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Retention by Vegetation of Radionuclides Deposited in Rainfall—A Literature Summary

L. R. Anspaugh

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LAWRENCE LIVERMORE NATIONAL LABORATORY
University of California • Livermore, California • 94550

MASTER

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PREFACE

This work was completed in 1972, and it was published as part of a larger report that was classified. Because this work received only very limited distribution and because of the current concern with reconstructing radiation doses received from the ingestion of radioiodine, this work is now being published as a separate report with unlimited distribution.

However, the reader is cautioned that the literature reviewed for this report only extended through 1972, and that the conclusions reached in this report were directed toward use in a predictive model. A somewhat different analysis would be appropriate for use in reconstructing doses already received.

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RETENTION BY VEGETATION OF RADIONUCLIDES DEPOSITED IN RAINFALL--A LITERATURE SUMMARY

ABSTRACT

This report is a review of the literature pertaining to the retention on vegetation of radionuclides deposited in rainwater. Data of two types were reviewed--the results of experiments with artificial tracers and data from direct measurements of naturally occurring and fallout-produced radionuclides washed out or rained out by storms. The data were considered from the viewpoint of choosing a value for use in a predictive model. Individual measurements of retention varied from negative to over 100%. The conclusion is reached that a value of 0.4 to 0.5 would be appropriate for average retention, and that a value of 1.0 would not be unreasonable if one wished to be conservative, particularly where high vegetation densities might be expected.

INTRODUCTION

A review of data on the retention by vegetation of fallout carried to the earth's surface by precipitation has been undertaken. The specific purpose is to examine whether the assumption of 25% retention as used in the 2BPUFF code (1-3) is reasonable. This value has been criticized as excessively conservative, but available data indicate that it may be low for purposes of a predictive model.

Data are available from many studies, which may be divided into two general categories. The first involves experiments where contamination is sprayed on vegetation and the retention is directly measured. The second involves studies of the deposition and retention of actual fallout debris brought down by precipitation. These latter studies are somewhat less satisfactory as many involve long-term measurements with fits to models to derive the necessary parameters. A few measurements of deposition following single rainstorms are available, however, but are compounded by simultaneous washoff of previously deposited debris.

Data from studies of both types are summarized below. Included are all such studies that we have been able to identify and locate at this time.

EXPERIMENTAL DATA

STUDIES USING ARTIFICIAL SPRAYS

Several experiments have been done using artificial sprays. Nearly all have utilized radionuclides in a soluble form. This may prejudice the results somewhat, although it has been observed that many radionuclides in fallout deposited by precipitation are water soluble (4). Also, one study (5), which will be discussed in more detail below, found that the retention of 1- μ m particles tagged with insoluble ^{51}Cr was essentially the same as the retention of radionuclides in solution. Only one study has examined the relationship between retention and the volume of solution applied (6). It is unfortunate, however, that only the relative magnitudes of the solution volumes are reported. Relevant data from each individual study are summarized below.

1. Middleton (7,8). Wheat, cabbage, potatoes, and sugar beets were grown in pots or boxes. Replicate groups of plants at various stages of growth were placed within a spraying apparatus and sprayed with solutions containing ^{89}Sr or ^{137}Cs in carrier-free, completely soluble form. The rate of application was 30 to 90 μCi in 6-7 mL of solution per square yard. The initial retention was determined by taking samples from duplicate batches of plants and comparing these results to the deposition per unit area determined by spraying strips of filter paper under similar conditions. Pots containing cabbage were placed under the spraying machine as close together as possible while still avoiding shading. The density of potato and sugar beet plants also exceeded that found in normal agricultural practice. The results of this study are summarized in Table 1. Values of the fractional initial retention calculated on an area basis range from 0.042 to 0.650. An unweighted average of all data in Table 1 is 0.31.

2. Milbourn and Taylor (9). Experiments were carried out on representative areas of pasture in different parts of the United Kingdom. Cultivation procedures were approximated as closely as possible to those used in normal practice. Carrier-free ^{89}Sr was applied as a spray using a spraying machine in most experiments. Each plot was sprayed twice with 10 to 30 minutes between sprayings. Each of the two sprayings deposited 85 mL of solution per m^2 . At one location the spraying machine could not be used, so the solution was applied in two equal portions from a can fitted with a sprinkling nozzle for a total application of 440 mL/m^2 . Initial retention was determined by

Table 1. Values of the initial retention of ^{89}Sr and ^{137}Cs applied in soluble form as a fine spray as determined by Middleton (7,8). The amount of solution applied was 6 to 7 mL/yd², which is equivalent to a rainfall of 0.007 to 0.008 mm. Values for the initial retention are the fraction retained by the plant on an areal deposition basis.

Plant	Year	Time from planting to contamination, days	Initial retention, fraction	
			^{89}Sr	^{137}Cs
Wheat	1957	100	0.330	0.204
		105	0.259	0.301
		149	0.273	0.289
	1958	86	0.400 $\pm$ 0.064	0.446 $\pm$ 0.084
		107	0.470 $\pm$ 0.068	0.523 $\pm$ 0.041
		136	0.540 $\pm$ 0.068	0.585 $\pm$ 0.052
Cabbage	1957	32	0.081	0.042
		88	0.086	0.087
	1958	58	0.085 $\pm$ 0.009	0.141 $\pm$ 0.026
		72	0.292 $\pm$ 0.026	0.281 $\pm$ 0.020
		87	0.309 $\pm$ 0.040	0.309 $\pm$ 0.036
		99	0.281 $\pm$ 0.016	0.280 $\pm$ 0.044
Potato	1957	55	0.480	0.480
		69	0.650	0.520
	1958	47	0.270	0.240
		74	0.320	0.450
		97	0.580	0.390
Sugar beet	1957	62	0.269	0.187
	1958	48	0.145	0.125
		71	0.185	0.315

sampling herbage immediately after the spray had dried (about 4 hours) and comparing this on an areal basis to the total deposition determined by measuring the deposition on strips of absorbent paper mounted on aluminum sheets. The purpose was to determine the contamination of herbage edible by cattle, so grass was sampled only down to 2 inches above the ground surface. The results of this study are summarized in Table 2. Values range from 0.167 to 0.301 with an average of 0.23.

3. Edvarson et al. (10). Experiments were carried out at a farm near Stockholm where a pasture field was divided into several pens of about 3000 m² each. Several pens were sprayed with a 250-L solution containing carrier-free ¹³¹I as NaI. Each pen was considered adequate to provide feed for two days for nine milk cows. Twelve hours after the contamination, the cows were allowed into the pens to graze for 48 hours. Six cows were "calibrated" with single oral doses of ¹³¹I, and from this it was calculated that the grazing cows had consumed in the 2-day period about 10% of the total ¹³¹I applied in the pen area. If we assume that the cows trampled or otherwise made unavailable for grazing one-half of the area (11), then the initial retention would be approximately 20%. With the further assumption that dairy cows in Sweden require 10 kg dry matter per day (12), the forage yield is calculated to be 120 g dry wt/m². The application of 250 L to a 3000 m² area is equivalent to a 0.08-mm rain.

4. Kirchmann et al. (6). Experiments were done by contaminating rye-grass and red clover grown in pots by "watering" with the radioactive solution, and by using a motor-sprayer to contaminate pasture. Unfortunately, many of the pertinent details of the experiments are not provided. The results of one laboratory experiment are shown in Table 3 where the effect of the concentration of the solution was studied. It is not possible to tell how much of the solution was applied, or exactly how it was applied. Another study, summarized in Table 4, investigated the effect of the volume of the radioactive solution applied. Again it is not possible to determine how the solution was applied or what the actual volumes applied were except in a relative way. The effect of plant density on initial retention was also investigated, but results are reported only in a very general manner. Three phases of increase with plant density were noted. A steep increase in retention with increasing plant density was noted as the density increased from about 20 to 22 g dry wt/m². Retention continues to increase at a slower rate as density increases from 22 g dry wt/m² until complete retention is found at a density of 610 g dry wt/m². No further details of this experiment are provided, but the authors state that similar results were obtained under natural conditions by Kirchmann and Boulenger (13). A copy of this report was not available at this writing.

Kirchmann et al. also performed a laboratory experiment where a volume of solution equivalent to 5 mm was applied every two days to rye-grass over a period of 28 days. The rye-grass was five-weeks old at the beginning of the experiment. The herbage density varied from about 25 to 75 g dry wt/m² from the beginning to the end of the experiment. The activity retained by the rye-grass on a unit-area basis closely paralleled the cumulative deposition with a retention value throughout the experiment of about 0.06 for both ⁸⁵Sr and ¹³⁴Cs.

Table 2. Values of the initial retention of ^{89}Sr applied in soluble form as a spray onto pasture grass as reported by Milbourn and Taylor (9). Values for the initial retention are the fraction retained by the grass on an areal deposition basis.

Yield of herbage, g dry wt/m ²	Rainfall equivalent of applied solution, mm	Initial retention, fraction
73	0.17	0.208 ± 0.030
229	0.44 ^a	0.256 ± 0.025
81	0.17	0.173 ± 0.024
50	0.17	0.173 ± 0.021
70	0.17	0.167 ± 0.029
130	0.17	0.259 ± 0.037
62	0.17	0.262 ± 0.017
58	0.17	0.301 ± 0.020

^a Solution applied with a sprinkling can.

Table 3. Values of the initial retention of ^{85}Sr and ^{134}Cs as reported by Kirchmann *et al.* (6). The effect of the concentration of the radioactive solution was studied; it is not possible to tell how much solution was applied or how it was applied. The density of vegetation (rye-grass) was ~130 g dry wt/m². Each result is the mean of three replications.

Concentration in solution, $\mu\text{Ci/L}$	Deposition, cpm/cm ²		Initial retention on shoots, fraction	
	^{85}Sr	^{134}Cs	^{85}Sr	^{134}Cs
2	361	969	0.102	0.065
10	2171	6216	0.082	0.049
25	5476	16865	0.071	0.033
50	8530	27593	0.077	0.033

Table 4. Values of the initial retention of ^{85}Sr and ^{134}Cs on rye-grass as a function of the volume of radioactive solution applied. It is not possible to determine what the actual volumes applied were or how they were applied. Density of the rye-grass was $\sim 130 \text{ g dry wt/m}^2$. Results are from Kirchmann *et al.* (6); each result is the mean of three replications.

Deposition, cpm/cm ²		Initial retention on shoots, fraction	
^{85}Sr	^{134}Cs	^{85}Sr	^{134}Cs
218	655	0.151 _•	0.132
512	1477	0.173	0.152
1314	3720	0.088	0.066

5. Ellis, Mercer and Milbourn (14). This paper is largely a continuation of similar work done by Milbourn and Taylor (9), which has already been discussed. The method of contamination and the amounts applied were apparently the same as those used previously (9). Experiments were done at seven different pastures in the United Kingdom over a three-year period. Pasture samples were cut to 10 cm above the ground when they reached a height of 30 cm, so the time period between contamination and harvest ranged from 3 to 8 weeks. Data from 15 experiments were fit with an exponential function; this resulted in a calculated decay constant of $0.055 \pm 0.017/\text{day}$. Extrapolation to time zero results in an average value for the initial retention of slightly more than 0.25. It is not possible to determine the herbage density at the time of application; the contaminating radionuclide was ^{89}Sr .

6. Aarkrog (15). ^{85}Sr , ^{134}Cs , ^{54}Mn , and ^{141}Ce were sprayed on spring varieties of rye, oats, barley, and wheat at four different stages of development. The plants were grown in circular pots of 40-cm diameter. A spraying chamber was used to apply 10 mL of the contaminating solution in 5 sec (the solution contained 25-ppm carrier of each nuclide) followed by a spray of 10 mL of distilled water. After the plants were sprayed they were covered with a wire cage covered with plastic foil for two days. At the end of this period, all plant material in the plot was harvested to determine the initial retention. The plant yields in this experiment were reported to be significantly greater than is usual under field conditions. The volume of solution applied is equivalent to a

0.08-mm rain followed by a relatively uncontaminated rain of an additional 0.08 mm. The results of this experiment are summarized in Table 5. The reported fractional initial retention varied from 0.301 to 0.976 with an average of 0.58.

7. Chadwick and Chamberlain (5). Experiments were done on an area of old grassland, which consisted of a mixture of grasses, clover, and weeds. Solutions containing carrier-free ^{85}Sr , ^{51}Cr , and ^{210}Pb were sprayed on plots. A suspension containing 1- μm diameter polystyrene particles tagged with ^{51}Cr was also sprayed. All sprays were delivered at the rate of 1 litre/ yd^2 , which is equivalent to a 1.2-mm rain. The initial retention was determined by clipping the grass close to the soil surface 2 hours after the application. The results are summarized in Table 6. Values of the initial retention range from 0.201 to 0.816 with a mean of 0.403. The data were interpreted in the form of a filtration model where it may be expected that the fraction initially retained, p , will be related to the herbage density, $w(\text{kg dry wt}/\text{m}^2)$ by $1 - p = \exp(-\mu w)$. The value of μ was determined to be $2.30 \pm 0.08 \text{ m}^2/\text{kg dry wt}$ for their work and from that of Milbourn and Taylor (9) $3.33 \pm 0.56 \text{ m}^2/\text{kg dry wt}$. The above results were derived using only the data for Sr retention. The unusually high retention of ^{210}Pb was unexplained although reference is made to other observations (16) that indicate a substantial retention by herbage of ^{210}Pb originating from natural fallout.

8. Douglas, Black and Barth (17). These experiments were performed at the dairy farm at the Nevada Test Site. Growing alfalfa was contaminated with a spray containing 23 mCi of ^{131}I and 5 g of KI as carrier per 55 gal. A power sprayer was used, which delivered an average activity deposition of $24.6 \mu\text{Ci}/\text{m}^2$, with an average of 0.18 mm of precipitation. In order that the effect of washoff could be studied, five plots were sprayed with uncontaminated water following the contamination at the levels of 0.08, 1.0, 0.81, 0, and 209 mm. No washoff effect was observed even at the extreme level of 209 mm, which was applied starting 4.5 hours after the contaminating event and continuing for 19.5 hours. The data are not reported in terms of initial retention, but a curve is provided showing the activity in $\text{pCi}/\text{kg dry wt}$ versus time following exposure. Extrapolation to time zero gives a value of $5.5 \times 10^7 \text{ pCi}/\text{kg dry weight}$. An appropriate value for the yield of the alfalfa is about $300 \text{ g dry wt}/\text{m}^2$ (18), which gives an initial retention value of 0.7.

Table 5. Values of the initial retention of radionuclides contained in a spray solution as determined by Aarkrog (15). The volume applied was equivalent to a 0.08-mm contaminated rain followed by 0.08 mm of rinse. The dry matter yields were reported to be significantly greater than is usual under normal field conditions.

Plant	Plant yield, g dry wt/m ²	Date	Initial retention on plants, fraction			
			¹⁴¹ Ce	⁸⁵ Sr	⁵⁴ Mn	¹³⁴ Cs
Rye (awns cut off)	2400	Aug 1	0.444	0.551	0.570	0.637
	2560	Sep 4	0.519	0.529	0.560	0.558
Rye (with awns)	1380	Jun 26	0.825	0.810	0.775	0.791
	1190	Jul 5	0.613	0.573	0.566	0.587
	2400	Aug 1	0.408	0.480	0.478	0.514
	2560	Sep 4	0.487	0.557	0.570	0.557
	570	Jun 26	0.661	0.612	0.595	0.595
Oats	600	Jul 5	0.615	0.629	0.619	0.648
	1740	Aug 1	0.418	0.461	0.441	0.481
	2060	Sep 4	0.417	0.533	0.488	0.497
	2350	Aug 1	0.494	0.568	0.588	0.588
Barley (awns cut off)	2130	Aug 16	0.608	0.748	0.776	0.770
	1040	Jun 26	0.957	0.916	0.976	0.875
Barley (with awns)	1110	Jul 5	0.792	0.795	0.822	0.793
	2350	Aug 1	0.495	0.583	0.584	0.629
	2130	Aug 16	0.655	0.708	0.720	0.734
	320	Jun 26	0.508	0.470	0.488	0.494
	390	Jul 5	0.510	0.498	0.448	0.519
Wheat (awnless variety)	883	Aug 1	0.301	0.349	0.352	0.398
	1130	Sep 4	0.431	0.495	0.490	0.497
	971	Aug 1	0.431	0.493	0.478	0.528
	971	Sep 4	0.474	0.550	0.454	0.538
Wheat (with awns)	420	Jun 26	0.807	0.793	0.788	0.761
	410	Jul 5	0.564	0.639	0.575	0.554
	971	Aug 1	0.415	0.508	0.509	0.536
	971	Sep 4	0.506	0.575	0.566	0.568

Table 6. Initial retention on grassland as determined by Chadwick and Chamberlain (5). Radioactivity was applied as a solution or as a suspension (particles) by spraying 1 litre/yd², which is equivalent to a 1.2-mm rain. Each value represents six replications.

Herbage density, g dry wt/m ²	Date	Initial retention on herbage, fraction			
		⁸⁵ Sr	⁵¹ Cr	²¹⁰ Pb	Particles
190	12/22/66	0.369 ± 0.010	0.484 ± 0.033		
228	5/25/67	0.383 ± 0.025	0.324 ± 0.025		
179	1/17/68	0.357 ± 0.015		0.816 ± 0.017	
100	5/23/68	0.201 ± 0.007		0.294 ± 0.028	0.217 ± 0.009

STUDIES UNDER NATURAL CONDITIONS

Several investigations on the retention of fallout by vegetation were conducted during periods when fallout levels were readily measurable. As might be expected, these studies show substantial variability, but do provide strong evidence that a very high fraction of fallout deposited with precipitation can be retained by vegetation although this is not always the situation. The studies are summarized individually below.

1. Morgan (19). Samples of soil were taken, the fallout ⁹⁰Sr quantified, and were then spiked with ⁸⁵Sr. Eighteen plots were then made up, sown with rye-grass and allowed to germinate in a greenhouse supplied with filtered air. A week after sowing nine plots were transferred to the outside and nine remained in the greenhouse. The crops were harvested 32 days after sowing, and the content of Ca, ⁹⁰Sr, and ⁸⁵Sr measured. The amount of ⁹⁰Sr taken up from the soil by the outdoor plants is calculated using the ⁹⁰Sr/⁸⁵Sr ratio of the indoor plants and the ⁸⁵Sr content of the outdoor plants. During the period of exposure rainwater was collected in two polythene rain gauges; 64 mm of rain fell during this period and the ⁹⁰Sr deposition was determined to be 30.7 pCi/ft². The retention of this deposition by the plant may be determined from the data supplied and is summarized in Table 7. The average retention for the nine samples was 0.23 with a range of 0.19 to 0.28. This experiment, however, has neglected the effects of dry deposition and the field loss rate; these effects would tend to cancel each other but to an uncertain extent.

Table 7. Retention of rain-deposited ^{90}Sr by rye-grass plots exposed for 25 days to fallout. Activity of 30.7 pCi/ft^2 was deposited in 64 mm of rain. Corrections were made for soil-root uptake calculated from an ^{85}Sr spike in the soil and $^{90}\text{Sr}/^{85}\text{Sr}$ ratios in plants grown in a greenhouse supplied with filtered air. The experiments were done in 1957 by Morgan (19).

Harvest yield, g dry wt/m ²	Total contamination, pCi/ft ²	Direct contamination, pCi/ft ²	Retention fraction
277	14.06	8.58	0.28
326	12.99	6.89	0.22
295	12.88	7.31	0.24
199	9.57	5.88	0.19
209	10.46	6.66	0.22
241	11.46	7.34	0.24
140	12.83	6.21	0.20
140	13.88	7.53	0.25
137	13.86	6.76	0.22

2. Hill (16). This study has already been referred to above. It was quoted by Chadwick and Chamberlain (5) as indicating a substantial retention by vegetation of natural ^{210}Pb fallout. Samples of grass were collected during December 1959 from twenty points in southern Great Britain and analyzed for ^{210}Pb . A good correlation ($r = 0.87$) was observed between the ^{210}Pb content as pCi/g and the mean annual rainfall. Based on measurements at Harwell, Hill estimates that ^{210}Pb deposition at any point in Great Britain is $2260 \text{ pCi/m}^2/\text{year}$ for each 100 cm of annual rainfall. Hill further estimates that 23% of the mean annual rainfall fell in the three months preceding the sample collections and the yields of grass were in the range of 20 to 200 g dry wt/m^2 . The retention values that can be calculated from these data are given in Table 8. Obviously, these data are of limited usefulness, but even the minimum estimate of retention for a yield of 20 g dry wt/m^2 averages 0.2. As in the previous study, the effects of dry deposition and the field loss factor were neglected.

Table 8. Estimates of the retention by grass of ^{210}Pb deposited in rainfall over a three-month period based upon the data of Hill (16). Estimates are based upon Hill's assumption that 23% of the mean annual rainfall was received during the three months preceding sample collection and that the yields were in the range of 20 to 200 g dry wt/m².

Mean annual rainfall, cm	^{210}Pb activity, pCi/g dry wt	^{210}Pb activity per 100 cm of annual rain, pCi/g dry wt/100 cm	Fractional retention calculated for	
			200 g dry wt/m ²	20 g dry wt/m ²
63	0.74	1.2	0.46	0.046
63	1.9	3.0	1.2	0.12
63	3.0	4.8	1.8	0.18
63	3.6	5.7	2.2	0.22
69	5.3	7.7	3.0	0.30
90	2.5	2.8	1.1	0.11
93	4.0	4.3	1.7	0.17
150	2.2	1.5	0.58	0.058
150	3.6	2.4	0.92	0.092
150	4.6	3.1	1.2	0.12
170	5.2	3.1	1.2	0.12
130	8.9	6.8	2.6	0.26
180	7.9	4.4	1.7	0.17
180	8.3	4.6	1.8	0.18
220	9.8	4.5	1.7	0.17
190	12.	6.3	2.4	0.24
190	12.	6.3	2.4	0.24
230	14.	6.1	2.3	0.23
240	16.	6.7	2.6	0.26
180	18.	10.	3.8	0.38
310	23.	7.4	2.8	0.28
380	23.	6.1	2.3	0.23

3. Peirson and Keane (20). Measurements were made at Chilton, England, of the ^{131}I and ^{140}Ba content of air, rain, and grass during September to November 1961. This was a period of tropospheric fallout. The data were averaged into 10 weekly periods, and analyzed according to the equation

$$\frac{dC_g}{dt} + \lambda C_g = v_g C_a + b R C_r - k R C_g$$

where C_g = Concentration per unit area of grass,

λ = Radioactive decay constant,

v_g = Velocity of dry deposition,

C_a = Concentration per unit volume of air,

b = Retention factor for rain deposited activity,

R = Rainfall rate, mm/week,

C_r = Concentration per unit volume of rain, and

k = Fraction of deposition on grass washed off by unit rainfall.

It was further assumed that b is dependent upon the rainfall rate so that $b = 1 - mR$. Ten equations with three unknowns ($\bar{v}_g$, m , and k) were solved by multiple regression analysis. The solution yielded the following values for ^{140}Ba and ^{131}I , respectively: $\bar{v}_g$, 0.26 ± 0.24 and 0.26 ± 0.18 cm/sec; m , -0.010 ± 0.027 and 0.015 ± 0.013 week/mm; and k , 0.009 ± 0.018 and 0.020 ± 0.028 /mm. The values of the retention factor b that can then be derived from the data are listed in Table 9. These values are found to be very high, the lowest value observed is 0.48 for ^{131}I with a rainfall rate of 34.8 mm/week. The authors state that the derived values for m (and therefore b) are "virtual" values because of the contribution of dry deposition on the funnel used to collect rainwater. Retention values shown in Table 9 are therefore expected to be somewhat in error.

4. Menzel et al. (21). Sweet corn, cabbage, potatoes, and snap beans were grown according to recommended commercial practices at Belle Grade, Florida, during the spring of 1962. ^{90}Sr content was determined on samples consisting of three randomly selected subplots of 4 m^2 each from which vegetation was cut from 5 to 7 cm above ground. Samples that were analyzed were taken immediately before and after three rainy periods and at nearly equal intervals between rains. Rainfall was collected in pans and the ^{90}Sr deposition in rainfall determined. Except when the crops were very small, the ^{90}Sr content increased greatly during each rain interval; comparatively little

Table 9. Values of the retention factor b derived from Peirson and Keane (20). Data were fit assuming b is a function of rainfall rate R so that $b = 1 - mR$. Values of m were -0.010 ± 0.27 week/mm for ^{140}Ba and 0.015 ± 0.013 week/mm for ^{131}I .

Week, mid-date	Rainfall (R), mm/week	Retention factor (b),	
		^{140}Ba	^{131}I
Sep 28, 1961	10.6	1.11 ± 0.29	0.84 ± 0.14
Oct 5, 1961	14.8	1.15 ± 0.40	0.78 ± 0.19
Oct 12, 1961	5.6	1.06 ± 0.15	0.92 ± 0.07
Oct 19, 1961	27.9	1.28 ± 0.75	0.58 ± 0.36
Oct 26, 1961	28.7	1.29 ± 0.77	0.57 ± 0.37
Nov 2, 1961	6.4	1.06 ± 0.17	0.90 ± 0.08
Nov 9, 1961	16.4	1.16 ± 0.44	0.75 ± 0.21
Nov 16, 1961	1.0	1.01 ± 0.03	0.98 ± 0.01
Nov 23, 1961	0	1.	1.
Nov 30, 1961	34.8	1.35 ± 0.94	0.48 ± 0.45

accumulation was observed during intervals without rain. The relevant data from this study are given in Table 10. The retention varied markedly as the crop densities increased, and also varied by an order of magnitude between crops.

5. Hull (22). The ^{131}I content of air, rain and settled dust (combined) and grass was determined in the vicinity of the Brookhaven National Laboratory from September to December 1962. The ^{131}I content in grass was reasonably well predicted from the air and rainfall values if a deposition velocity of 0.5 cm/sec and a rainfall-retention value of 0.5 were used.

6. Ward, Johnson, and Stewart (23). Precipitation samples were collected at Colorado State University and the ^{137}Cs deposition/ m^2 was determined. Samples of forage were analyzed at harvest and the ^{137}Cs content in forage was determined from which the overall retention of ^{137}Cs could be determined. These data are given in Table 11. Soil uptake is stated to be on the order of 0.0001; the cumulative soil activity was about 80,000 pCi/ m^2 so this route may be assumed to be negligible. Additional samples were

Table 10. The retention of ^{90}Sr in rain by various crops as determined by Menzel et al. (21) during the spring of 1962. The sweet corn density of April 6 was 45 g dry wt/m². The densities of May 16 were sweet corn, 840; snap beans, 340; cabbage, 230; and potatoes, 150 g dry wt/m². Densities at other periods are not provided.

Date	Rainfall, mm	^{90}Sr Deposition, pCi/m ²	Retention, fraction			
			Corn	Beans	Cabbage	Potatoes
Apr 7-8	61	310	0.09	~0	~0	~0
Apr 26	38	280	0.22	0.10	0.02	0.09
May 4-6	36	410	0.76	0.32	0.06	0.23

Table 11. Retention of ^{137}Cs in rainfall by forage crops grown in Colorado as reported by Ward, Johnson, and Stewart (23). These data give the net retention of the total amount deposited over the entire growing period with no consideration of the field loss factor.

Crop	Cutting	Date	Yield, g dry wt/m ²	Rainfall, mm	^{137}Cs deposition ^a in rainfall, pCi/m ²	Fractional retention
Alfalfa hay	1st	Jun 15, 1963	340	111	3424	0.48
Alfalfa hay	2nd	Jul 20, 1963	225	5.3	754	0.70
Alfalfa hay	3rd	Aug 30, 1963	210	12	1079	0.21
Corn silage	---	Sep 15, 1963	1013	120	5239	0.32
Alfalfa hay	1st	Jun 15, 1964	340	105	4174	0.33
Alfalfa hay	2nd	Jul 20, 1964	225	9.7	1147	0.13
Alfalfa hay	3rd	Sep 10, 1964	210	17	889	0.09

^a Integrated over the relevant growth period.

taken during the growth period. When relatively long intervals occurred between rains, plant growth diluted the activity present, so it may be presumed that dry deposition was not substantial. The average retention over the growth period was 0.32.

An attempt was also made to determine the retention by forage of ^{137}Cs deposited by individual rains. Samples were not always taken immediately before and after each rain storm, but were corrected to the appropriate date using an empirically determined field loss factor. Data from this study are given in Table 12. The values range from 0.26 to 1.79 with a mean of 0.80.

7. Chamberlain and Chadwick (24). Values of the daily levels of ^{131}I in air and rain were deduced from published values of daily or weekly levels of ^{131}I in air, weekly levels of ^{131}I in rain, and daily levels of ^{140}Ba in air and rain for the British Isles (25). The period covered is September to November 1961. These data were used in conjunction with an effective half-time of 5 days for ^{131}I on herbage to derive the most likely values of deposition velocity and rain-retention factor to predict herbage activity (pCi/m^2). The herbage activity values used in the least squares fit were taken from the same report (25). The result of this analysis gave the following values: Deposition velocity, 0.54 ± 0.09 cm/sec and rain-retention factor, 0.51 ± 0.10 .

3. Krieger, Kahn and Cummings (26). Studies were conducted in Ohio during the growing season of 1965. Kentucky bluegrass was grown in a 1500 m^2 pasture, which contained ten uncovered sampling plots of 20 m^2 area and four 5 m^2 sampling plots that were covered with sloping plastic sheets. Between April and September, bluegrass cuttings were collected seven times at intervals ranging from 2 to 7 weeks. Sampling was done when the grass was between 15 and 25 cm high and was collected using a rotary blade mower set at a 5-cm cutting height. Fallout deposition during each sampling period was accumulated in two 0.33 m^2 trays. During the period of greater ^{90}Sr and ^{137}Cs deposition, almost all of the deposition was by precipitation. The contribution of dry fallout during the other periods is not stated unequivocally, but appears not to have been substantial. The data from this study are given in Table 13. The effect of soil uptake has been eliminated by using ratios of stable Sr^{++} and Cs^+ determined in the soil and grass (soil-tracer studies gave nearly identical corrections). The retention values vary widely; for the uncovered plots the ^{90}Sr retention varied from 0.051 to 0.33 with a mean of 0.13 and the ^{137}Cs retention varied from 0.015 to 0.25 with a mean of 0.072. The covered plots showed wide variations in retention and in some cases approached the retention found in the uncovered plots. Several explanations might be offered for this effect; dry

Table 12. Retention by pasture of the ^{137}Cs deposited by individual rains in Colorado. Data are from Ward, Johnson, and Stewart (23). No explanation is offered for the retention value that is more than 1.0.

Date, 1964	Rainfall, mm	^{137}Cs deposition in rainfall, pCi/m^2	Fractional retention
May 7	5.3	640	0.83
May 26	8.4	480	0.69
May 28	48.5	1017	0.56
Jun 8	3.8	280	0.67
Jun 21	2.8	700	0.26
Jun 30	2.3	182	1.79

deposition may have contributed a substantial part of the measured deposition, the covers might have been inadequate to protect against wind-driven rain, or substantial redistribution of rain-deposited debris could have occurred. The latter is an attractive possibility as it has been repeatedly documented that plants lose radioactivity even under dry conditions (27, 28) and a small relatively uncontaminated plot surrounded by a large, more highly contaminated pasture would be expected to accumulate some of this activity.

9. Bergstrom (12). No relevant data are actually provided in this paper, but Bergstrom quotes unpublished data by Svensson indicating that the retention by the edible (by cows) portion of vegetation is 0.17. The data pertain to ^{131}I deposited in precipitation in Sweden during the 1961 fallout period. In the conclusion of this paper, Bergstrom states that "the following preferred data are in agreement with the measurements of fallout iodine in Sweden: Wet deposition: 0.3 of iodine deposited by light rains is retained on vegetation. This factor decreases to 0.1 to 0.2 in heavy rains."

10. Svensson (29). Measurements were made of the ^{137}Cs deposition in Sweden, and samples of moss were collected and analyzed for ^{137}Cs . During the time period from 1962 to 1965 $35 \text{ nCi}/\text{m}^2$ were deposited in rainfall and $32 \text{ nCi}/\text{m}^2$ were retained by the moss. This gives a retention value of 0.91 over a three-year period. The annual rainfall at the sampling plot varied from 500 to 600 mm.

Table 13. Retention by Kentucky bluegrass of rain-deposited fallout during the 1965 growing season. Data are from Krieger, Kahn, and Cummings (26). Retention values given are the net retention over the period with no consideration of field loss. Seeding was in March.

Sampling		Deposition,		Pasture	Plant activity, ^b pCi/m ²				Fractional retention			
date,	Rain, ^a	pCi/m ²		yield,	⁹⁰ Sr		¹³⁷ Cs		⁹⁰ Sr		¹³⁷ Cs	
1965	mm	⁹⁰ Sr	¹³⁷ Cs	g ash/m ²	Open	Covered ^c	Open	Covered ^c	Open	Covered ^c	Open	Covered ^c
Apr 16	73.2	730	1000	2.9	47.3	43.7	32.2	15.1	0.065	0.060	0.032	0.015
Apr 30	59.7	440	850	9.9	146.7	116.0	97.8	50.8	0.33	0.26	0.12	0.060
May 12	2.0	330	150	3.6	53.2	8.7	38.1	7.1	0.16	0.026	0.25	0.047
May 28	12.2	300	480	1.7	25.3	7.7	17.0	2.8	0.084	0.026	0.035	0.0058
Jul 2	63.8	410	300	1.0	25.9	25.5	9.8	7.6	0.063	0.062	0.033	0.025
Aug 8	90.2	490	550	1.1	25.1	5.9	8.3	2.1	0.051	0.012	0.015	0.0038
Sep 17	277.1	230	340	2.0	32.8	3.4	7.2	0.8	0.14	0.015	0.021	0.0024
Average									0.13	0.066	0.072	0.023

^a Over the period.

^b At each cutting, corrected for soil-root uptake, using stable element plant-to-soil ratios.

^c The cover was a sloping plastic sheet, 0.6 m above ground on one side and 0.9 m on the other.

11. Osburn (30). Measurements of fallout activity were made in a Colorado mountain watershed. One experiment was conducted in 1963 to determine what fraction of the total deposition was due to dry fallout. The results over a six-week time period indicated that 20% of the total was due to dry deposition. This period had the most rain of any like period measured over 12 years, so in this environment at least dry deposition is substantial relative to wet deposition. The fallout retention by three species of alpine vegetation was measured during the growing seasons of 1962 and 1963. The dry and wet deposition was not separated and no field loss effect was considered. The data are given in Table 14. Substantial variation was observed between species and was quite different for the two years. Rainfall was also quite different in the two years. In 1962 summer rains were very light, rarely greater than 10 mm/day with a total summer precipitation less than 100 mm. In 1963 nearly 330 mm of rain fell during the summer with half occurring one month prior to the sampling of Kobresia. Net retention values varied from 0.081 to 0.77.

12. Wilson (31). The retention by pasture grass and alfalfa of ^{137}Cs deposited by rain was measured at Colorado State University. During 1962 to 1964, the ^{137}Cs deposition in individual rains was measured and the level of ^{137}Cs in pasture grass was determined before and after the rains. Data from this study are shown in Table 15. A large discrepancy was found for deposition values measured using the HASL collector as opposed to the CSU collector; measurements using the HASL collector were nearly twice as high. Deposition values in Table 15 are based on CSU collector measurements but were corrected to correspond to HASL collector values using an equation given by Wilson. This minimizes the retention values. Wilson analyzed these data according to the equation

$$A_F = A_I - qA_I + pF,$$

where A_F is the final pasture activity (nCi/m^2), A_I is the initial activity, F is the deposition, and q and p are wash-off and retention coefficients. With the CSU collector deposition data, analysis using the above equation gave a significant correlation with $r = 0.59$, $p = 1.24 \pm 0.36$, and $q = 0.28 \pm 0.20$. When the CSU collector deposition data were normalized to correspond to data derived using the HASL collectors, analysis gave $p = 0.70$ and $q = 0.30$, which Wilson interpreted as the most logical choice to express the effects of rain deposition on pasture grass.

Table 14. Retention of fallout by alpine tundra vegetation at the end of the 1962 and 1963 growing seasons. Data are from Osburn (30) and are measurements of gross beta activity. The effect of dry deposition is not separated, nor is a field loss factor considered.

Vegetation	Year	n	Yield, g dry wt/m ²	Vegetation activity, ^a nCi/m ²	Deposition during growing season, nCi/m ²	Fractional retention
<u>Kobresia</u>	1962	20	122 ± 13	60 ± 6	204	0.29
<u>myosuroides</u>	1963	9	64 ± 4	100 ± 11	1220	0.081
<u>Carex</u>	1962	15	234 ± 27	48 ± 6	130	0.37
<u>scopulorum</u>	1963	12	244 ± 40	210 ± 52	1180	0.18
<u>Deschampsia</u>	1962	25	190 ± 17	90 ± 15	115	0.77
<u>caespitosa</u>	1963	12	176 ± 20	220 ± 26	1070	0.21

^a At end of growing season.

Studies were also done using alfalfa in 1965; these data have been published in the open literature (32). These data are not as detailed because rains in 1965 extended for several days at a time, and the time interval between the samples was greater. These data are presented in Table 16; the retention values given do not separate the deposition rate from the effect of wash-off of previously deposited activity. It is not specifically stated whether the deposition values were recorded using CSU or HASL collectors, but they probably represent CSU collector data. The trend is similar to that observed with grass. At the beginning of the growing season when accumulated deposition on plants is low and deposition per rain period is high, high retention values are generally observed. Later, when an equilibrium of sorts has been established between deposition and loss mechanisms and the ¹³⁷Cs activity per volume of rainwater is much lower, the effect of precipitation is more significant in washing off previously deposited activity than in depositing new activity.

Table 15. The retention by pasture grass of ^{137}Cs deposited in individual rains. Data are from Wilson (31) and were recorded at Fort Collins, Colorado. Deposition values are those recorded using CSU collectors normalized to correspond to measurements using HASL collectors.

Date of rain	Rainfall, mm	Deposition, nCi/m ²	Sampling dates	Change in grass activity, nCi/m ²	Fractional retention
<u>1962</u>					
May 4-7	2.	445	May 3, 19	62	0.14
May 23-27	20.	899	May 16, 30	583	0.65
Jun 1	9.1	974	May 30, Jun 2	662	0.68
Jun 6	8.4	273	Jun 5, 8	-365	-1.3
Jun 16	4.8	359	Jun 15, 18	53	0.15
Jun 20	3.8	322	Jun 18, 20	96	0.30
Jun 20	3.8	322	Jun 20, 22	-102	-0.32
Jun 30	13	365	Jun 27, 30	-211	-0.58
Jun 30	13	365	Jun 30, Jul 6	-24	-0.066
Jul 8	9.9	937	Jul 6, 9	80	0.085
Jul 16	6.1	807	Jul 11, 19	-107	-0.13
Jul 23	8.6	381	Jul 19, 24	98	0.26
Jul 26-27	10	398	Jul 24, 26	-220	-0.55
Jul 26-27	10	398	Jul 26, 28	23	0.058
Aug 1	3.0	55	Jul 30, Aug 2	307	5.6
Aug 4	2.5	21	Aug 2, 6	-139	-6.6
Aug 23	2.8	108	Aug 22, 27	-268	-2.5
Sep 9	3.8	77	Sep 5, 12	-196	-2.5
Sep 18	0.3	15	Sep 12, 20	-40	-2.7
<u>1963</u>					
May 16	0.8	597	May 15, 16	-612	-1.0
May 19	2.5	454	May 16, 20	110	0.24
May 23	5.1	959	May 22, 24	301	0.31
Jun 6	4.3	3291	May 30, Jun 10	279	0.085
<u>1964</u>					
Jun 8	7.6	2134	Jun 5, 9	-338	-0.16

Table 16. Retention by alfalfa of rain-deposited ^{137}Cs measured at Fort Collins, Colorado, in 1965. Data are from Wilson, Ward, and Johnson (32).

Time interval	Rainfall, mm	^{137}Cs deposition, pCi/m ²	Change in alfalfa activity, pCi/m ²	Fractional retention
May 21-28	21.4	212	161	0.76
May 31-Jun 19	131.4	880	179	0.20
Jul 1-6	4.8	150	-394	-2.63
Jul 10-14	1.0	23	-123	-5.41
Jul 17-19	3.3	151	52	0.34
Jul 26-31	2.8	40	-31	-0.78
Aug 2-6	0.8	17	18	1.06
			Mean -0.92	

Some alfalfa was also grown in the same field under a polyethylene canopy supported 0.75 m above ground. Activity in the sheltered alfalfa was generally lower, but not substantially so, which Wilson *et al.* interpreted as indicating the importance of dry deposition. Other explanations for this effect may be offered, however, and have been discussed previously.

13. van der Stricht, Gaglione, and de Bortoli (33). A mathematical analysis was performed to derive predictive models for ^{137}Cs and ^{90}Sr activity in herbage and milk. The data used were collected during 1961 to 1968 at Ispra, Italy. The model used was

$$C' = P'_R F_R + P'_D F_D$$

where C' is the mean contamination (pCi/g dry weight) of all herbage samples taken monthly from May to October, F_R is the total fallout deposit of 6 months (mCi/km²) starting from 1 April, F_D is an adequate fraction of the cumulative fallout (mCi/km²) deposited during the years preceding the period considered for F_R , and P'_R and P'_D are proportionality coefficients. This analysis differs from most in that F_D is corrected to take into account the fraction of fallout removed with grass, which thus never reaches the soil, and is also corrected for the annual depletion of soil activity due to transfer to herbage by soil-root uptake. The results of a multiple correlation analysis are the following:

$$^{137}\text{Cs: } C' = 0.0843 F_T + 0.0166 F_d \quad (r = 0.9845)$$

$$^{90}\text{Sr: } C' = 0.130 F_T + 0.0322 F_d \quad (r = 0.9751)$$

The authors use the figure of 1 kg dry weight/m² for the herbage yield during the growing season, which indicates that the net retention values for ⁹⁰Sr and ¹³⁷Cs are 0.130 and 0.0843. As in many studies, the value for F_T is total dry and wet deposition. However, the monthly deposition values are much more strongly correlated with the amount of rainfall than they are with air activity (34). The amount of rainfall for the growing season from April to September varied from 497 to 1446 mm during the years 1961 to 1968 (33).

14. Chamberlain (35). Reference has already been made to the filtration model used by Chadwick and Chamberlain (5) to fit data derived from spraying experiments. The concept is considerably expanded by Chamberlain, who analyzed data from several different sources. Both acute and chronic exposures were considered. The model for chronic exposure conditions included a field loss factor. The results of his analysis were very successful in giving uniform results for many studies on the contamination of herbage under natural conditions, if a value of $\mu = 3 \text{ m}^2/\text{kg dry weight}$ was used. As a function of herbage yield (g dry wt/m²), representative retention values are therefore

<u>Yield</u>	<u>Retention</u>
20	0.06
50	0.14
100	0.26
250	0.53
500	0.78
1000	0.95.

CONCLUSIONS

The primary purpose of this literature summary is to derive a value for the initial retention by forage of rain-deposited radionuclides for use in predictive models. Data for grass and alfalfa are summarized in Table 17. The retention by most types of other vegetation is not greatly different from these values. For use in a predictive model, it seems clear that a value of 0.4 to 0.5 would be appropriate for the average retention. A value of 1.0 would not appear to be unreasonable if one wished to be conservative, particularly where high vegetation densities might be expected.

Table 17. Summary of reported retention values for grass and alfalfa, which may be considered as forage for dairy cows.

Reference	Exposure, days	Herbage yield, g dry wt/m ²	Fractional retention		
			Mean	Range	
9	a	50 - 229	0.23	0.167	0.301
10	a	~120	~0.2		
6	a	~130	0.091	0.033	0.173
6	28	25 - 75	0.06 ^b		
14	a		0.25		
5	a	100 - 228	0.40	0.201	0.816
17	a	~300	~0.7		
19	25	137 - 326	0.23 ^b	0.19 ^b	0.28 ^b
16	90?	20 - 200	>0.2	?	?
20	c		0.96	0.48	1.35
22	c		0.5		
23	30 - 50	210 - 340	0.32 ^b	0.09 ^b	0.70 ^b
23	d		0.80	0.26	1.79
24	c		0.51	0.41	0.61
26	30 - 180		0.10 ^b	0.021 ^b	0.33 ^b
12	?		0.3		
31	c,d		0.70		
32	e		-0.92	-5.41	1.06
33	30 - 180	1000 ^f	0.11 ^b	0.0843 ^b	0.130 ^b
35	g	250 ^g	0.53 ^g		
Average of					
All mean values above			0.31		
All, except -0.92			0.38		
All, except those with superscript b			0.38		
All, except those with superscript b and -0.92			0.47		

^a Acute contaminating spray.

^b Net retention over the indicated period.

^c Acute retention value derived from model of chronic exposure.

^d Single rainstorm.

^e Rainstorms lasting several days.

^f Over entire growing season.

^g $0.53 = 1 - \exp(-0.003 \times 250)$.

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