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ECOLOGICAL SAMPLING AND METEOROLOGICAL CALCULATION OF FALLOUT ON FORESTS NEAR OAK RIDGE

J. S. Olson

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ORNL-3181

Contract No. W-7405-eng-26

HEALTH PHYSICS DIVISION

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Jerry S. Olson

DATE ISSUED

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Jerry S. Olson

ABSTRACT

Spatial patterns of radioactive contamination on forest foliage were measured by gamma spectrometry and are discussed with respect to local vs. world-wide origin of the fallout and implications for ecology, health physics and management of nuclear facilities. In September 1959, I^{131} on dogwood leaf samples varied from over 500 μμc/g dry wt near Oak Ridge National Laboratory stacks to 1-7 μμc/g near the margins of the Oak Ridge Reservation. Stack fallout tended to occur closer to the source than was calculated from hourly wind data by an IBM 610 computer program based on Culkowski's adaptation of the Sutton-Chamberlain theory of atmospheric diffusion and deposition.

Over most of the Reservation, levels of Ru^{106}, Cs^{137}, Zr^{95}-Nb^{95} and Ce^{144} were similar to levels found elsewhere (2-9, 1-3, 2-9 and 10-20 μμc/g respectively) and were presumably controlled by weapons fallout. Higher levels were found in small areas and indicate the need for attention to localized contamination, even though indirect estimates of concentration per unit area did not yet approach levels considered hazardous from the standpoint of health physics.

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1. INTRODUCTION

Hazards of environmental contamination by radioactive materials already dominate much public discussion of the location and operation of nuclear facilities, and will attract even more attention as nuclear energy grows into a significant role in the world's economy. While high-level radioactivity poses an obvious threat to a few individuals, the chance of less intense but more widespread contamination might affect many people. Accidental releases could interfere with continued use of expensive facilities and tend to undermine public confidence in nuclear industry.

During the present stage of technology, existing experience must be used to evaluate problems of environmental contamination. Larger nuclear installations like the Oak Ridge National Laboratory provide opportunities for gaining this experience and comparing facts with theory.

The Health Physics Division of ORNL has therefore carried on not only the legally-required monitoring of the environment (1) but also investigations of the processes governing environmental dispersal of specific isotopes. The Ecology Section has previously studied the biological uptake of long-lived isotopes in small acreages contaminated by liquid wastes (2). The present report is a preliminary study of gamma-emitting isotopes on foliage over the much larger parts of the Oak Ridge Reservation not affected by liquid waste disposal. Sampling of foliage was carried on in conjunction with basic research on the movement of isotopes in forests.

One objective was to measure the amounts of contamination which might hamper forest ecology tracer experiments on the Oak Ridge Reservation (3). The samples of foliage collected for this exploratory purpose were also useful for a second objective--to evaluate the relative contributions of local stack fallout and regional weapons fallout to the natural vegetation. Rapid deposition of the medium-lived fission products (Figure 1) from bomb tests was occurring in 1959 following the moratorium on nuclear testing in late October, 1958. Yet I^{131} and, locally, at least four other nuclides or mother-daughter pairs of nuclides exceeded the levels ascribed to stratospheric fallout (Figure 2).

For I^{131}, which showed the clearest pattern of spatial distribution around ORNL, it was possible to compare the observed geographic distribution of concentrations on foliage samples with calculated values based on meteorological theory (4). These calculations utilized estimates of the amount of iodine release for each of the four weeks preceding the collection of samples (5). Whereas most calculations of stack effluent which are oriented toward single accidental releases are based on some single meteorological situation or average condition, the present calculations of environmental build-up of radioactivity were based on a new procedure of Culkowski (4), using data on hourly variations not only in wind direction but also in wind speed. His formulas were translated into a simple computer program.

While the sampling of natural vegetation was not designed for health physics problems involving iodine contamination of pasture forage, milk and thyroid glands, it is possible to infer indirectly that recent levels of contamination did not exceed currently accepted maximum

permissible concentrations. Nevertheless, the detection of several fission products above levels ascribable to weapons fallout, and the possibilities of accidental release of larger quantities by the same avenues to the environment, confirm the need for further research on the subsequent movement of any radionuclides released.

I want to thank T. H. Handley for initial ash analyses, and especially C. L. Burros and J. H. Moneyhun of the Low Level Radio-analytical Laboratory of the Analytical Chemistry Division for scheduling and carrying out the two main series of spectrometric analyses promptly enough so that relatively short-lived I^{131} could be detected on fresh samples. J. C. Hart and J. W. Youngblood made available tentative estimates of stack releases with the understanding that these estimates are subject to revision. J. A. Gillcrist of the Y-12 IBM Facility kindly helped program the IBM 610 electronic computer for calculating the meteorological pattern of contamination. Walter Culkowski and Frank Gifford, U. S. Weather Bureau, AEC Oak Ridge Operations, provided many helpful suggestions and I am grateful to them and to several members of the Health Physics Division, ORNL, for comments on the manuscript.

2. MATERIALS AND METHODS

Preliminary analyses (Table 1) were made by gamma spectrometry of plant ash from samples of pine needles and twigs from large forest trees 1 mile southeast of ORNL, and from freshly fallen deciduous litter from more distant locations (3, pp. 45-47, 53-54). June 1959 samples (Table 2) were chosen from small trees, mostly growing with open crowns, well exposed to the sky. Fresh foliage was counted in 7 cm plastic dishes, 2 cm deep.

Flowering dogwood (Cornus florida L.) was selected for the main study of geographic dispersal of fallout in September 1959. Dogwood is widely distributed and typically develops full crowns near the ground, suitable for convenient collecting. Its thin leaves have a large ratio of absorptive surface to mass and are flexible. Up to 200 g fresh wt (32 to 92 g dry wt) of leaves were compressed into paper bags in pint plastic containers. Aberrantly low values for I^{131} for a few samples which were partially air-dried prior to sealing suggested the possibility that some iodine may be vaporized unless samples are sealed promptly after collection. Samples were oven dried later for adjustment of counts to a dry weight basis.

Pulses of varying energy from a 3 x 3" Tl-activated NaI crystal were sorted into 100 channels of a 200 channel RIDL pulse-height analyzer. Background counts previously stored in the other 100 channels were subtracted before printing data for spectra. Standard solutions of each detectable isotope were pipetted through layers of cellulose sponge in plastic containers, to provide standards with counting

geometry and density similar to that of the samples. Allowance for Compton scatter and other interference by secondary spectral peaks of gamma radiation was made by using graphs of standard spectra on semi-log paper to estimate the contribution of each isotope in channels outside the energy range of its photopeak. A 1.23 g/cm^2 beryllium absorber was used to minimize contributions of beta radiation.

Radioactivity of parent-daughter pairs of isotopes is expressed in terms of the parent except in the case of Zr^{95}-Nb^{95} for which it represents the sum of both nuclides.

3. ISOTOPES ON TREE FOLIAGE

3.1 Preliminary Samples and Comments

A few major isotopes of fallout were identified on the several kinds of plant material collected in autumn, 1958 (Table 1), and June, 1959 (Table 2), prior to the main sampling in September, 1959 (Tables 3-10).

The 1958 pine needles and twigs and fallen deciduous leaves (Table 1) showed a preponderance of isotopes of medium half-lives and their associated shorter-lived daughter products: Zr^{95} and Nb^{95} (65 and 35 days) and Ce^{144} and Pr^{144} (282 days, 15 minutes). Changes in count rates suggest that Ce^{144} (32 days) also may have been prominent initially, but a lapse of several months prior to gamma spectrometry prevented estimation of this isotope and the shorter-lived Ba^{140} and La^{140} (13 days, 40 hours) from the spectra. These isotopes were typical of fresh fallout from weapons tests (8-12) and could not necessarily be attributed to laboratory sources, even though the levels of radioactivity seemed higher than were expected from stratospheric fallout. Failure to detect Ru^{106} and Rh^{106} (365 days, 30 seconds) might be explained by volatilization during ashing. The level of Cs^{137} was low relative to that of several other isotopes, and could not be detected in their presence.

In June, 1959, collections of fresh foliage were taken at several locations (1 mile southwest, 1 mile north, 4 miles northeast, and 30 miles east of ORNL). Samples were assayed as promptly as possible so that I^{131} (8 day half-life) could be detected.

Table 1. Fallout Contamination on Plant Materials, Autumn, 1958.

Micromicrocuries per gram oven dry leaves ($\mu\mu c/g$)[2]

Sample	Zr^{95}-Nb^{95}	Ce^{144}	Cs^{137}
Shortleaf pine (October 1)			
New needles	50	28	5
Old needles	< 160	78	17
Branches	< 50	43	13
Deciduous leaves (November 10)			
Mulberry	180	61	7
Sugar maple 1	180	45	< 2
Sugar maple 2	155	57	< 3
Sugar maple 3	165	18	< 4
Shumard oak	400	60	< 2
Scarlet oak	148	5	< 7
Beech	148	60	< 5
Sycamore	170	49	< 2

[a]These results are based on ORNL-2806, Table 18, corrected for radioactive decay between collection and analysis.

Table 2. Fallout Contamination of Fresh Foliage, June 1959 (μμc/g)

Sample	Zr^{95}-Nb^{95}	Ru^{106}	I^{131}
Shortleaf pine			
SW 1[a]	68;16[b]	28;137[b]	106;121[b]
N 1	82;14	52;---	52;---
NE 4	72;---	---;---	---;---
E 30	188;36	117;21	---;---
Dogwood			
SW 1	23	141	211
N 1	28	90	13
NE 4	15	16	--
E 30	36	21	--
Red maple			
SW 1	43	--	232
N 1	49	22	7
NE 4	22	--	22
E 30	27	19	11
Red mulberry			
SW 1	43	71	545
NE 4	17	--	9
Sycamore			
SW 1	20	32	391
N 1	21	6	6
Post oak			
SW 1	16	13	170
N 1	12	15	3
Redbud			
SW 1	9	15	32
NE 4	11	15	10

[a]Distance from ORNL, in four compass directions, in miles.

[b]For pine needles, left figure is for old needles, and right figure is for new needles. Blanks represent values below a variable limit of detection.

Table 2 shows a difference in geographic pattern among three nuclides. For Zr^{95}-Nb^{95} there is no consistent difference between locations near or far from Oak Ridge. For Ru^{106}, samples near ORNL (particularly 1 mile SW) tend to be higher than others, but there are exceptions. For I^{131} the samples 1 mile SW of ORNL show far higher activity than other locations, and there is only one uncertain case in which I^{131} might be recognizable 30 miles east or ORNL. Wind directions from the NE or ENE for June 8 and 9, preceding the collection, suggested the ORNL stacks as a plausible source, to be investigated in the later sampling.

The great variability in radioactivity between kinds of leaves collected at a single site (like SW 1 on Table 2) was represented by values of I^{131} ranging from only 32 μμc/g for smooth redbud leaves to 545 μμc/g for hairy, glandular mulberry leaves. Therefore the September 1959 sampling concentrated on one species of leaf in order that geographic patterns would not be obscured.

3.2 Sampling of Dogwood Leaves in September, 1959

Local dispersal of radionuclide was estimated (Tables 3-10) from dogwood tree foliage collections radiating in four lines from ORNL in the direction of the two dominant winds (NE and SW) and at right angles (NW and SE). Most samples were distributed in a regular network at 1, 2 and 4 miles from the stack. The time required for collection and analyses prevented filling in many intermediate points, but an additional sample was taken 4 miles north and others were taken at 8, 16 and 32 miles NE of the Laboratory, in the direction where greatest horizontal

movement was expected. Several samples were taken within the ORNL grounds.

Agreement of independent duplicate analyses for three of the most radioactive samples taken at ORNL is shown on Table 3. Ranges of only 6-9 μμc/g for I^{131} imply a coefficient of variation of only 2.4 to 1.1 per cent. Four or possibly five other gamma emitters could be estimated by graphical "stripping" of their spectra, but the lower counting rates and overlap of the spectra led to less certain estimates. Except for a single large difference between duplicates (89 vs. 138 μμc/g for Ru^{106}), differences were less than 7 μμc/g. Coefficients of variation of course were larger for samples with low means. Dashed lines on the table indicate that a given isotope was below limits of detection, but the lower limit of detection cannot be specified because it depended on the sample weight and on the other isotopes present. Nevertheless, the differences shown on Tables 3-10 are great enough that the main pattern of isotope distribution was not obscured by analytical limitations.

Maximum values for I^{131}, Ru^{106} and Cs^{137} (583, 113, 22 μμc/g, respectively) were found 0.15 mile northeast of the Graphite Reactor and the large stack of the Isotope Processing Area (spectrum of this area in Figure 2). The maximum for Zr^{95}-Nb^{95} (68 μμc/g) was found 0.08 mile west of the stacks, south of building 3019 (Table 4A). In most samples the photopeak of Ce^{144} was obscured by scattered gamma radiation from all the other isotopes so that the estimates are only tentative: approximately 100 to 200 μμc/g near ORNL, as compared with 10-20 elsewhere. The identification of Co^{60} is uncertain because of the low level of activity (generally lower than 1 μμc/g; perhaps locally 2.5 to 5 μμc/g).

3.3 Distribution as a Function of Distance and Direction

Comparison of Tables 4A, B, and C shows little difference for Zr^{95}-Nb^{95} between samples 0.5 to 1 mile from the Laboratory and those of distant parts of the Oak Ridge Controlled Area or outside this area. The range of values found in late September (4-9 μμc/g) is below values of 15-36 μμc/g for dogwood recorded in June (Tables 2, 8), and far below values of 150-400 μμc/g found on fallen leaves of other species in November of 1958 (Table 1).

Tentative estimates of Cs^{137} and Ru^{106} (Tables 6 and 7) are from 2-3 μμc/g and from 2-9 μμc/g, respectively, at points more than 1 or 2 miles from the Laboratory. Slightly higher values (4.5 and 30 μμc/g) at station SE 1 above the White Oak Lake bed suggest possible contamination by dust or organisms moving from the lake bed or from the liquid waste pits across the lake bed, or by deposition of contaminants from the HRE stack (Figure 4).

While high values for Cs^{137} are restricted to station SE 1 and to the grounds of the Laboratory proper, Ru^{106} values also slightly exceed those of the general surroundings for 1 mile northeast (12 μμc/g) and 2 miles southwest (17 μμc/g), thus extending furthest from the stack in the directions of the two dominant winds.

Confirming earlier analyses, I^{131} contributed more radioactivity than any other gamma emitter for 1 to 4 miles in every direction except the southeast (Tables 3-6, Figure 4a). Furthest extension of detectable I^{131} was to the northeast (5 μμc/g, 8 miles), next was to the southwest (7 μμc/g, 4 miles), next northwest (5 μμc/g, 4 miles), and least to the southeast (1 μμc/g barely detectable, 4 miles). These stations are all

Table 3. Duplicate analyses of gamma emitting isotopes in foliage of flowering dogwood trees, NE of ORNL, September 1959.

Direction and miles from Bldg. 3001	Gamma-emitting isotopes (μμc/g dry wt of leaves)[a]					
	I^{131}	Ru^{106}	Cs^{137}	Zr^{95}-Nb^{95}	Ce^{144}[b]	Co^{60}[b]
NE 0.15	578	89	20	28	209	3
	587	138	25	25	216	2
NE 0.5	172	17	8	8	-	-
	178	18	6	7	33	-
NE 1.0	185	11	3	6	-	-
	191	14	6	6	-	1

Table 4. Summary of Additional Analyses of Dogwood Foliage Around ORNL.

Direction and miles from Bldg. 3001	Gamma-emitting isotopes (μμc/g dry wt of leaves)[a]					
	I^{131}	Ru^{106}	Cs^{137}	Zr^{95}-Nb^{95}	Ce^{144}[b]	Co^{60}[b]
A. Additional samples in or near ORNL						
SW 0.08	194	15	16	68	214	5
NW 0.25	102	27	-	14	111	-
SW 0.35	23	-	3	7	-	-
B. Oak Ridge Reservation						
1 SW,NW,SE	28-59	7-30	3-4.5	5-6.5	~ 18	-
2 (4 dir.)	7-27	6-16	1-3.6	6.6-7.8	~ 12	-
4 (4 dir.)	1-24	2-9	1-1.3	5.1-5.7	13-17	0.4-0.7
8 NE	5	9	-	3.7	-	-
C. NE from Oak Ridge Reservation						
16 NE	-	8	2	6.4	-	-
32 NE	-	6	3	9.0	~ 20	-

[a]Lower limits of detection vary due to counting errors of all the isotopes in mixture. Activities of parent-daughter paris of isotopes expressed in terms of parent only, except for Zr^{95}-Nb^{95}.

[b]All estimates of Ce^{144} and Co^{60} are uncertain.

near the edge of the Oak Ridge Reservation and levels are so low that it is not surprising that I^{131} was not detectable at the two stations northeast of the Reservation. Between the extremes of the maximum value near ORNL and undetectable levels outside the Reservation, there was a fairly regular progression of I^{131} concentration decreasing with distance from ORNL stacks (Table 5, Figure 4).

4. METEOROLOGICAL INTERPRETATION OF DISTRIBUTION OF I^{131}

The spatial pattern of I^{131} as measured on the foliage (Figure 4a) shows a striking resemblance to the generalized annual deposition pattern calculated by Culkowski (Figure 3). Of course, the 8.1 day half-life of I^{131} implies that annual wind distribution would not be so relevant in explaining the observed contamination pattern as would the wind distribution of the last four weeks immediately preceding collection. Hence data for separate wind roses were tabulated for all stations in the area for each of these weeks. Figure 4b summarizes in map form the values calculated for these four weeks, with weighting by Youngblood's provisional estimate of the amount of I^{131} emitted from the stacks during each week. The similarities and discrepancies between observed and calculated patterns will be discussed after a review of the calculations.

4.1 Calculations of Environmental Deposition of Atmospheric Contamination

Culkowski's calculation of average air concentrations (4) develops directly from common formulations of Sutton's turbulent atmospheric diffusion theory and Chamberlain's theory of aerosol deposition (14,15). This theory can be developed from the assumption that a plume of smoke emanating from a stack would expand so that the density of radioactive particles is approximated by a Gaussian probability distribution having widths measured in terms of standard deviations σ_y and σ_z in the horizontal and vertical directions from the mean axis of the smoke plume.

$$\chi = \frac{Q}{2\pi\sigma_y\sigma_z\bar{u}} e^{-\frac{1}{2}\left(\frac{y^2}{\sigma_y^2} + \frac{z^2}{\sigma_z^2}\right)} \tag{1}$$

where

χ = concentration per cubic meter,

Q = output per second,

$\bar{u}$ = mean wind velocity,

y and z = horizontal and vertical distances from the cloud center,

σ_y^2 and σ_z^2 = corresponding variances of the particle distribution.

Recent meteorological work is beginning to evaluate small-scale and large-scale components of these variances for different weather conditions as a function of x, the distance downwind (16). Sutton's theory of turbulent dispersion implies that variances increase slower than x^2:

$$\sigma_y^2 = \frac{1}{2} C_y^2 x^{2-n} \quad \text{and} \quad \sigma_z^2 = \frac{1}{2} C_z^2 x^{2-n} \tag{2}$$

Here n is a measure of the vertical thermal stability of the air and the terms C^2 are coefficients of dispersion (14). Substitution of (2) in (1) results in the Sutton expression for a source of particles at ground level. (A factor of 2 enters Sutton's expression from the assumption that particles cannot be adsorbed in the ground, except insofar as deposition is allowed for in a later stage of calculation.)

The concentration at ground level for a source elevated at height h (e.g., stack height + height of plume rise) is given by:

$$\chi = \frac{2Q_o}{\pi C^2 x^{2-n} \bar{u}} e^{-\left(\frac{h^2 + y^2}{C^2 x^{2-n}}\right)} \tag{3}$$

with the simplification of neglecting the difference between horizontal and vertical coefficients of diffusion. Culkowski (4) integrated equation (3) with respect to y to get the total deposition at a given distance x due to winds from a given compass direction (22.5^o sector of a circle). He then assumed for simplicity that all this material was distributed evenly in that sector toward which the wind blew, i.e., in 1/16 of the total circumference $2\pi x$ of a circle x meters away from the source. The <u>long-time average deposition for the hours when wind was blowing in this sector</u> is the total deposition divided by this circumference, or:

$$\frac{16}{2\pi x} \int_{-\infty}^{+\infty} \chi dy = \frac{16}{2\pi x} \int_{-\infty}^{+\infty} \frac{2Q_o}{\pi C^2 x^{2-n} \bar{u}} e^{-\left(\frac{h^2 + y^2}{C^2 x^{2-n}}\right)} dy$$

$$= Q_o \frac{16}{\pi^{\frac{3}{2}} C x^{\frac{4-n}{2}} \bar{u}} e^{\left(\frac{-h^2}{C^2 x^{2-n}}\right)}$$

$$= Q_o B_i. \tag{4}$$

Here B_i expresses the concentration as a fraction of the stack emission rate for that fraction of time when the wind was blowing in the specified direction with a specified velocity.

Several earlier workers had made calculations of expected long-term average air concentrations using frequencies of winds for each direction

and mean wind speed for each direction. Culkowski went a step further in using the mean wind speed for each of R (=6) speed groups for each sector, which are recorded in the Oak Ridge Weather Bureau telemeter records, and the fraction F_i of total time the wind is represented in each class. The average air concentration at distance x in direction D is then:

$$\chi_D = \sum_{i=1}^{R} F_i B_i Q_o. \tag{5}$$

In the present application Culkowski's computational procedure was programmed on punched tape for a small electronic computer, the IBM 610, so that partial calculations could be repeated for separate weeks and be multiplied by estimated emission rates Q_o for these weeks.

All these calculations are preliminary to the calculation of deposition rates, following Chamberlain's method (15,17,18) of specifying a certain "velocity of deposition" which removes a fraction of the material from the cubic meter above the ground onto the underlying square meter of ground surface or of foliage growing above the ground:

$$V_g = \frac{\text{deposition rate per } m^2 \text{ of ground}}{\text{concentration per } m^3 \text{ of air}} \tag{6}$$

The value of V_g is affected by terminal or settling velocity in the case of very large particles, by the diffusion of gases in the case of unadsorbed I^{131}, and by the impaction of sub-micron aerosols on which iodine tends to be adsorbed (19). It is also affected by the characteristics of the surface on which deposition is occurring.

As a result of deposition, there is a depletion of material left

suspended in the cloud. The fraction Q/Q_o, left in suspension at any distance x was applied as a correction factor in adjusting the downwind, air concentration, and hence further deposition (20,4).

In the present application, deposition ϕ was calculated (first relative to unit effluent, and then relative to estimated iodine effluent, corrected by a factor d for radioactive decay) immediately following the uncorrected air concentration for each of 16 wind directions in turn, and for each of several distances.

$$\phi = \chi_D V_g \frac{Q}{Q_o} d \qquad (7)$$

Calculations were made separately for day (taking n = 0.25) and night (n = 0.33) and combined in Figure 4b. Coefficients of turbulence were determined by nomogram (14).

Of the wind stations available, that for the Tower Shielding Facility was recommended by Culkowski (21) as most likely to approximate the winds at the height of the stack plus rise of plume through buoyancy, h, approximately 100 meters. Present calculations were made for winds from the same station and same choice of h = 100, V_g = 0.04 meters/sec and the graph of Q/Q_o which had previously been used by Culkowski (4), so that results (Figure 4b) could be compared with his (Figure 3). The value of V_g = 0.04 is near the upper limit of Chamberlain's original estimates (.018 to .042, mean .025) based on the uptake of I^{131} by grass at Harwell (17,18). Hence, the present choice would tend to predict slightly higher deposition near the source and slightly lower deposition at a distance than Harwell work. Alternative choices of several other parameters would give results differing in some details but similar in

general to those which follow, concerning the pattern of wind dispersal near Oak Ridge.

4.2 Distant Deposition

The hours when wind was not calm should result in most stack fallout on areas one to several miles from the Laboratory because the plume of stack effluent is carried away before it spreads enough to reach the ground. Comparison of Figure 4a with predicted contours in Figure 4b shows a gross similarity between observed and calculated deposition pattern for such distances. Greatest total deposition appears to be distributed in a broad area from ENE to NNW directions, as expected from the high frequency of WSW to SSE winds (13). High deposition SW of the Laboratory is expected from frequent and occasionally strong northeasterlies. The relatively lower deposition to the W and NW and, more especially, to S through ESE clearly reflects the low frequency and velocity of winds that blew in these directions, at right angles to the Ridge and Valley topography (13). Obviously the limited number of field points prevented comparison with many details of the calculated wind pattern.

The agreement in general pattern of predicted deposition and observed contamination seems fair, considering (1) the short period (4 weeks) for averaging hourly wind values into a smoothed pattern, (2) the possibility that iodine releases are irregularly timed during the individual weeks to which the filter estimates apply, and (3) the simplified nature of the mathematical model and choice of parameters. Discrepancies between prediction and observation do exist and these might be explained in several ways.

Figure 5 shows a cross-section along the NE-SW axis separating several theoretical contributions to total deposition: (a) the lowest curve shows the contributions from night winds; their association with stable air and low coefficients of diffusion account for low deposition rates, especially within 3 miles of the source. (b) daytime winds should contribute more deposition within 0.5 to 3 miles of the source, because greater turbulence disperses the plume and carries parts of it near the ground more promptly. (c) high deposition near the source might occur during calm weather or due to special causes noted below.

During September, 1959, a high frequency of light night breezes from the southwest should have led to a peak deposition 4 miles NE, explaining the reading of 24 μμc/g. Northeasterly night winds tended to be stronger and should have led to a relatively more distant transportation to the SW. A high frequency of NE daytime winds of medium velocity (5 to 20 mph) led one to expect slightly greater deposition SW of the source than to the NE. Since observed deposition appeared to be greater to the NE, this discrepancy needs an explanation.

The distribution of I^{131} in several directions shows that it was not all released in one or two episodes, but was released at many different times during many wind conditions. Yet it is possible that releases of I^{131} did not occur frequently enough to represent faithfully the average distribution of winds for the month prior to sampling. One or more major releases which happened to occur when the wind was from the SW might have been enough to shift the balance in favor of more deposition in the NE quadrant than would be expected from the actual average September wind distribution. The average annual wind distribution

(Figure 3) would tend to give stronger representation of SW winds (NE deposition) and lesser representation of S to SSE winds (N to NNW deposition) than the winds which happened to be observed in September, 1959. The observed deposition pattern thus happens to resemble more closely the pattern to be expected for the whole year than it does the pattern based on the month when data were actually taken.

Accelerated deposition rainout due to washing by rain might help to explain the extra deposition to the northeast, if this occurred during a period of southwesterly wind (15,19). Another special condition that could have enhanced deposition during such a period is the adsorption of I^{131} on particles that were coarse enough to settle out more rapidly than theory predicts (i.e., leave fewer particles suspended for transport to great distances).

4.3 Anomalous Deposition Near the Stacks

The tendency for highest concentrations of I^{131} and other isotopes to occur in the samples <u>near</u> the stacks (Figure 4a, Tables 4-10) does not fit the simple meteorological model which assumes that a plume of effluent must travel many hundred meters before it finally expands enough to intersect the ground. The above-mentioned possibilities of (a) rainout, and (b) fallout of iodine adsorbed on large particles, might help explain this anomaly. Hypothesis (c) is that of morning "fumigation" of the area below the stack by downward movement of air that had been trapped aloft by a night-time inversion (13,14).

All three mechanisms of close-in deposition would be most effective during calm hours (winds less than 1 mph) when dispersal and dilution

by wind would be least. There is no completely satisfactory method of predicting the distribution of effluent under such calm conditions. Culkowski (4, equ. 8) used the simple approximation of assuming all effluent during calm hours was deposited uniformly within a circle.

An alternative model considered here avoids the assumption of a sharp boundary by assuming a bivariate Gaussian distribution, although the choice of standard deviation (or deviations) is still arbitrary. For trial, the standard deviation was here taken as 1000 meters, a distance which would be traversed in 41.6 minutes by a particle falling 100 meters at a speed of 0.04 m/sec during a wind blowing 1.3 m/sec (0.9 miles per hour). Many particles would not spread this far, especially if the wind was even slower than this threshhold of measurement or was truly calm. Some particles might move further in light winds if their settling was notably slower, but the density per unit area would be diminished drastically 2 or 3 miles from the source. Most deposition would occur within the cross-hatched circle of Figure 4b, 1 mile around the plants, by either Culkowski's or the present model.

The uppermost (dashed) curve in Figure 5 is based on the hypothesis that <u>all</u> effluent during a given number of hours of calm was indeed deposited in the area indicated by the Gaussian probability distribution. If only 1/2 or 1/4 of the material was so deposited and the rest carried far away, there would still be a conspicuous peak of deposition near the source (indicated by the light solid lines in Figure 5), which would be greater than the ground concentration expected at more remote points due to deposition during non-calm hours. If such a substantial fraction of

the effluent released during "calm" hours was not deposited locally, it would be necessary to assume that a small fraction of the effluent released during other hours was deposited more rapidly than the Sutton-Chamberlain deposition theory predict, in order to account for peak concentrations observed near the stacks.

5. ECOLOGICAL PROBLEMS

5.1 Inferences on Iodine Deposition per Unit Area

A comparison of observed contamination of I^{131} per unit weight of dogwood leaves and predicted contamination per unit area of ground indicates the general agreement of pattern with that predicted by meteorological theory. However, further comment on the magnitude of certain discrepancies requires an estimate of tree foliage per unit area, so that I^{131} observations can be converted to a unit-area basis. Data for such estimates are being obtained as part of the forest studies on the Oak Ridge Reservation (3), but the present comment will be restricted to a nominal value of 400 g/m^2 (oven dry) which is typical (or minimal) for many forests in the southeastern United States.

On Figure 4a, the contour of 25 $\mu\mu c/g$ would correspond with a total iodine burden of 10^{-2} $\mu c/m^2$ using this conversion, and the other four contours plotted have similar correspondence with the contours of calculated contamination given in Figure 4b. Comparison shows that the observed area within most contours is smaller than the calculated area.

In Figure 5, the scale of $\mu\mu c/g$ on the right axis was placed so that the approximate equivalent $\mu c/m^2$ could be read directly on the left axis, on the same scale as the predicted curves based on the meteorological model. The observed deposition tends to be lower than calculated values several miles from the stacks. However, the most radioactive sample (0.15 miles NE of stacks) is equivalent to 0.23 $\mu c/m^2$, which is almost identical with the calculated value under the extreme assumption that all the I^{131} emitted during calm weather was deposited

in a small area (mostly within 1 mile radius) around the Laboratory.

One possible interpretation is that certain mechanisms enhance deposition near the stacks and thereby deplete the amount of I^{131} that remains in suspension and which otherwise could be deposited at greater distances. However, such a conclusion is not yet warranted, for several reasons. Better estimates of I^{131} per unit area are obviously needed in order to allow for that quantity which fell past or was washed off the foliage or was translocated into the plants. Such estimates would probably lead to the conclusion that true deposition is <u>at least</u> as great as the values calculated for expected curves in the areas remote from the stacks.

Allowance for deposition unaccounted for by foliage samples would also increase the estimated concentrations near the stacks. Here the estimated concentrations already approach or exceed predictions based on meteorological assumptions of almost complete deposition near the stacks during calm weather. However, not all of the I^{131} vented during calm hours could be expected to settle near the stacks. If the 25 per cent line of Figure 5, for example, applied to the deposition during calm hours, then all of the calculated levels would fall below the estimates actually made from near-source observations. One way of explaining low calculated values is the interpretation that the original estimates of iodine released from the stacks may have been low. Technical problems of stack sampling are being studied by Youngblood in order to evaluate this possibility and to provide the more complete records of I^{131} release through time which would be needed for more exact calculations.

5.2 Ecological Movement of Iodine

Selders and Hungate (22) showed that bean and geranium leaves absorb gaseous I^{131} and stable I^{127} in almost direct proportion to atmospheric concentration over a millionfold range of air concentration (up to the equivalent of 10 μc/cc, which is 10^{-7} mg of iodine)--many orders of magnitude above the concentrations in air at ground level at Oak Ridge. Bean and geranium differed by a factor of almost two, just as the tree leaves on Table 2 differed severalfold in their iodine activity because of differences such as surface hairs, glands, thickness, and physiological behavior. In geranium, 35-40 per cent of the I^{131} in the leaf reached the mesophyll in 1 hour, but only 2 per cent was translocated into other parts of the plants. It is not yet clear whether translocation could be expected to continue at this rate, so that most of the tissues of any one species would become contaminated within a few days after exposure to I^{131}. However, differences between plant tissues as well as differences between species in rates of intake (Table 2) could lead to significant differences in the contamination levels attained when income was balanced by decay or by other losses.

Animals eating the foliage that was directly exposed to fallout would ingest the large fraction of I^{131} that was adsorbed on the plant epidermis, as well as that which had already been incorporated inside the plants. Experience with biological monitoring at Hanford, Washington (23) and Arco, Idaho (24) indicates that rabbits quickly reach a predictable stable level of internal contamination in areas of persistent iodine activity and quickly respond to unusual increases in fallout.

Comparable studies have not yet been made for plants and animals under conditions of the eastern United States. Ecological sampling at ORNL has been concerned with longer-lived isotopes, especially in waste disposal areas more highly contaminated by liquid radioactive wastes. The present sampling indicates that other areas within a mile or less surrounding ORNL would be suitable for measuring the natural ecological redistribution of local fallout. Not only I^{131} but several of the other fission products occur at levels more readily measurable in a variety of components of the ecological system than is the case for areas affected only by weapons fallout.

5.3 Possibility of Interference with Ecological Tracer Experiments

For some environmental tagging programs it is necessary to follow a known amount of experimentally introduced radioactive tracer(s) through different parts of the natural ecological cycle, as well as to observe the end-product of redistribution of isotopes which may have been introduced irregularly at various times through the past. The original concern that locally-produced aerial contamination might interfere with such experiments has been diminished by the present study.

Convenience in sample processing and counting makes it desirable that experimentally introduced tracers be introduced at levels several orders of magnitude above background levels. Stack fallout would have little effect on measuring directly tagged material. However, for experiments in which it is particularly desirable to detect a tracer isotope after it has been diluted in large amounts of organic material or soil, it would seem desirable to avoid portions of Bethel Valley within 1 or 2 miles of ORNL.

Even further away, samples may have to be inspected by gamma spectrometry in order to see whether I^{131} or other aerial contamination is detectable. Detailed spectrometric analyses rather than integral counts or single-channel differential counts of gamma activity may be needed in order to avoid contributions from I^{131}. Except in tracer experiments with short-lived isotopes like K^{42}, one might let I^{131} activity decay by holding the samples several weeks before assay of other isotopes or volatize it during processing procedure.

6. IMPLICATIONS OF ATMOSPHERIC CONTAMINATION FOR HEALTH PHYSICS

6.1 The Radioiodine Problem

Radioactive iodine provides a classic case of increasing concentration at each stage of the food-chain transfer, from air to forage to cows' milk to man (in particular to the thyroid gland). Standards for environmental contamination by I^{131} have become more exacting in recent years, and therefore invite some kinds of comparison with present data, even though the present sampling was not oriented toward problems of human consumption.

In 1946 Parker (26) evaluated the maximum permissible concentration on vegetation with special reference to the hazard to the cow instead of man. Assuming (a) that a dose to the thyroid equivalent to 1 r/day could be tolerated by the cow, and (b) that a cow eats 10 lb of forage per day, 2×10^{-4} $\mu c/g$ was estimated to be the MPC for forage. In 1952, Chamberlain and Chadwick (17,18) concluded that this consumption figure was too low. This criticism is supported by consumption estimates of 18.6-22.3 lbs dry weight for a 1000 lb dairy cow under conditions of favorable management (27). Accordingly, Chamberlain recommended 1×10^{-4} $\mu c/g$ for the MPC; assuming a standing crop or biomass of only 200 g/m^2 for a "rather thin covering of grass", grazed closely, this implies a concentration of 2 $\mu c/m^2$ as an MPC on a unit area basis.

The accidental release of fission products from a graphite reactor at Windscale in October 1957 provides the most notable case history involving hazard from I^{131}. Because medical evidence indicated that cancer of the thyroid had sometimes followed radiation doses of 200 rad to

thyroids of children, emergency standards were chosen with a safety factor to restrict the dose to 20 rad. Assuming the thyroid gland weighs 5 g and retains 45 per cent of the ingested dose of I^{131}, with an effective half-life of 8 days, milk consumption of 1 liter per day was calculated to attain the 20 rad limit if contamination reached 0.15 μc per liter (28). Other figures available at the time of the incident, assuming a 1.5 g thyroid for a small child and retention of 20 per cent of the ingested dose, led to setting a limit of 0.1 μc/l, the level assumed for control. This concentration was exceeded and milk was confiscated over most of the area of 20 x 40 km in northwestern England, where deposition exceeded 1 $\mu c/m^2$. This limit thus constituted another restriction by a factor of two in the level considered permissible.

In considering selection of sites for reactors still further restriction was implied by Farmer and Fletcher (29) who considered limits for emergencies of 0.065 μc/liter of milk and approximately 0.5 $\mu c/m^2$ on forage. While Chamberlain's original recommended limit was 1×10^{-12} μc/cc of air, Chamberlain's mode of calculation and the stricter criteria would suggest a maximum permissible air concentration of only 0.25×10^{-12} μc/cc, or 0.25 $\mu\mu c/m^3$, for air over dairy pasture.

Different assumptions about the deposition velocity parameter for various kinds of suspensions and absorbing surfaces might modify this indirect estimation of maximum permissible air concentration considerably, but would nevertheless lead to limits with respect to food contamination which are stricter than those required with respect to inhalation exposure. The MPC for air recommended by the International

Commission on Radiological Protection (30) is 9×10^{-9} μc/cc for occupational exposure during a 40 hour week, and 9×10^{-10} μc/cc for neighboring populations.

Until systematic comparisons are made between forest foliage and pasture forage, the present data cannot be transformed into the same terms as these health physics standards. If both kinds of foliage were similar in uptake, the highest measured value (584 μμc/g) might approximate a deposition of 0.23 $\mu c/m^2$, which would be almost half the maximum permissible concentration. However, the nearest cattle pasture or other vegetation which might enter the food-chain leading to man is several miles away where iodine levels are 1 or 2 orders of magnitude below the maximum. These comparisons suggested that the Oak Ridge National Laboratory has not been approaching an emergency situation with respect to aerial contamination by I^{131}. Nevertheless, additional air-cleaning equipment is being installed in order to keep routine release as far below MPC values as is feasible, and to provide extra precautions against accidental releases (31).

Culkowski's method which was used here for calculating long-term build-up may provide a useful method of comparing the relative expectation of contamination at any distance along each of sixteen 22.5° arcs. But, in case of a single accidental release, this method (equ. 4) would underestimate the air concentration and deposition under that portion of the arc which happened to be crossed by the pathway of the radioactive cloud. For this case equation 3 could be used directly (13,14). The discrepancy between the chronic contamination case and the case of a single accident becomes greater with distance, because the width of the plume increases

only as some fractional power of x (equ. 2), instead of the first power of distance. This discrepancy is greatest under inversion conditions represented by high values of the atmospheric stability parameter n, and n could be greater than the approximate average night-time value of 0.33 which as assumed in the present calculations. Barring rainout and gravitational fallout, night-time conditions would lead to less deposition near the source and relatively more at a distance than day-time conditions (Figure 5), as a result of slower dispersal, particularly in the vertical dimension.

Whereas slow dispersion tends to postpone the time of contact of contaminated air with ground level, it means that the cloud has had less opportunity for dilution than it would in a less stable, more turbulent atmosphere. If the relatively undiluted plume of radioactive material should reach a particular spot of ground, as during the turnover of air due to ground heating in early morning, after drifting for many miles during the night, then the local area where this happened could have higher activity than would be expected for an area chosen at random at a given distance and general direction from the source (13,19). The deposition pattern on the Hanford Reservation illustrates distant dispersal under inversion conditions (32).

Health physics requirements emphasize consideration of these extreme, or pessimistic, situations in evaluating hazards to people or areas of habitation, particularly for evaluating accidents or other unusual situations. On the other hand, for chronic environmental contamination under variable conditions, calculations such as those of Figures 3 and 4b, deserve wider application. Problems of I^{131} may involve

either chronic or high short-term releases, or some mixture of both (32).

6.2 Hazards from other Radioisotopes

It was noted that Ru^{106} showed a pattern suggesting traces of wind dispersal up to 2 miles from ORNL, over a period of several months prior to the notable stack release of November, 1959. This November release (33) was most interesting as an example of very rapid deposition of coarse particles within a few stack-heights of distance from the source. Such behavior of coarse particulates minimizes the danger to distant populated areas. However, prompt deposition poses another risk of potential occupational hazard in a small area where there is minimum opportunity for dilution and where there exists high investment in facilities and where costs of decontamination are great.

The Cs^{137} was even more localized than Ru^{106}. This localization suggests the possibility of adsorption on particulates. While presently observed values were low (Table 7) the possibility of accidental release of this long-lived Cs^{137} warrants research on its movement in the environment.

Unlike the other nuclides, Zr^{95}-Nb^{95} appeared to be highest south of Bldg. 3019, to the SW instead of NE of the isotope stacks. Concentrations were moderate in September 1959, by comparison with 1958 levels from weapons fallout. Nevertheless, further attention may be deserved in order to define local patterns and changes in these fission products.

6.3 Land Management Around Nuclear Sites

The routine release of low levels of radioactive contamination, or possible accidental release of higher levels, raises long-range questions of land management around nuclear installations. In a humid, moderately populated area like that surrounding Oak Ridge, the problem of preventing run off of fallout materials into the surface water supplies of the region has been emphasized (34). Because soils, litter and native vegetation tend to hold isotopes and prevent runoff into surface drainage, efforts to enhance this natural protection deserve to be considered as part of the preparation of reactor sites and operation of reactor areas.

The much larger buffer areas surrounding a reactor site will intercept more fallout and will contribute more runoff than the immediate site. For example, because of the history of the Oak Ridge Reservation, many farms were abandoned suddenly in 1942 with no ground cover left on the fields. Extensive gullying and removal of topsoil occurred in many areas within the few years following, and gullying continues today in some places. In other old fields, gullying evidently contributed to abandonment of agriculture long before 1942. Secondary ecological succession normally involves invasion of these fields by Virginia pine and shortleaf pine, cedar and pioneer hardwoods. Concurrent development of natural litter and humus have healed the scars on the land to varying degrees. More study is needed to determine how the vegetation cover and the soils of the Reservation control the runoff to surface waters.

In general, long range policy in the management of Reservations around nuclear facilities will need to stress the importance of protection

of the landscape, and the utilization of the protected lands for research. Certain kinds of productive land use for forestry or agriculture are presumably consistent with these main functions of the land. However, these have to be evaluated individually for (1) their impact on the protective cover of the land, and (2) the risk of chronic or accidental contamination for these economic activities. Even if certain land uses are finally deemed consistent with chronic build-up of environmental radioactivity, the demonstrated possibilities of an unusually high release must be considered. The gradual accumulation of experience will have to guide the decision between the cautious course of reserving and regulating the use of large areas around nuclear sites, vs. the alternative of reserving only smaller areas but accepting greater risk of someday having to impose emergency restrictions on the public activities outside these Reservations.

7. CONCLUSIONS AND RECOMMENDATIONS

Tree foliage can provide a convenient natural collector for estimating spatial patterns of fallout contamination if precautions are taken to standardize on species and tree-crown position of samples. The mixtures of several gamma-emitting isotopes require analysis by gamma spectrometry. Results of Oak Ridge indicate different spatial patterns of contamination for I^{131} and 4 or 5 other isotopes:

1. Concentration of Zr^{95}-Nb^{95} on fallen litter reached 150-400 μμc/g after October 1958 nuclear weapons tests. In 1959, levels in living foliage were decreasing over most of the Oak Ridge Reservation and elsewhere (to 6-9 μμc/g in dogwood leaves in September 1959). Local high values (27 to 68 μμc/g within 0.15 mile of ORNL) were lower than weapons fallout levels of a year earlier.

2. Similarly, Ce^{144} showed highest abundance in late 1958 and early 1959, a notable decrease (~ 20 μμc/g) in most areas by September, with very localized high values (~ 200 μμc/g).

3. Samples in which Cs^{137} was detectable showed 2 to 4 μμc/g outside as well as most areas inside the Oak Ridge Reservation, except for very local values (16 to 20 μμc/g near ORNL).

4. No indication of Co^{60} was present in most samples, but a few uncertain estimates (2.7 to 5 μμc/g) for samples near ORNL warrant further investigation.

5. A range of 2 to 9 μμc/g for Ru^{106} suggests origin by weapons fallout, except for local values (12 to 100 μμc/g in September 1959) that might be due to small stack releases affecting areas 1 or 2 miles

NE and SW of ORNL.

6. As at many other reactor sites, I^{131} (up to 583 μμc/g 0.15 mi NE of ORNL in September 1959) was the main radionuclide attributable to local sources. Levels decreased with distance from ORNL almost to limits of detection around the perimeter of the Reservation (1 μμc/g 4 mi SE, 7 μμc/g 4 mi SW, 5 μμc/g 4 mi NW and 5 μμc/g 8 mi NE) and contributed only a small fraction of the fallout radioactivity to the surrounding area.

The elogation of the I^{131} (and Ru^{106}) distribution patterns along a NE-SW axis was predictable from the dominance of southwesterly and northeasterly winds in this region. Culkowski's meteorological formula for calculating environmental build-up of radioactivity was used with an IBM 610 computer program which allowed for hourly variations in speed as well as direction of local winds. For routine use, programming for a faster computer would be needed.

The most conspicuous discrepancy between calculations and observed spatial distribution involved relatively high deposition near the stacks, instead of a mile or two downwind where the plume of stack effluent would theoretically be expected to lead to most deposition on the ground. The possibilities that this pattern could be accounted for by rainout, or fallout of coarse particulates, or "fumigation" of the area by the turnover of an inversion (especially in calm air) deserve further attention, preferably with refined estimates of amount and time of stack release.

More complete ecological sampling on a unit-area basis (soil and organic litter, as well as vegetation) is needed before critical

comparisons can be made between observed and predicted levels of contamination. Forage samples need to be included for comparisons with health physics standards involving the food-chain leading to main (grass-cattle-milk-man).

Apparently I^{131} was well below levels which are currently considered hazardous. Concentrations were locally high enough to require special precautions in certain kinds of low-level ecological tracer experiments in the field, but in most of the Oak Ridge Reservation this was not a serious problem. Ecological tracer experiments are needed in order to evaluate the possible consequences of higher-level accidental contamination, to determine the fate of long-lived isotopes in the environment around future nuclear industries, as well as to carry out basic research on environmental processes.

The existence of aerial contamination surrounding a nuclear installation and the possibility of accidents indicates the desirability of managing the site and the reserved area surrounding the site so that land uses will minimize the processes of runoff and erosion which could carry radioactive fallout into surface water supplies. The prediction of patterns of contamination, for the present type of chronic build-up of radioactivity as well as for accidental release in the environment, should also be taken into account in determining policy on land use for areas around a nuclear facility.

REFERENCES

(1) H. H. Abee et al., Environmental levels of radioactivity for the Oak Ridge Area, Report for 1959. Report from Union Carbide Nuclear Company to AEC for public distribution, May 28, 1960. ORNL-3073 See also J. C. Hart, Applied Health Physics Annual Report for 1959.

(2) S. I. Auerbach et al., "Ecological Research", Health Physics Div. Ann. Prog. Rept., July 31, 1958, ORNL 2590, p. 27-52. Oak Ridge National Laboratory.

(3) J. S. Olson, "Forest Studies", Health Physics Div. Ann. Prog. Rept., July 31, 1959, ORNL 2806, p. 37-54. Oak Ridge National Laboratory.

(4) W. M. Culkowski, "Estimates of accumulated exposures and environmental build-up of radioactivity", Proc. Sixth AEC Air Cleaning Conference, Idaho Falls, Idaho, July 7-9, 1959. TID 7593, AEC Technical Information Service Extension, Oak Ridge (1961).

(5) J. W. Youngblood, Personal communication, October 1959.

(6) C. C. Harris, D. P. Hamblen, J. E. Francis, "Basic principles of scintillation counting for medical investigators", ORNL 2808, Oak Ridge National Laboratory (1959).

(7) P. Iredale and D. L. O. Humphreys, "The analysis of low level gamma ray activity by scintillation spectrometry", AERE El/R 2326. Atomic Energy Research Establishment, Harwell, Berkshire (1957).

(8) R. W. Perkins, "Gamma-ray spectrometric systems of analysis, Part II. I^{131} and other fission products on vegetation", .15/P/2377

(June 1958) Second UN International Conference on the Peaceful Uses of Atomic Energy, 28, 445-461.

(9) P. F. Gustafson, L. D. Marinelli, S. S. Brar, "Natural and fission-produced gamma-ray emitting radioactivity in soil". Science, 127, 1240-1242 (1958).

(10) P. F. Gustafson, Radioactivity measurements and dose calculations of soil collected at Argonne. Radiological Physics Division simi-annual Report, January-June 1959. ANL-6049.

(11) H. W. Patterson, A. R. Smith and L. D. Stephens, Fallout and natural background in the San Francisco Bay Area, UCRL-8401, University of California Radiation Laboratory, Berkeley, California (Aug. 4, 1958).

(12) B. V. Andersen, "Fallout Data--March 1958 through December 31, 1958". General Electric Company, Hanford Works. In: Radioactive Fallout Data Collected from Eleven Atomic Energy Commission Installations for 1958. G. M. Dunning, Ed. U. S. Atomic Energy Commission (1959).

(13) U. S. Weather Bureau, Meteorology of the Oak Ridge Area. ORO 99. AEC Technical Information Service Extension, Oak Ridge.

(14) _____, Meteorology and Atomic Energy. U. S. Govt. Printing Office, Washington. (1955).

(15) A. C. Chamberlain. Aspects of travel and deposition of aerosol and vapour clouds. AERE HP/R 1261. Harwell, Berks. (1955).

(16) F. A. Gifford, Jr., "Atmospheric Dispersion". Nuclear Safety 1, 56-62 (1960).

(17) A. C. Chamberlain and R. C. Chadwick, Experiments on the uptake of iodine 131 vapour by grass. AERE HP/R 993. Harwell, Berks. (1952).

(18) _____, "Deposition of airborne radioactive vapor". Nucleonics 11, 22 (1953).

(19) W. J. Megaw, Personal communication, July 1960.

(20) W. M. Culkowski, Calculation of the deposition of aerosols from elevated sources. ORO-171. AEC Technical Information Service Extension, Oak Ridge. 1958.

(21) _____, Personal communication, March 1960.

(22) A. A. Selders and F. P. Hungate, The foliar absorption of iodine by plants, HW-44890, Hanford Atomic Products Operation, Richland, Washington (1956).

(23) J. J. Davis, "Accumulation of Radioisotopes from Fallout by Terrestrial Animals at Hanford". In: Radioactive Fallout Data Collected from Eleven Atomic Energy Commission Installations for 1958. G. M. Dunning, ed. U. S. Atomic Energy Commission (1959).

(24) J. R. Horan, "Radioactive Waste Disposal Management at the National Reactor Testing Station." In: Hearing before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, Congress of the United States, 1, 584-98. 1959.

(25) J. S. Olson, et al., "Forest Studies", Health Physics Div. Annual Prog. Rept., July 31, 1960. ORNL-2994, 167-85.

(26) H. M. Parker, Tolerable Concentration of Radio-iodine on Edible Plants. AECD 2907 (1946).

(27) F. B. Morrison, Feeds and Feeding: A Handbook for the Student and Stockman, p. 1088. Morrison Publishing Company. 1956.

(28) H. H. Dunster, H. Howells and W. L. Templeton. "District Surveys following the Windscale Incident, October 1957". P/316 UK. Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, 1958, 18, 296-308.

(29) F. R. Farmer and P. I.Fletcher, "Siting in Relation to Normal Reactor Operation and Accident Conditions". Symposium on Safety and Location of Nuclear Plants. Rome. 1959.

(30) International Commission on Radiological Protection, "Report of Committee II on Permissible dose for Internal Radiation". Health Physics 3, 61 (1960).

(31) F. R. Bruce, "Radiation-Safety Improvements to ORNL Facilities", Nuclear Safety, 2, 40-44 (March, 1961).

(32) J. W. Healy, B. V. Andersen, H. V. Clukey and J. K. Soldat, "Radiation Exposure to People in the Environs of a Major Production Atomic Energy Plant". P/743 USA. Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, 1958, 18, 309-18.

(33) H. N. Culver, "Activity Releases at Oak Ridge National Laboratory", Nuclear Safety 1, 83-88 (June 1960).

(34) W. de Laguna, "Geologic and Hydrologic Considerations in Power Reactor Site Selection", Nuclear Safety, 1, 64-68 (June, 1960).

Appendix. Concentrations of Radioactive Isotopes in Dogwood Leaves as a Function of Distance and Direction from Oak Ridge National Laboratory (September 1959).

Table 5. Iodine-131 on Dogwood Foliage
(μμc/g dry wt.)

Miles from ORNL	Direction				
	NE	SE	SW	NW	N
0-0.3	583	23	191	102	
0.5	175				
1	189	2.4[a]	59	28	
2	[a]	7	7	27	
4	24	1	7	5	7
8	5				
16	-				
32	-				

[a]Possibly low because of volatilization.

Table 6. Ruthenium-106 on Dogwood Foliage
(μμc/g dry wt.)

Miles from ORNL	Direction				
	NE	SE	SW	NW	N
0-0.3	113	-	15	27	
0.5	17				
1	12	30	13	7	
2	-	6	17	6	
4	9	7	6	2	-
8	9				
16	8				
32	6				

Table 7. Cesium-137 on Dogwood Foliage
(μμc/g dry wt.)

Miles from ORNL	Direction				
	NE	SE	SW	NW	N
0-0.3	22	3	16	-	
0.5	7				
1	4.5	4.5	2.7	-	
2	-	0.9	3.6	2.2	
4	-	1.3	-	0.9	-
8	-				
16	2.2				
32	3.1				

Table 8. Zirconium-Niobium-95 on Dogwood Foliage (μμc/g dry wt., as Zr-95, assuming Nb-95 is in equilibrium).

Miles from ORNL	Direction				
	NE	SE	SW	NW	N
0-0.3	26	7	68	14	
0.5	7.5				
1	6.4	6.5	5.4	5.3	
2	-	7.8	6.6	6.6	
4	5.1	5.2	5.1	6.1	2.4
8	3.7				
16	6.4				
32	9.0				

Table 9. Cerium-Praseodymium-144 on Dogwood Foliage (μμc/g dry wt., as Ce-144; all estimates tentative).

Miles from ORNL	Direction				
	NE	SE	SW	NW	N
0-0.3	213	-	214	111	
0.5	30				
1	-	18	-	-	
2	12	-	-	-	
4	17	13	16	-	18
8	-				
16	-				
32	20				

Table 10. Cobalt-60 (?) on Dogwood Foliage
(μμc/g dry wt., very tentative)

Miles from ORNL	Direction				
	NE	SE	SW	NW	N
0-0.3	2.6?	-	5.5?	-	
0.5	-				
1	-	-	-	-	
2	-	-	-	-	
4	0.7?	-	0.4	0.7	-
8	-				
16	-				
32	-				

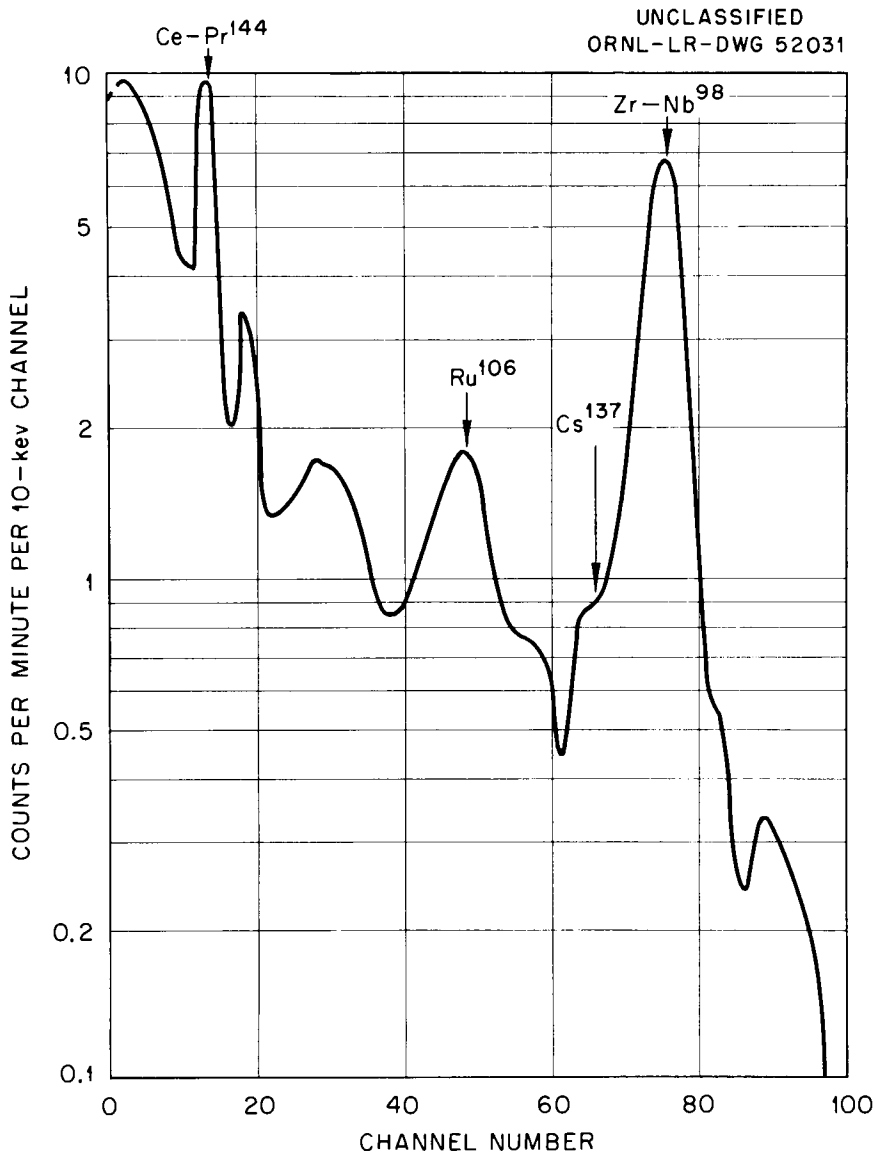

Fig. 1. Typical Spectrum of Gamma Radiation from Weapons Fallout on Dogwood Leaves (30 Miles East of Oak Ridge, June 1959).

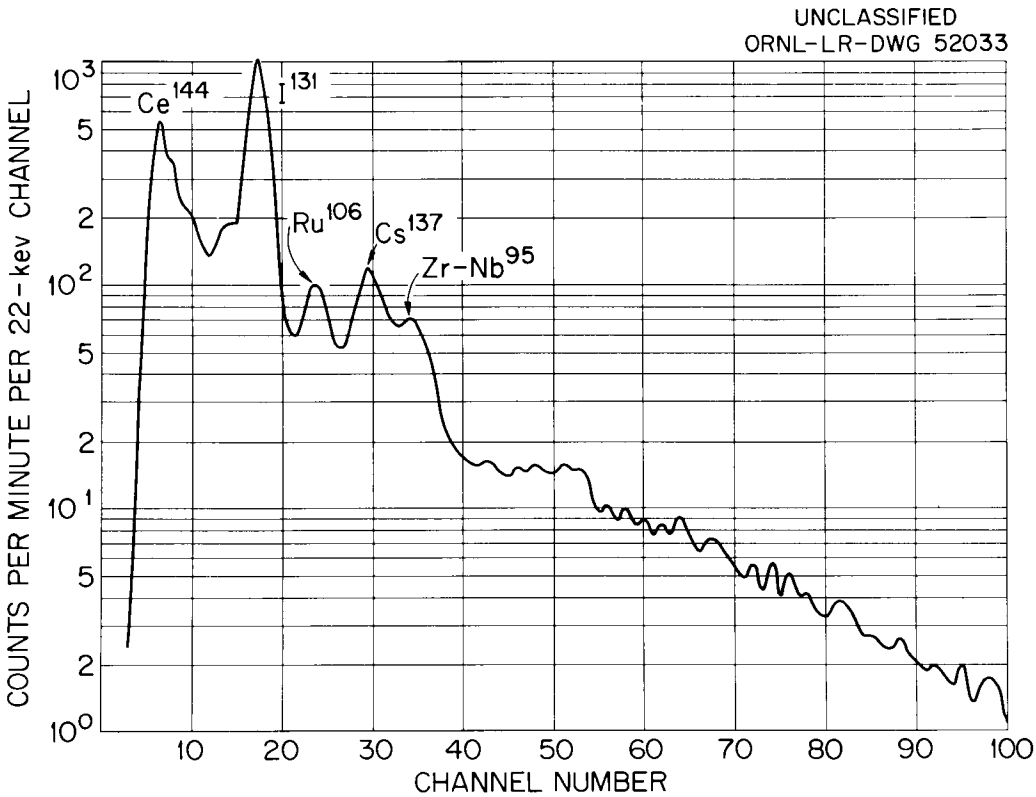

Fig. 2. Spectrum of Gamma Radiation Including I^{131} and Other Isotopes in Addition to Weapons Fallout on Dogwood Leaves (Oak Ridge National Laboratory, September 1959).

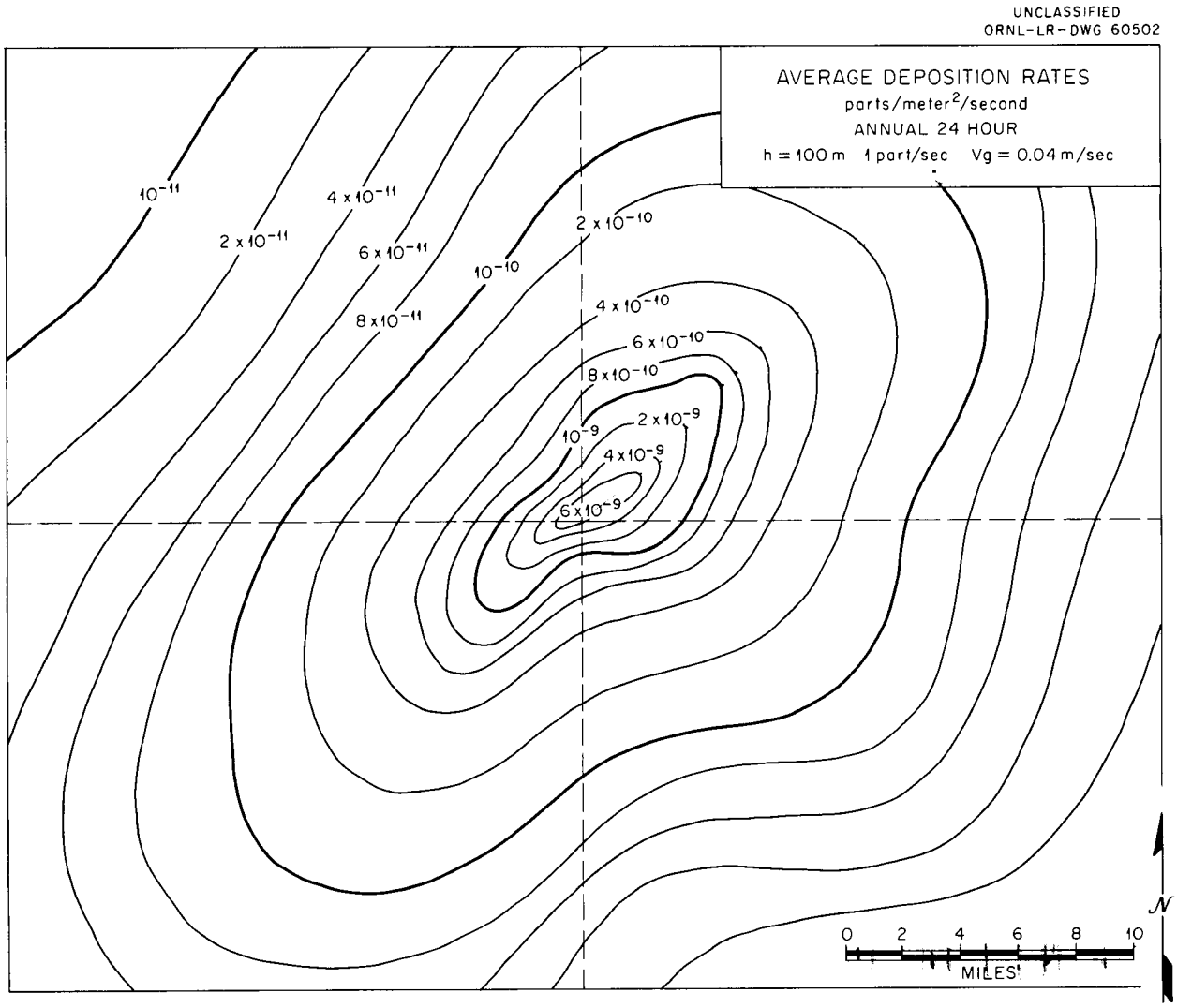

Fig. 3. Average Annual Deposition Rates Calculated by W. Culkowski for Particles Having Effective Deposition Velocity of 4 cm/sec, from Stack Having Effective Height of 100 Meters, Actual Height + Plume Rise (by Permission).

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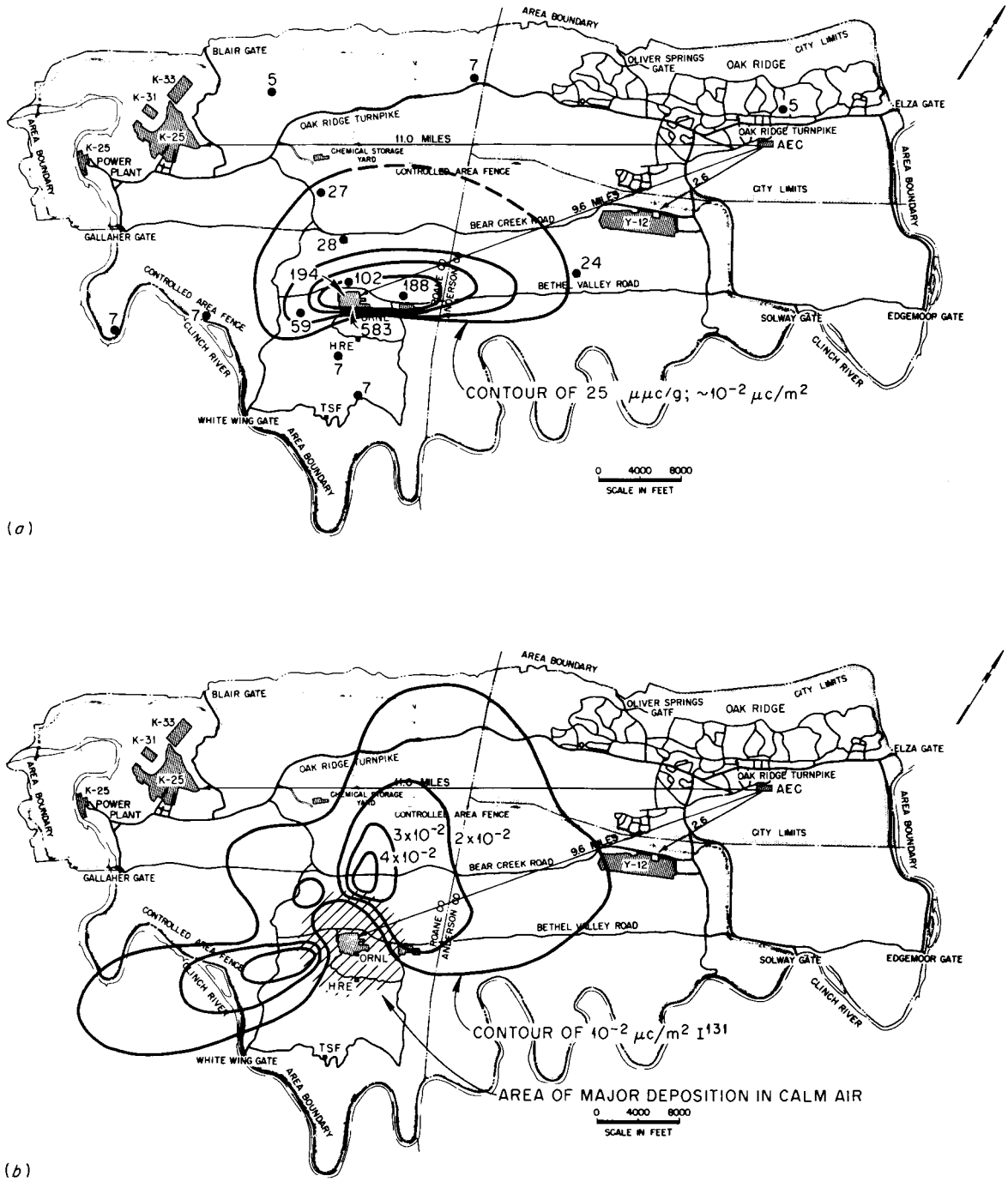

Fig. 4. Dispersal of I^{131} Around Oak Ridge, September 1959; (a) Observed on Sampling of Foliage of Dogwood Treets; (b) Calculated from Wind Distribution by Procedure of W. Culkowski, Allowing for Variations in Velocity and Air Temperature Stability.

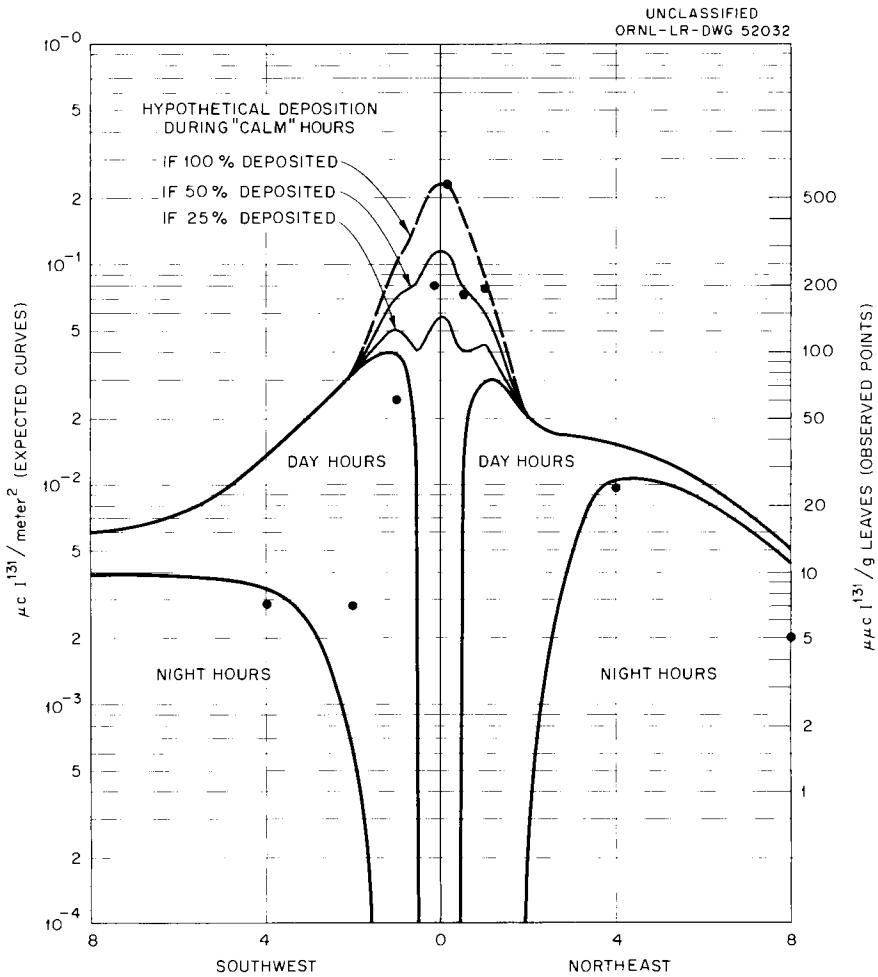

Fig. 5. Comparison of Expected Deposition of I^{131} from Winds During Night Hours (Lowest Curves), Day Hours (Main Curves), and from Air During Calm Hours (Three Curves for Hypotheses that 25, 50, or 100% of Effluent if Deposited Locally in Bivariate Normal Spatial Distribution).

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