

## MOVEMENT OF RADIONUCLIDES IN TERRESTRIAL ECOSYSTEMS BY PHYSICAL PROCESSES

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**Abstract**—Physical processes that effect the movement of radionuclides in the temperate environments post-deposition are considered in this paper. The physical processes considered include the interception of radionuclides by vegetation, resuspension, and vertical migration in soil. United States and Russian results on the interception of radionuclides are reviewed and defined in terms of models that are currently undergoing evaluation and revision. New results on resuspension are evaluated, and a preliminary new model for the time-dependent resuspension factor is proposed. Chernobyl-related results on the movement of radionuclides into the soil column are presented, as is a revised model for this process based upon recent results from Ukraine.

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**Key words:** fallout; radionuclide; radioactivity, environmental; National Council on Radiation Protection and Measurements

### INTRODUCTION

THERE ARE many physical, chemical, and biological processes that govern the movement of radionuclides through terrestrial ecosystems to humans. In general, these processes can be considered as (1) physical processes that are independent of the radionuclide or (2) chemical and biological processes that are strongly dependent upon the element and its chemical form. This paper is concerned only with the physical processes; Whicker and Pinder (2002) discuss the chemical and biological processes in a companion paper.

The physical processes of interest include:

1. Interception of airborne and waterborne radionuclides by vegetation;

2. Loss of radionuclides from vegetation;
3. Resuspension of radionuclides from soil and redeposition onto vegetation; and
4. Weathering of radionuclides from the soil surfaces into deeper soil layers.

This paper is focused on items 1, 3, and 4. The loss of radionuclides from vegetation has been treated by many authors (e.g., Martin 1963, 1964; Thompson 1965; Miller and Hoffman 1983; Mück et al. 1994; Pröhl and Hoffman 1996).

The interception of airborne or waterborne (rainfall) radionuclides by vegetation is a poorly understood process. Empirical observations (Romney et al. 1963; Martin 1965; Anspaugh 1987; Hoffman et al. 1989, 1992; Pröhl and Hoffman 1996) of this factor have been noted to vary substantially, and uncertainty in this factor is a primary contributor to uncertainty in calculations of dose from the ingestion of terrestrial foodstuffs (Whicker et al. 1990).

Resuspension of radionuclides deposited on soil is the result of many complicated processes that defy accurate description or modeling. Empirical observations of resuspended concentrations of radionuclides in air have been noted to display a strong time dependence at early times, but correlations with meteorological variables remain elusive. This pathway may be important at times early after deposition, particularly for radionuclides of low biologic availability or for re-occupation of contaminated property.

The final pathway to be discussed here is the weathering of radionuclides into deeper soil layers from the soil surface. The rate of this process has been determined to be remarkably similar for radionuclides of different chemical characteristics, and this implies that the weathering is mainly a physical process, at least in normal soil in temperate climates. The process is important for two reasons. Perhaps the more significant in the shorter term is that this process determines the time dependence of external gamma-exposure rate after radionuclides are deposited on soil. The second reason is that the process of weathering into deeper soil layers has an impact on the long-term availability of radionuclides to plant roots.

This paper is a summary of work on the three subjects mentioned above. There is no attempt to provide

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the details concerning the original work, which has been done by many investigators over many decades, and which may be found by consulting the publications listed in the Reference Section.

### INTERCEPTION OF RADIONUCLIDES BY VEGETATION

#### Fallout deposited without rainfall ("dry" fallout)

An important early systematic investigation of the interception of dry fallout by vegetation was reported by Chamberlain (1970). He observed an empirical relationship between the interception of particles or vapors by vegetation and the biomass of the vegetation:

$$1 - p = e^{-\mu w}, \quad (1)$$

where

$p$  = interception fraction, unitless;  
 $\mu$  = interception parameter,  $\text{m}^2 \text{kg}^{-1}$ ; and  
 $w$  = biomass (dry weight),  $\text{kg m}^{-2}$ .

This logically implies that the greater the biomass, the larger is the fraction of the radionuclide deposit that is retained by vegetation rather than being deposited on soil.

Chamberlain performed experiments with small particles and vapors and reported that the value of  $\mu$  varied from 2.3 to 3.3  $\text{m}^2 \text{kg}^{-1}$ . This same value of  $\mu$  ( $\sim 3 \text{ m}^2 \text{kg}^{-1}$ ) may also be applied to dry global fallout, but the use of this value in the late 1970's for reconstructing doses from the ingestion of fallout by residents downwind of the Nevada Test Site (NTS) showed that the estimates of dose were too high to be biologically plausible. Thus, attempts were made to find measured data that would be more applicable to the NTS situation. Unfortunately, there were no measurements made of  $p$ . However, some historical data were found of the total deposition density,  $d$  ( $\text{Bq m}^{-2}$ ), and of the concentration,  $c$  ( $\text{Bq kg}^{-1}$ ), in vegetation. By definition  $p = cw d^{-1}$ , if  $d$  is measured in such a way that it reflects the total deposition, and

$$\frac{p}{w} = \frac{c}{d}. \quad (2)$$

Further, eqn (1) can be approximated by expanding the exponential function to

$$1 - p = 1 - \mu w + \frac{(\mu w)^2}{2!} - \frac{(\mu w)^3}{3!} + \frac{(\mu w)^4}{4!} - \dots \quad (3)$$

If  $\mu w \ll 1$ , then  $\mu$  is approximately  $p w^{-1}$  or  $c d^{-1}$ . Values of  $p w^{-1}$  have frequently been called the "mass-interception fraction" with units of  $\text{m}^2 \text{kg}^{-1}$ . Through use of eqn (2) the mass-interception fractions can be derived from measurements of  $c$  and  $d$ ; through the approximation based upon eqn (3) approximate values of  $\mu$  can also be derived from measurements of  $c$  and  $d$ . The error in the approximation of  $\mu = c d^{-1}$  is about 20% at  $\mu w = 0.4$  and less than 5% at  $\mu w = 0.1$ .

Data pertaining to measurements of  $c$  and  $d$  do not appear to be available for the early tests in Nevada. The first two known data points were reported by Lindberg et al. (1954) for one point each for shots Nancy and Simon in 1953. The derived values of mass-interception fraction are for native desert vegetation and are given in Table 1. Beginning with Operation Teapot in 1955, more measurements were made and reported in Lindberg et al. (1959); a further refinement was that the deposited activity was collected in trays and was then partitioned according to size fraction. Thus, it was possible to examine the retention of fallout on vegetation that was associated with soil deposition on particles of  $\leq 44 \mu\text{m}$ . Where such particle-size information is available for associated measurements on native vegetation, it is also reported in Table 1. Extensive sets of measurements were also performed for Operation Plumbbob in 1957, but the original data have not been found. Much of the pertinent data has fortunately been summarized by Miller (1963) and by Romney et al. (1963). Values from these two sources for native vegetation are also shown in Table 1. Martin (1965) has also reported measurements following Project Sedan, which was a 104 kt cratering event conducted on 6 July 1962; however, those data are not included here due to the strong difference between a large cratering experiment and the atmospheric nuclear tests.

Beginning with Operation Teapot in 1955 some measurements were also reported for pasture-type vegetation. Such measurements found in or derived from Lindberg et al. (1959) and Romney et al. (1963) are shown in Table 2. The data from Tables 1 and 2 are summarized in Table 3 and in Fig. 1. There are several notable features of these data; the data span nearly five orders of magnitude, but there is only one value more than 10  $\text{m}^2 \text{kg}^{-1}$ , and only three values are  $< 0.001 \text{ m}^2 \text{kg}^{-1}$ . Restricting the analysis to soil deposition of radioactive materials associated with particles of  $\leq 44 \mu\text{m}$  diameter reduces the variance [as suggested by Miller (1963)], and provides a strong indication that the smaller particles are retained preferentially by vegetation. The behavior of the two types of vegetation appears to be very similar, especially for the larger data sets normalized to total deposition. The apparently higher retention by pasture-type vegetation as opposed to native vegetation when normalized to the deposition of particles of diameter  $\leq 44 \mu\text{m}$  (noticeable in Fig. 1) is partly an artifact due to relatively more measurements on cultivated vegetation having been made at distances further from the point of origin as compared to the measurements on native vegetation (Tables 1 and 2). Of course, difference between the two types of vegetation would be expected due to the differences in the leaf surfaces as well.

Most of the data in Tables 1 and 2 are drawn from three relatively obscure sources and have not been published in the open literature, except for the few measurements provided in Romney et al. (1963). Most of the data were available in a draft report (Anspaugh et al.

**Table 1.** Values of mass-interception fraction (dry weight) for native vegetation.<sup>a,b</sup>

Distance, km	$c d^{-1}$ , $m^2 kg^{-1}$	$c d^{-1}$ , $m^2 kg^{-1}$ for $\leq 44 \mu m$	Distance, km	$c d^{-1}$ , $m^2 kg^{-1}$	$c d^{-1}$ , $m^2 kg^{-1}$ for $\leq 44 \mu m$
			Shot Nancy, March 24, 1953, 5:10 a.m., 24 kt, 91 m tower		
30	.013				
			Shot Simon, April 25, 1953, 4:30 a.m., 43 kt, 91 m tower		
26	.0023				
			Shot Tesla, March 1, 1955, 5:30 a.m., 7 kt, 91 m tower		
19	0.00077	0.21	96	0.0086	0.046
19	0.00047	0.024	126	0.0042	0.021
19	0.00078	0.020	126	0.042	0.21
96	0.038	0.20	154	0.049	0.19
			Shot Apple I, March 29, 1955, 4:55 a.m., 14 kt, 152 m tower		
21	0.0015	0.012	64	0.041	0.12
21	0.033	0.26	128	0.044	0.17
21	0.020	0.19	128	0.047	0.19
64	0.097	0.38	128	0.057	0.23
64	0.13	0.49	264	0.40	0.58
64	0.076	0.22	264	0.36	0.52
			Shot Met, April 15, 1955, 11:15 a.m., 22 kt, 122 m tower		
32	0.058	1.2	93	0.15	1.7
32	0.18	0.52	93	0.80	7.5
93	0.22	2.4	224	0.25	5.1
93	0.48	5.2	224	0.33	1.6
			Shot Apple II, May 5, 1955, 5:10 a.m., 29 kt, 152 m tower		
11	0.0021	0.22	77	0.083	0.85
11	0.0027	0.29	77	0.016	0.17
77	0.039	0.30	77	0.084	0.55
77	0.013	0.10	77	0.011	0.071
77	0.068	0.44	170	0.18	0.92
77	0.018	0.12	170	0.082	0.11
77	0.034	0.21	170	0.70	0.70
77	0.0048	0.030	170	0.78	0.78
77	0.039	0.24	170	0.70	0.70
77	0.052	0.32			
			Shot Priscilla, June 24, 1957, 6:30 a.m., 37 kt, 213 m balloon		
11	0.40	0.47	246	0.48	0.60
134	0.21	0.67	304	0.40	0.57
206	0.48	0.60			
			Shot Diablo, July 15, 1957, 4:30 a.m., 17 kt, 152 m tower		
19	0.18	2.2	64	0.58	1.1
24	0.17	3.6	99	1.2	1.5
32	0.15	1.7			
			Shot Shasta, August 18, 1957, 5:00 a.m., 17 kt, 152 m tower		
24	0.14	1.1	122	0.26	0.52
70	0.17	0.72	275	0.75	0.88
			Shot Smoky, August 31, 1957, 5:30 a.m., 44 kt, 213 m tower		
77	0.043	0.28	253	0.13	0.29
128	0.035	0.23	280	0.12	0.27
160	0.023	0.26	330	0.087	0.30
218	0.149	0.26			

<sup>a</sup> Information on shot characteristics is from Collison (1953), Sanders et al. (1955), Placak et al. (1957), and DOE (1994).

<sup>b</sup> Information on mass-interception fractions was taken, computed, or derived from Lindberg et al. (1954, 1959), Miller (1963), and Romney et al. (1963).

1986). Some of those data (13 measurements<sup>††</sup> on herbaceous vegetation at distances of 129 to 416 km from NTS following shots Apple II, Smoky, Tesla, and Met) were reviewed by Whicker and Kirchner (1987) for use in the PATHWAY food-chain model for dose-assessment purposes within the domain nearby NTS. Although Whicker and Kirchner noted that there is evidence that interception decreases with an increase in particle size, they chose to use the geometric mean of the

13 measurements, which was  $0.39 m^2 kg^{-1}$ . This use was for limited application to dose reconstruction nearby NTS and where it was known that the particle sizes of the deposited fallout tended to be quite large (e.g., Cederwall et al. 1990).

Simon (1990) considered the 28 values reported here in Table 2 and derived an empirical fit to the data with the independent parameter being either the distance (km),  $x$ , from the shot site or the time of transit from the shot site, which was referred to as the time of arrival (TOA in hours). The relationships derived by Simon (1990) are

$$c d^{-1} = 7.02 \times 10^{-4} \times x^{1.127} (r^2 = 0.63) \quad (4)$$

<sup>††</sup> There are only 11 such measurements in Table 2. It is possible that two measurements for Apple II that appear in two citations [Romney et al. 1963; Anspaugh et al. 1986 (based on Lindberg et al. 1959)] were counted twice.

**Table 2.** Values of mass-interception fraction (dry weight) for pasture-type vegetation.<sup>a,b</sup>

Distance, km	$c d^{-1}$ , $m^2 kg^{-1}$	$c d^{-1}$ , $m^2 kg^{-1}$ for $\leq 44 \mu m$	Distance, km	$c d^{-1}$ , $m^2 kg^{-1}$	$c d^{-1}$ , $m^2 kg^{-1}$ for $\leq 44 \mu m$
Shot Tesla, March 1, 1955, 5:30 a.m., 7 kt, 91 m tower					
126	0.30	1.5	154	0.32	1.3
Shot Met, April 15, 1955, 11:15 a.m., 22 kt, 122 m tower					
224	0.33	1.7			
Shot Apple II, May 5, 1955, 5:10 a.m., 29 kt, 152 m tower					
11	0.021	2.4	77	0.030	0.19
11	0.040	4.7	77	0.066	0.40
11	0.19	23	77	0.068	0.41
11	0.0054	3.6	77	0.051	0.52
11	0.0054	3.6	77	0.064	0.65
11	0.0046	0.50	77	0.13	0.89
11	0.0022	0.24	77	0.15	0.98
77	0.066	0.51	170	0.078	0.11
77	0.069	0.54	170	0.17	0.23
77	0.047	0.30	170	1.3	1.3
77	0.056	0.36	170	0.86	0.86
77	0.039	0.25			
Shot Smoky, August 31, 1957, 5:30 a.m., 44 kt, 213 m tower					
211	0.24	1.1	414	0.39	—
328	0.62	1.5	414	0.30	—

<sup>a</sup> Information on shot characteristics is from Collison (1953), Sanders et al. (1955), Placak et al. (1957), and DOE (1994).

<sup>b</sup> Information on mass-interception fractions was taken, computed, or derived from Lindberg et al. (1959) and Romney et al. (1963).

**Table 3.** Summary of computed values of mass-interception fractions from measurements following nuclear tests at the Nevada Test Site. The values have been computed by using the dry weights of the vegetation considered.

Parameter	Computed values of mass-interception fractions, $m^2 kg^{-1}$			
	Native desert vegetation		Pasture-type vegetation	
	Total fallout	$\leq 44 \mu m$ fallout	Total fallout	$\leq 44 \mu m$ fallout
Number	70	68	30	28
Arith. mean	0.18	0.81	0.20	1.9
Std. deviation	0.25	1.3	0.29	4.29
Geo. mean	0.062	0.37	0.081	0.82
Geo. std. dev.	6.3	3.7	4.8	3.2

and

$$c d^{-1} = 0.0417 \times TOA^{1.063} (r^2 = 0.61). \quad (5)$$

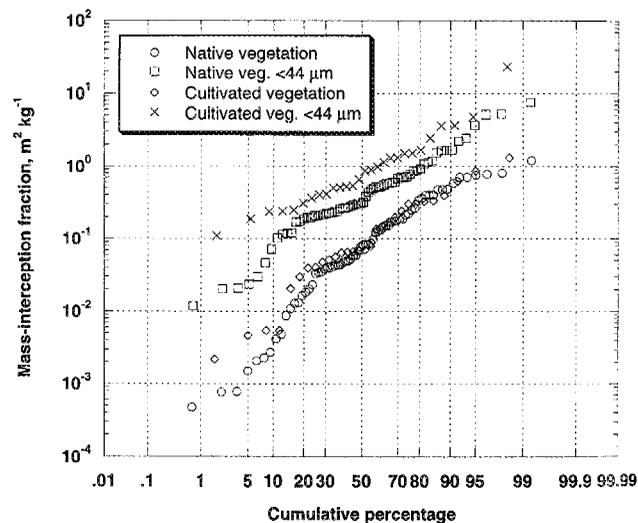
Simon also fit a function to the mass-interception fraction for native vegetation as a function of downwind distance. The result was

$$c d^{-1} = 19 \times 10^{-4} \times x^{0.882} (r^2 = 0.22), \quad (6)$$

which was less successful due to the larger amount of scatter in these data (Table 1 and Fig. 1).

The equations fit by Simon used the variables distance ( $x$ ) and time-of-arrival ( $TOA$ ) as surrogates to the actual (but unknown) distribution of particle sizes deposited. Theory predicts that large particles would be deposited earlier (or at shorter distances) due to gravitational settling.

An interesting opportunity has arisen recently to exchange data related to dose-reconstruction methods



**Fig. 1.** Summary of results for the mass-interception fraction for radionuclides retained by vegetation as measured near NTS. Separate curves are given for native vegetation and for cultivated pasture-type crops. Data are normalized either to total deposition on the ground or the fraction of the total deposition that is associated with particles of diameter  $\leq 44 \mu m$ .

among United States investigators and those working within the former Soviet Union. Comparison of the two methodologies has indicated that results for external dose are similar, as are results for thyroid dose at relatively far distances. However, results calculated for thyroid dose for persons at very close distances differ. Some of the latter difference is due to varying assumptions about lifestyle and food-consumption habits, but much of the difference seems to be due to the formalism used to

**Table 4.** Characteristics of the Soviet tests at the Semipalatinsk Test Site for which measurements have been made and used for the purpose of deriving a method to estimate contamination of vegetation (Gordeev 1999).

Parameter	Values			
Number of test <sup>a</sup>	242	148	2	4
Date	14 Oct. 65	7 Aug. 62	24 Sept. 51	12 Aug. 53
Time		9:00 a.m.	1:10 p.m.	7:30 a.m.
Yield, kt	1.1	9.9	38	400
Height of detonation, m	-48	0	30	30
Cloud top height, $H_{max}$ , km	0.55	5.7	11.6	16.1
Average wind speed, $\bar{u}$ , km h <sup>-1</sup>	40	16	26.4	64.6
$H_{max} \times \bar{v}$ , km <sup>2</sup> h <sup>-1</sup>	22	91.2	306	1040
Critical distance, $x_c$ , km	30	120	420	1426

<sup>a</sup> According to Mikhailov et al. (1996).

estimate the "biologically active fraction" of fallout in the Russian methods compared to the "interception fraction" in the United States methods. Availability in this case essentially refers to the efficiency of retention by vegetation but also includes consideration of the lack of solubility of radionuclides associated with large particles.

A recent report by Gordeev (1999) discusses the issue of the "biologically active fraction" of fallout. The approach used by Gordeev was to normalize the concentration of gross beta activity on vegetation to the external gamma-exposure rate.<sup>††</sup> Specifically, the formulation of this ratio,  $A_{gr}$ , is

$$A_{gr} = \frac{Q_{gr} \sum t_s}{P_{t_s}}, \quad (7)$$

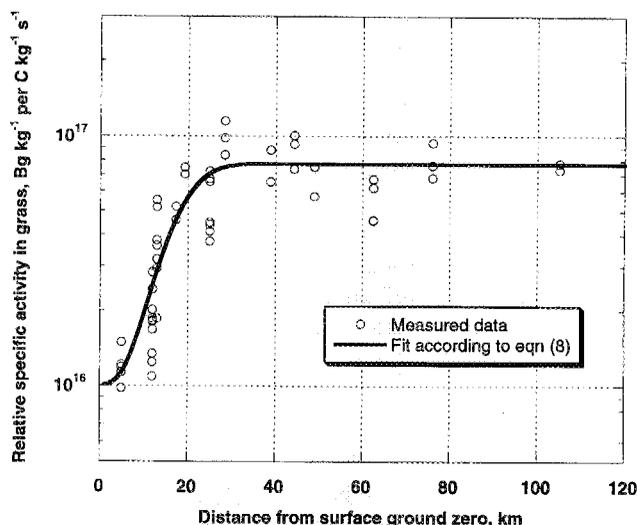
where

- $t_s$  = Time of sampling, h;
- $Q_{gr} \sum t_s$  = Gross specific activity of grass, which is decay corrected to  $t_s$ , Bq kg<sup>-1</sup>; and
- $P_{t_s}$  = External gamma-exposure rate, which is decay corrected to  $t_s$ , C kg<sup>-1</sup> s<sup>-1</sup>.

Measured values of  $A_{gr}$  have been tabulated and examined by Gordeev (1999) for four different tests. The characteristics of the tests and other parameter values to be used later are listed in Table 4. The most studied test, No. 242, was a 1.1 kt test that was placed 48 m underground. Another feature of all four tests was that they occurred in August or later, and it was noted that there seemed to be no loss of activity from the vegetation other than the loss due to radioactive decay. In contrast, much of the similar kinds of data in the United States were associated with tests in the spring or early summer (Table 2).

An example of the Soviet data for Test No. 242 is shown in Fig. 2. This explosion had a yield of only 1.1 kt,

<sup>††</sup> This is essentially the same approach as used by United States investigators, as there is a known correlation between the deposition density of radionuclides and the external gamma-exposure rate, both of which parameters vary with time. These correlations have been derived by Hicks (1982, 1990) for every United States test at NTS.



**Fig. 2.** Russian results (Gordeev 1999) for the mass-interception fraction of fallout retained by vegetation following Test No. 242. Measured data are indicated by circles; a fit to the data is indicated by the solid line.

but measurements were made at sampling points as far as 105 km. What is observed is a 10-fold increase in the mass-interception fraction out to about 30 km and then a plateau beyond that point. This plateau has been rationalized on the basis that only smaller particles are retained on plants, and that there is a critical distance,  $x_c$ , where the plateau occurs that is equal to the distance beyond which no particles of physical diameter  $>50 \mu\text{m}$  exist in the cloud. The critical distance for Test No. 242 is noted to have occurred at 30 km. The critical distance for the largest test of 400 kt is noted to have occurred at 1,426 km.

The curve in Fig. 2 resulted from a fit to the data and is given by the following:

$$A_{gr} = \frac{Q_{gr} \sum t_s}{P_{t_s}} = 7.74 \times 10^{16} [1 - 0.87 \exp(-1.48 \times 10^{-4} x^3)], \quad (8)$$

where  $x$  is the distance of the sampling point from the point of the explosion. It was noted by Gordeev (1999) that eqn (8) is for dry steppe grass. For the typical wet weight of grass, it was suggested that a more appropriate value for the numerical coefficient in eqn (8) would be  $3.10 \times 10^{16} \text{ Bq kg}^{-1} \text{ per C kg}^{-1} \text{ s}^{-1}$ .

One of the useful features of this methodology is its general application to many different types of events, both very small and quite large. The critical distance,  $x_c$ , has been defined as the distance that it would take a particle with a physical diameter<sup>§§</sup> of  $50 \mu\text{m}$  to fall from the top of the cloud to the land surface. This is based

<sup>§§</sup> For Test No. 242, which was detonated underground, it is assumed that the relevant particles consist mainly of soil with a density of  $2.5 \text{ g cm}^{-3}$ . Further, it is assumed that for atmospheric tests the density of the particles is about two times higher.

upon considerations that larger particles are not effectively retained on vegetation, and consideration has been given to related United States results concerning the selected retention of particles of  $\leq 44 \mu\text{m}$  diameter. A general formula describing the settling velocity ( $\text{km h}^{-1}$ ),  $v_d$ , of a particle is given by Gordeev (1999) as

$$v_d = 0.025(d - 22), \quad (9)$$

where  $d$  is the physical diameter of the particle in  $\mu\text{m}$ . Accordingly,  $v_{50}$  is  $0.7 \text{ km h}^{-1}$  for Event No. 242 and  $1.4 \text{ km h}^{-1}$  for atmospheric tests. Thus, the critical distance (km),  $x_c$ , is given by

$$x_c = \frac{H_{\max} \bar{u}}{v_{50}}, \quad (10)$$

where  $H_{\max}$  = maximum height of cloud, km; and  $\bar{u}$  is the average wind speed throughout the cloud layer,  $\text{km h}^{-1}$ . In order to provide a general solution for the retention of radionuclides by vegetation, it has been useful to define a dimensionless reduced distance,  $x_r$ , which is given by

$$x_r = \frac{x}{x_c} = \frac{xv_{50}}{H_{\max} \bar{u}}. \quad (11)$$

For the four shots considered, the average value of the numerical coefficient in eqn (8) for *wet* vegetation [the value in eqn (8) was for dry vegetation] was determined to be  $3.07 \times 10^{16} \text{ Bq kg}^{-1}$  per  $\text{C kg}^{-1} \text{ s}^{-1}$ . Thus, with the normalization of the material retained on vegetation and of the distance to the reduced distance, a general predictive equation for the dimensionless "biologically active fraction,"  $\eta_{d \leq 50}$ , is

$$\eta_{d \leq 50} = 1 - [1 - (H_{\max} \bar{u})^{-0.9}] \exp(-4x_r^3). \quad (12)$$

The parameter  $\eta_{d \leq 50}$  would be more appropriately called the "retained fraction," as Gordeev (1999) defines another parameter to describe the solubility of the fallout retained on the vegetation. The product of those two parameters more closely defines the true "availability" of the radioactive material. The solubility is also described as a function dependent on distance where the change in solubility reflects the differences in particle size and amounts of refractory elements present:

$$\beta_{d \leq 50}(x_r) = 0.0734 + 0.331x_r^{0.55}. \quad (13)$$

The formalism of Gordeev appears to offer a very useful approach for a general method for assessing the fraction of the radioactive materials that are available to be transferred through biological systems. Its use is currently undergoing further evaluation and application to United States and Russian tests. The generic value of  $0.39 \text{ m}^2 \text{ kg}^{-1}$  applied for the mass-interception fraction for dose reconstruction for United States nuclear tests probably resulted in overestimates of thyroid doses at near distances. Some combination of the Simon and Gordeev formulations would likely produce more credible results.

### Fallout deposited with rainfall ("wet" fallout)

Much of the early work on fallout was concerned with dry deposition, as shots were not deliberately fired during periods of expected rainfall. However, as debris clouds traveled far downwind, it was inevitable that rain was encountered and fallout debris contaminated vegetation. Unfortunately, there has not been as much attention given to this parameter, and little work has been done in recent times. Further, there are no comparable Russian methodologies or data for comparison.

Anspaugh (1987) reviewed 14 reports that yielded 20 results for the fractional retention by vegetation of fallout deposited with rainfall. The mean value was 0.31, but results varied from negative values (i.e., when fallout was washed off by rain) up to 0.96. Thus, one suggestion for evaluating the retention,  $p_r$ , of fallout in rain by vegetation is

$$p_{\text{wet}} = 0.3 \times 3^{\pm 1}, \quad (14)$$

meaning that it is reasonable to expect variation from 0.1 to 0.9.

Voillequé (1986) in another review considered 30 values that were found to have a median value of 0.35 with a geometric standard deviation of 2.9—results that are quite similar to those indicated in eqn (14). Voillequé also fit the literature values of mass-interception fraction to a modified form of the Horton (1919) model, which was originally formulated to describe the interception and initial retention of water by plants as a function of the rainfall storage capacity per unit biomass density of vegetation,  $S$ , the amount of rain deposited per storm,  $P_s$ , and the in-storm evaporation fraction per unit biomass density,  $E$ :

$$\frac{P}{w} = \frac{S}{P_s} + E. \quad (15)$$

Voillequé's modified values of  $S$  and  $E$  were  $16 \text{ mm m}^2 \text{ kg}^{-1}$  and  $1.3 \text{ m}^2 \text{ kg}^{-1}$ .

A general drawback to most values found in the literature reviewed by Anspaugh (1987) and Voillequé (1986) is that many values are derived from experiments where dissolved radionuclides have been applied in fine sprays over short periods of time.

More recent attempts to measure the retention of fallout in artificial rain have been reported by Hoffman et al. (1989, 1992, 1995) and by Kinnersley et al. (1997). The results of Hoffman et al. are particularly relevant for fallout from nuclear tests, as Hoffman et al. considered both dissolved  $^{131}\text{I}$  and  $^7\text{Be}$  and radionuclides attached to particles of various sizes; they also attempted to simulate both moderate intensity storms ( $1$  to  $4 \text{ cm h}^{-1}$ ) and high-intensity storms ( $4$  to  $12 \text{ cm h}^{-1}$ ). Kinnersley et al. considered only dissolved cesium. Hoffman et al. (1992) examined fractional retention by vegetation and mass-interception fraction in multiple regressions against biomass,  $w$ , rain amount,  $P$  (mm), and rain intensity,  $I$  ( $\text{cm h}^{-1}$ ). For the fractional retention of materials on vegetation the Hoffman et al. (1992) results indicated that biomass was by far the most important variable for

particle-bound activity and for dissolved  $^7\text{Be}$ . For dissolved  $^{131}\text{I}$  the most important variable was the amount of simulated rain with less  $^{131}\text{I}$  being retained as the rain amount increased. For the mass-interception fraction biomass still tended to be the most important variable, but rain amount was also important and was most important for the dissolved radionuclides. Examples of the regressions\*\*\* found by Hoffman et al. for radionuclides bound to 3  $\mu\text{m}$  and 25  $\mu\text{m}$  particles, respectively, depositing on mixed pasture grass are

$$\begin{aligned} \frac{P}{w} &= 1.54w^{-0.299}P^{-0.207}I^{-0.119} \\ \frac{P}{w} &= 1.38w^{-0.303}P^{-0.124}I^{-0.089} \end{aligned} \quad (16)$$

Hoffman et al. (1992) noted that their results were substantially less than the results derived with the use of eqn (15) with parameters derived by Voillequé from the analysis of values found in the literature. In general, the amounts of  $^{131}\text{I}$  in solution retained on vegetation were much less than the amounts of radionuclides attached to particles.

The Hoffman et al. (1989) and other results were used by the National Cancer Institute (NCI 1997) in their study of thyroid doses to the entire country from the nuclear tests in Nevada. The formalism adopted by the NCI is a complicated combination of theoretical results, experimental measurements, and a necessary constraint that retention not be more than 100%.

For rainfall rates of more than 5  $\text{mm d}^{-1}$  the mass-interception fraction was calculated with the use of eqn (15) multiplied by a factor of 0.7. For rainfall rates between 2.5  $\text{mm}$  and 5  $\text{mm d}^{-1}$ , the mass-interception fraction was a constant value of 3.1  $\text{m}^2 \text{kg}^{-1}$  (dry). For rainfall rates of <2.5  $\text{mm d}^{-1}$ , a value was interpolated according to rainfall rate between the predicted dry deposition at that distance predicted by the Simon (1990) equation [eqn (4) in this paper] and the value of 3.1  $\text{m}^2 \text{kg}^{-1}$  (dry).

Obviously, these rather crude formulations are less than desirable, but nothing more reliable and sophisticated has yet been proposed. More work on evaluating the retention of waterborne radionuclides by vegetation is needed in order to refine the estimates of dose to the residents of the contiguous United States from the tests in Nevada.

## RESUSPENSION

The resuspension of radionuclides after their initial deposit on the ground surface is another physical pathway of interest. There has not been universal agreement that resuspension is an important pathway, but it is now generally accepted that there are a few situations where the pathway could be the dominant one. Many observations have shown that the rate of resuspension decreases

\*\*\* Although this formalism was used for the regression calculations, it may be possible to derive a more useful functional form.

very rapidly with time, and that for accident situations, resuspension is only of importance (compared to the inhalation exposure from the initial cloud passage) over short time periods. For cleanup and property-release situations, resuspension can sometimes be a dominant pathway, especially for transuranic radionuclides or for other radionuclides that do not readily cross biological barriers. Historically, the concern with resuspension has been with isotopes of plutonium, which do not readily cross biological barriers but stay in the lung for long periods if inhaled.

## Relative importance of resuspension

One interesting way to assess the importance of resuspension in an accident situation is to compare the integrated air concentration from resuspension to that of the original integrated air concentration from the passing cloud. If the initial integrated air concentration is  $IAC$  ( $\text{Bq s m}^{-3}$ ), the deposition onto soil and vegetation would be equal to  $IAC \times v_g$ , where  $v_g$  is the deposition velocity ( $\text{m s}^{-1}$ ). The resuspended air concentration would be equal to  $IAC \times v_g \times S_f(t)$ , where  $S_f(t)$  is a time-dependent resuspension factor ( $\text{m}^{-1}$ ). Then, the ratio of the integrated air concentrations would be

$$\text{Ratio} = \frac{\int_0^{\infty} IAC \times v_g \times S_f(t) \times dt}{IAC} \quad (17)$$

A simple, but reasonable model of resuspension with time early after deposit is that the initial value is  $10^{-5} \text{m}^{-1}$  and that it decreases exponentially with increasing time. Under these conditions the ratio becomes

$$\text{Ratio} = \frac{v_g \times S_f(0)}{\lambda} \quad (18)$$

and with the reasonable assumptions that  $v_g$  is  $10^{-2} \text{m s}^{-1}$  and the half time of decrease is 5 wk, then the value of the ratio is equal to  $\sim 0.5$ . Thus, the importance of resuspended air concentration is very roughly equivalent to that of the integrated concentration due to the initial passage of the original cloud. This calculation also points out that most concern about resuspension in accident situations is limited to very early times (the first few weeks) after initial cloud passage. The results of this calculation are also generally consistent with what has been observed over the years from the study of global fallout, although it is typically quite difficult to distinguish between the end of the original "cloud" and the beginning of resuspended air activity.

## General types of resuspension models

Broadly, there are three different types of models that have been used to describe the resuspension process. The first is the time-dependent resuspension factor,  $S_f(t)$ , already introduced above. It is simply the quotient of the observed resuspended air concentration,  $C_a(t)$ , divided by the total deposition density (activity per unit area) of a

radionuclide. Some attempts have been made to redefine the deposition as that down to some level from which resuspension occurs, but this depth is not known.

The second type of model is that of the resuspension rate, which is simply the fraction of the deposited activity that is resuspended per unit time. Although theoretically attractive, this concept has not gained wide use, perhaps because it must be used with some additional model to define the quantity of interest—that of the resuspended concentration in air.

The third type of model is that of “mass loading.” The fundamental assumption is that the suspended mass at any local point is strongly correlated with the local soil. Thus:

$$C_a = C_m C_s E, \quad (19)$$

where  $C_m$  is the mass loading in air ( $\text{kg m}^{-3}$ ),  $C_s$  is the concentration of the contaminant in the soil ( $\text{Bq kg}^{-1}$ ), and  $E$  is an “enhancement” factor. The concept of an enhancement factor has been added to the basic mass-loading model to allow for the possibility that the concentration of the contaminant might be higher in the small soil particles likely to be resuspended.

The resuspension-factor model has been widely used to predict the concentration of resuspended contaminant at times early after the initial deposition. The mass-loading model has generally been preferred for times long after the deposition. However, at times long after deposition and in situations where there is a real and legitimate concern about resuspension, it is always preferable to rely on actual measurements that are performed over long time periods.

### Types of resuspension-factor models

Over the years several different types of resuspension-factor models have been proposed. Perhaps the first was due to Langham, who carried out experiments at NTS during 1956. His observations, reported later in Langham (1969), were of the form

$$S_f = S_f(0)\exp(-\lambda t), \quad (20)$$

and for which Langham gave values of  $S_f(0) = 10^{-6} \text{ m}^{-1}$  and  $\lambda = 0.693 (45 \text{ d})^{-1}$ . A similar, more conservative model was used by Kathren (1968) to propose interim standards for surface contamination of plutonium oxide. Kathren's proposal included values of  $S_f(0) = 10^{-4} \text{ m}^{-1}$  and  $\lambda = 0.693 (35 \text{ d})^{-1}$ .

Another early analysis was given by Shreve (1958) that described a time-dependent resuspension model as a power function:

$$C_a(t) = C_a(0) \times 0.177 \times t^{-0.75}. \quad (21)$$

This provided a fairly good fit to data associated with a field release of plutonium near NTS in 1957 (Wilson et al. 1961), but a simple exponential function with a half life of 35 d provided better estimates at later times. A power function has also been used by more recent investigators. A variant of eqn (20) was proposed by Anspaugh et al. (1975) of the form

$$S_f = S_f(0)\exp(-\lambda t^{0.5}) + C, \quad (22)$$

where  $S_f(0) = 10^{-4} \text{ m}^{-1}$ ,  $\lambda = 0.15 \text{ d}^{-0.5}$ , and  $C = 10^{-9} \text{ m}^{-1}$ . The feature of a long-term constant value was needed in order to describe the results of measurements made 15 to 17 y after a contaminating event at NTS. This feature of a long-term constant value has been used in several other studies (AEC 1974; NRC 1975).

### Recent results in evaluating resuspension

There have been many reviews of resuspension (e.g., Linsley 1978; Smith et al. 1982; Nicholson 1988). Unfortunately, there are probably more reviews than there are measurement sets to be evaluated. The Chernobyl accident provided the opportunity for new measurements, but with rare exception (Garger et al. 1997) such measurements were undertaken too late in time to be of real value in resolving the issue of resuspension at early times.

Recently, Maxwell et al.<sup>††</sup> have undertaken an extensive review of all resuspension measurements with the goal of adding new results to the existing data bank. The addition of new data is possible, because new results can be added by recalculating old data that may have been referenced to measured external gamma-exposure rate. With the results of Beck (1980) and Hicks (1982, 1990), it is possible to derive ground-deposition values from such measured external gamma-exposure rates. This has resulted in an extensive addition to the databank of evaluated measurements.

Several things are clear from this expanded database. One is that the scatter in the data is enormous; this indicates that the resuspension process is very complicated. Clearly, the resuspension process cannot be understood in terms of saltation models, as some of the higher concentrations of resuspended materials are observed at times of very low wind speed. The other factor of interest is that none of the current resuspension models adequately describes the empirical results. Some models severely over predict the measured results at intermediate times, while others severely under predict the same results. A preliminary suggestion of a predictive model has been derived from this expanded data set:

$$S_f = [10^{-5}\exp(-0.07t) + 6 \times 10^{-9}\exp(-0.003t) + 10^{-9}] \times 10^{\pm 1} \text{ m}^{-1}. \quad (23)$$

This proposed model more accurately describes the observed results over the entire span of the data set. It also offers an explicit statement of the uncertainty in the model, which is taken to be a factor of 10. This is a large uncertainty, but it is indicative of the dispersion in the data sets.

<sup>††</sup> Maxwell RM, Anspaugh LR, Shinn JH, Kercher JR. An evaluation of resuspension data and derivation of an improved model to be used for predictive purposes. Livermore, CA: Lawrence Livermore National Laboratory; in preparation; 2001.

## VERTICAL MIGRATION OF RADIONUCLIDES INTO SOIL

The final topic to be considered is the rate of migration of radionuclides into the soil column. This topic is of interest for several reasons. The migration determines the long-term external gamma-exposure rate from deposited gamma-emitting activity, as more shielding is achieved as the radionuclides penetrate into soil. The migration also has a long-term effect on the uptake of radionuclides by plants through their roots. Other considerations relate to cleanup issues, such as the validity of sampling to determine original depositions for  $^{129}\text{I}$ ,  $^{137}\text{Cs}$ , and  $^{239+240}\text{Pu}$  and how deeply a proposed cleanup should be done.

The discussion here on vertical migration is intended to apply only to temperate climates and to normal soil. The rate of movement of  $^{137}\text{Cs}$  in soil at tropical atolls, for example, would be very different due to the much larger amounts of rainfall and the very different soil system that contains very little potassium or clay.

Historic considerations (Beck 1966; UNSCEAR 2000) have been that depositions of radionuclides on soil immediately experience a soil-roughness effect that is equivalent to the shielding afforded by 1 mm of soil; then radionuclides move to an average depth of about 1 cm within 1 mo. And finally, within a year radionuclides move to a depth of 3 cm and stay there indefinitely. More recent measurements made many years after the deposition of fallout from global and Nevada sources indicate that once  $^{137}\text{Cs}$  reaches the depth of 5 cm it does not stay there indefinitely but does move very slowly to deeper levels (Miller and Helfer 1985; McArthur and Miller 1989).

Efforts to quantify this process more precisely by measuring the penetration of global fallout into soil have not been very successful, because fallout occurred over many years and it has not been possible to distinguish the effects of the yearly impulses. Experiments designed to capture the process (Gale et al. 1964) were helpful but gave very limited results.

The large pulse of radionuclides deposited within a short period of time by the Chernobyl accident has afforded a new opportunity to examine this process in more detail. One interesting result is that of Straume et al. (1997), who collected soil samples in 1993 at Pogonoe, Belarus, a location 20 km north of the Chernobyl Nuclear Power Plant, and measured three radionuclides of different chemical properties. The results indicated that  $^{129}\text{I}$ ,  $^{137}\text{Cs}$ , and  $^{239+240}\text{Pu}$  had all moved into the soil at the same rate, as the distributions with depth in soil were essentially identical. This provides further credence that the process of vertical migration into soil is physical, rather than chemical, at least in the types of soil considered.

Other findings that are of greater relevance to radiological assessment are those presented in Likhtarev et al. (2002). The evaluations in that publication are based upon the results of measuring more than 400 soil-depth profiles and measurements of external

gamma-exposure rate at more than 20 meteorological stations. The evaluated results for the "reference Ukrainian attenuation function" for the decrease in external gamma exposure rate in air due to the weathering of  $^{137}\text{Cs}$  into soil are given by

$$r_{\text{Cs}}(t) = 0.82[0.4 \exp(-0.46t) + 0.6 \exp(-0.024t)], \quad (24)$$

where the 0.82 factor is due to the immediate shielding effect of soil roughness and the two rate constants correspond to half lives of 1.5 and 50 y. These half lives describe the time dependence of ecological processes and do not include the additional attenuation due to radioactive decay. The long-term ecological half life must be considered as uncertain due to the relatively short period of observation and is subject to future revision.

## CONCLUDING REMARKS

This paper has reviewed and discussed the present state of knowledge with respect to environmental processes that influence and partially govern movement, as well as retention, of radionuclides in terrestrial ecosystems. The data presented here pertain mainly to continental sites and soils of volcanic origin rather than to tropical ecosystems and carbonate-based soils. The processes in tropical ecosystems would not be different, but would require site specific data. Much of the data reviewed became available from studies following the deposition of radioactive fallout from nuclear weapons testing. Because there are only a few instances, e.g., the Chernobyl accident, that have produced new data since the active decades of above ground nuclear testing (primarily 1950's to early 1960's), few new data have emerged in recent years. Nevertheless, the need for more high quality data still exists to refine assessment models for both retrospective dosimetry and for assessing future accidents.

The three processes discussed here—interception and retention of radioactive particles on vegetation, resuspension of radioactive materials, and weathering of radioactive materials from surface to deep soil layers—have varying importance depending on the time period after the release. Moreover, evaluation of each of the three processes is supported by different amounts of descriptive data available for modeling purposes.

The process likely to have the greatest importance to food-chain transport is that of interception and retention of radioactive materials on vegetation surfaces. Progress in conceptualizing that process has distinguished immediate interception (primarily governed by physics of particles and gravitational settling) from that of longer term retention (governed additionally by chemical factors such as solubility). However, the scatter of interception data, like that for resuspension, indicates that the processes are complex. Consequently, models of either process are empiric and do not capture details of the mechanisms. While empirical captures are useful for assessments under familiar scenarios, models that would

account for fundamental principles would ultimately be more useful for new contamination situations and/or environments.

In closing, we note that the last decade and a half has emphasized two points about furthering the state of knowledge of environmental transport. First, accidental releases sometimes do occur and it is imperative that when such unfortunate circumstances occur measurements be made quickly and efficiently such that our knowledge base is broadened. Secondly, it is clear that historic data can sometimes be revisited and modeled in ways not previously considered. For example, an international collaboration to compare interception models, devised independently by Russian and United States scientists, is now underway. That is one example of how improvements in models and understanding can be made even when new data are not available.

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