Transport of iodine from atmosphere to ground

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ABSTRACT

Three methods have been used to measure the velocity of deposition of iodine to grassland. For average conditions the results are: field experiments, $v_g = 1$ cm/sec; analysis of fallout data, $v_g = 0.5$ cm/sec; analysis of stable iodine measurements $v_g = 0.16$ cm/sec. The varying physical-chemical state of the iodine accounts for the variation.

An analysis is made of the contributions of various parts of the boundary layer to the resistance to transport of gaseous elemental iodine to grass.

1. Introduction

Iodine is naturally present in the atmosphere at concentrations of order 0.1 μg/m^3, though not all of this is in elemental form. The mode of release of natural iodine to the air is uncertain, but two mechanisms which certainly play a part are the oxidation of plant residues, particularly of seaweed, and the evaporation of sprayed droplets over the oceans.

The radioactive isotope, iodine-131, has been released to air by the oxidation of irradiated fissile material in chemical processing plants (PARKER, 1956; HEALY *et al.*, 1958; MARTER, 1963) in reactor accidents (CHAMBERLAIN, 1959; ISLITZER, 1962) and in nuclear explosions (KNAPP, 1964; MARTELL, 1964).

In the atmosphere generally iodine diffuses in the same way as any other gas, and the physical or chemical form has no important effect on the rate of diffusion. The only exception to this is if the iodine is contained in particles of several microns size, such that the terminal velocity and momentum of the particles causes deviation from the stream lines.

The deposition at the earth's surface, however, is affected by the physical and chemical nature. Elemental iodine vapour is chemically active and is readily sorbed on most surfaces. Iodine which has reacted with other substances to form compounds is generally less readily sorbed, and the rate of transport to the surface may then be limited by the rate of surface reaction. However, all molecular forms of iodine are subject to the same aerodynamic laws, with perhaps very slight variations caused by dependence of molecular diffusion rates on molecular weight.

When iodine is present in particulate form, the diffusion to surfaces is controlled by the mechanisms which determine the transport of particles across boundary layers. For very small particles, say less than 10^{-2} μ diameter, diffusion or Brownian motion is still the mechanism by which the last part of the boundary layer is traversed. Particles of diameter order 0.1 μ and upwards, have such small rate of Brownian motion that other mechanisms must be invoked to account for transport across sub-laminar layers.

A study of the transport of iodine to grass and other surfaces has been in progress at Harwell for many years (CHAMBERLAIN & CHADWICK, 1953, CHAMBERLAIN, 1960; CHAMBERLAIN, EGGLETON, MEGAW & MORRIS, 1960). In the course of this work, it has been realised that the physical and chemical nature of iodine in the atmosphere is a function of the mode of release, the presence of other contaminants and the time since the iodine was released. The uptake of iodine to plant leaves in laboratory conditions has been found to depend also on the environmental conditions and the physiological state of the plant leaves. Attempts have been made to determine the effect of these variables on the rate of transport to the ground. This is important for the estimation of the hazard of the release of radioactive iodine, since, in general, the contamination of vegetation and thence of milk, is of greater potential hazard than the contamination of air as such.

2. Deposition velocities and resistances

The rate of deposition of a gas or aerosol to ground may be considered in terms of the transport velocity v_g (units cm/sec) or its inverse the resistance r (units sec/cm). If F is the flux to ground (grams or curies per cm² of ground surface per second), χ_1 the volumetric concentration in the free air at a reference height z_1 (grams or curies per cm³), and χ_0 the concentration at the surface:

$$r = \frac{1}{v_g} = \frac{\chi_1 - \chi_0}{F}. \tag{1}$$

The advantage of expressing rate of transport as a resistance rather than as a velocity, is that the resistances of different parts of the boundary layer may be considered to be additive, and analogous to electrical resistance in series. This concept has proved useful in discussing the evaporation of water from vegetation and other surfaces (Penman & Schofield, 1951; Monteith, 1963). Thus the total resistance may be written;

$$r = r_a + r_b + r_s. \tag{2}$$

Where the suffixes a, b and s refer to the outer and inner parts of the boundary layer, and the surface or stomatal resistance.

r_a is the resistance in the region of eddy diffusion between the reference height and the top of the vegetation cover. In this region molecular properties play no part and Reynolds' Analogy applies. If conditions are not too far from adiabatic, and the reference height is not more than say 100 cm, r_a can be equated to u_1/u_*^2, where u_1 is the wind speed at the reference height and u_* is the friction velocity. This is the resistance to momentum transport, since by definition of u_*,

$$\frac{u_1}{u_*^2} = \frac{\varrho u_1}{\tau} = \frac{\text{momentum at reference height}}{\text{flux of momentum.}}$$

r_b is the additional resistance which applies to transport of matter (or heat) as compared with momentum by virtue of the fact that there is no analogy in mass transport to the "bluff body effect" by which momentum is destroyed at the surface (Sheppard, 1958; Owen & Thomson, 1963). Molecular properties enter here, since molecular diffusion determines the rate of transport across the laminar flow very near the surface. Owen & Thomson (1963) define a non-dimensional number B by the equation

$$\frac{1}{K_g} = \frac{u_1}{u_*}\left(\frac{u_1}{u_*} + \frac{1}{B}\right), \tag{3}$$

where $K_g = V_g/u_1 = F/u_1(\chi_1 - \chi_0)$ is the Stanton number of mass transport. If for the moment we take r_s to be zero (i.e. take χ_0 to be the concentration at the outer surface of the roughness elements), it follows from (1), (2) and (3) that

$$B^{-1} = u_* r_b.$$

Thus B^{-1} as defined by Owen & Thomson is a non-dimensional form of the inner boundary layer resistance r_b.

From consideration of a number of experiments in wind tunnels and roughened pipes, Owen & Thomson conclude that B^{-1} has the form

$$B^{-1} = \alpha \left(\frac{u_* h}{\nu}\right)^m \left(\frac{\nu}{D}\right)^n. \tag{4}$$

In (4), α is a constant for a given type of roughness, h is the "equivalent sand roughness" ($h = 30 z_0$, where z_0 is the roughness length). ν and D are the kinematic viscosity and molecular diffusivity, in cm² sec⁻¹ units. m and n are constants to which the values 0.45 and 0.8 respectively are provisionally assigned. Chamberlain (1966) has done wind tunnel experiments similar in principle to those of Owen and Thomson but with two important differences

(*a*) grass and grass-like surfaces were used, giving roughness elements of a more fibrous character than applied to any of the experiments on which (4) is based;

(*b*) a radioactive tracer gas (Pb-212, otherwise known as thorium-B) was used. For this gas $D = 0.054$ cm² sec⁻¹.

From these experiments it appears that both m and n have smaller values when the roughness elements are fibrous rather than bluff. It seems that values $B^{-1} = 8 \pm 2$ for a gas such as ThB or I_2 of low molecular diffusivity, and $B^{-1} = 5 \pm 2$ for water vapour, apply over a considerable range of u_* and z_0.

r_s is the resistance to transport at or in the cuticle of the leaf. For the transport of water outwards from a leaf, r_s can be related to the number and size of the stomatal openings, through which the water passes by molecular diffusion. PENMAN & SCHOFIELD (1951) show that a reasonable value for the resistance length L_s of the open stomata is 0.08 cm. This means that the stomatal resistance is equivalent to the resistance to transport by molecular diffusion across a stagnant film of air 0.08 cm thick. For a resistance like this, $r_s = L_s D^{-1}$, and the stomatal resistance is thus inversely proportional to molecular diffusivity.

There is some evidence, which is not conclusive, that the uptake of iodine vapour by leaves may be controlled by stomatal resistance (BARRY & CHAMBERLAIN, 1963). It is known that waxy substances are poor adsorbers of iodine, and there is evidence that an intact cuticle acts as a barrier to deposition (CHAMBERLAIN, 1960). However, some sorption on the cuticle may occur, and iodine is also known to be adsorbed at points where the cuticle is damaged. The term r_s will be used to refer to a resistance at the surface which may be compounded of several factors of which stomatal resistance is one.

To have a full understanding of the factors governing the transport of a gas such as iodine one would need to know the variations of the resistance with the meteorological parameters, the physical and chemical state of the iodine, and the physiological state of the vegetation. In fact, these factors enter into the data in a partially controlled, or uncontrolled manner.

3. Short range field experiments on uptake of iodine

Over a period of years, a number of field experiments have been done at Harwell to determine the rate of uptake of radio-iodine by grassland. The original purpose of these experiments was to obtain data for assessment of the permissible output of radio-iodine from nuclear installations. Details of the earlier experiments have been given by CHAMBERLAIN & CHADWICK (1953) and CHAMBERLAIN (1960).

The experimental releases were made on the former airfield at Harwell, an area of rather smooth terrain with fetches of at least 200 m over grass. The vegetation consists of a mixture of grasses and broad leaved species such as dandelion, clover and vetch. At the time of the experiments the herbage was about 10 to 15 cm long, except in two cases (experiments 8 and 9) where it was 30 to 40 cm long. The wind velocity was measured during the experiments with Casella cup anemometers at 6 heights above ground. The lapse rate of temperature was measured as the difference in temperature between ½ and 13½ metres above ground. The zero displacement d, and roughness length z_0 were determined by measurements of the velocity profile in adiabatic conditions, at the time of the experiments, or as soon as possible afterwards.

The iodine was prepared in elemental form by oxidation of sodium iodide with potassium dichromate. The iodine vapour released on oxidation was dried by passing over magnesium perchlorate and stored as crystalline elemental iodine in a cold trap. It was released by warming the trap to ambient temperature by passing air through it. 50 mg of stable iodine was used for each source with about 30 mc of iodine-131. From the measured activities at the sampling positions it can be calculated that the added iodine concentration in the air passing over them was not more than about 1 $\mu g/m^3$.

The iodine was released at a height of 50 cm above ground upwind of a number of sampling positions. In experiments 1–3, the sampling positions were arranged in two arcs, one at 50 and the other at 100 metres from the point of release. In experiments 4–8, the sampling positions were placed close together in the centre of the field, and the release was made 50 metres upwind so as to allow the centre of the cloud to pass over the sampling positions.

It was not the purpose of these experiments to study the lateral or vertical diffusion of the plume, but only the relation between the volumetric concentration, or dosage, near the ground and the activity deposited on the ground. Volumetric measurements were made at 4 heights, 100, 75, 50 and 25 cm above ground. The method of measurement was to draw air at known rates through the sampling heads containing a charcoal loaded filter paper backed by a glass fibre filter paper. Tests of the efficiency of this system showed that negligible amounts of iodine penetrated under the conditions of the experiment.

The activity deposited on the herbage was measured by taking the vegetation and underlying matt from a known area immediately in front of the volumetric samplers. The herbage was placed in polythene bags and weighed. It

TABLE 1. *Field experiments on deposition of iodine to grass.*

Run No.	1	2	3	4	5	6	7	8	Mean
Date	12/7 62	18/7 62	12/11 62	21/10 63	4/3 64	22/4 64	25/5 64	15/6 64	
Time (G.M.T.)	1030	1630	1210	1414	1240	1040	1320	1900	
Cloud cover	6/8	8/8	5/8	8/8	8/8	7/8	6/8	4/8	
Temp. °C	17	18	6	17	1	11	20	14	
RH %	64	67	84	70	81	74	70	78	
$\Delta T/\Delta z \times 10^4$ °C per cm	−8.6	−2.6	−11.0	−1.6	−1.2	−4.4	−1.4	+0.5	
u_{200} cm/sec	315	560	300	550	490	609	480	230	
u_{100} cm/sec	270	480	265	470	440	561	380	175	
u_{50} cm/sec	230	395	220	395	375	465	280	120	
u_*	24	44	28	45	36.5	54	57	30	40
d (cm)	5	5	5	5	5	5	20	25	
z_0 (cm)	1.1	1.4	2.5	1.5	0.8	1.5	7.0	9.5	
Herbage mg/cm² (wet wt.)	26	24	30	26	38	30	117	173	
r_a $(=u_{100}/u_*^2)$ (sec/cm)	0.469	0.247	0.344	0.232	0.331	0.192	0.117	0.195	
r $(=1/v_g)$ (sec/cm)									
at $x=50$ m	1.04 ± 0.13	0.82 ± 0.10	0.87 ± 0.02	0.77 ± 0.23	0.62 ± 0.03	0.54 ± 0.08	0.34 ± 0.02	0.34 ± 0.02	
at $x=100$ m	0.88 ± 0.09	0.76 ± 0.08	1.07 ± 0.07						
Mean	0.98 ± 0.09	0.79 ± 0.06	0.95 ± 0.05	0.77 ± 0.23	0.62 ± 0.03	0.54 ± 0.08	0.34 ± 0.02	0.34 ± 0.02	0.66
r_b+r_s $(=r-r_a)$	0.51	0.54	0.61	0.54	0.29	0.35	0.23	0.15	
χ_z/χ_{100} at $z=100$	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
75	0.85	0.96	1.06	1.06	1.07	0.99	1.03	0.96	1.00
50	0.88	1.02	0.96	1.02	1.04	0.93	1.06	0.85	0.97
25	0.79	0.92	0.82	0.93	1.00	0.98	0.91	—	0.91

was then placed, still within the bag, in a container designed to surround a 4″ sodium iodide crystal. The iodine-131 activity was assayed by γ-spectrometry. The counting efficiency of the system was assessed by a series of counts with a source of known activity placed among sheets of strawboard in the same geometry as was used for the grass.

4. Results of field experiments

Results of field experiments on the deposition of iodine done in 1962 and 1963 are summarized in Table 1. Results of earlier trials (in which the procedure differed in some respects from that described above) have been given by CHAMBERLAIN & CHADWICK (1953) and CHAMBERLAIN (1960).

The first point to notice is that the deposition velocities determined at the 50 and 100 m distances are not significantly different in the trials 1 to 3, where this comparison is possible. The same was true in the trials previously reported (CHAMBERLAIN, 1960). This indicated that a source to sample distance of 50 m is sufficient for the development of the boundary layer to the extent of determining the deposition velocity.

At the bottom of Table 1 is shown the reciprocal transport velocity or resistance r. In practice r was determined from $r = C/qA$, where C = activity in volumetric sampling filters, q = flow rate through filters in cm³/sec, A = activity of herbage per cm² of ground. The difference between r and the aerodynamic resistance r_a is equal to $r_b + r_s$, where r_b and r_s are the boundary layer and surface (or stomatal) resistances respectively.

In theory, an independent measure of the transport velocity of iodine can be obtained from the gradient of χ with height. By analogy with the Thornthwaite-Holzmann equation for evaporation, the flux F to the surface in near neutral conditions is given by:

$$F = k^2 \frac{(\chi_1 - \chi_2)(u_1 - u_2)}{(\log_e z_1/z_2)^2}, \tag{5}$$

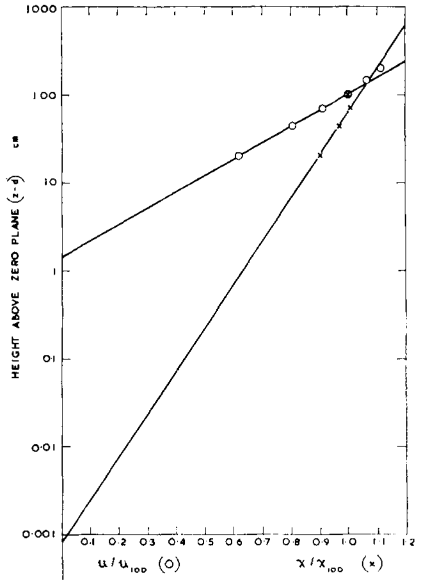

FIG. 1. Logarithmic profiles of wind speed and I^{131} activity.

where the suffixes refer to two heights at which measurements of χ and u are made.

The profiles of χ determined from measurements at heights of 100, 75, 50 and 25 cm are shown in Table 1, in the form of values of χ_z/χ_{100}. The measurements from each individual experiment are insufficiently accurate for determination of F from equation (5). The mean values of the ratios of χ_z/χ_{100} for experiments 1–6 are shown in Fig. 1, plotted against $\log(z-d)$. The ratio χ_z/χ_{100} for $z-d=70$ $(z=75)$ is almost exactly unity, i.e. there is no gradient of χ between $z=75$ and $z=100$. This is a consequence of the limited distance of travel and the fact that the height of release was 50 cm. The line drawn through the points at $z-d=70$, 45 and 20 cm in Fig. 1 has a slope which, when taken in conjunction with mean values of wind speed from the 6 experiments, yields from equation (5) a mean value of $r=\chi_{100}/F=0.77$ sec/cm, compared with a mean value 0.75 sec/cm from Table 1. This shows that the profile of χ in the range $z=25$–50 cm is not inconsistent with equation (5) and the observed values of deposition.

The determination of the mean ratio of volumetric concentration χ_1/χ_2 at two heights, with simultaneous measurements of u_1 and u_2 (or of u_1 and z_0) is a possible method of determining the rate of dry deposition of a naturally occurring aerosol. To give reasonably accurate results the lower height z_2 of measurement of χ must be as near the surface as possible and the surface must be sufficiently uniform over a sufficiently long fetch for the profile to be developed to the upper height z_1.

The mean value of r from all the experiments of Table 1 is 0.66 sec/cm ($v_g=1.5$ cm/sec). The mean of an earlier series of 7 experiments was $r=0.54$ sec/cm (CHAMBERLAIN, 1960).

A series of experiments in the U.S.A., in which iodine was allowed to diffuse over a series of grass grown in trays and exposed on the ground at distances from 1000 to 32,000 m, gave a mean resistance 0.65 sec/cm ($v_g=1.53\pm0.59$ cm/sec) (GIFFORD & PACK, 1962). Earlier, an analysis of deposition of iodine released from a chemical plant at Hanford, U.S.A., gave $r=0.36$ sec/cm ($v_g=2.8$ cm/sec) (PARKER, 1956).

5. Surface resistance to uptake of iodine

The difference between the observed values of r in Table 1 and the aerodynamical resistance r_a (taken equal to u_1/u_*^2) is equated to the sum of the terms r_b and r_s. In an attempt to obtain an independent estimate of r_s, a comparison was made in experiments 5 to 8 of the uptake of iodine to grass, and in some cases clover leaves in the field, with the uptake to artificial leaves of approximately the same size and shape made of silver foil (Fig. 2).

The theory of this method of determining surface resistance has been given by BARRY & CHAMBERLAIN (1963). It is similar in principle to the method of determining the stomatal resistance to the evaporation of water by comparing the transpiration from a leaf with the evaporation from an artificial leaf of wet blotting paper (see for example Raschke, 1956).

The resistance r_L to a leaf, or r_{AL} to an artificial leaf is defined by,

$$r_L=\frac{\chi_1-\chi_0}{F}, \tag{6}$$

where χ_1 is the volumetric concentration in the free air, χ_0 at the surface of the cells of the mesophyll, and F the flux to the leaf per unit area counting both sides of the leaf. For the transport of iodine it is assumed that the concentration χ_0 at the mesophyll surface is zero, the overall resistance being the sum of an aerodynamic and surface (= stomatal?) resistance.

For the artificial silver leaf the surface resistance is zero, silver acting as a perfect sink for elemental iodine vapour at low concentrations (CHAMBERLAIN, 1953; CHAMBERLAIN, EGGLETON, MEGAW & MORRIS, 1963). Hence:

$$r_L = r_{AL} + r_S. \qquad (7)$$

Observed values of r_L and r_{AL}, and deduced values of r_S, from the experiments in which artificial leaves were attached to grass and clover in the field, are given in Table 2. The results vary from $r_S = 2.2$ sec/cm for experiment 5 (conducted when the temperature was 1°C) to 0.5 sec/cm for experiments 7 and 8. The standard errors on the mean resistances are rather large in comparison with the differences used to deduce r_S.

Whilst its not certain whether the surface resistance for uptake of iodine does in fact represent the resistance to diffusion through the stomata, the values found for r_S in Table 2 are reasonable in relation to the stomatal resistance found for transpiration.

The average values of r_S from the last column of Table 2 is 1.0 sec/cm^{-1}, and since D for iodine is 0.08 cm^2 sec^{-1}, the corresponding resistance length L_S is 0.08 cm, in agreement with PENMAN & SCHOFIELD's (1951) estimate.

The surface or stomatal resistance r_S of an individual leaf is not the same as the contribution of surface or stomatal resistance to the resistance of the herbage as a whole, which is denoted by r_s. There is much more than one cm^2 of leaf surface (counting both sides of the leaf) on each cm^2 of ground carrying a reasonable weight of herbage. If the effective leaf surface per unit area of ground is S and the resistances to various leaf surfaces are in parallel, the contribution to the overall resistance can perhaps be written r_S/S. S can be estimated by measuring the mean weight per unit surface of leaf, and the mean weight of herbage per unit area of ground. The difficulty is to select repre-

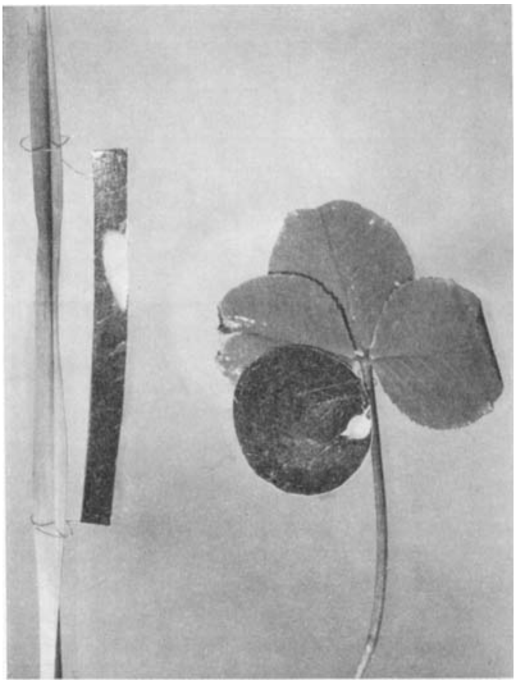

FIG. 2. Leaves and silver replicas.

TABLE 2. *Comparison of resistances to leaves and artificial leaves.*

Experiment	No. of leaves and replicas	Resistance (sec/cm) Leaves (r_L) Mean	S.E.	Replicas (r_{AL}) Mean	S.E.	$r_S = r_L - r_{AL}$
			Grass leaves and silver replicas			
5	8	4.63	1.02	2.46	0.47	2.2
6	14	2.71	0.40	1.34	0.16	1.4
7	18	2.89	0.27	2.38	0.35	0.5
8	11	2.52	0.50	2.04	0.32	0.5
			Clover leaves and silver replicas			
7	18	3.09	0.25	2.09	0.18	1.0
8	12	4.2	0.8	3.7	0.7	0.5

TABLE 3. *Analysis of resistances.*

Expt. No.	u_* cm/sec	S cm²/cm²	Resistances (sec/cm) r	r_a	r_b	r_s
5	36	7.5	0.62	0.33	0.00	0.29
6	54	5.9	0.54	0.19	0.08	0.23
7	57	8.5	0.34	0.12	0.16	0.06
8	30	7.4	0.34	0.19	0.09	0.06
Mean	44	7.3	0.44	0.21	0.08	0.16

In the above table, the last four columns are derived as follows:

r: measurements of F and χ (Table 1).
r_a: measurements of u and u_* (Table 1).
r_s: Table 2 and equation (8).
r_b: from $r_b = r - r_a - r_s$.

sentative leaves to weigh. Also the lower part of the herbage may be non effective in absorbing iodine.

Another method of estimating S is from the ratio r_L/r, which is the resistance to individual leaves divided by the resistance of the herbage as a whole. The values of S so deduced shown in Table 3, vary from 7.5 to 5.9 cm² of leaf (both sides) per cm² of ground. The effective specific surface does not appear to increase very much as the herbage grows, presumably because the lower part is shielded by the upper.

If now r_s is calculated from

$$r_s = \frac{r_S}{S} = \frac{(r_L - r_{AL})r}{r_L} \qquad (8)$$

and the result subtracted from $r_b + r_s$ which is equal to $r - r_a$ and is given in Table 1, values of r_b can be deduced, as set out in Table 3. There is however a possible fallacy in the method, which would lead to the values of r_b derived in Table 3 being too low. Equations (2), (6) and (7) can be used to derive,

$$r_a + r_b = r - r_s = r - (r_L - r_{AL})\frac{r}{r_L} = \frac{r_{AL}}{S}. \qquad (9)$$

Now $r_a + r_b$ is the resistance which would apply if the herbage as a whole was a perfect sink for iodine. This would be the case if all the leaves were replaced by silver replicas. r_{AL}/S is the experimental resistance of isolated silver leaves, divided by the specific surface. However, the deposition to an isolated silver leaf, surrounded by somewhat less iodine-attractive real leaves, is likely to be greater than would be if all the leaves were silver. This source of error is probably small when the ratio r_L/r_{AL} is near unity, as in experiments 7 and 8, but not otherwise.

By multiplying the values of $r_b + r_s$ in Table 1 by the corresponding values of u_*, estimates of B^{-1} are obtained. The results vary from B^{-1} equals 4.5 to B^{-1} equals 24, with mean 15. This estimate of B^{-1} includes in it the surface, or stomatal resistance as well as the boundary layer resistance. The answer is about twice the value derived from Chamberlain's (1966) experiments with a vapour of Pb-212, for which the surface resistance is zero. Hence this analysis independently of Table 3 suggests that the orders of magnitude of r_b and r_s are the same.

6. Removal of iodine from herbage

Several determinations have been made of the rate of removal of radioiodine from vegetation, and these are summarised in Table 4 in terms of the apparent half-life of the iodine. In some instances the apparent half-life was determined by taking serial samples of grass or other vegetation from adjoining areas of ground. In some cases all the herbage was taken from a known area, and the results expressed in activity per unit area, but in others only the activity per kg weight was recorded. In some instances the apparent half-life was determined indirectly by observing the rate of diminution in the I-131 contamination of milk. The mean of all determinations is 4.9 days and using the expression $1/T_e = 1/T_r + 1/T_b$, where T_e, T_r and T_b are the effective, radioactive and biological half-life respectively, a biological half-life of about 13 days is deduced, corresponding to a mean residence time of 20 days.

Several mechanisms may contribute to the difference between the effective and radioactive half-lives:

(*a*) Activity may be washed off herbage by rain.

(*b*) Iodine may be re-volatilised from the herbage.

(*c*) It may be translocated downwards in the plant.

(*d*) The herbage may die back, become prostrate on the ground, and escape sampling.

(*e*) Plants may shed and re-generate parts of their cuticle.

TABLE 4. *Removal of radio-iodine from vegetation.*

Date	Location	Source of iodine	Material contaminated	Effective half-life (days)	Remarks	References
1950–51	Harwell	Experimental release	Grass	5–6		CHAMBERLAIN & CHADWICK (1953)
May/June 1961	Savannah River	Fuel processing	Grass	5^a	No effect of washing	MARTER (1963)
May/June 1961	Savannah River	Fuel processing	Milk	4–5	—	MARTER (1963)
July 1962	Yucca Flats, Nevada	Ground burst Nuclear explosion	Sagebrush and shadescale	5.5^a	No rain	MARTIN (1963)
Sept./Oct. 1962	Hanford	Distant nuclear explosion	Grass	6^a	—	SOLDAT (1963)
July/Aug. 1962	Utah	Nuclear explosions	Milk	5.8	—	PENDLETON *et al.* (1963)
July/Aug. 1962	Nevada	Nuclear explosions	Milk	5.0		KNAPP (1964)
Oct./Nov. 1957	N.W. England	Windscale accident	Milk Milk	3 5.3	4–13 days after release 13–32 days after release	BURCH (1959)
Oct./Nov. 1957	Seascale, Cumberland	Windscale accident	Milk Grass	4.7 4.9		BOOKER (1958)
May 1963	Idaho Falls	Experimental release (CERT Expt.)	Grass	3.5^a	Probably affected by new growth	HAWLEY *et al.* (1964)

[a] These values were determined from the activity/g of herbage, the remaining grass values from activity/m^2.

There is evidence that wash off by rain is not the main mechanism. MARTER (1963) reported no effect of rain on the effective half-life, and MARTIN (1963) found a 5.5 day half-life in a region where rain hardly ever occurs.

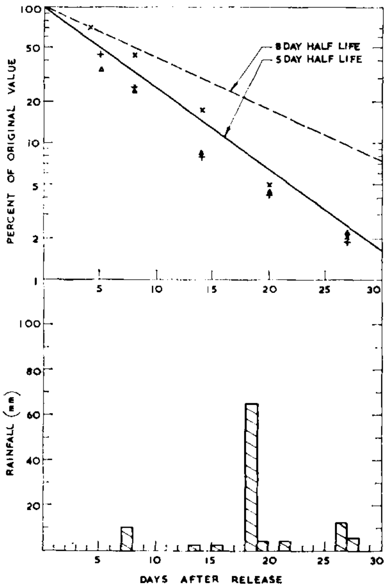

FIG. 3. Activity of I^{131} per m^2 grass as percentage of initial activity following release of 18.7.62.

Measurements of effective half-lives in herbage were made after several of the field experiments already described. Fig. 3 shows a fall off of activity after experiment No. 2. At the bottom of Fig. 3 the daily rainfall is shown. The effective half-life was 5 days, and the rate of decay does not appear to be affected by the heavy rainfall on the 19th day. The same value was found in the earlier series of experiments of CHAMBERLAIN & CHADWICK (1953).

Measurements by BOOKER (1958) of the iodine content of herbage following the Windscale accident of October, 1957, show a 4.8 day half-life of the activity per unit area of ground, but when the ratio of I-131 to Cs-137 activity of the grass was plotted, an apparent half-life of 9.5 days was found. This suggests that the process by which iodine is lost does not depend on the volatility of iodine.[1]

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[1] A contrary conclusion has been recorded by CLINE, WILSON & HUNGATE (1964) following an experiment in which the volatilisation of iodine was measured from a geranium plant placed under a bell-jar fitted with a filter trap. About 50 % of the radioiodine originally deposited on the plant was found in the trap after eight days.

Following this report, a similar experiment was performed at Harwell in December 1964. A source of radioiodine was released over the grassland used for the deposition experiments. After the release,

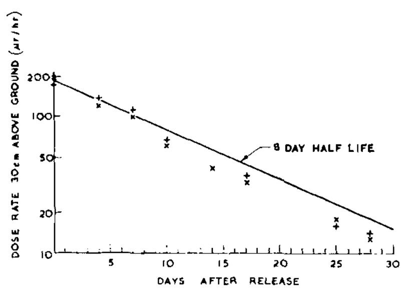

FIG. 4. Gamma dose rate after release of 4.3.64 measured at position 8 m down wind of source.

Following experiment 5 of the present series, measurements were made of the γ dose rate above the experimental area due to the deposited iodine. Two instruments were used, one of the Geiger and the other of the γ scintillation type. The results are plotted in Fig. 4, and the apparent half-life is between 7 and 8 days. This again suggests that re-volatilisation is not rapid. Although the reason for the effective half-life of 5 days on herbage is not known, it appears to be sufficiently reproduceable to be used in calculation of iodine deposition rates from measurements of cumulative activity as in the next paragraph.

7. Calculation of rate of deposition of iodine from distant nuclear explosions

During the autumn of 1961 a number of nuclear tests were performed, many of them in the troposphere. During this period there was a considerable fallout of iodine in the northern hemisphere. FISHER, KEANE, TOUREAU & CAMBRAY (1962) have given details of the activity of I-131 in air, rain and herbage in the British Isles. The figures for air are given on a daily basis for September and October 1961 and on a weekly basis for November. The concentration in rain is also given on a weekly basis. It is, therefore, possible to deduce daily values by making use of the daily concentration of Ba-140 in air and rain, which are given on a daily basis for the whole period.

In order to calculate the daily increment to the iodine activity of herbage values must be assumed for the following two constants:

v_g = velocity of dry deposition of I-131 from air to grass, and
p = proportion of iodine falling in rain which is retained on herbage.

The procedure adopted was to assign arbitrary, but constant, values to v_g and p for the period 10th September to 2nd December 1961. The daily increment of iodine in herbage was calculated by multiplying the air concentration by v_g and adding the amount falling in rain multiplied by p. Making use of an effective half-life of 5 days, the cumulative totals were calculated day by day. A least squares fit was made to the observed activities in herbage, to determine the most likely values of the parameters v_g and p. This procedure led to the following means and standard errors:

$v_g = 0.54 \pm 0.09$ cm/sec ($r = 1.85$ cm^{-1} sec).
$p = 0.51 \pm 0.10$.

In Fig. 5 the contributions of dry and wet deposition, and the sum of the two, in the autumn of 1961, are shown. The circles represent the measured activity of iodine on herbage. The first part of the period was comparatively rainless, whereas from 20th October to 12th November there was heavy rain, thus alternately dry and wet deposition predominated. For this reason fairly good accuracy is obtainable on the values of v_g and p, even though there were two or three observed activities in herbage which are difficult to explain. The results of the Harwell analysis agreed very well with those obtained by HULL (1963) who made a similar analysis of the fallout of iodine at Brookhaven, Long Island. HULL's results are:

$v_g = 0.5$ cm/sec.
$p = 0.5$.

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a transparent plastic cover, maintained at negative pressure by the extraction of 20 litres/min of ventilation air through iodine traps, was placed over an area of one square yard of grass 5 yards downwind from the point of release.

After 14 days the grass within the cover was harvested and its radioiodine content measured. Correcting for radioactive decay, the total activity found in the iodine traps was 0.16 % of that remaining on the grass and top soil after 14 days. From the distribution of the activity within the traps, following the method of Chamberlain *et al.* (1963), it was concluded that most of the desorbed iodine was in non-elemental form. The discordance of this result with that of CLINE *et al.* must presumably be attributed either to differences in the mode of fumigation (e.g. stable iodine carrier, which in the Harwell experiment amounted to about 20 μg on the grass sampled) or to differences between plants grown indoors and out of doors.

When the dry deposition velocity of 0.5 cm/sec obtained by this method is compared with the mean deposition velocity of 1.5 cm/sec obtained in the field trials, allowance must be made for two factors, (*a*) an estimate of the mean wind speed over the period Sep.–Nov. 1961 at Harwell is 4.4 m/sec, the anemometer being at height 13.5 m. Assuming a mean roughness length of 2 cms, this would give a mean friction velocity of about 27 cm/sec, compared with a mean of 40 cm/sec in Table 1. Since, other things being equal, v_g is approximately proportional to friction velocity, the deposition velocity for elemental iodine at the mean wind speed would be about 1 cm/sec, although this makes no allowance for possibly greater stomatal resistance at night. (*b*) It is almost certain that a considerable proportion of the iodine from nuclear tests is in particulate form. From an analysis of the distribution of activity between different parts of a sampling train, for the same period EGGLETON, ATKINS & COUSINS (1963) deduced that 75% was particulate, and further that the v_g of the I-131 to grass in a wind-tunnel was similar to that of total fission products. An effective diameter for fall-out particles in September 1961 of 0.7 μ has been deduced by PEIRSON & KEANE (1962) from consideration of filter efficiencies.

It is remarkable that the physical nature of the iodine makes so little apparent difference to the deposition velocity, since from (4) B and hence r_b depends on D. Presumably other modes of transport (impaction, and possibly terminal velocity) compensate for the greatly reduced molecular diffusion. This is convenient from the point of view of estimating transfer rates of substances which may be in either gaseous or sub-micron particulate form.

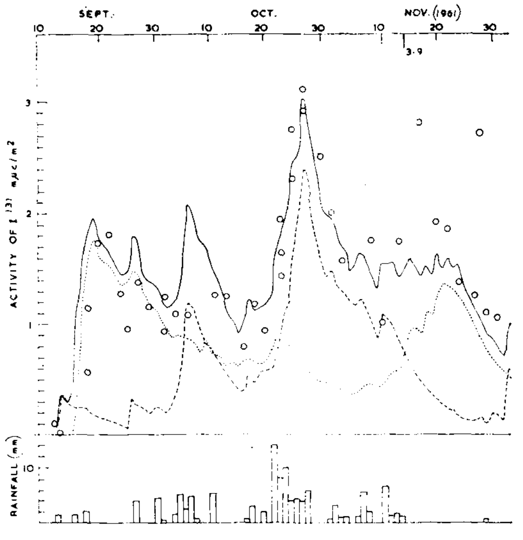

FIG. 5. Deposition of iodine 131 in autumn 1961. ------, dry deposition; ----, deposition in rain; ——, total deposition; ○, activity on grass.

8. Exchange of stable iodine between the atmosphere, grass and soil

Bolin (1959) has made use of some of the earlier estimates of deposition velocity of radio-iodine to deduce the mean residence time of stable iodine in vegetation, and concludes that this must be very short. This conclusion is at variance with the observed fairly long biological half-life of radio-iodine in herbage.

EGGLETON & COUSINS (1966) made measurements of the stable iodine content of air, rain and grass at Harwell. Although the number of grass determinations was small they were made with care and they have the advantage that they refer to the same place as the more extensive air and rain measurements. In Table 5 EGGLETON and COUSINS' measurements are shown, and compared with the values used by BOLIN.

EGGLETON and COUSINS' value of the concentration of iodine in air, 0.08 μ gram/kilogram, which is the mean of measurements showing a wide scatter is considerably lower than the values reported in the older literature. However, there is evidence, summarised by JUNGE (1963) that the concentration of iodine in European air has decreased substantially since the 1930's

TABLE 5. *Measurements of stable iodine at Harwell 1960–63.*

Sub-stance	No. of samples	Iodine content (μg/kg) Range	Mean	Values used by BOLIN (1959)
Air	97	Nil–0.25	0.08	0.4
Rain	23	Nil–6.9	2.0	0.4–4
Grass	3	330–660	380[a]	50

[a] Per kg wet weight. On average 850 g wet wt. per m² ground.

owing to the use of imported iodine instead of iodine made by burning seaweed.

If the mean biological residence time of 20 days in grass found for radioiodine is assumed to apply to stable iodine, the rate at which iodine must be deposited to maintain an equilibrium concentration of 320 $\mu g/m^2$ is 16 $\mu g/m^2$ day. If it is further assumed that 50% of what falls in rain is retained on the herbage, a daily rainfall of 0.2 cm with iodine concentration 2.0 $\mu g/kg$ will contribute 2.0 $\mu g/m^2$ day. If the remaining 14 $\mu g/m^2$ day is attributed to dry deposition, and the average air concentration is 0.08 $\mu g/kg$ or 0.10 $\mu g/m^3$, the dry deposition velocity required is 0.16 cm/sec. Although this is lower than would be expected from the data on deposition of radio-iodine, it is not an unreasonable figure, having regard to the uncertain form of stable iodine in air, and the limited number of measurements.

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ПЕРЕХОД ИОДА ИЗ АТМОСФЕРЫ В ПОЧВУ

Для измерения скорости отложения иода в луговую почву использовались 3 метода. Для средних условий получены следующие результаты:

1) из полевых экспериментов — $V_g = 1$ см/сек.

2) из анализа данных о радиоактивных осадках — $V_g = 0{,}5$ см/сек.

3) из анализа измерений устойчивой формы иода — $V_g = 0{,}23$ см/сек.

Это различие объясняется изменением физико-химического состояния иода. Сделан анализ вклада различных частей пограничного слоя в сопротивление перехода газообразного иода в траву.