, Am and Cm to liver.

<sup>c</sup> Average concentration in soil top-layer.

Table 8

Summary of critical deposition densities on soil and grazing areas required to exceed the generic action levels for foodstuffs

Nuclide(s)	Deposition density (kBq/m <sup>2</sup> )				
	Soil		Direct deposition on grazing area		
	Plant products		Pasture vegetation	Cattle and sheep	Reindeer
	No action taken	Ploughing after deposition	Ploughing after deposition	Worst-case scenario	
Sr-90	9	135	5400	0.7	10
Pu, Am, Cm <sup>a</sup>	60	900	4500	0.3	10
			90 <sup>b</sup>	0.0014 <sup>b</sup>	0.1 <sup>b</sup>
Cs-137	60	900	1800	0.3	1

<sup>a</sup> The values refer to the activity level of each of the  $\alpha$ -emitting isotopes.

<sup>b</sup> For the transfer of Pu, Am and Cm to liver.

of feed and soil by grazing animals are shown in the footnote of Table 5. All values were taken from the NRPB, except the one for cattle feed which was roughly averaged from the values used by the NRPB and the National Council on Radiation Protection and Measurements (NCRP, 1984). We have assumed that the radionuclides in the soil are fully available for animal uptake which, probably, is a conservative assumption. The intake of soil strongly effects the intake of Pu, Am and Cm because of their low soil-to-pasture TFs.

Table 5 shows that the limiting critical activity concentrations in soil for the transfer from well-mixed soil to animal products is about 12 kBq/kg for <sup>90</sup>Sr (milk), about 1 kBq/kg for the group of  $\alpha$ -emitters (liver) and about 4 kBq/kg for <sup>137</sup>Cs (sheep products). The limit for each of the  $\alpha$ -emitters becomes about 0.2 kBq/kg. It is useful to derive an additional critical concentration for the  $\alpha$ -emitters where liver is neglected because (1) liver lowers the critical concentration considerably, (2) liver is a less important type of food in many countries and (3) the liver can be discarded after slaughter. Neglecting liver, the critical soil concentration for the group of  $\alpha$ -emitters increases to 83 kBq/kg (sheep meat). For the individual nuclides, the limit becomes about 16 kBq/kg.

### 2.2.1. Deposition densities

With the same plough layer depth and soil density as previously, the critical deposition densities for ploughing the soil, growing pasture vegetation and letting animals graze, would be 5400, 90, 7200 and 1800

kBq/m<sup>2</sup> for <sup>90</sup>Sr, each of the  $\alpha$ -emitters with respect to liver, each of the  $\alpha$ -emitters neglecting liver and <sup>137</sup>Cs, respectively.

### 2.3. Transfer to animal products from direct deposition on pasture vegetation

Radionuclides deposited on pasture vegetation will be distributed between parts of the vegetation consumed by grazing animals, parts that are not consumed and the top-layer of the soil. The amount of radionuclides ingested by grazing animals depends therefore on this distribution, which is difficult to predict. Initially, we assume that the deposition is completely retained in the part of the vegetation consumed by grazing animals. As in the previous section, we also assume that the animals get their full feed intake by grazing on contaminated land and that the consumption of contaminated feed continues for a sufficiently long time to reach up to the equilibrium TFs given in Table 4. Under these conditions, the animals consume the complete deposition, square-meter by square-meter. Accordingly, the resulting deposition densities should reflect the lowest possible values that could result in exceeded action levels for foodstuffs. The critical animal intakes, vegetation activity concentrations and deposition densities for the transfer to animal products under the assumptions above are shown in Table 6. The values used in the calculations are given below Table 6. The limits for the animal intake and the activity concentrations apply also to e.g. stored feed.

According to Table 6, the limiting critical activity concentrations in ashed pasture vegetation are 18 kBq/kg for  $^{90}\text{Sr}$  (milk), 0.18 kBq/kg for the summed  $\alpha$ -activity of Pu, Am and Cm (cattle liver) and about 7 kBq/kg for  $^{137}\text{Cs}$  (sheep products). The critical concentration for each of the  $\alpha$ -emitting isotopes would be about 0.04 kBq/kg. For the same reasons as in the previous section, it is useful to derive an additional limit for the  $\alpha$ -emitters where liver is neglected. This limit becomes 36 kBq/kg (cattle meat) for the group of  $\alpha$ -emitters and about 7 kBq/kg for each of the nuclides.

Since the analyses of  $^{137}\text{Cs}$  and  $^{131}\text{I}$  do not require ashing, the critical activity concentration in dry pasture vegetation is more relevant for these nuclides. According to Table 6, the limiting critical value for  $^{137}\text{Cs}$  is 1.3 kBq/kg (sheep products) and for  $^{131}\text{I}$ : 1.4 kBq/kg (milk). Measurements of  $^{131}\text{I}$  (and at the same time  $^{137}\text{Cs}$ ) are normally carried out prior to drying because of the volatility of iodine. With a dry weight of 20%, the critical concentrations in fresh vegetation become 0.26 and 0.28 kBq/kg for  $^{137}\text{Cs}$  and  $^{131}\text{I}$ , respectively.

However, the critical activity concentrations listed in Table 6 are not valid if the distribution of radionuclides is such that the ingestion of soil by grazing animals significantly increases the daily animal intakes. To obtain the critical concentrations in the pasture vegetation and the soil top-layer for any distribution of the radionuclides, one may take the limiting values for pasture vegetation (derived above) and well-mixed soil (derived from Table 5) and reduce them by 50%. This way, the daily animal intake of radionuclides will sum up to a maximum of 100% of the critical value, corresponding to the action levels for foodstuffs. The critical concentrations in ashed pasture vegetation and soil become 9 and 6 kBq/kg for  $^{90}\text{Sr}$ , 0.02 and 0.1 kBq for each of the  $\alpha$ -emitters, 3.5 and 8 kBq/kg for each of the  $\alpha$ -emitters neglecting liver and 3.5 and 2 kBq/kg for  $^{137}\text{Cs}$ . For dry vegetation and soil, the values become about 0.7 and 2 kBq/kg for  $^{137}\text{Cs}$  and 0.7 and 20 kBq/kg for  $^{131}\text{I}$  (the last value corresponds to a daily intake of 10 kBq via 0.5 kg soil). The critical concentration in fresh vegetation can be set to 0.14 kBq/kg for both  $^{137}\text{Cs}$  and  $^{131}\text{I}$ .

### 2.3.1. Deposition densities

According to Table 6, the limiting critical deposition densities on pasture vegetation are about 0.7 kBq/m<sup>2</sup> for  $^{90}\text{Sr}$  (milk), 0.007 kBq/m<sup>2</sup> for Pu, Am and Cm (cattle liver), 1.4 kBq/m<sup>2</sup> for the same nuclides if liver is neglected (cattle meat) and 0.3 kBq/m<sup>2</sup> for both  $^{137}\text{Cs}$  (sheep products) and  $^{131}\text{I}$  (milk). The values for each of the  $\alpha$ -emitters become 0.0014 kBq/m<sup>2</sup> for cattle liver and 0.3 kBq/m<sup>2</sup> with liver neglected. The critical deposition density for both the individual  $\alpha$ -emitters and  $^{137}\text{Cs}$  with respect to milk, which is one of the

most important foodstuffs, is about 3 kBq/m<sup>2</sup>. It is likely that there is a fairly large margin to more realistic values because of the conservative assumptions (e.g. complete retention in the part of the vegetation consumed by animals).

Studies on the Chernobyl fallout of  $^{137}\text{Cs}$  in Sweden may be used to estimate the magnitude of this margin. The highest measured concentration of  $^{137}\text{Cs}$  in milk produced in an area with a deposition density of 1 kBq/m<sup>2</sup> was 17 Bq/L (Wallström et al., 1998). The contribution of grazing to the daily animal intake was about 70%. This suggests the transfer of  $^{137}\text{Cs}$  to milk to be less than 0.03 Bq/L per Bq/m<sup>2</sup>, which would lead to an action level of more than 30 kBq/m<sup>2</sup>, i.e. more than one order of magnitude higher than the 2.9 kBq/m<sup>2</sup> presented in Table 6. It is possible that the margin to more realistic critical deposition densities is in the same range also for the other nuclides and foodstuffs.

### 2.3.2. Transfer to reindeer

The critical deposition densities for reindeer meat are of particular interest in some countries. From an investigation on the Chernobyl fallout in Sweden, one can estimate the transfer of  $^{137}\text{Cs}$  to reindeer meat to be less than 1 Bq/kg per Bq/m<sup>2</sup> (Eriksson et al., 1991). Assuming similar TF ratios for different elements to reindeer meat as to cattle and sheep meat, one can use Table 4 to estimate the TFs for  $^{90}\text{Sr}$  and the group of  $\alpha$ -emitters to reindeer meat to be about 100 and 500 times lower than that of  $^{137}\text{Cs}$ , respectively. The minima deposition densities required to exceed the action levels for reindeer meat would thus be 10 kBq/m<sup>2</sup> for  $^{90}\text{Sr}$  and 5 kBq/m<sup>2</sup> for Pu, Am and Cm. From a study regarding the bomb-fallout (Holm, 1977), one can derive a TF of about  $1.7 \cdot 10^{-4}$  (Bq/kg per Bq/m<sup>2</sup>) for the transfer of  $^{239}\text{Pu}$  to reindeer meat. The critical deposition density with this TF becomes approximately 60 kBq/m<sup>2</sup>. This suggests that the value derived above for the group of  $\alpha$ -emitters can be increased with an order of magnitude and set to 50 kBq/m<sup>2</sup>. The activity concentration of the  $\alpha$ -emitters in the reindeer liver can be expected to be about two orders of magnitude higher than in the meat (Holm, 1977; Jaakkola, 1989). The action levels for reindeer liver may thus be exceeded if the deposition density for the group of  $\alpha$ -emitters is higher than around 0.5 kBq/m<sup>2</sup>.

### 2.4. General discussion

The critical activity concentrations and deposition densities obtained are gathered in Tables 7 and 8, respectively. It is likely that a safety margin is included in these figures because (1) there is no reason to believe that the transfer factors used in the calculations are underestimated, (2) the highest TFs have been selected where possible and (3) part of the calculations are

based on conservative assumptions. It should be noted that direct deposition on plant products may lead to lower critical deposition densities depending on the crop yield.

As a comparison to the activity concentrations in well-mixed soil given in Table 7, many soils contain up to 0.5 kBq/kg  $^{226}\text{Ra}$  and 1 kBq/kg  $^{40}\text{K}$  (UNSCEAR, 1993). Ra-226 is, owing to its chemistry, more available for root uptake than the  $\alpha$ -emitters considered in this work, whereas the radiotoxicity (Sv/Bq, ingestion) is about similar. In the case of  $^{40}\text{K}$ , both the chemistry and the radiotoxicity are, roughly, similar to that of  $^{137}\text{Cs}$ . With respect to the transfer to foodstuffs, the hazard posed by the soil concentrations listed in Table 7 is thus in about the same range as the hazard posed by natural radionuclides in many soils. This suggests that the critical soil concentrations calculated in this work are of a reasonable magnitude.

Consequently, lower activity concentrations than those presented in Table 7 do not have to be detected to determine whether or not the generic action levels for foodstuffs produced in a certain area will be exceeded. These concentrations may therefore be regarded as the required detection limits in analyses of soil and pasture vegetation shortly after nuclear accidents (if the detection limit is defined to be the activity concentration that actually can be detected in practice). According to Table 7, the lowest detection limits required are: 300 Bq/kg for  $^{90}\text{Sr}$ , 2000 Bq/kg for Pu, Am and Cm neglecting the transfer to liver and 20 Bq/kg for Pu, Am and Cm including the transfer to liver.

The detection limits for the  $\alpha$ -emitting isotopes of Pu, Am and Cm achieved with thorough radioanalytical procedures have been reported to be below 0.07 Bq/kg soil (Moreno et al., 1997; Hiatt and Hahn, 1979). Others report a rather good accuracy for concentrations below 1 Bq/kg (Bunzl and Kracke, 1994; Godoy et al., 1995; Smith et al., 1995) suggesting a detection limit of a few tenths of a Bq per kg, or less. In consequence, these detection limits could be increased by about a factor of 100 and, if liver is neglected, by about a factor of 10,000, in analyses of soil and pasture vegetation after nuclear accidents. Common detection limits for  $^{90}\text{Sr}$  are 10–100 times higher than that of the  $\alpha$ -emitters, i.e. 1–10 Bq/kg (Cooper et al., 1992; Moreno et al., 1997; Friberg, 1997). These limits could be increased 10–100 times for the same type of analyses.

### 3. Conclusions

We have shown that the detection limits of many of the currently employed methods for environmental analysis of  $^{90}\text{Sr}$ , Pu, Am and Cm are orders of magnitude lower than the critical activity concentrations in

soil and ashed pasture vegetation which correspond to the action levels for foodstuffs recommended by the IAEA. One may therefore allow much higher detection limits in analyses of such materials if the aim is to determine whether or not these action levels will be exceeded. Since a higher detection limit justifies shorter counting times and/or the use of smaller samples, these findings can be used in the development of more simple and rapid methods. Such methods could, for instance, be used in the first stage after a nuclear accident to determine whether or not any grazing or agricultural restrictions are required. The more time-consuming and labor intensive methods required for a thorough analysis can thus be left to a later, less urgent, stage.

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