Resuspension and Deposition of Radionuclides Under Various Conditions

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Abstract: The resuspension of Cs-137 and Pu-239+240 has been assessed at sites within and outside the 30 km exclusion zone around Chernobyl. Measurements were made during periods of wind-derived resuspension and during simulated and real agricultural activity. From these data, resuspension rates (fraction of deposit removed in unit time) or emission rates (fraction of deposit removed in unit time or unit area) have been calculated. Resuspension rates of Cs-137 have declined by at least an order of magnitude 7 years after the accident and were found to be of the order of 10^{-10} s^{-1}. During agricultural activity, the resuspension rate may exceed background levels by four orders of magnitude.

Introduction

During experimental investigations of " Experimental Collaboration Project 1(ECP 1)" in 1992-1994 detailed information about the airborne concentration and size distribution of the radioactive atmospheric aerosol was obtained for the resuspension by wind and during anthropogenic activity. Measurements were made at sites with different surface and soil characteristics within and outside the 30 km exclusion zone around Chernobyl. These measurements have allowed a number of important parameters to be calculated which will help to predict the spread of contamination and the inhalation dose.

Resuspension as a source of Cs-137 in the atmosphere

The data presented in Figs.1a and 1b demonstrate how the atmospheric concentrations of Cs-137 and Ce-144 have changed with time since the accident and how resuspension of deposited material has affected the airborne concentrations of these two radionuclides. The measurements were made at two sites, Chernobyl, and Pripyat [1]. The two straight lines show the expected atmospheric concentrations of Cs-137 and Ce-144, allowing for radioactive decay, standardised to the activity concentration in June 1989. After 1989, the majority of the decontamination work in the 30 km zone was complete, and resuspension by mechanical activity would have become less important. The figures show the monthly mean atmospheric concentrations of Cs-137 and Ce-144 have declined by one or two orders during the four year measurement period. During 1987 and 1988, there was a large effort made to decontaminate the area surrounding the Chernobyl NPP, and there is some evidence that this work enhanced the atmospheric concentrations of Cs-137 and Ce-144. The decrease in atmospheric concentration of Ce-144 and Cs-137 is higher than would be expected from radioactive decay alone and this is particularly well illustrated in Figure1a. This feature demonstrates that there are processes which are responsible for the reduction of the activity concentration in the air,

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for example, vertical migration in soil, and run-off of radionuclides with rain or melting water from snow cover.The atmospheric concentrations are apparently influenced by the levels of anthropogenic activity with atmospheric concentrations attributable to: wind-driven resuspension, ~ 10 to 100 μBq m^{-3}; light agricultural activity, ~ 200 to 400 μBq m^{-3}; and a forest fire 17 km from the measurement site, ~ 1000 to 2000 μBq m^{-3}.

Studies in Sweden have illustrated the potential for long range transport of Cs-137 from the heavily contaminated region around Chernobyl. There are events in Sweden where Cs concentrations increased simultaneously over a large region, and on these occasions, the weather situation in northern Europe has been dominated by an anticyclone over Russia. The receptor orientated trajectories calculated for these periods indicate that the Chernobyl area is a possible source for the extra atmospheric activity.

Range of measurements made

European and CIS collaborators made measurements of atmospheric concentration and deposition using a wide variety of equipment. The institutes represented by their initials in the text and tables are shown in the footnote at the bottom of the first page of this article. The devices used to measure atmospheric concentrations included: passively aspirated samplers (fabric 'cone' sampler , UAAS; gauze screen; IRBAS) and actively aspirated size selective samplers (impaction surfaces in a wind tunnel, AEAT; Andersen PM10, CIEMAT; 'GRAD', high volume, and 'PK' impactors, UAAS; rotating arm impactor (RAI),GSF and Aerodynamic Particle Sizer (APS), GSF).

The teams made measurements of the atmospheric concentration of Cs-137 at six sites; three within the 30 km zone and three outside the zone. In some cases, these measurements have been combined with parameters from a meteorological station (atmospheric stability, wind direction) to allow the calculation of specific resuspension parameters, for example, wind-driven resuspension rates and emission rates from agricultural activity.

Intersite variability in the resuspension factor.

A simple, easily measured estimate of the level of resuspension is given by the resuspension factor [2]:

$$R = \frac{c_{am}}{\sigma_a} \qquad \text{(Equation 1)}$$

where: R = resuspension factor (m^{-1})
c_{am} = atmospheric concentration (Bq m^{-3})
σ_a = soil activity to a specified depth (Bq m^{-2})

The data in Table 1 summarises the mean resuspension factors for Cs-137 and Pu-239+240 at the measurement sites. The Cs-137 resuspension factors are mean values calculated from a range of atmospheric samplers. The Pu-239+240 resuspension factors are for particles < 10 μm in diameter (data from the PM10 sampler).

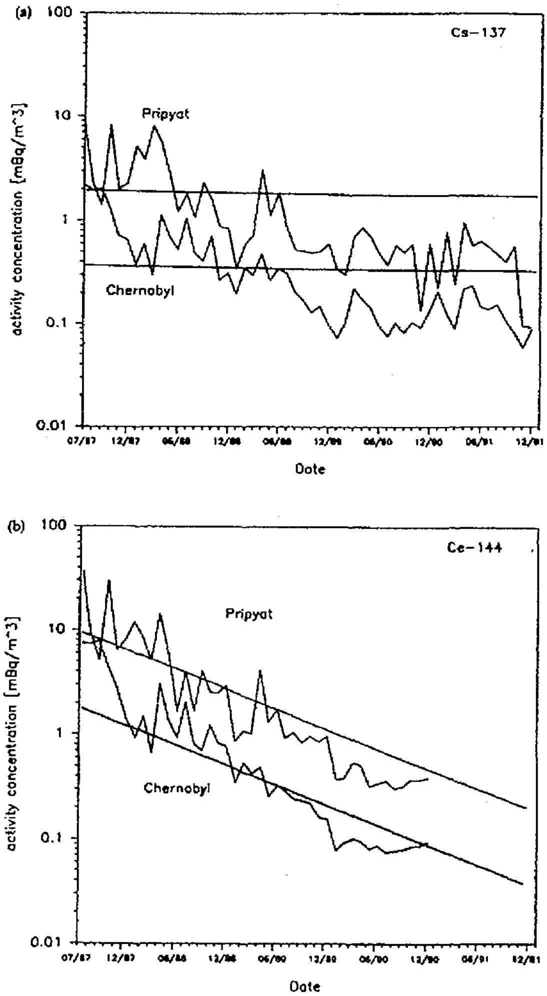

Figure. 1: *Air concentrations of Cs-137 and Ce-144 at Pripyat and Chernobyl (July 1987 to December 1991)*

The table shows that the highest resuspension factor was recorded at Novozybkov and this may be because the surface at this site was bare soil which would have been relatively erodable. Despite the range of environmental conditions during the work and the variability of the sites examined, there is only a four-fold difference between the lowest and highest measured mean values of the resuspension factor. A relatively small value was recorded at the Beach-Pripyat, although this site has the highest surface contamination. This is probably because the surface particles on the beach were larger than at the other sites, and the radioactivity had penetrated to greater depths. Localised activity at the Beach site, for example people moving around, enhanced the resuspension rate by a factor of ten.

Table 1: *Mean values of the resuspension factor of Cs-137 and Pu-239+240 measured between 1992 and 1994*

	Beach-Pripyat	Zapolye (1993)	Kopachi	Novozybkov	Mikulichi	Kovali
Surface characteristics	> 90 % sand	grassland	grassland	bare soil	rye field	barley field
Direction [distance (km)] from Chernobyl NPP	W [4]	S [14]	S [2]	NE [150]	N [45]	N [45]
Cs-137 contamination (Bq m^{-2})	$3.3x10^6$	$5.9x10^5$	$2.3x10^6$	$1.1x10^6$	$4.2x10^5$	$5.3x10^5$
Mean resuspension factor of Cs-137 ± SD ($x10^{-10}$ m^{-1})	2.2 ± 1.9	4.4 ± 2.3	2.0 ± 1.4	7.7 ± 3.5	3.1 ± 3.1	6.3 ± 7.2
Pu-239+240 contamination (Bq m^{-2})	$5x10^4$	$5.7x10^3$	$3.9x10^4$			$3.4x10^2$
Mean resuspension factor of Pu-239+240 ($x10^{-10}$ m^{-1})	$1.1x10^{-10}$	$1.8x10^{-10}$	$2.4x10^{-11}$			$1.1x10^{-9}$

The resuspension factors of Pu-239+240 are comparable to those measured for Cs-137. The highest mean value was recorded at Kovali, although it is difficult to ascribe reasons for this.

I Resuspension factor in relation to particle size

Measurements of the atmospheric concentration of Cs-137 according to particle size were made by several samplers. Figure. 2 shows the resuspension factor of Cs-137 according to these two particle size fractions at all the field measurement sites. The figure clearly shows the large contribution of particles greater than 15 µm in diameter to the total resuspension factor at the heavilycultivated sites of Novozybkov, Mikulichi and Kovali. At Zapolye, during periods of wind driven resuspension, there was a good linear correlation between the resuspension factor of Cs-137 and particles below 20 µm in diameter. This is shown in Figure 3. In general, the resuspension factor increases with particle diameter.

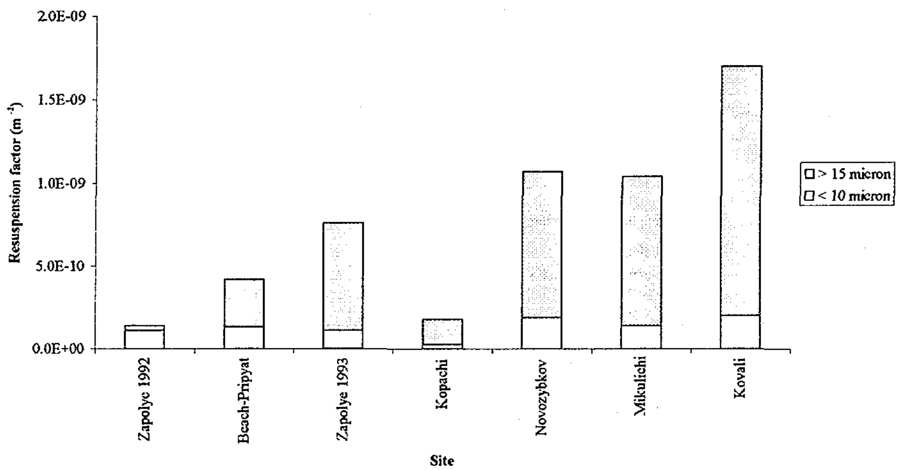

Figure 2: *Resuspension factors for Cs-137 associated with airborne particles < 10 μm and > 15 μm at all the measurement sites*

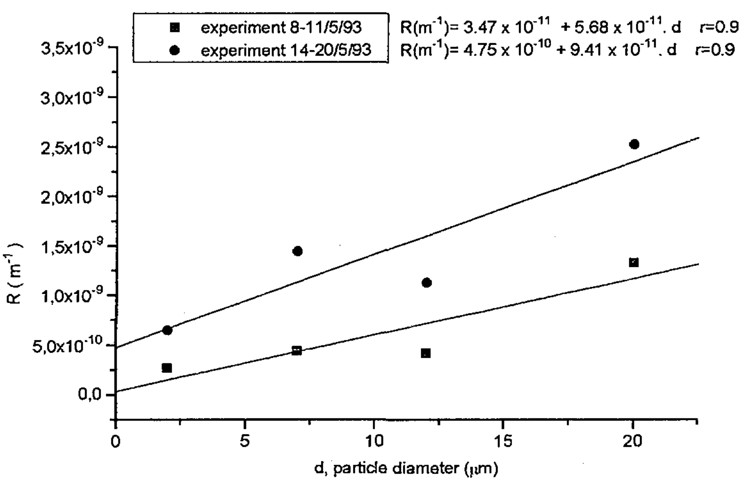

Figure 3: *Resuspension factor as a function of particle diameter during wind resuspension experiments. Zapolye field site. Atmospheric concentration data from PK impactor.*

Intersite variability in the resuspension rate

The resuspension rate, potentially, is a more useful measurement of resuspension than the resuspension factor, since it enables downwind inhalation doses and deposition levels to be predicted. However, it is difficult to measure in practice. It is defined as [2]:

$$\beta_{\uparrow} = \frac{j_{\downarrow m}}{\sigma_a} \qquad \text{(Equation 2)}$$

where:	$\beta_{\uparrow}$	=	resuspension rate	(s^{-1})
	$j_{\downarrow m}$	=	resuspension flux	(Bq m^{-2} s^{-1})
	s	=	soil activity to a specified depth	(Bq m^{-2})

Shortly after the Chernobyl accident in 1986, it were made several measurements of the resuspension rate within 30 km of the Chernobyl plant in [3]. These measurements were made at sites with very different surface characteristics. All the measurements relate to periods when advection of material from upwind sources was considered to be small, the ground surface dry and with a moderate wind (~2 m s^{-1} at 1.0 m). Table. 2 presents these data.

Table. 2: *Resuspension rate according to surface type in 1986*

Site	Resuspension rate ± SD x 10^{-9} (s^{-1})		
	Ce-144	Cs-137	Zr-95+Nb-95
Zapolye	0.3 ± 0.1	1.0 ± 0.7	0.4 ± 0.2
Forest	2.1 ± 0.9	2.1 ± 0.8	3.7 ± 0.9
Beach-Pripyat	2.2	3.7	2.4

The work suggests that resuspension rates of between 1 and 4×10^{-9} s^{-1} were appropriate for a fresh deposit of Cs-137 at all the sites, but, the resuspension rates of Ce-144 and Zr-95+Nb-95 were approximately three to four times lower at Zapolie. Table. 3 summarises the resuspension rates of Cs-137 measured at sites during the field campaigns of the ECP1 programme.

The resuspension rates determined are highly variable, both with respect to the magnitude of estimates at individual sites using the same measurement technique and at the same site using different measurement techniques. It must be noted that the determination of the resuspension rate in this way is prone to high levels of uncertainty. Factors such as systematic measurement differences, including different measurement heights and integration periods could account for some of the differences between techniques also. The resuspension rate would be expected to vary with time, anyway. The technique used by IRBAS provided the highest estimates of resuspension rate at Novozybkov and Mikulichi .The values reported are of the same order as those recorded shortly after the accident, although the resuspension rate would have been expected to decline sharply in the first year or two after the accident (see Cs-

Table. 3: *Resuspension rate of Cs-137*

Site	Date	Cs-137 mean β_f x 10^{-10} ± SD (s^{-1})			
		cone	GRAD	impactor+PM10	gauze screen
Zapolye	13/5/92 to 11/8/92	4.20± 0.16		0.0011	
Beach-Pripyat	14/7/92 to 11/8/92	0.08 ± 0.07		0.11	
Zapolye	06/5/93 to 01/6/92	1.60 ± 1.20	0.8 ± 0.7	0.017	
Kopachi	28/7/93 to 03/8/93				0.44
Novozybkov	17/5/94 to 24/5/94			0.0044	4.6
Mikulichi	13/7/94 to 29/7/94			0.13	0.09

137 data in Figure. 1). Although these measurements of resuspension have been made in the absence of agricultural activity, personnel moving whilst preparing equipment has enhanced the resuspension rate by up to an order of magnitude.

Resuspension during agricultural activity

At Zapolie and Kopachi in 1993 approximately half of the experiments were designed to simulate agricultural activity (harrowing) while the others consisted of vehicles being driven along a dirt track. Line sources were prepared (raked bare soil) around the measurement site to cater for different wind directions. Prior to carrying out the experiments, both of these sites were undisturbed grassland, sparsely vegetated, with generally dry conditions prevailing throughout. Vehicles used included two different sizes of tractor (pulling a spiked harrow) and a large, six-wheeled army truck. In 1994 the emphasis of the fieldwork was on real agricultural practice and measurements were undertaken for a variety of operations at three different sites. At Novozybkov fertilisation, cultivation and planting were carried out on working agricultural land, i.e. the soil was bare and well mixed. Whole field areas were worked as opposed to a single strip as at Zapolie. At Mikulichi a rye field was harvested and ploughed. Harvesting (of barley) was also carried out at Kovali. To maintain a strict quality control over the results, only data from experiments satisfying certain criteria have been interpreted. The basic criteria were as follows. Experiments were only selected when:
(1) The wind direction was consistently blowing across the line (or area) source towards the measurement site;
(2) The wind speed was sufficiently strong to provide a steady, well-mixed plume. Experiments conducted during variable, thermally generated winds were rejected;
(3) The type of vehicle, it's speed and operation must have been constant throughout the duration of the experiment;
(4) The meteorological conditions should be the same for the whole experiment.

The Aerodynamic Particle Sizer provided a detailed picture of the dust concentration of the different particles sizes during the experiments (see Figure 4). The data obtained demonstrated that the agricultural activity increased the atmospheric concentrations by a factor of several thousand in comparison to the background concentrations at distances of 20 to 30 m from the dust sources and 10 to 100 times at 100 m or more, depending on conditions. The increase in particle concentration was not uniform for the whole particle size range. The increase due to agricultural activity was the highest in the giant particle fraction. This finding makes the assessment of the activity connected to the giant particles important.

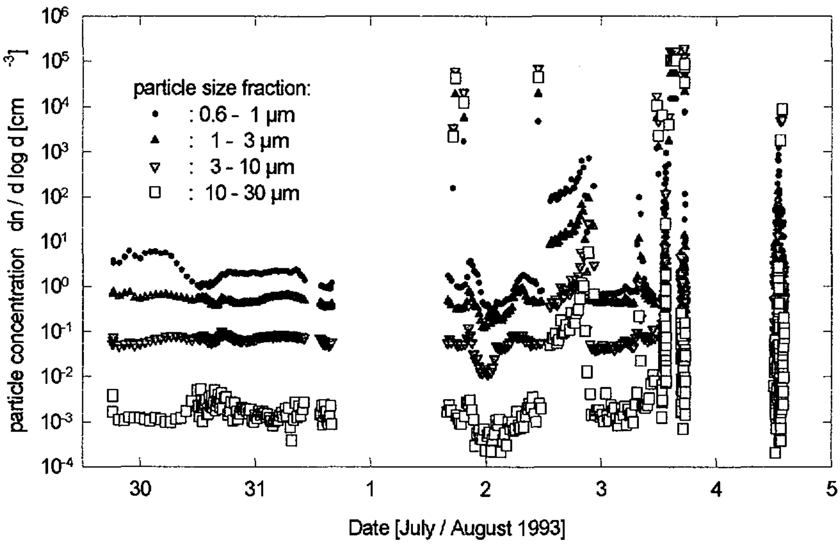

Figure 4: *Normalised particle number concentration in four different size ranges as measured by an Aerodynamic Particle Sizer*

The measurement of the number concentrations of hot particles showed an increase of three orders of magnitude, reaching 0.7 to 1.0 hot particles per m³ with a maximum activity of 1.5 to 2.0 Bq per particle. The atmospheric concentration of resuspended Cs-137 was found to depend strongly on soil humidity r_{sh}. The relationship is roughly exponential i.e. $R=R_0 \exp(\lambda r_{sh})$ with $R_0 \approx 3\times10^{-7} m^{-1}$ and $\lambda \approx 60$. This means that a soil humidity of 10% reduces R by a factor 300 as compared to dry soil.

1 Emission rates during agricultural activity

The analitical solution of the semi-empirical diffusion equation for a stationary infinite crosswind strip source was used for the estimation of the emission rates during agricultural activity [4]. These experiments have been considered as approximating to a finite dust strip. In reality, the source was neither instantaneous nor stationary as the agricultural equipment moved many times during an experimental period. The emission rates reached very high values during the work (see Table 4). These rates exceed background resuspension rates by four orders of magnitude for the whole particle size spectrum, and by three orders of magnitude for the respirable size range.

Table 4: *Emission rates for various types of agricultural vehicles and agricultural activity derived from PK impactor measurements and Equation (4.9)*

Date (Zapolye)	Kind of vehicle	Emission rate, 10^{-6} s^{-1}	Emission rate for d≤12 μm, 10^{-6} s^{-1}
12.05.93 morning	MTZ-82	0.027	0.004
13.05.93 morning	T-150	1.5	0.16
13.05.93 afternoon	T-150	2.0	0.20
22.05.93 morning	ZIL-131	1.0	0.09
22.05.93 afternoon	ZIL-131	1.4	0.13
24.05.93 afternoon	ZIL-131 and ZIL-130	0.48	0.09

Results calculated for the agricultural work at Novozybkov, Mikulichi and Kovali using a box model are shown in Table 5. Here, the results are expressed in terms of an emission rate per unit area. The mean value for all the sites was 9.2 mBq s^{-1} m^{-2} (s.d. = 7.8) for >15μm particles. For an area source the emission rate can be considered equivalent to a resuspension flux and this enables calculation of the resuspension rates presented in Table 5. Soil contamination densities of 1.2, 0.42 and 0.56 MBq m^{-2} have been used for Novozybkov, Mikulichi and Kovali respectively . Overall, the mean resuspension rate (for >15 μm particles) was 1.4 x 10^{-8} s^{-1} (s.d. = 1.5 x 10^{-8}). This is several orders of magnitude greater than the background values for these areas.

6.2 Deposition velocity

The values of deposition velocity measured during agricultural work are systematically higher (by nearly a factor of three) compared with natural conditions. Close to the source (x=22m) the mean deposition velocity is (0.144± 0.136) m s^{-1} while at greater distances (x ≥130m), a value of (0.113±0.087) m s^{-1} has been calculated. The large scatter in values of $v_\downarrow$ does not permit detailed discussion of the dependence of deposition velocity on distance. If Stoke's formula is used to estimate the mean radius of particles with observed $v_\downarrow$ = 0.115 m s^{-1}, then a value of r ≈ 20 μm is obtained. Using the equation $v_\downarrow = bu_* + v_{sed}$ [5], where b=0.01-0.08 for particles of diameter greater than 5 μm depositing to a dry grass surface and with a soil density of 2.3 g cm^{-3}, gives a value for the mean radius of 18 μm. From impactor measurements it was found that particles of this order of size had the second highest activity.

Table. 5: *Summary of results from area source equation (Novozybkov and Bragin 1994)*

Experiment (reference)	c (mBq m^{-3})	Q (<10 μm) (mBq s^{-1} m^{-2})	Q (>15 μm) (mBq s^{-1} m^{-2})	Resuspension rate (total) (s^{-1})
Novozybkov[a]				
13.5 (Agric 2/Nov 94)	19.3	-	8.1	6.8×10^{-9}
14.5 (Agric 3/Nov 94)	43.5	-	20.4	1.7×10^{-8}
14.5 (Agric 4/Nov 94)	39.9	-	16.3	1.4×10^{-8}
17.5 (Agric 7/Nov 94)	11.7	-	5.3	4.4×10^{-9}
Mikulichi				
30.7 (Agric 1/Mik 94)	25.6	0.2	5.5	1.4×10^{-8}
31.7 (Agric 2/Mik 94)	8.7	0.4	1.2	3.8×10^{-9}
01.8 (Agric 3/Mik 94)	30.7	1.6	4.7	1.5×10^{-8}
02.8 (Agric 4/Mik 94)	127.0	0.9	20.6	5.1×10^{-8}
Kovali				
06.8 (Agric 1/Kov 94)	5.6	0.3	0.8	2.6×10^{-9}

Note: [a] Novozybkov data are for particles >15 μm only

7. Contamination of the atmospheric surface layer and yield for agricultural work

Measurements of the temporal change of the mean concentration of Cs-137 showed a sharp increase in concentration at the start of harvesting. Concentrations for agricultural work were higher by a factor of ten over background concentrations. The mean background deposition flux density was 0.003 mBq $m^{-2}s^{-1}$ at Mikulichi and 0.009 mBq $m^{-2}s^{-1}$ at Kovali. The maximum deposition flux densities were 1.4 mBq $m^{-2}s^{-1}$ and 0.53 mBq $m^{-2}s^{-1}$ for rye harvesting and barley harvesting respectively. Thus, for anthropogenic activities the deposition flux density was 60-400 times higher than natural background levels.

Summary

The atmospheric concentrations of Cs-137 have declined by over two orders of magnitude since the accident. However, long term measurements extending to some years after the accident show clear evidence that resuspension influences the atmospheric concentrations. The data highlight the potential for events such as forest fires to raise the atmospheric concentrations by up to 100 times.

Team members of have made extensive measurements of atmospheric concentrations several sites in the CIS and have combined these with meteorological measurements to provide estimates of the resuspension in these areas. This work has included calculating resuspension factors and also resuspension rates.

Atmospheric concentrations of Cs-137 were assessed in all the field campaigns, but only a few measurements of Pu were made. Resuspension factors were found to be in the range 2.0 to $8x10^{-10}$ m^{-1} for ^{137}Cs for the particle size range covering a few microns up to tens of microns in diameter. Resuspension factors were similar for Pu, ~ 10^{-9} to 10^{-10} m^{-1}, but these measurements only relate to particle sizes less than 10 μm in diameter.

Despite the wide range of soil types and environmental conditions at the sites where resuspension was assessed, there was only a four-fold difference between the lowest and highest estimates of the mean resuspension factor of Cs-137. The inter-equipment variability in the resuspension factor was around an order of magnitude, and this needs to taken into account when comparing data from a range of types of equipment. It is not an easy task to relate the magnitude of the resuspension factor to environmental parameters such as soil moisture content and wind, since the effect of individual parameters cannot be controlled. However, large scale human movement, for example, preparing the field sites, increased the resuspension factor of Cs-137 by up to an order of magnitude.

Resuspension rates of Cs-137 have declined by at least one order of magnitude 7 years after the accident. For example, at a grassland site, the resuspension rate has fallen from $1.0x10^{-9}$ s^{-1} in 1986 to $1.6x10^{-10}$ s^{-1} in 1993 for the particle size range covering a few microns up to tens of microns in diameter. The variability in the resuspension rates was much greater than in the resuspension factors. Values of the resuspension rate, calculated from the same instrument, varied by up to two orders of magnitude at individual sites and a similar variability was observed in the values of the mean resuspension rate calculated from different instruments.

The magnitude of the measured resuspension rates was of the same order as those found during a review of the literature, taking into account the age of the Chernobyl deposit. From the limited data available, the resuspension rate for Pu appears comparable to that of Cs-137 but these Pu measurements only relate to particle sizes less than 10 μm in diameter.

The data from field based measurement campaigns of agricultural work have been used in conjunction with models to predict the airborne radioactive contamination in the atmosphere at ground level. The fraction of material removed from the surface in either unit time or per unit area have been calculated. Values varied from $2.7x10^{-8}$ to $2.0x10^{-6}$ for all particle sizes and from $4.0x10^{-9}$ to $2.0x10^{-7}$ for particles of 12 μm in diameter.

Harvesting, harrowing, cultivation, planting, etc., are operations which give an increase in secondary contamination of the underlying ground surface at distances of between 50-200 m from the agricultural work being undertaken. On the ground the contamination is increased by a factor of between 60-200 while in the surface layer of the atmosphere levels are increased by 10-20 times.

The creation of a buffer zone (an untreated agricultural area) of 200 m width between a settlement and nearby farmland would allow a decrease of approximately two orders of magnitude in the transfer of secondary radioactive substances into the settlement during agricultural work on the bordering fields.

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