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**CONTROLLED ENVIRONMENTAL RADIOIODINE TESTS
PROGRESS REPORT NUMBER TWO**

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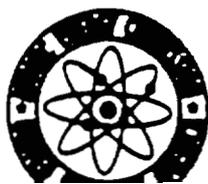
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SUMMARY

The Controlled Environmental Radioiodine Test (CERT) program consists of a series of field releases of various types of radioiodine over different vegetation and under various meteorological conditions with two major objectives: (a) to obtain data on the hazard of iodine and (b) to obtain data on the quantitative movement of iodine through the air-vegetation-cow-milk-human chain. The series was begun in the spring of 1963. This report contains the results of the sixth and seventh tests in the series. It contains the results of recently initiated laboratory experiments and also the status of some of our analytical efforts. Laboratory experiments to support the field studies were begun in 1964. These experiments consist of measuring the deposition and retention of the various forms of radioiodine on vegetation in an environmental chamber. They are also used to develop sampling techniques.

The sixth test was originally designed to be a full-scale study of the behavior of CH_3I (methyl iodide) in the milk-food chain. The vials containing the CH_3I were broken during shipment, and the program had to be reduced drastically. However, the results indicate that the deposition of CH_3I is at least a factor of 100 less than I_2 (iodine vapor) under similar conditions and that the dose potential (defined as the ratio of standard-man ingestion dose to inhalation dose) is at least a factor of 400 less. The metabolic behavior of CH_3I in humans appears to be very similar to other chemical forms of iodine.

The seventh test was designed to provide information on the movement of radioiodine under typical late-fall or early-winter conditions. The dose potential was a factor of 2.7 greater than the 150 observed in CERT-2. The increase was largely a result of two parameter changes:

- (1) Grass half-life -- 6.5 days (CERT-2 -- 5.5 days)
- (2) Fraction of iodine secreted in milk -- 0.20 (CERT-2 -- 0.09).

Other significant results for CERT-7 were:

- (1) Deposition velocity -- 0.8 cm/sec
- (2) Peak milk activity to initial grass activity ratio -- 230 ($\text{Ci}/1:\text{Ci}/\text{g}$)
- (3) Adult human thyroid uptake -- 0.30 of that inhaled.

The major accomplishments of the laboratory experiments have been the development of high-volume air samplers to characterize the forms of iodine and the finding of a relationship between deposition velocity and areal vegetation density.

An analytical model is derived which makes possible rapid and reasonably accurate assessment of levels of radioiodine activity in milk as well as the expected thyroid dose. The dose potential has been evaluated as a function of season and age groups, and it is estimated that the probable ingestion dose to children under typical circumstances is

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**CONTROLLED ENVIRONMENTAL RADIOIODINE TESTS
PROGRESS REPORT NUMBER 2**

I. INTRODUCTION

The Controlled Environmental Radioiodine Tests (CERT) were originally proposed as a project "to determine the fate of radioiodine in the environment from the point of release to its ultimate deposition in humans." As in other areas of interest in the fields of nuclear safety and radiation safety, little information has been available to allow reliable extrapolation to all conditions. The CERT program consists of field releases of various types of radioiodine over different vegetation and various meteorological conditions. We are attempting to gather data, over a spectrum of conditions, to derive functional models that describe quantitatively the movement of iodine through the air-vegetation-cow-milk-human chain. With these models, the potential hazard of radioiodine could be meaningfully assessed.

II. CERT-6

1. INTRODUCTION

1.1 General Information

In recent years, much attention has been given to the characteristics of CH_3I (methyl iodide). At a recent symposium [1], a number of papers and discussions indicated that different chemical forms of radiiodine may exhibit markedly different behavior. Since a significant fraction of the iodine released to the atmosphere from a reactor accident might be in the form of CH_3I , it is of considerable interest to examine its movement through the food chain.

To this end, a field test was scheduled for the late summer of 1965, and the methyl iodide was ordered. Unfortunately, several of the vials, each containing 2 curies of methyl iodide-131, were accidentally broken in transit or were leaking when received. Those that were not broken were subsequently opened in the hot cell of the Idaho Chemical Processing Plant (ICPP), and the methyl iodide (2 to 6 curies) escaped to the atmosphere from a 75-meter stack. The stack was located approximately 4 kilometers upwind of the test grid at the Experimental Dairy Farm (EDF).

1.2 Test Sampling

It had been planned that the test sampling at the EDF would be similar to that used in the CERT-2 test [2], as shown in Figure 1. However, because of the leaking sources, it was not possible to conduct the test as originally conceived. Prior to the release at the ICPP, the high-volume air samplers in the C arc were turned on so that some field measurements could be taken. Each air sampler was equipped with a BM-2133 prefilter and a BM-2306[a] carbon cartridge, the same system used in previous tests [3].

Grass samples were taken prior to and after the release. These were collected near the base of each high-volume air sampler and at selected locations on the A and D arcs. Sagebrush was sampled at 2 and 5 kilometers downwind from the release point. Grass and alfalfa were sampled about 40 kilometers downwind (the distance to the nearest farm areas downwind).

Six cows were allowed to graze over the entire 27-acre pasture. The cows were grazing during the time of release and remained on pasture for two days. They were milked twice daily. The iodine concentration in the milk was determined by counting 900-ml samples in conventional well counters. Activity in grass samples was determined in the same counters. Several individuals were inadvertently exposed to airborne radiiodine from the leaking and broken containers, and efforts were made to obtain data on the retention of this form of iodine in humans.

1.3 Meteorological Conditions

A summary of the meteorological conditions during CERT-6 is shown in Table I. The atmospheric stability at the EDF was estimated to be slightly unstable to near neutral (Pasquill's stability class C to D).

[a] Bureau of Mines Approval Number 2306.

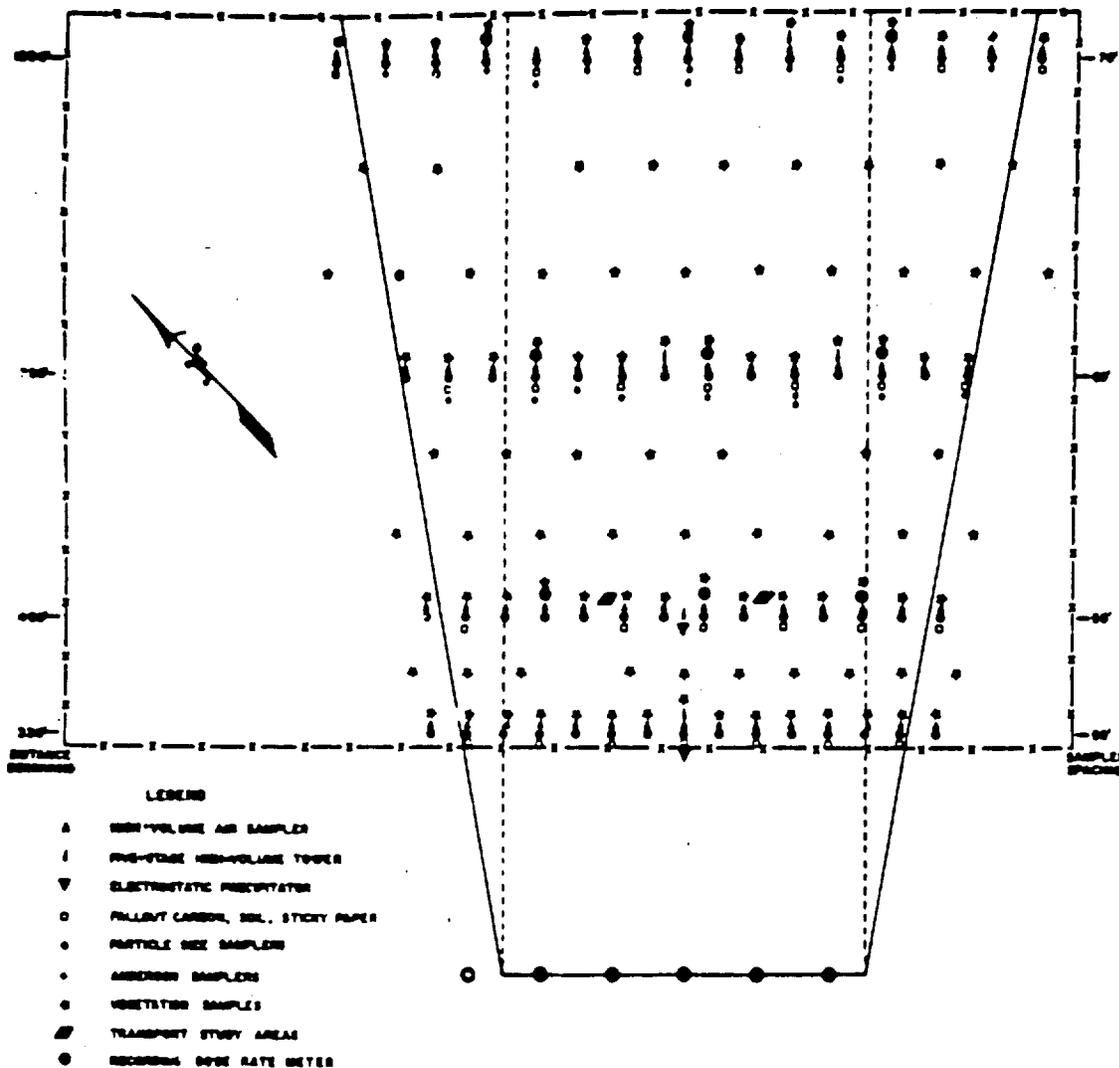


FIG. 1 SAMPLING SYSTEM AT THE EXPERIMENTAL DAIRY FARM.

2. DEPOSITION AND MILK STUDIES

TABLE I

METEROLOGICAL CONDITIONS
DURING CERT-6

Temperature	17.8°C
Relative humidity	33 per cent
Wind speed at 4m	15 m/sec
Stability	Slightly unstable

2.1 Deposition Analyses

Iodine-131 was not detected on the grass samples collected from the pasture, on the sagebrush samples, or on the grass and alfalfa samples collected 40 kilometers downwind. Based on background activity of 10 d/m/g (wet weight) and a grass density, at the time, of 300 g/m² (wet weight), it is estimated that iodine activity was no greater than

$$1 \times 10^{-4} \mu\text{Ci}/\text{m}^2 \text{ or } 4 \times 10^{-7} \mu\text{Ci}/\text{g.} \quad (1)$$

The average time-integrated air concentration was about $1 \mu\text{Ci-sec}/\text{m}^3$. Based on laboratory tests (described in Section IV of this report), the actual concentrations may have been 2 to 4 times greater because the filter system was not 100 percent efficient. At any rate, it is estimated from this data that the deposition velocity is no greater than

$$\frac{1 \times 10^{-4} \mu\text{Ci}/\text{m}^2}{1 \mu\text{Ci-sec}/\text{m}^3} = 10^{-4} \text{ m/sec} = 0.01 \text{ cm/sec} \quad (2)$$

This value is about a factor of 100 less than that observed under similar conditions in CERT-2. Although there is no conclusive proof that the material passing over the EDF was in fact CH_3I , the ratio of carbon to prefilter activity was 10 as contrasted to a ratio of 1 or 2 for elemental iodine releases. This difference in filter ratios follows the same trends found in CH_3I and I_2 studies in our laboratories. These results suggest that methyl iodide does not readily deposit on vegetation. They further suggest the methyl iodide does not readily dissociate in the atmosphere, at least not over the time it took to reach the sampling arcs. At a wind speed of approximately 15 m/sec, the material reached the grid in about five minutes and the off-site farmland in about one hour. The dissociation of CH_3I in the atmosphere was of interest because even if methyl iodide did not deposit at short distances, at some distance downwind, the dissociated iodine component might create an ingestion hazard.

2.2 Milk Analyses

Milk samples collected for three days after the release did not show any iodine-131 above background levels (20 pCi/l). On this basis, the net transport of CH_3I from air to milk was at least a factor of 400 less than I_2 in the CERT-2 test. The results of the milk sampling confirm the fact that there was no significant deposition of CH_3I on pasture, since milk is a more sensitive indicator of iodine-131 than vegetation. If the activity associated with the air samplers had been elemental iodine rather than methyl iodide, a peak activity of approximately 8×10^3 pCi/l should have been detected in the milk, based on the 130 pCi/l peak milk activity to 1 pCi/g initial grass activity found in CERT-1 and -2 [2].

3. HUMAN UPTAKE STUDIES

While preparations were being made for the planned CH_3I test, an estimated 2 curies of iodine-131 escaped from a ruptured glass ampoule which led to significant uptake of iodine by several individuals. The exposures occurred over a four-day period with a few people receiving multiple exposures. Because of the interest in the retention characteristics of this form of iodine, a number of "in vivo" counts were taken over a period of a month.

It was found that the behavior of this material in humans was virtually identical to that observed in studies using other forms of iodine [4,5], which conforms with results of a recent study by Morgan [6]. The curve for the individual receiving the greatest exposure is shown in Figure 2. The solid lines represent the equations drawn to fit the data listed in this figure.

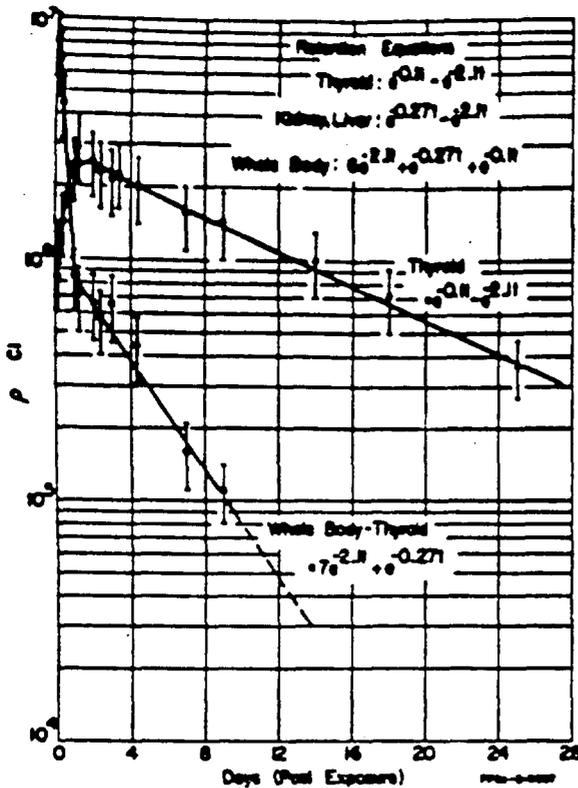


FIG. 2 RETENTION CURVES FOR CH₃I INHALATION, CASE SJ.

While the actual amount inhaled is not known, the iodine remaining in the body reasonably conforms to the equation derived by Lushbaugh et al [4];

$$R(t) = 0.8 \exp - 2.1t + 0.2 \exp - 0.094t$$

4. CONCLUSIONS

On the basis of the above information, we have drawn the following conclusions:

- (1) The dose potential of CH₃I-131 (as the ratio of ICRP standard-man [7] ingestion to inhalation dose) is less than 0.3 -- a factor of 400 less than observed for elemental iodine, as determined by comparison with CERT-2 data
- (2) The deposition velocity of CH₃I is less than 0.01 cm/sec in a slightly unstable atmosphere
- (3) Inhaled CH₃I, once it is in the bloodstream, behaves the same as other forms of iodine.

III. CERT-7

1. GENERAL INFORMATION

1.1 Test Objectives

Previous CERT experiments have been conducted in spring and summer months, and the results are not necessarily applicable to other seasons of the year. More data are needed before models can be derived to confidently estimate such factors as deposition, residence time, and transfer from grass to milk over a wide range of conditions. Therefore, a full-scale test was carried out over dry pasture containing a mixture of grasses typical of late fall pastures in this area.

1.2 Test Grid and Sampling

On November 22, 1965, at 1410 MST, 1.1 curies of iodine-131, as I_2 , were released over the pasture at the EDF. The CERT-7 test grid, with instrumentation, is illustrated in Figure 3. The general design of the grid was similar to those of past tests. The major change was that the BM-2133 paper of previous

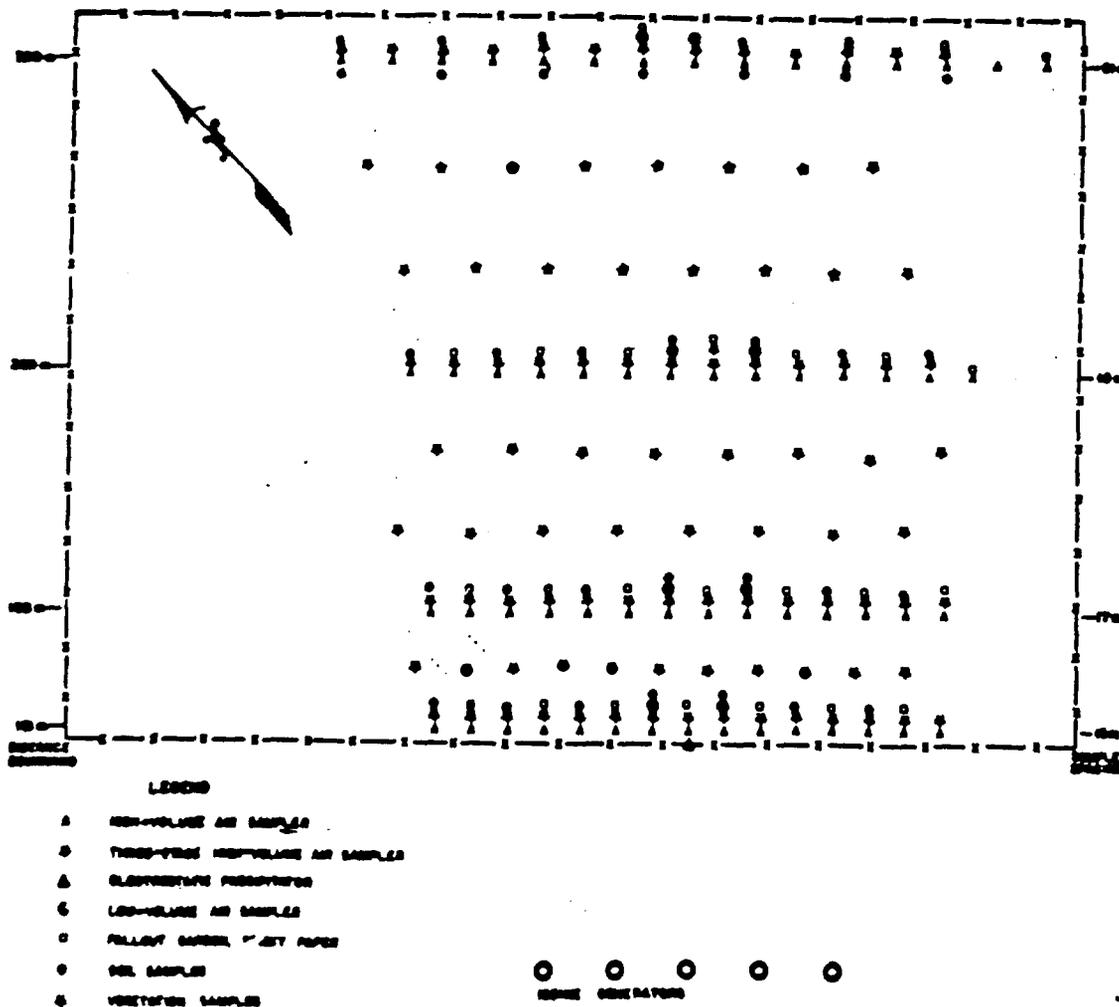


FIG. 3 SAMPLING GRID FOR CERT-7.

tests was replaced by Gelman AC-1, carbon-impregnated filter papers. Charcoal-covered sticky paper (fallout carbon) and plain sticky paper, each having an area 645 cm², were located at every other high-volume air sampling station. Low-volume air samplers were equipped with two-inch, GM-4 membrane filters, and the electrostatic precipitator was equipped with an electron microscope grid. A three-stage high-volume air sampler was tested. It was equipped with three graded filters in tandem.

As in past tests, many grass samples were collected immediately after the release to determine the initial iodine deposition on the pasture. Grass samples from an area of 1/2 m² were collected near the base of each high-volume air sampler on the instrumented arcs (A, B, C, and D), and grab samples (randomly clipped handfuls of vegetation) were collected at other arcs. Ten 1/2 x m² samples were collected daily for a total of 21 days after the test to obtain estimates of the residence time of iodine on the pasture. In addition to the grass samples, soil and sagebrush samples were taken to provide estimates of total deposition per unit area. The data from these samples will be used in estimating depletion of airborne activity (by deposition) over terrain similar to that at the National Reactor Testing Station. In order to estimate the grass-to-milk transport factors, the contaminated pasture was divided into 150- by 12-meter strips for grazing. Each strip was surrounded by an electric fence to confine the cows. The six cows at the EDF were placed on a different pasture strip each day, after the morning milking, and allowed to graze for the 24-hour period. After the cows were removed from the strip, forage utilization was measured. Grass samples were collected each morning from each strip that the cows were to graze (to obtain estimates of the amount of activity consumed). Moisture content of samples was estimated for the purpose of minimizing variations in consumption estimates.

As before, the cows were milked twice daily (7 a.m. and 5 p.m.). A 900-ml sample was taken from each cow at each milking. These samples were used for gross gamma analyses; the contribution from K-40 was subtracted from the gross count before conversion to pCi of iodine-131 per liter of milk.

To obtain data on the retention of I₂ by inhalation, seven volunteers were seated next to high-volume air samples on the D arc during the release period. The breathing rate of the individual volunteers, in a sitting position, was determined prior to the test. Since reliable air concentration data were available from the air samplers, it was possible to make good estimates of the iodine inhaled by each volunteer. Two of the volunteers were wearing lapel samplers [a] equipped with a GM-4 membrane filter backed by a charcoal-impregnated filter paper to obtain particle size data on the inhaled iodine.

1.3 Meteorological Conditions During the Test

A summary of the meteorological conditions during CERT-7 is shown in Table II.

[a] Mine Safety Appliances, Pittsburgh, Pa.

TABLE II

SUMMARY OF THE METEOROLOGICAL CONDITIONS DURING CERT-7

Temperature	9.3°C
Relative humidity	34 percent
Wind speed at 4m	5.4 m/sec
Richardson number (Ri) 4 to 16m	-0.20

The Richardson number indicates the degree of atmosphere stability; in CERT-7 it was slightly unstable.

2. DEPOSITION AND HALF-LIFE STUDIES

Figure 4 shows the distribution deposition velocity, V_g , estimates for vegetation at the EDF. These were calculated, after Chamberlain [8], by the following:

$$V_g = \frac{\text{total deposition (Ci/m}^2\text{)}}{\text{time integrated concentration (Ci-sec/m}^3\text{)}} .$$

The distribution appears to be log-normal with a median value of 0.82 cm/sec. This value does not appear to be significantly different from results obtained in previous tests.

The observed half-life of iodine on pasture grasses is shown in Figure 5. The 6.5-day effective half-time is longer than the effective half-time observed in either CERT-1 (3.5 days) or CERT-2 (5.5 days) and longer than the 4- to 6-day

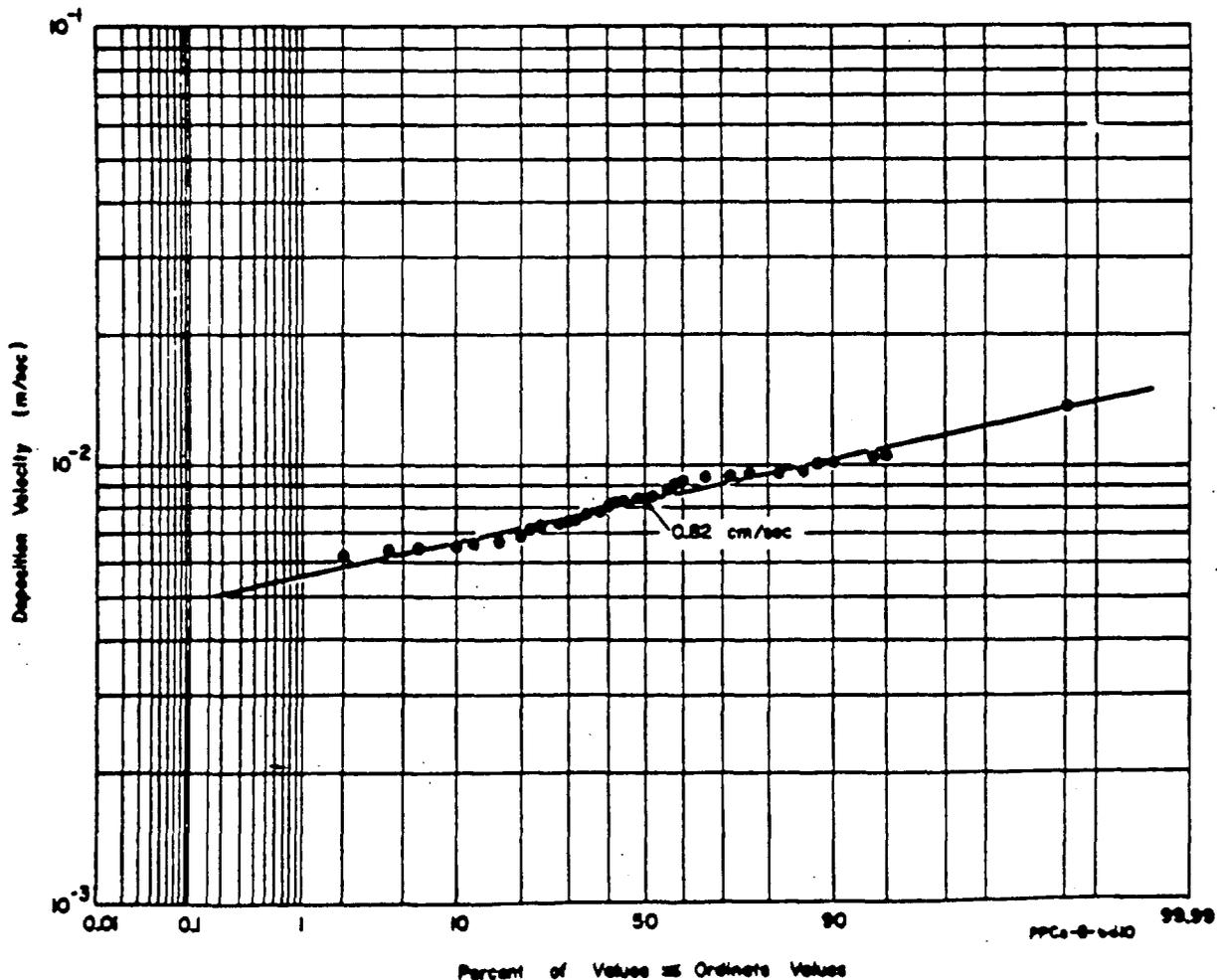


FIG. 4. DEPOSITION VELOCITIES IN CERT-7

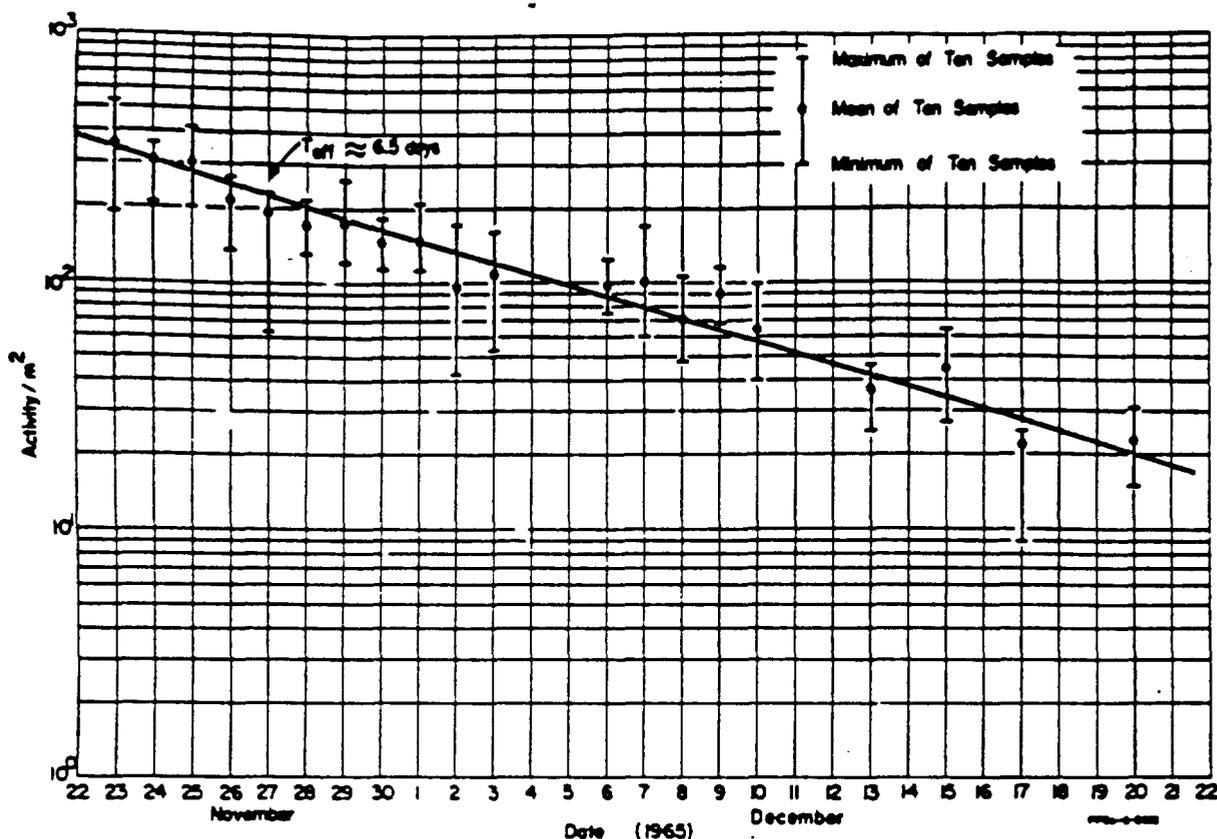


FIG. 9 EFFECTIVE HALF-LIFE OF IODINE-131 ON VEGETATION DURING CERT-7.

half-times frequently reported in the literature [9]. Further consideration of the problems in predicting both deposition and half-life is discussed in Section V.

3. TRANSFER OF ACTIVITY FROM GRASS TO MILK

As indicated earlier, the activity on the areas where the cows were to graze was measured just prior to cow entry on the particular area. The results of these measurements are shown in Table III in terms of pCi/g fresh weight and pCi/g dry weight.

Because it rained and snowed in the days succeeding the test, the fresh weight of vegetation (g/m^2) varied during the test. As a result, estimates of the total activity consumed per day were based on total activity per gram of dry weight. These estimates are found in Table IV.

These numbers in Table IV were obtained by estimating the total forage consumed in a given area. While this method is usually a reasonably accurate method of estimating mean consumption of grass, we are of the opinion that the grass consumption was underestimated. The cows were observed to choose small spots to graze cleanly, consuming all standing grass and old grass clippings. The cows did not consume enough forage to maintain milk production or body weight. This was apparently due to the decreased palatability of the grass. These factors make the utilization and consumption estimates subject to some question.

TABLE III

MEAN DAILY PASTURE ACTIVITY

Date	pCi/g (fresh)	pCi/g (dry)
11/23	740 ± 120[a]	1350 ± 150
11/24	440 ± 60	940 ± 80
11/25	360 ± 40	720 ± 80
11/26	410 ± 50	520 ± 60
11/27	1040 ± 80[b]	—
11/28	520 ± 60	550 ± 60
11/29	530 ± 100	560 ± 100
11/30	450 ± 90	460 ± 90
12/1	480 ± 80	500 ± 80

[a] Ninety-five percent confidence interval.

[b] Apparent error, data point discounted.

TABLE IV

ESTIMATED ACTIVITY CONSUMED BY COWS

Date	Total Consumption by All Cows (μCi)	Mean Consumption per Cow (μCi)
11/23	72	12.0
11/24	52	8.7
11/25	40	6.7
11/26	29	4.8
11/27	29[a]	4.8
11/28	30	5.1
11/29	31	5.2
11/30	26	4.3
12/1	28	4.6
Total	340	56.0

[a] Interpolated estimate based on initial pasture contamination.

Average iodine-131 concentrations in milk are shown in Table V and Figure 6. The peak activity was reached on the fifth milking, 54 hours after the cows went on contaminated pasture. Thereafter, activity in the milk remained at or near the peak level of activity until the cows were removed from the contaminated pasture. It was planned that the cows would be moved on more highly contaminated areas to counterbalance the loss of activity by weathering. After removal of the cows from the contaminated pasture, the milk activity decreased with a half-time of 0.6 days for 3 days, a half-time of 2.4 days from the fourth to the ninth day, and finally decreased with a half-time of 6.8 days thereafter. Mathematically, this is expressed as follows:

$$A_t = A_0 (0.98 \exp [-1.15t] + 0.009 \exp [-0.88t] + 0.009 \exp [-0.102t])$$

where A_0 is the concentration in milk when the cows were removed from pasture. (This decay rate is about the same as observed in earlier CERT tests and is shorter than that reported by Tamplin [10].)

The ratio of peak milk activity (pCi/liter) to initial grass activity (pCi/g) at the start of grazing was 230, some 70 percent higher than was observed in CERT-1 or -2. CERT-7 was conducted in November and December with dry

TABLE V
MILK DATA FOR CERT-7

<u>Date</u>	<u>Average μCi per Liter</u>	<u>Total Liters Produced per Day</u>	<u>Average Liters per Cow per Day</u>	<u>Total μCi Excreted in Milk per Day</u>	<u>Average μCi Excreted in Milk per Day per Cow</u>
11/23/65	0.015	33.5[a]	5.6[a]	0.510	0.0851
11/24/65	0.097	68.8	11.5	6.68	1.11
11/25/65	0.138	63.2	10.5	8.72	1.45
11/26/65	0.149	56.2	9.4	8.24	1.37
11/27/65	0.132	57.8	9.6	7.64	1.27
11/28/65	0.133	55.1	9.2	7.33	1.22
11/29/65	0.149	55.9	9.3	8.31	1.38
11/30/65	0.154	56.7	9.5	8.72	1.45
12/1 /65	0.139	54.4	9.1	7.55	1.26
12/2 /65	0.091	57.2	9.5	5.22	0.871
12/3 /65	0.029	60.6	10.1	1.76	0.294
12/4 /65	0.0108	62.4	10.4	0.674	0.112
12/5 /65	0.0045	61.8	10.3	0.272	0.0453
12/6 /65	0.0025	59.7	10.0	0.148	0.0247
12/7 /65	0.0012	62.3	10.4	0.0764	0.0127
12/8 /65	0.0009	60.7	10.1	0.0560	0.0093
12/9 /65	0.0008	61.8	10.3	0.0478	0.0080
12/10/65	0.0007	62.4	10.4	0.0421	0.0070
12/11/65	0.0006	60.9	10.2	0.0336	0.0056
12/12/65	0.0005	51.7	8.6	0.0244	0.0041
12/13/65	0.0003	57.5	9.6	0.0194	0.0032
12/14/65	0.0003	60.2	10.0	0.0191	0.0032
12/15/65	0.0003	59.4	9.9	0.0173	0.0029
12/16/65	0.0003	58.9	9.8	0.0148	0.0025

[a] Only one-half output included (p.m. milking)

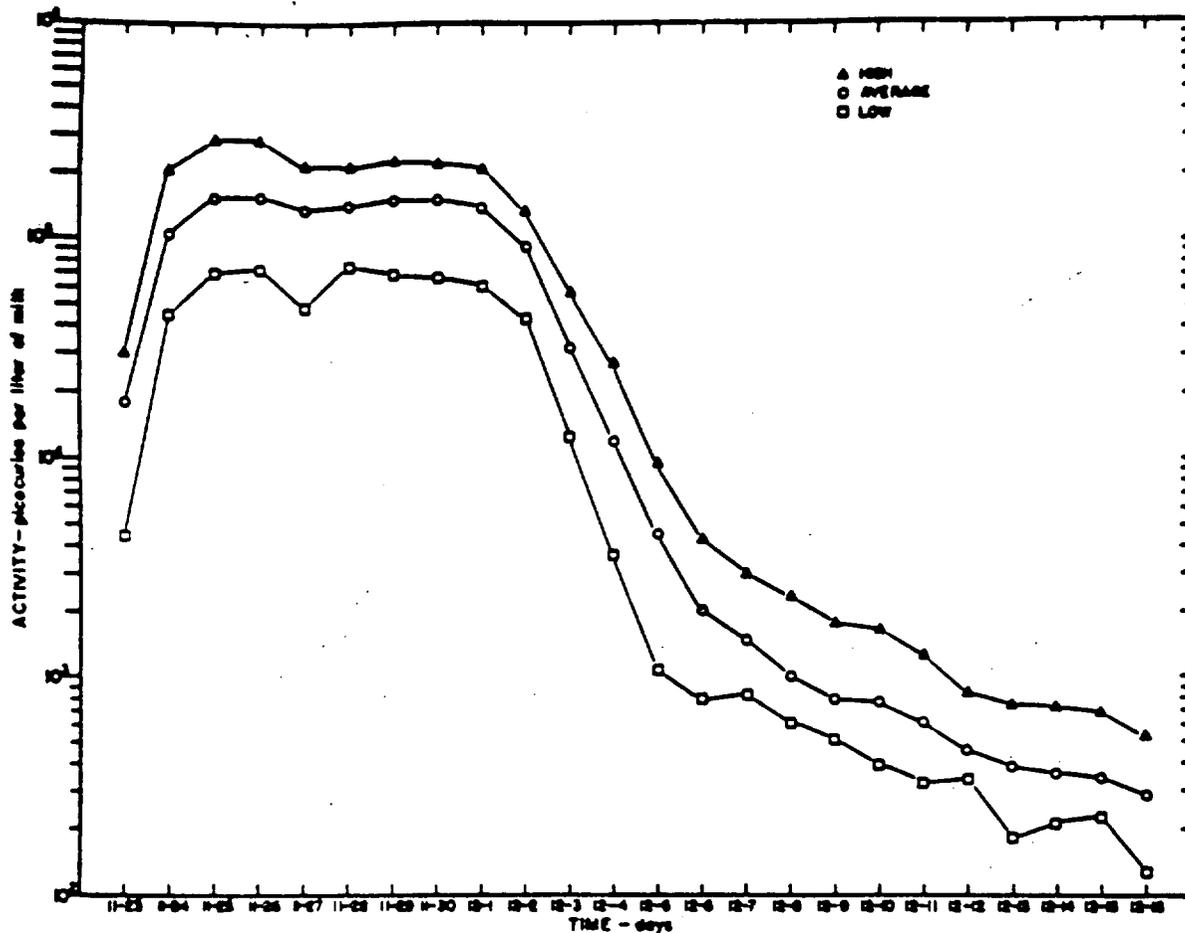


FIG. 6 MEAN MILK ACTIVITY DURING CERT-7.

pasture of less nutritive quality than in earlier tests conducted in May, June, and September. In this regard, it has been suggested that a relationship exists between the nutritive quality of pasture and the levels of iodine in milk; the highest iodine-131 levels were reached when pasture quality was poorest [11].

This is borne out in other comparisons; the mean recovery in milk was approximately 20 percent of the mean total iodine-131 ingested. This compared with mean recoveries of about 10 percent for CERT-1 and -2. The mean daily ingested iodine-131 secreted per liter of milk also reflected this increase; this was less than 1 percent per liter in CERT-1 and -2 and slightly more than 2 percent per liter in CERT-7. At this time, it is not known whether the greater recovery is related to (a) the difference in pasture quality, (b) if other factors are working with the pasture factor to produce a net increase in iodine-131 secretion in milk, or (c) if the consumption data were biased enough to cause a difference in results.

4. HUMAN INHALATION STUDIES

As in previous tests, attempts have been made to make a direct comparison of the dose from ingestion and inhalation. In CERT-7, as in CERT-2, seven

volunteers were seated next to air samplers on the D arc during the iodine release. In CERT-2, a "standard" breathing rate was assumed since no data were available at that time on the individual variations or the mean breathing rate of these individuals in a sitting position. In CERT-7, respiration rates were measured and estimated with the results shown in Table VI.

Based on the above breathing rates and on counts of thyroid activity following the test, the estimates of f_a (fraction of the amount inhaled that was deposited in the thyroid) are listed in Table VII.

TABLE VI

MEASURED BREATHING RATES

Volunteer	Respiration Rate	
	(liters/min)	m ³ /sec
DP	8.4	1.4 x 10 ⁻⁴
JTC	6.5	1.1 x 10 ⁻⁴
LO	8.5	1.4 x 10 ⁻⁴
JO	6.7	1.1 x 10 ⁻⁴
JPC	16.0	2.7 x 10 ⁻⁴
KB	8.3	1.4 x 10 ⁻⁴
NH	9.5	1.6 x 10 ⁻⁴

TABLE VII

FRACTIONAL UPTAKE OF INHALED IODINE

Volunteer	f_a
DP	0.27
JTC	0.32
LO	0.25
JO	0.33
JPC	0.32
KB	0.21
NH	<u>0.44</u>
Average	0.30

In CERT-2, the concentrations to which volunteers were exposed were virtually identical to those in CERT-7. The activity in the thyroids was also about the same. This indicates that the fractional uptakes were about the same and that the standard breathing rate used in the CERT-2 data analysis was too high.

5. CONCLUSIONS

On the basis of data from this test, it appears that iodine concentrations in milk may be relatively greater in fall than in spring or summer months, probably due to an increased rate of secretion of iodine in milk, since changes in half-life were not great enough to account for observed differences. Since CERT-7 was conducted in late fall, these results may not be representative of normal fall conditions. However, there is indication [12] that there is an increased rate of secretion in milk in fall; this can only be proved by further studies.

The median deposition velocity, as measured, was 0.82 cm/sec, not markedly different from previous tests. The observed half-life on vegetation was 6.5 days. The average thyroid uptake of iodine was 30 percent of that inhaled.

IV. CERT LABORATORY EXPERIMENTS

The CERT Laboratory Experiments (CERTLE) were begun in late 1964 and have two major functions:

- (1) To provide preliminary data to enable better system design in field tests
- (2) To complement studies in the field to enable better interpretation of field data.

To this end, a plexiglass exposure chamber approximately 3 by 3 by 5 feet was built and installed in a controlled environment laboratory in the ID Health and Safety Laboratory Building (Figure 7). In this chamber the temperature, humidity,

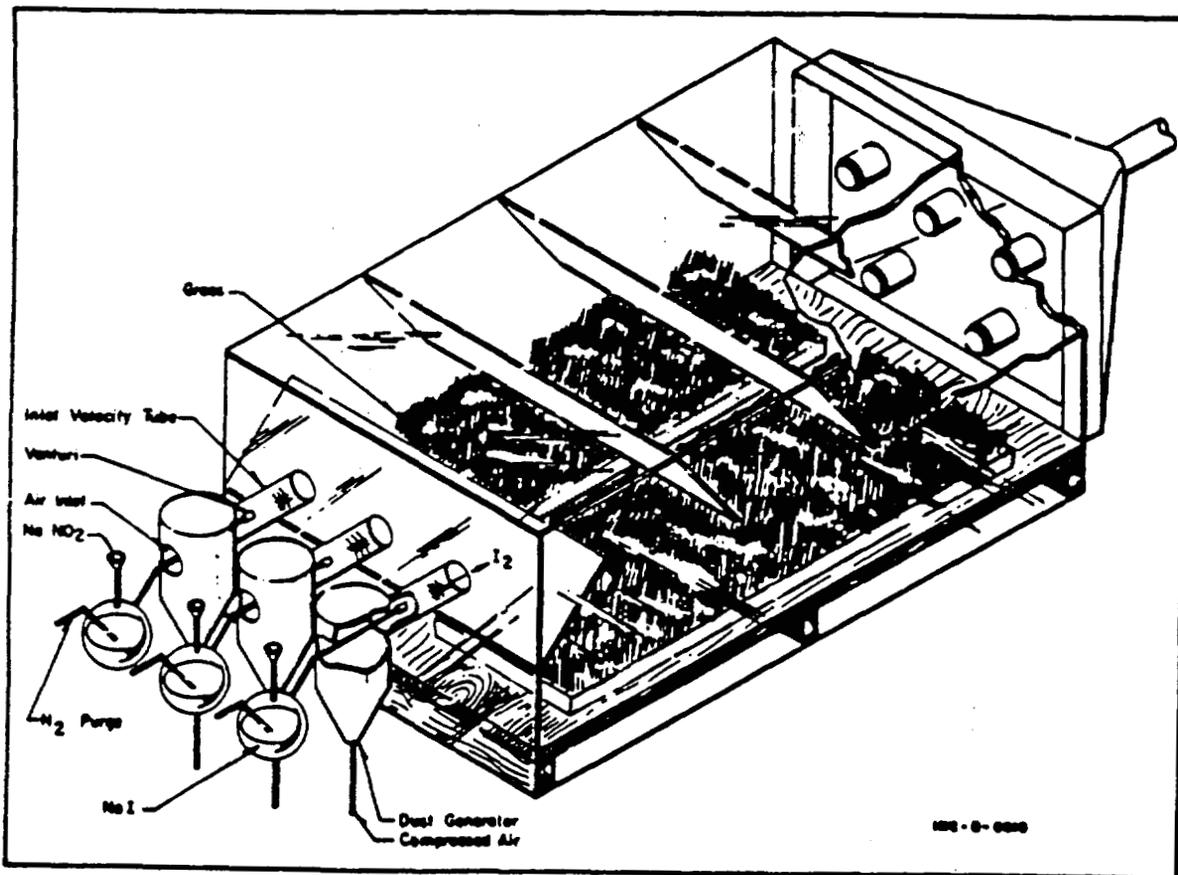


FIG. 7 SYSTEM DESIGN IN THE CERT LABORATORY EXPERIMENTS (CERTLE).

quality, quantity, and duration of light and air speed can be controlled. Experiments are completely contained within the chamber. Off-gases are filtered and vented through a five-foot stack on top of the building. The stack vent is monitored during every operation.

1. DEVELOPMENT OF AN IODINE SAMPLER FOR FIELD TESTING

Over the years, a number of attempts have been made to devise a field sampler that will discriminate between the various forms of radiiodine. In

order to have a sampler that would adequately collect airborne CH_3I in future tests, developmental work and testing have been started on a May pack type sampler. The present sampler design consists of the following:

- (1) A Staplex High-Volume Air Sampler to operate (loaded) at about 10 cfm
- (2) First filter -- Microsorban
- (3) Second filter -- two Gelman E glass fiber filters upon which finely divided silver metal has been precipitated
- (4) Third filter -- Gelman AC-1 carbon-impregnated paper
- (5) Fourth filter -- MSA 46727 (carbon charcoal No. 85851) carbon cartridge.

While the testing has not been completed, we have found rather marked separations of collection between I_2 and CH_3I (Table VIII). Furthermore, there appears to be no appreciable migration of iodine from one filter to another, at least over a period of a few hours.

This system will allow reasonably high flow rates (10 cfm) and still provide data on the material form. Any change in material form with distance traveled can be detected by the relative change in the ratio of collected activities on each of the filters.

2. DEPOSITION OF IODINE ON GRASS

Experiments to date have only dealt with deposition on grass as a function of released form and areal density of vegetation (g/m^2). In these tests, day time (85°F, relative humidity 50 percent), night time (65°F, relative humidity 100 percent), and dusty conditions (2 m/g liter) were simulated. For I_2 , the results indicate that the night time deposition is about twice that of day time deposition. Additions of particulate aerosols eliminated any apparent differences. The deposition of CH_3I on grass appears to be about three orders of magnitude less (10^{-3}) than I_2 under the same conditions.

Deposition studies were conducted in this box over three different types of grass (crested wheatgrass, bluegrass, pasture grass) and a range of areal densities. While there were no significant variations among grass types, a very distinct trend was observed with regard to deposition as a function of density.

The later section on the meteorological aspects of turbulent deposition indicates a linear relationship of deposition velocity/density, V_g/D , to friction velocity, u_* . CERTLE studies have carried these measurements up to densities

TABLE VIII

PERCENT COLLECTION OF ACTIVITY ON HIGH-VOLUME IODINE SAMPLERS[a]

Filter	I_2	CH_3I	HI
Microsorban	2.0	<0.02	46.0
Silvered Paper	87.0	<0.01	4.2
Carbon Paper	9.0	2.0	4.6
Carbon Pack	1.0	88.0	45.0

[a] Estimates of impurities in HI release not known at this time.

of 2000 g/m² wet weight; the results of which are shown in Figure 8. The slope of the line is the same as found in field data from CERT-2. As can be seen, the true relation is somewhat less than linear; as densities increase, transfer of iodine to lower parts of the plants is inhibited. Hence, the rate of increase of V_g with an increase in D, becomes smaller. The deposition velocity in this case is determined from the ratio of the collected activity on filters and the total activity on vegetation. Autoradiographic studies have shown a definite progression toward relatively greater deposition on upper parts of the leaves as grass density increases. It should be noted, however, that the assumption of a linear relation is reasonable over the range of densities encountered in typical pastures in the Western United States (D < 500 g/m² wet weight or 200 g/m² dry weight). Studies are continuing on this problem as well as studies on half-life.

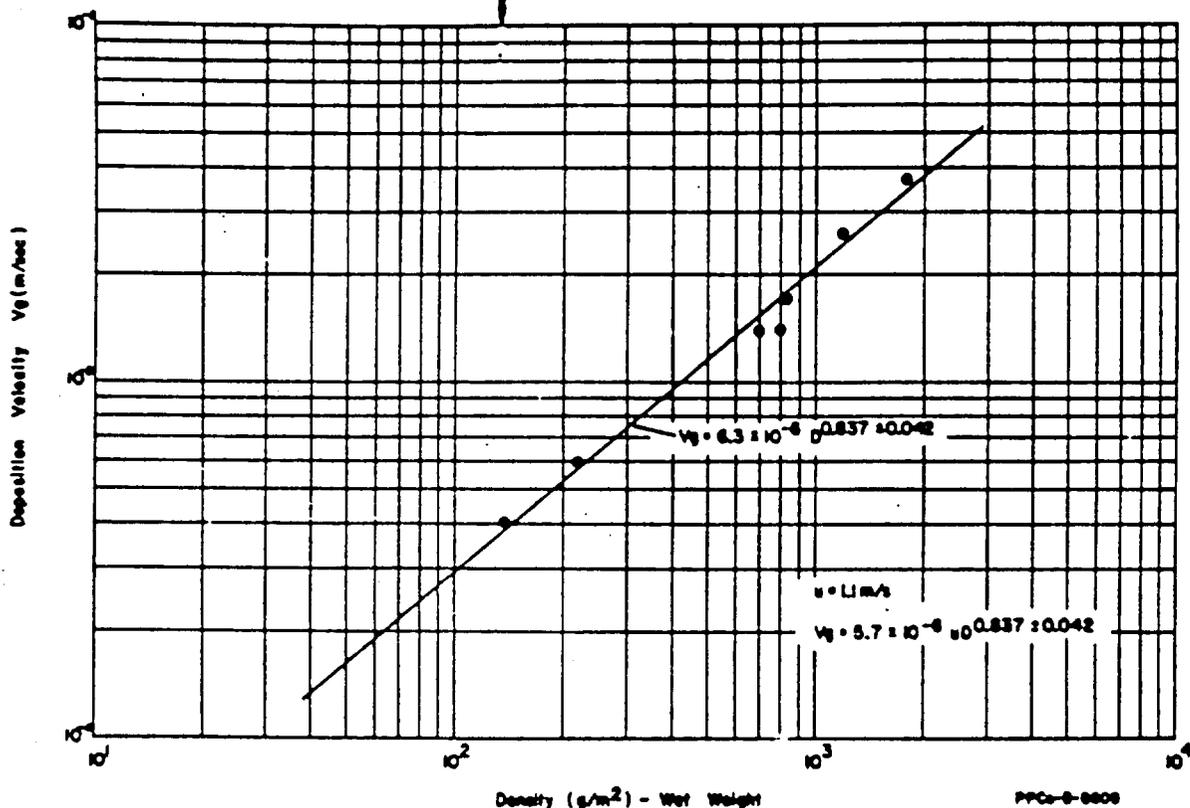


FIG. 8 DEPOSITION VELOCITY AS A FUNCTION OF AREAL DENSITY.

V. CERT ANALYTICAL EFFORTS

1. INTRODUCTION

The general objectives stated in the summary and introduction indicated the scope and nature of the project. More specifically, there are four primary objectives of the CERT project:

- (1) To determine quantitative estimates of the thyroid dose due to ingestion of iodine-131
- (2) To derive a working model for deposition of iodine
- (3) To derive a working model to estimate the effective residence time (half-time) of iodine on vegetation
- (4) To derive a working model to estimate the transfer of iodine from grass to milk.

Obviously, none of these objectives can be met after a single test. As the program continues, efforts are made to continually upgrade the analytical effort and preliminary models in order to meet these objectives. Over the last three years, a great deal of progress has been made in meeting these objectives, and this section contains information on the present status of several of these efforts.

2. METEOROLOGICAL ASPECTS OF TURBULENT DEPOSITION

In trying to develop a working model for iodine deposition in the milk-food chain, we are concerned with variables such as wind speed and atmospheric stability which determine the transfer from air to ground surface and the kind and density of vegetation and form of the depositing material. To derive such a model, the relative affect of the variables on the net deposition must be considered.

There have been many attempts to derive models that would adequately describe the deposition of iodine onto surfaces [13 to 18]. Of these, two [17,18] are directly concerned with deposition from the free atmosphere.

If, as is assumed by these theories, the primary transport mechanisms are eddy diffusion and molecular diffusion, the equation for the vertical transport of matter to a surface can be represented as

$$\left[D_m + K(z) \right] \frac{\partial x}{\partial z} = V_{g_0} x_0 \quad (3)$$

where

V_{g_0} = deposition rate per concentration at the interface of plant and atmosphere (m/sec)

- x_0 = concentration at the interface of plant and atmosphere ($C1/m^3$)
 x = concentration at a height z ($C1/m^3$)
 $K(z)$ = eddy diffusivity (m^2/sec)
 D_m = molecular diffusivity (m^2/sec)
 z = height above plant surface.

In this case, x is taken to be x_0 when $z = 0$ since most surfaces are not "perfect sinks." Three tests have been selected to compare results of deposition measurements with theoretical calculations. CERT-1, -2, and -7 were selected because they represent deposition in an unstable atmosphere, the stability condition, under which the condition,

$$K(z) = k u_* z \quad (4)$$

is most likely to be valid [19]. Where u_* equals the friction velocity derived from the wind velocity profile (m/sec), k = von Karman's constant = 0.41,

and

$$u_* = \frac{k \bar{u}}{\ln(z/z_0)} \quad (5)$$

where

z_0 = the roughness length (m)

\bar{u} = the mean wind speed (m/sec).

After substituting Equations (4) and (5) into Equation (3), Equation (3) can be solved with the following result:

$$V_{g_0} = \left(\frac{x - x_0}{x_0} \right) \frac{k u_*}{\ln \left(1 + \frac{k u_* z}{D_m} \right)} \approx \frac{x - x_0}{x_0} \frac{k u_*}{\ln \left(\frac{k u_* z}{D_m} \right)} \quad (6)$$

By definition,

$$V_{g_0} = \frac{C}{x_0} \text{ and } V_{g_1} = \frac{C}{x_1} \quad (7)$$

where

C = amount deposited/sec V_{g_1} and x_1 as determined at some reference height z_1 so that

$$V_{g_0} x_0 = V_{g_1} x_1$$

$$V_{g_1} = \left(\frac{x_1 - x_0}{x_1} \right) \frac{ku_s}{\ln \left(\frac{ku_s z}{D_m} \right)} \quad (8)$$

or

$$V_{g_1} = \left(1 - \frac{x_0}{x_1} \right) \frac{ku_s}{\ln \left(\frac{ku_s z}{D_m} \right)} \quad (9)$$

This transformation is made since V_g and x are more easily measured than x_0 and V_{g_0} . In this case it is important to note that the percent retention of material by a surface is

$$100 \left(1 - \frac{x_0}{x_1} \right)$$

and that the "resistance" at the surface termed by Chamberlain [17] is also related to the transport characteristics of the atmosphere as long as the surface is not a perfect sink.

The retention and the calculated and measured deposition velocities are shown in Table IX (assuming all airborne radioactivity was in the form of I_2). It can be seen that grass is far from a perfect sink. There also appears to be a very definite relation between deposition and the mass or surface of the plant as represented by grams of vegetation per meter² (dry weight).

TABLE IX

VALUES OF DEPOSITION AND RETENTION FACTORS ON GRASS
FOR SELECTED CERT RELEASES

Test	$V_g(z_1)$		Percent Retention	
	Measured (m/sec)	Perfect Sink (m/sec)	Total	Per Gram (dry weight)
CERT-1	0.0058	0.0242	24	0.44
CERT-2	0.0098	0.0270	36	0.45
CERT-7	0.0082	0.0185	44	0.40

Empirical analyses of several of the tests (CERT-1, -2, -5, and -7) indicate, as do the laboratory experiments described earlier, that grass deposition can be estimated with a knowledge of time-integrated concentration of radioactivity in air and either u_s or \bar{u} . The formulas are

$$A = 1.76 \times 10^{-4} u_s x \quad (10)$$

and

$$A = 1.81 \times 10^{-5} \bar{u} x \quad (11)$$

where

$$A = \text{amount of activity deposited per gram of vegetation (dry weight)} \\ = \frac{Vg \times X}{D}$$

The data points and the lines representing the above two equations are shown in Figure 9.

We have found that estimates of deposition based on dry weight are much less variable than those based on fresh weight. This is to be expected since dry weight is a more realistic indicator of plant area. Again, a linear relationship with grass density has been assumed. All the tests conducted so far have been with mean dry weight densities of 150 grams per square meter or less. In this region, the assumption of a linear relationship produces reasonably accurate results. When more lush pastures are encountered, a correction from D^1 to $D^{0.8}$ in Equations (10) and (11) should be made.

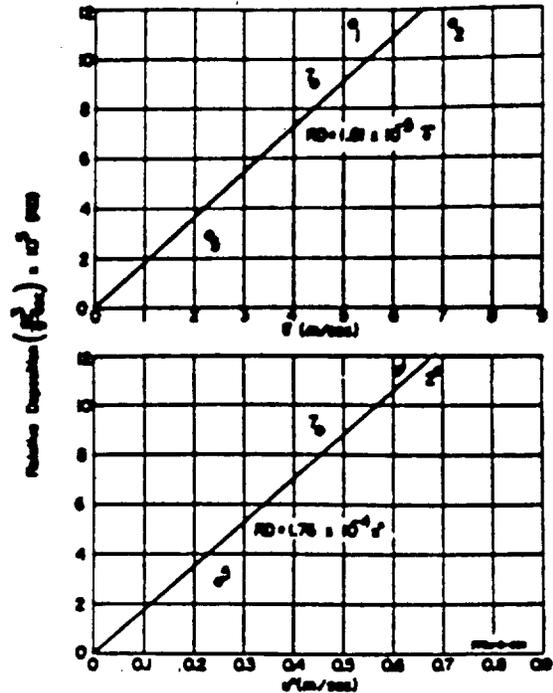


FIG. 9 RELATIVE DEPOSITION VERSUS FRICTION VELOCITY AND WIND SPEED.

3. RESIDENCE OF IODINE ON GRASS

Up to the present time, there appears to have been no serious attempts to explain the loss of iodine on vegetation. In the absence of satisfactory explanations, we have endeavored to identify the major variables associated with iodine loss on vegetation.

3.1 Growth Factors

The concentration of iodine-131 on grass is reduced by simple dilution when the grass grows. This being the case, the possibility of grass growth after a contaminating event must be considered [9].

Previous CERT experiments have shown that there is no significant difference in activity on various parts of the leaves of the pasture grasses (grass density 500 g/m² wet weight). This being the case, changes in consumed activity per unit mass of vegetation should be reasonably approximated by the rate of increase in mass, all other factors being constant. A recent report [20] has indicated a relationship of dilution with the rate of growth. If it is assumed that the cows continue to graze a pasture so that the total grams per unit area remains reasonably constant and it is also assumed that the rate of new growth remains constant over the period of interest, then an assumed exponential rate of loss, per gram, should be appropriate.

In CERT-1, these conditions were met. The average rate of grass growth was 5.2 percent per day. This would nearly account for the difference between half-lives in CERT-1 and -2.

3.2 Weathering Factors

The resuspension experiment in CERT-2 indicated an exponential rate of resuspension, corresponding to a rate of loss proportional to $e^{-0.03t}$. Various studies have dealt with the problem of resuspension, wind pickup, and evaporation [21,22,23].

Several of these have indicated a rate of pickup that is proportional to the square of the mean wind speed. In the report by Healy and Fuquay [22], the concentration over an infinite area source is constant and proportional to the areal density of particles on the ground and the rate of resuspension. If the CERT deposition area is regarded as effectively infinite in size, then the rate of loss of iodine from ground may be described as

$$\lambda_u \propto ku^2$$

If "k" is regarded as a constant and "u" is taken as the average wind speed over the first five days of the test (CERT-1, $u = 7$; CERT-2, $u = 7$; CERT-7, $u = 5$), then the rate of loss by resuspension should be on the order of two times greater in CERT-1 and -2 than in CERT-7. Since wind weathering appears to be a significant factor in iodine loss from vegetation (as indicated by CERT-2 resuspension studies), we have assigned a weathering rate of loss of activity on vegetation in Cert-7 of 2 percent per day based on comparative wind speed.

3.3 Plant Factors

In speculating on possible reasons for differences in residence time of iodine on vegetation, two obvious possibilities have been mentioned -- grass growth and weathering effects. There should be at least one other set of phenomena leading to iodine loss on vegetation, which is related to the physiology of the plant itself.

In analyzing CERT-2 data, there appears to be about a 2 percent loss per day due to factors other than wind weathering and radioactive decay. One study [24] has indicated a relationship of half-life to temperature, and another [25] has indicated that the rate of loss could be associated with the seasonal variation in the rate of loss of waxy particles from the grass. Either of these factors could account for the 2 percent per day loss in CERT-1 or -2, and either of these factors would probably have little effect in CERT-7.

3.4 General

We have attempted to account for the differences in observed half-lives in CERT-1, -2, and -7 on the basis of the above factors. In this, we have assumed that all factors are exponential by nature and that

$$\lambda_E = \lambda_G + \lambda_R + \lambda_u + \lambda_P \quad (12)$$

where

λ_E = effective rate of loss

λ_R = radioactive rate of decay

λ_u = rate of loss by weathering

λ_P = rate of loss by plant factors.

This influence is shown in Table X.

TABLE X
HALF-LIFE FACTORS ON VEGETATION

	<u>CERT-1</u>	<u>CERT-2</u>	<u>CERT-7</u>
λ_R	0.086	0.086	0.086
λ_G	0.052	0.000	0.000
λ_u	0.030[a]	0.030	0.02[a]
λ_P	0.020[b]	0.020[b]	0.000
λ_E	0.188	0.136	0.106
T_{eff} (days)	3.7	5.1	6.5

[a] Extrapolated from CERT-2.

[b] Assumed.

These results, if a "P" of 0.02 is assumed for CERT-1 and -2, are not significantly different from those actually observed.

Although these are rather gross estimates, it certainly seems possible to account for observed half-life on the basis of a few parameters.

Further tests are being planned to refine these estimates and to provide a more sound theoretical basis upon which to make these assumptions.

4. TRANSFER FROM GRASS TO MILK

In the CERT program, we are more interested in predicting the transfer of ingested iodine to milk (and the confidence with which it can be predicted) rather than in deriving a mathematical model which describes the physiological transfer process in cows. From CERT data presently available, it appears that an empirical model can be derived which will adequately predict the activity levels in milk after a single contaminating event.

In estimating dose from iodine in the thyroid via the ingestion route, it is of importance to examine the interdependence of the various parameters. In

many cases, the combination of two conservatively estimated parameters may lead to an unduly conservative result. Therefore, in deriving a model, we have attempted to combine interdependent terms where possible.

From a mathematical standpoint, it appears that the data can be best represented by referring to secreted activity as a percent of the daily ingested activity per liter.

The average time from ingestion to arrival in milk appears to be on the order of 4 to 8 hours so that the initial assumptions made are as follows:

- (1) In the cow, the transfer time from the GI tract to blood is short enough so that it is assumed the material is instantly absorbed in blood upon ingestion [26].
- (2) Milk activity is assumed to be a constant fraction of blood activity [27] so that any decrease of iodine in blood will be marked by the same relative loss in milk concentration. This presupposes that a diffusion compartment exists between blood and milk [28]. On this basis, for an acute exposure,

$$\frac{dM}{dt} = k_m I_o C \exp [-\lambda_E t] - \lambda_B M \quad (13)$$

where

k_m = the fraction of the daily ingested dose per liter of milk (l⁻²)

I_o = the initial pasture activity per gram of vegetation (Ci/g)

C = the rate of consumption of vegetation in gram/day

λ_E = the effective rate of loss of activity on vegetation (day⁻¹)

M = the activity per liter of milk (Ci/l)

λ_B = the effective rate of loss of activity from the blood (day⁻¹).

The solution is

$$M = \frac{I_o C k_m}{\lambda_B - \lambda_E} \left(\exp [-\lambda_E t] - \exp [-\lambda_B t] \right)$$

and the total activity in μ Ci-days/liter is

$$\int_0^{\infty} M dt = \frac{I_o C k_m}{\lambda_B - \lambda_E} \quad (15)$$

An alternate form that can be used to estimate delivered dose as a function of time is

$$\int_0^t M = \frac{I_0 C k_B}{\lambda_B - \lambda_E} \left(\frac{1 - \exp[-\lambda_E t]}{\lambda_E} - \frac{1 - \exp[-\lambda_B t]}{\lambda_B} \right) \quad (16)$$

The value of k_m may be related to F_m (the fraction of that ingested that ultimately is secreted in milk) by the following relationship:

$$F_m = \frac{L \int M dt}{I_0 C \int \exp[-\lambda_E t] dt} = \frac{L k_m}{\lambda_B} \quad (17)$$

where

L = the average milk output (l/day).

Values of k_m were computed from the measured values of F_m , L , and β and compared graphically to the ratio of the activity per liter of milk and activity consumed per day (Figure 10). The results are obvious. A second check was made by comparing calculated and measured values of peak milk activity to initial grass activity. The results are shown in Table XI.

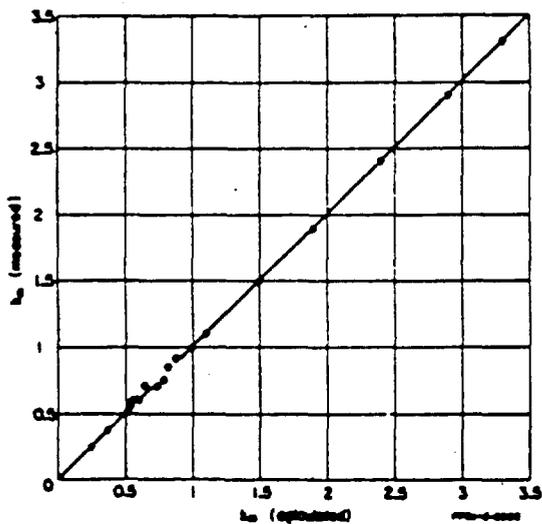


FIG. 10 MEASURED VERSUS COMPUTED VALUES OF PER LITER IODINE SECRETION.

TABLE XI

VALUES OF PEAK MILK ACTIVITY TO INITIAL GRASS ACTIVITY (pCi/l/pCi/g)

	CERT-1	CERT-2	CERT-7
Calculated	135	140	240
Measured	130	130	230

It should be noted that calculated estimates are in all cases high (although not significantly). This is due to the assumption of no transfer time to blood. The closeness of fit illustrates that this is not an unwarranted assumption.

One of the questions that is always posed in any evaluation of dose is the accuracy and/or possible variation. Since, in the CERT tests and in most hazards evaluations, a single consumption estimate is used, it seems possible to combine variations in k_m and C to evaluate the net variation in estimates of the transfer of activity from grass to milk. This has been done, and the results are shown in Table XII. At this point in time, it appears that the equations presented earlier adequately describe the transfer of activity from grass to milk. Flexibility is still maintained in that k_m may be varied as a function of the stage of lactation or time of year.

TABLE XII
VALUES OF FRACTIONAL SECRETION TO MILK

<u>Test</u>	<u>Cow Number</u>	<u>L</u>	<u>F_m</u>	<u>k_m</u>	
CERT-1	1	10.2	0.057	0.0071	
	2	9.3	0.057	0.0079	
	3	16.3	0.113	0.0100	
			15.8	0.077	0.0061
	5	15.5	0.101	0.0110	
	6	16.6	<u>0.144</u>	<u>0.0110</u>	
		Average		0.092	0.0089
CERT-2	1	12.3	0.160	0.0075	
	2	12.2	0.106	0.0100	
	3	14.8	0.032	0.0025	
	4	13.7	0.071	0.0060	
	5	10.4	0.046	0.0051	
	6	12.3	<u>0.058</u>	<u>0.0055</u>	
		Average		0.079	0.0061
CERT-7	1	3.8	0.064	0.019	
	2	6.2	0.058	0.011	
	3	24.3	0.264	0.012	
	4	11.4	0.289	0.029	
	5	2.0	0.042	0.024	
	6	11.9	<u>0.351</u>	<u>0.033</u>	
		Average		0.178	0.021

5. INGESTION DOSE ESTIMATES

On the basis of the discussed models and other sources of information, it is possible to estimate the thyroid dose from ingested iodine-131. The general equation to estimate mean infinity dose to the thyroid is

$$\text{Dose (rem)} = \left(\int_0^{\infty} M dt \right) \times I \times \text{CONV} \quad (18)$$

where

$\left(\int_0^{\infty} M dx\right)$ = the total per liter output in $\frac{\text{Ci-day}}{\text{l}}$

I = the rate of intake by humans in l/day

CONV = a conversion constant from curies ingested to mean thyroid dose.

This equation may be modified to estimate the dose from the point of release and may be expanded to include the major variables as follows:

$$\text{Dose (rem)} = Q \times \frac{(Xu)}{Q} \times \frac{Vg}{D} \times C \times \frac{k_m}{\lambda_B \lambda_E} \frac{(\exp[-\lambda_R t_1])^{PI} f_w E T_E}{M} \times 7.38 \times 10^7 \quad (19)$$

where

Q = the source in curies

$\frac{Xu}{Q}$ = the relative dilution in m^{-2}

$\frac{Vg}{D}$ = the mass deposition in $m^3/g\text{-sec}$ (deposition velocity in $m/\text{sec} \div$ mass density in g/m^2)

C = the rate of consumption of forage in g/day

k_m = the per liter milk fraction

λ_B = the rate of removal of activity in milk

λ_g = the rate of loss of activity in pasture

λ_R = the radioactive decay rate

t_1 = the time from production to consumption

P = the percent of forage obtained from pasture

f_w = the fraction of that ingested that reaches the thyroid

E = the effective absorbed energy per disintegration

T_E = the effective half-life in the thyroid

M = the mass of the thyroid

7.38×10^7 = a conversion constant to dose.

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DOE/HQ

The terms, $\frac{\lambda_H}{Q}$ and $\frac{V_R}{D}$, are used to reflect the dependency of wind speed on both dilution and mass deposition. Representative mean parameter values were chosen so that estimates of dose potential as a function of season and age group could be made. These are shown in Tables XIII and XIV. Unless otherwise referenced, the values in the table are extrapolated from CERT results or were determined from data from Reference 30.

TABLE XIII
TRANSPORT FACTORS

<u>Parameter</u>	<u>Spring</u>	<u>Summer</u>	<u>Fall</u>	<u>Winter</u>
V_E/D	$1.8 \times 10^{-5} \bar{u}$			
C	1.3×10^4	1.3×10^4	1.3×10^4	6×10^3
k_m	0.016	0.008	0.016	0.016
λ_B	0.90	0.90	0.90	0.90
λ_E	0.176	0.136	0.136	0.116
λ_R	0.086	0.086	0.086	0.086
τ_1	0.5	0.5	0.5	0.5
P[30]	0.05	0.75	0.35	0.02

TABLE XIV
UPTAKE FACTORS

<u>Age Group</u>	<u>I[31]</u>	<u>f_v</u>	<u>E[32]</u>	<u>T_E[33]</u>	<u>M[34]</u>
0-1	0.5	0.2	0.21	4.0	2.0
2-5	0.5	0.2	0.21	4.5	2.5
6-12	0.5	0.2	0.21	5.5	3.0
13-19	0.75	0.2	0.22	6.0	5.0
>20	1.0	0.2	0.22	6.5	15.0
	0.75	0.2	0.23	7.0	20.0

On the basis of these parameters, the dose potential in Table XV was derived. This was computed on the basis of 1 Ci-sec/m³ over the pasture.

TABLE XV

INGESTION DOSE POTENTIAL (rem)
FROM 1 Ci-sec/m³ OF I-131 [a]

Age	Winter	Spring	Summer	Fall
0-1	1000	3600	34,000	32,000
2-3	900	3200	31,000	29,000
4-6	900	3200	31,000	29,000
7-12	900	3200	31,000	29,000
13-19	450	1600	15,000	14,000
>20	290	1000	10,000	9,400

[a] All values must be multiplied by the mean wind speed.

where

Q = the source in curies

$\frac{X_u}{Q}$ = the relative dilution

F₁ = a dose conversion constant and takes the form of

$$1.8 \times 10^{-5} \left(\frac{\int_0^{\infty} Mdt}{I_0} \right) \times \frac{\text{rem}}{\text{Ci-day/liter}}$$

If Q and $\frac{X_u}{Q}$ are known, only the numerical values in Table XV need to be used. (Do not multiply by the wind speed.) In this fashion, consideration of wind speed in the dose calculation is eliminated. This is because an increase in wind speed will increase the values of the conversion factors, but this will be exactly counter-balanced by a corresponding decrease in concentration at the point of interest. Tables XVI and XVII contain conversion constants for Ci/g of grass (dry weight) and Ci-day/liter to mean dose in rem. -

The constant, F₁, or any of its three component parts can be evaluated separately for any particular group of interest. Since this will not normally

For nominal inversions (stable conditions) with a 4-m/sec wind speed in summer, the maximum ingestion dose to any age group would be about 400 times a standard adult dose of 340 rem from inhalation of 1 Ci-sec/m³, at a breathing rate of 20 m³/day. From this information, it further appears that action taken (such as removing cows from pasture) within two days after a contaminating event would eliminate over 90 percent of the potential dose.

It should be noted that the use of the relationship of deposition with wind speed allows rapid evaluation of dose. A typical computation involves

$$\text{Dose} = Q \times \frac{(X_u)}{Q} \times F_1 \quad (20)$$

TABLE XVI

CONVERSION CONSTANTS
(μCi-day/liter/μCi/g)[a]

Spring	64
Summer	620
Fall	580
Winter	18

[a]
$$\frac{\int_0^{\infty} Mdt}{I_0} = P \left(\exp [-\lambda_R t_1] \right) \frac{C_k}{\lambda_B \lambda_E}$$

TABLE XVII
DOSE CONVERSION CONSTANTS

<u>Age</u>	<u>rem/μCi-day/liter</u>	<u>rem/μCi/g[a]</u>			
		<u>Spring</u>	<u>Summer</u>	<u>Fall</u>	<u>Winter</u>
0-1	3.1	200	1900	1800	56
2-3	2.8	180	1700	1600	50
4-6	2.8	180	1700	1600	50
7-12	2.8	180	1700	1600	50
13-19	1.4	90	850	800	25
>20	0.9	60	560	520	16

[a] Initial activity on grass (dry weight).

vary from case to case, dose for any given meteorological conditions or for any given distance can be easily estimated.

While some variation may be expected from these mean estimates, these three tables (XV, XVI, XVII) allow reasonably accurate prediction of probable dose if data are gathered at any point in the transfer chain.

6. CONCLUSIONS

It now appears that the major questions regarding the gross transport characteristics of radiiodine can be resolved in the near future. There still are areas of concern in estimating deposition of different chemical and physical forms of iodine, in estimating iodine secretion into milk, and in estimating the retention of iodine on surfaces. The preliminary models presented in this section satisfactorily account for the data observed to date but, being largely of an empirical nature and gathered at a single environment (NRTS), do not allow a satisfactory theoretical accounting for the quantitative transport of iodine under all conditions and for any type of iodine.

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