SWRHL-103r

RADIOIODINE FIELD STUDIES WITH SYNTHETIC AEROSOLS

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Radiological Research Program
Southwestern Radiological Health Laboratory

ENVIRONMENTAL PROTECTION AGENCY

February 1971

This report was presented at the Eleventh AEC Air Cleaning Conference, August 31 through September 3, 1970, Richland, Washington.

This research performed under Memorandum of
Understanding (No. SF 54 373)
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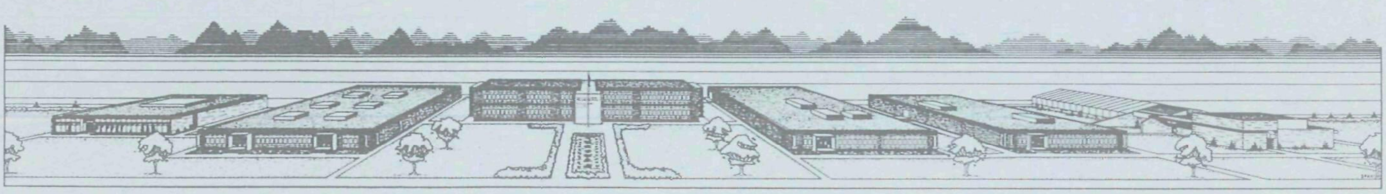

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*Formerly part of the U. S. Department of Health, Education, and Welfare, Public Health Service, Environmental Health Service, Environmental Control Administration, Bureau of Radiological Health.

ABSTRACT

The radioiodines are the principal source of human thyroid exposure at early times following the detonation of fission devices. The major pathway for this exposure is the forage-cow-milk chain. With the cessation of atmospheric testing and the reduction in Plowshare activities, simulation remains the primary method to study this pathway. To do this, a series of experiments was conducted in which a radioactive contaminant was released under controlled conditions to simulate the passage of a radioactive cloud over cow forage. Forage is the initial link in the forage-cow-milk-man food chain and in these studies the interrelationship of the physical, chemical and biological variables concerning the transport through that link were investigated. Sodium iodine-131 labeled diatomaceous earth aerosols with count median diameters ranging from 0.2 to 24.0 μm, a ^{131}I labeled hydrosol and elemental iodine were generated for these studies. The aerosol preparation, labeling, generation and analytical procedures are discussed.

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INTRODUCTION

The radioiodines are recognized as the principal source of human exposure at early times following the detonation of a fission device. This is due, primarily, to the relatively high fission yield, rapid transport in the forage-cow-milk pathway, and concentration in the thyroid.

One approach to the study of radioiodine exposures is to set up experiments in the fallout pattern of nuclear tests. If more control of the variables is required, synthetic aerosols may be used. This latter approach has become useful since atmospheric tests have been terminated and since Plowshare cratering tests have been so infrequent.

For controlled studies with radionuclides, a Grade A dairy farm [1] was established at the Nevada Test Site (Figure 1.). The generation and assessment of a variety of aerosols used to contaminate the cow forage at this farm are the subject of this report.

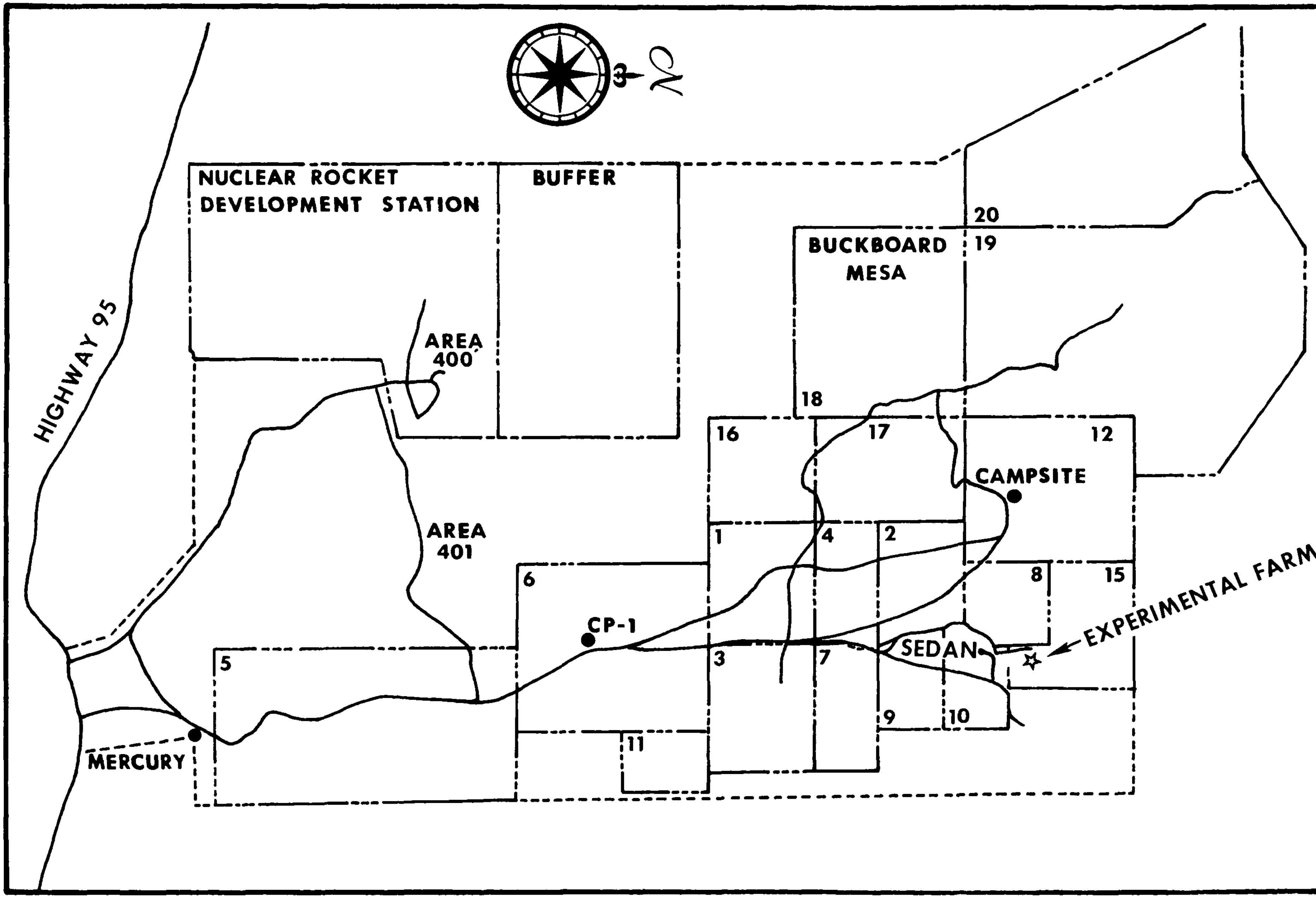

FIGURE 1 — LOCATION OF USPHS FACILITIES ON THE NEVADA TEST SITE

PROCEDURES

The procedures used in the studies to be described were designed to meet certain objectives, such as:

(1) generation of an aerosol with a known particle size distribution,

(2) deposition on a certain area sufficient to yield a predetermined $\mu Ci/m^2$ contamination, and

(3) determination of the physical parameters associated with a given deposition of the aerosols.

Five experiments, using aerosols tagged with ^{131}I, have been conducted at the farm as shown in outline in Table 1. The experimental requirements along with some of the actual data in the table indicate the success in meeting those requirements. The desired deposition, in all cases, was 100 nCi/kg or more on the cow forage.

In these experiments, careful consideration was given to the meteorological conditions to minimize their effect on the aerosol generation and deposition. Wind velocity and the relative humidity were prime considerations. Full use was made of the early morning drainage winds characteristic of the area because of their predictability and reproducibility. Wind speeds of less than 4 m/s (9 mph) and a wind direction with an azimuth between 315^o and 15^o were considered acceptable for these experiments.

The aerosol generator used in these experiments was developed during a series of preliminary tests and has proved satisfactory for our purposes. Figure 2 is a schematic drawing of the generator which was constructed with common laboratory supplies. The glass beads aid in the removal of the aerosol as well as in breaking-up clumps of material. The stainless steel screens at the outlet also helped to reduce clumps of material. Clumping was more of a problem when the relative humidity was high.

Table 1. Outline of Five Experiments Using ^{131}I Aerosols.

Project Name	Carrier Aerosol	Desired Size (μm)	Measured (CMD-μm)	Forage Type	Deposition μCi/m^2
Hayseed	Diatomaceous earth	20	23	Sudan grass	3.1
Alfalfa	Diatomaceous earth	1-5	2.0	Alfalfa	4.7
Hare	Diatomaceous earth	<1	.60	Alfalfa and Sudan grass	1.3
Sip	Diatomaceous earth	<1	.13	Alfalfa	1.6
Mice	Air	gas		Alfalfa	.66

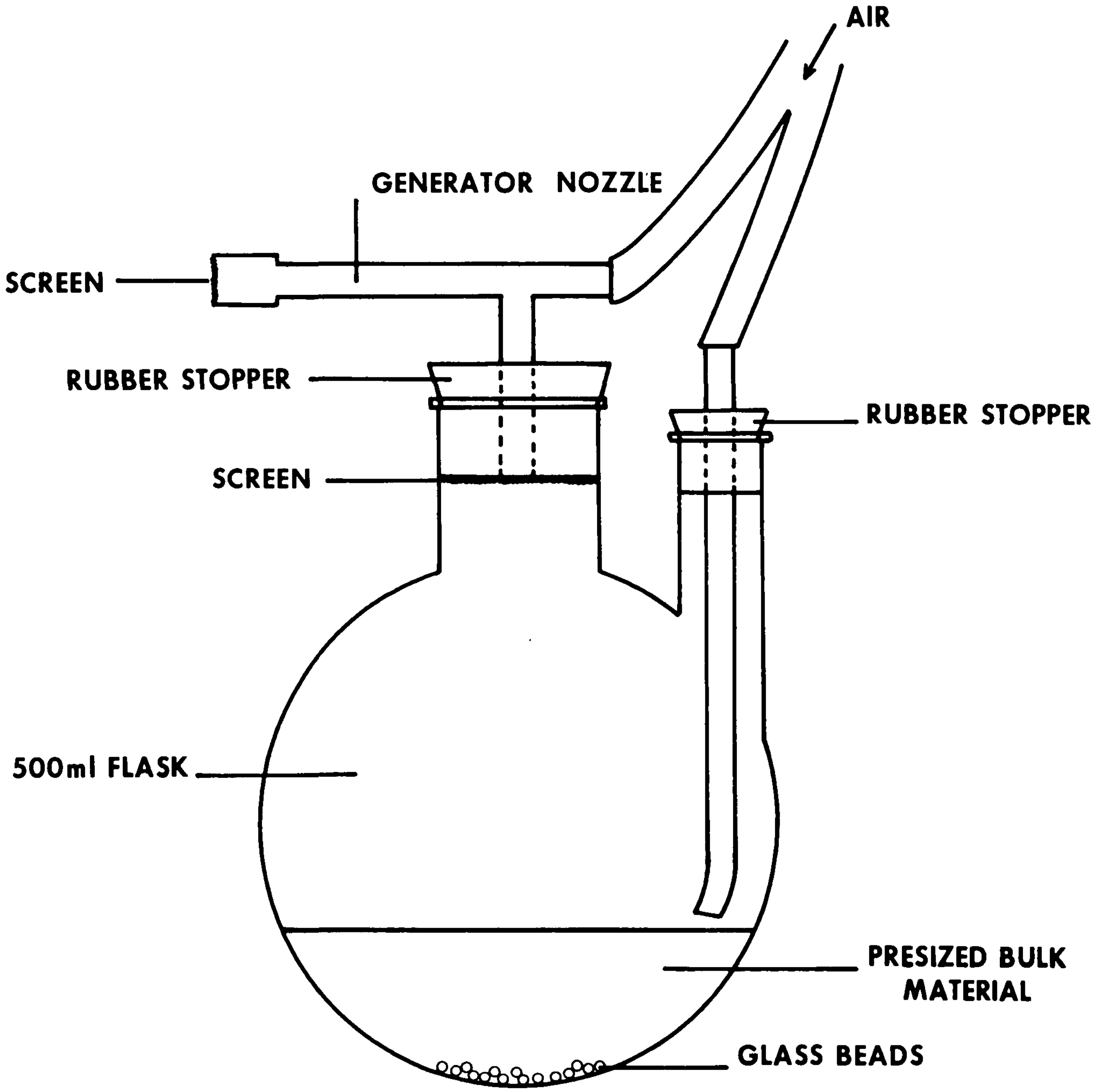

FIGURE 2 – AEROSOL GENERATOR

The dry aerosols were prepared using a wet-labeling and vacuum-evaporation drying system. A typical preparation procedure was that as used in Project Hare.[2] For this exercise, 10 ml of a source solution, containing 100 mCi of ^{131}I, was added to 7 ml of an aqueous solution containing 0.7 mg of NaI. Then 0.5 ml aliquots were transferred to each of 32 two-ounce bottles containing 1 ml of 1M $Na_2S_2O_3$, 5 ml of 2M Na_2CO_3 and 25 ml of water. The contents of these bottles were added to approximately 30 ml of water, 450 ml of isopropanol, 7 g of glass beads and 150 g of the prepared diatomaceous earth(DE). After mixing, the material was dried using a vacuum-evaporation system. The evaporation rate of the flasks' contents was controlled to prevent the boiling action from becoming too violent. The total drying time for this study was 40 hours with the flasks being heated by hot tap water for the last 18 hours to insure efficient drying. Depending on the particle size desired, mechanical screening or ball-milling or a combination are used to pretreat the particulates.

The preparation for the Mice experiment, the gaseous $^{131}I_2$ release, was similar to that employed by the National Reactor Testing Station (NRTS).[3] Four vials containing various solutions were added in sequence to the generating flasks. The first contained 1.5 mg of NaI carrier, 0.5 ml of H_3PO_2, and 10 ml of distilled water along with the ^{131}I activity. The second contained 130 ml of 2N H_2SO_4. The third was used to initiate the reaction and consisted of 30 mg of $NaNO_2$ and 10 ml of distilled water. The fourth contained the post-generation reducer solution (to stop the I_2 generation) which consisted of 5 ml of 30% H_3PO_2 and 5 ml of distilled water. The generating flasks for this release were modified to include a fritted glass sparging tube at the inlet and had the exhaust tube loosely packed with glass wool. To generate I_2, the acidic solution of $Na^{131}I$ was oxidized and the $^{131}I_2$ was sparged from the solution with nitrogen gas. Generation of the gaseous materials was regulated to last for approximately 30 minutes.

To obtain the many deposition parameters required to adequately describe the aerosol releases, many types of samples were collected. These included:

(1) Planchets - stainless steel 0.01 m^2 planchets coated with a non-setting resin. These are used to determine the $\mu Ci/m^2$ deposited and for deposition velocity calculations.

(2) Glass slides - specially cleaned glass microscope slides used for particle size distribution studies.

(3) Air samplers - 10 cfm air samplers equipped with prefilters and charcoal cartridges for determining air concentration and particulate to gaseous ratios.

(4) Special air samplers - modified sampling train, e.g., charcoal impregnated paper, used for special aerosol measurement.

(5) Cascade impactors - Unico* type used for certain particle size measurements.

(6) Planchet racks - racks holding planchets at various heights and angles and used to determine the radioactive profile of the aerosol cloud.

A typical sampling grid is shown in Figure 3 which is the actual experimental arrangement for Project Hare.[2]

The particle size distribution was determined from the Feret diameter measurement,[4] using optical and electron microscopes, and was characterized by the count median diameter (CMD). The filter/charcoal ratio used in Table 2 is just the ratio of the total activity collected on each. The data from the air samplers were used to calculate the integrated air concentration (units of $\mu Ci\text{-}s/m^3$) by dividing the sum of the prefilter and charcoal activities by the air flow rate. The planchet deposition ($\mu Ci/m^2$) divided by the integrated air concentration gives the deposition velocity (m/s).

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*Unico Cascade Impactor for Simplified Particle Size Analysis, manufactured by Union Industrial Equipment Corporation, Port Chester, New York.

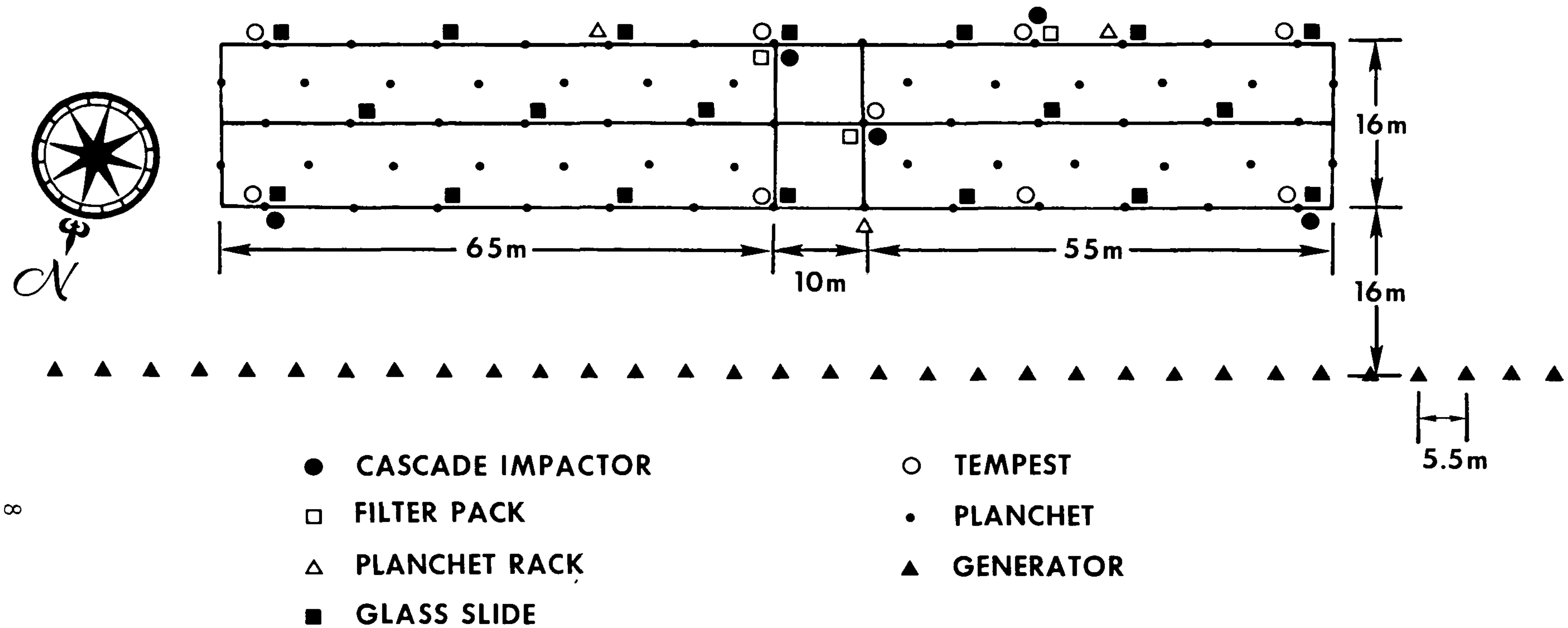


FIGURE 3 — TYPICAL SAMPLE GRID

Table 2. Summary of Results from Five Controlled Experiments.

<table>
<tr><th rowspan="2">Measured or Calculated Parameter</th><th colspan="6">Experiment Title</th></tr>
<tr><th>Hayseed</th><th>Alfalfa</th><th colspan="2">Hare</th><th>Sip</th><th>Mice</th></tr>
<tr><td>Particle size - μm</td><td>23</td><td>2.0</td><td></td><td>.60</td><td>.13</td><td>Gas</td></tr>
<tr><td>Forage type</td><td>Sudan</td><td>Alfalfa</td><td>Sudan</td><td>Alfalfa</td><td>Alfalfa</td><td>Alfalfa</td></tr>
<tr><td>Average deposit - $\mu Ci/m^2$</td><td>3.1</td><td>4.7</td><td>1.2</td><td>1.4</td><td>1.6</td><td>.66</td></tr>
<tr><td>Percent deposit</td><td>21</td><td>13</td><td colspan="2">5.0</td><td>4.0</td><td>1.8</td></tr>
<tr><td>Air concentration - $\mu Ci\text{-}s/m^3$</td><td>322</td><td>33.4</td><td colspan="2">88.0</td><td>157</td><td>129</td></tr>
<tr><td>Filter/Charcoal ratio</td><td>4.9</td><td>3.5</td><td colspan="2">1.0</td><td>3.2</td><td>.12</td></tr>
<tr><td>Deposition velocity - cm/s</td><td>.96</td><td>1.4</td><td>1.4</td><td>1.6</td><td>1.0</td><td>.51</td></tr>
<tr><td>Peak in milk - nCi/liter</td><td>75.9</td><td>237</td><td>21.6</td><td>50.8</td><td>68.4</td><td>151</td></tr>
<tr><td>Milk/Forage ratio</td><td>.028</td><td>.070</td><td>.021</td><td>.066</td><td>.061</td><td>.058</td></tr>
</table>

RESULTS AND DISCUSSION

Selected results from the five experiments under consideration are shown in Table 2 and are briefly discussed in the following paragraphs.

The equal deposition lines (isopleths) for Project Hayseed [5] are shown in Figure 4, and are based on the planchet data. The CMD as determined from the optical sizing of the particles on the glass slides was 23 μm. The small areas shown at the leading edge of the test grid were for piled green chop, piled hay, and cows in stanchions and were used for special studies. The major crop for this study was Sudan grass and the average deposition for the entire grid was 3.1 $\mu Ci/m^2$. Approximately 21% of the total radioiodine labeled aerosol released was deposited on the 600 m^2 test grid. The lateral distribution as can be seen from this figure appears quite uniform and the drop in activity from the front of the plot to the rear is consistent with the large CMD of the particulates.

The contribution to the radioactivity in the milk resulting from air uptake amounted to only about 1% for this study. This is in good agreement with the results found in Project Alfalfa where the air uptake contribution was calculated to be about 2%.[6] Subsequent tests (Projects Sip and Mice) confirm the contribution via this pathway to be minimal.

The deposition results for Project Alfalfa are shown in Figure 5. The CMD of this aerosol was calculated as 2.0 μm from the size distribution data which are shown in Figure 6. The planchet data yielded an average deposition value of 4.7 $\mu Ci/m^2$ and approximately 13% of the activity generated was deposited on the test grid. As would be surmised, the distribution for the smaller sized aerosol is more uniform over the study plot, as compared to Hayseed.

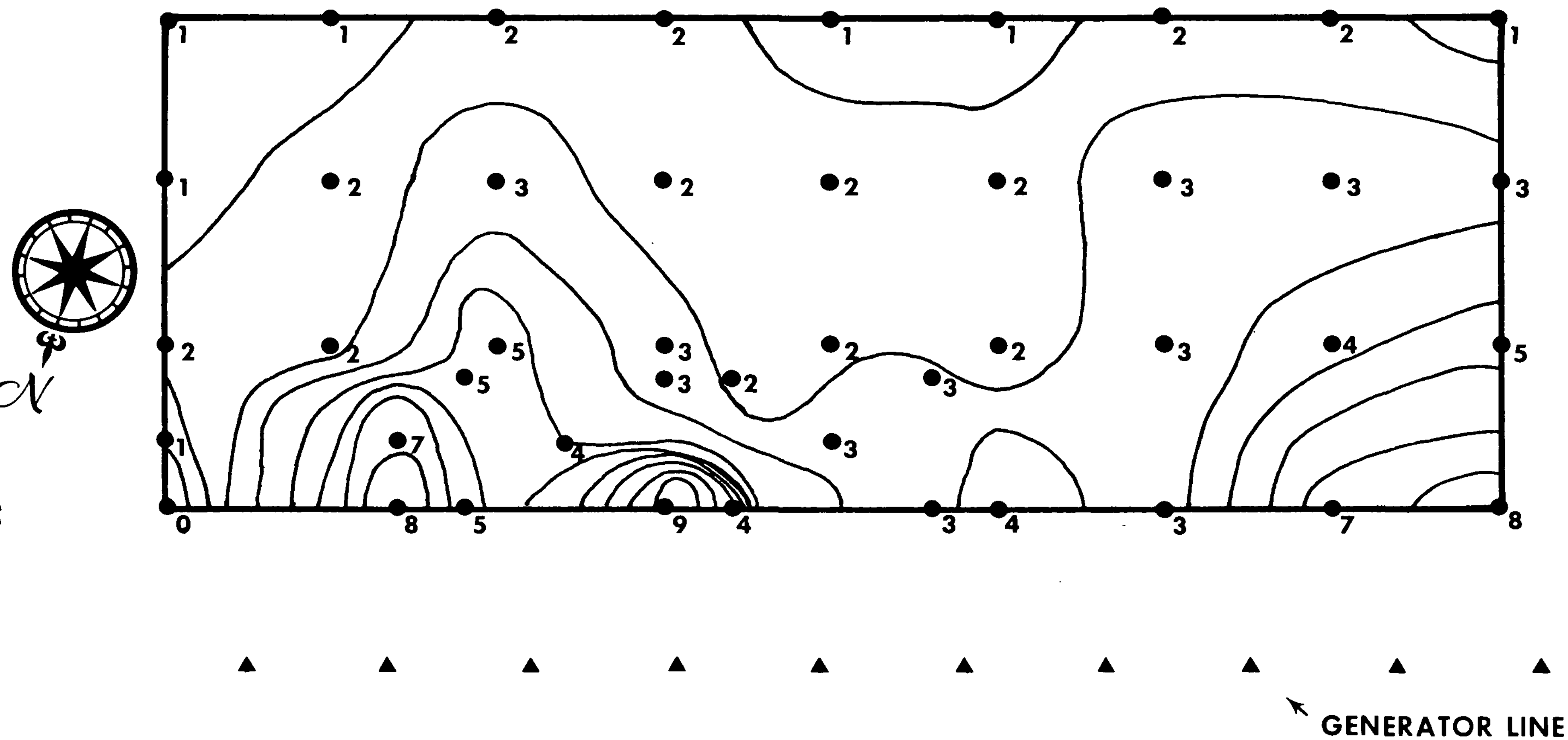


FIGURE 4 — PROJECT HAYSEED ^{131}I ACTIVITY ($\mu Ci/m^2$) ISOPLETHS FROM PLANCHET DATA

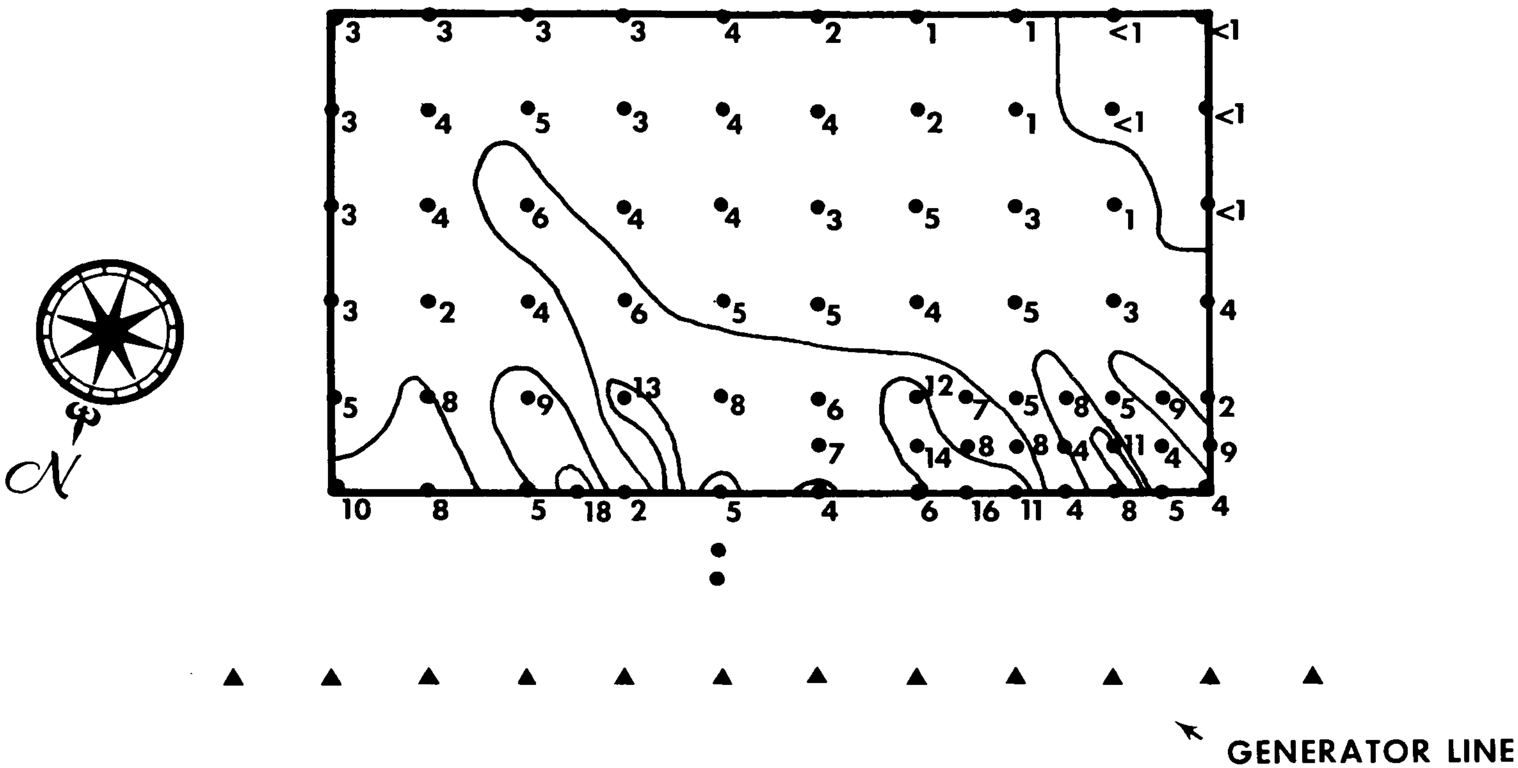


FIGURE 5 — PROJECT ALFALFA ^{131}I ACTIVITY (μCi/m^2) ISOPLETHS FROM PLANCHET DATA

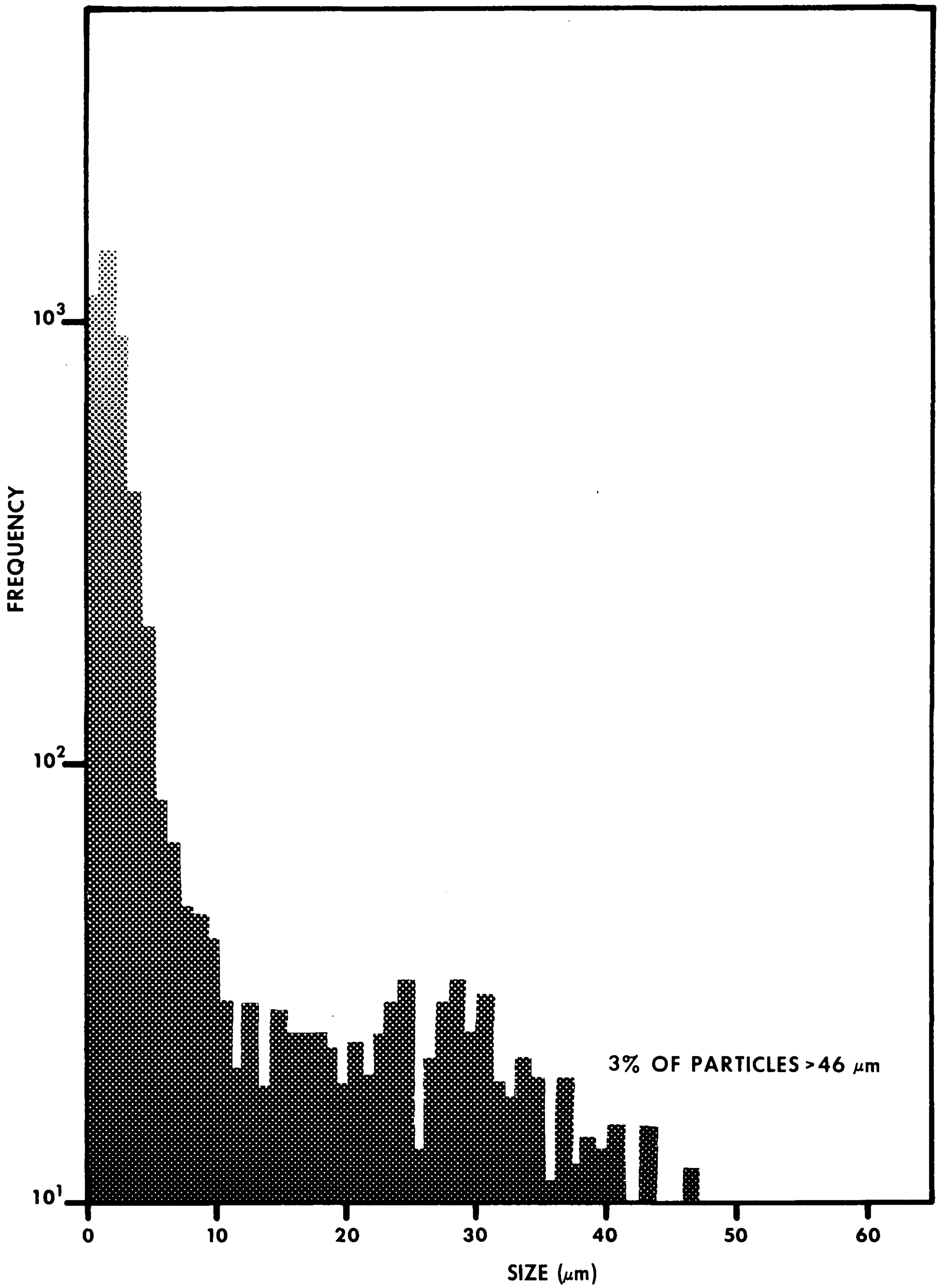


FIGURE 6 — PARTICLE SIZE DISTRIBUTION HISTOGRAM

Project Hare, the third study in the series of decreasing particle sizes, had as an additional objective, determining whether aerosol particle size or forage species was the main contributor to the difference in the milk to forage ratios from the first two experiments. This ratio for Project Alfalfa was approximately four times the ratio for Project Hayseed.

The deposition isopleths are shown in Figure 7, for the two test grids used in Hare, one crop being Sudan grass and the other alfalfa. The activity deposited on the field averaged 1.25 $\mu Ci/m^2$ for the Sudan grass plot and 1.43 $\mu Ci/m^2$ for the alfalfa study plot. Approximately 5% of the activity in the generator flasks was deposited on the test grid as estimated from the planchet data. This lower percentage deposited conforms to the decrease in particle size. The CMD of this aerosol was 0.6 μm.

The milk-to-forage ratio and percent in milk were both lower for Sudan-fed cows than for alfalfa-fed cows and the relative magnitudes agreed with similar data in the other experiments. These results suggest that forage type rather than particle size is responsible for the lower milk-to-forage ratio and lower percent in milk in Sudan-fed cows.

The smallest CMD aerosol studied in this series was 0.13 μm used during Project Sip.[7] Electron microscopy was used in addition to the optical microscopy to size this fine aerosol. The planchet values showed that the average deposition over the study area was 1.6 $\mu Ci/m^2$ and was by far the most uniform of any of the releases. The isopleths drawn from the planchet data (Figure 8) indicate that 96% of the test grid was contaminated at levels of from 1-3 $\mu Ci/m^2$ and the remaining 4% at a level of 4.5 $\mu Ci/m^2$. Approximately 4% of the total activity was deposited on the study area. The data from the planchet racks together with the respective "deposition vectors" are shown graphically in Figure 9. The deposition vector is defined as the resultant of the two vectors calculated from the activity on the horizontal and vertical planchets. These data demonstrate that the active cloud remained close to the ground, i.e., in the first meter

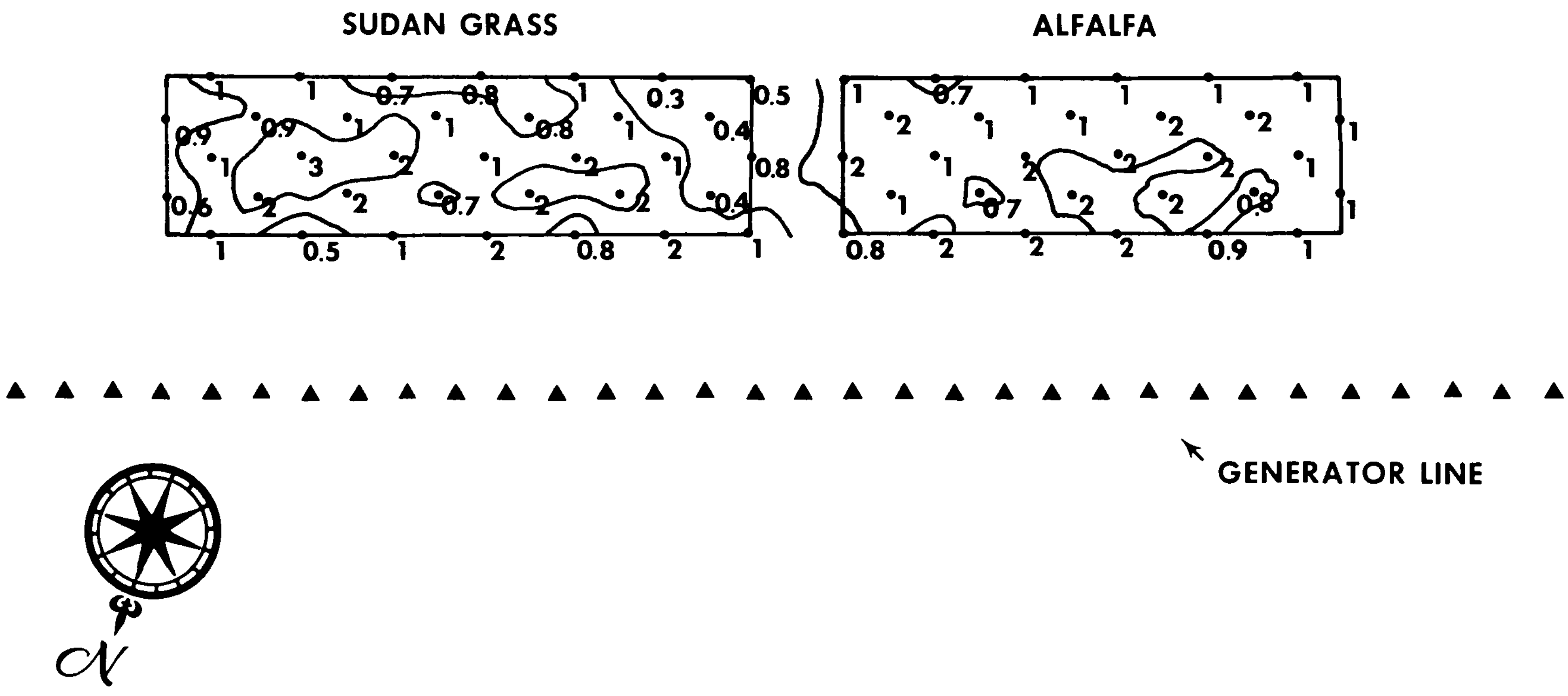


FIGURE 7 — PROJECT HARE ^{131}I ACTIVITY ($\mu Ci/m^2$) ISOPLETHS FROM PLANCHET DATA

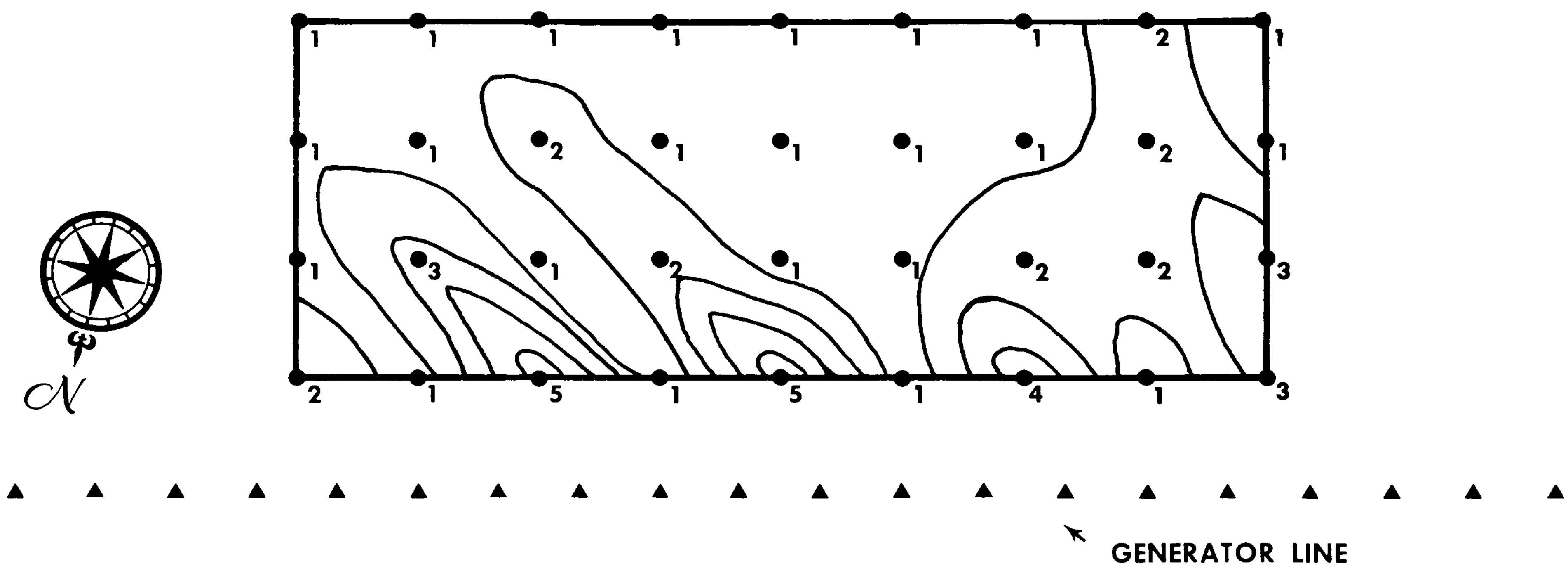


FIGURE 8 — PROJECT SIP ^{131}I ACTIVITY (μCi/m^2) ISOPLETHS FROM PLANCHET DATA

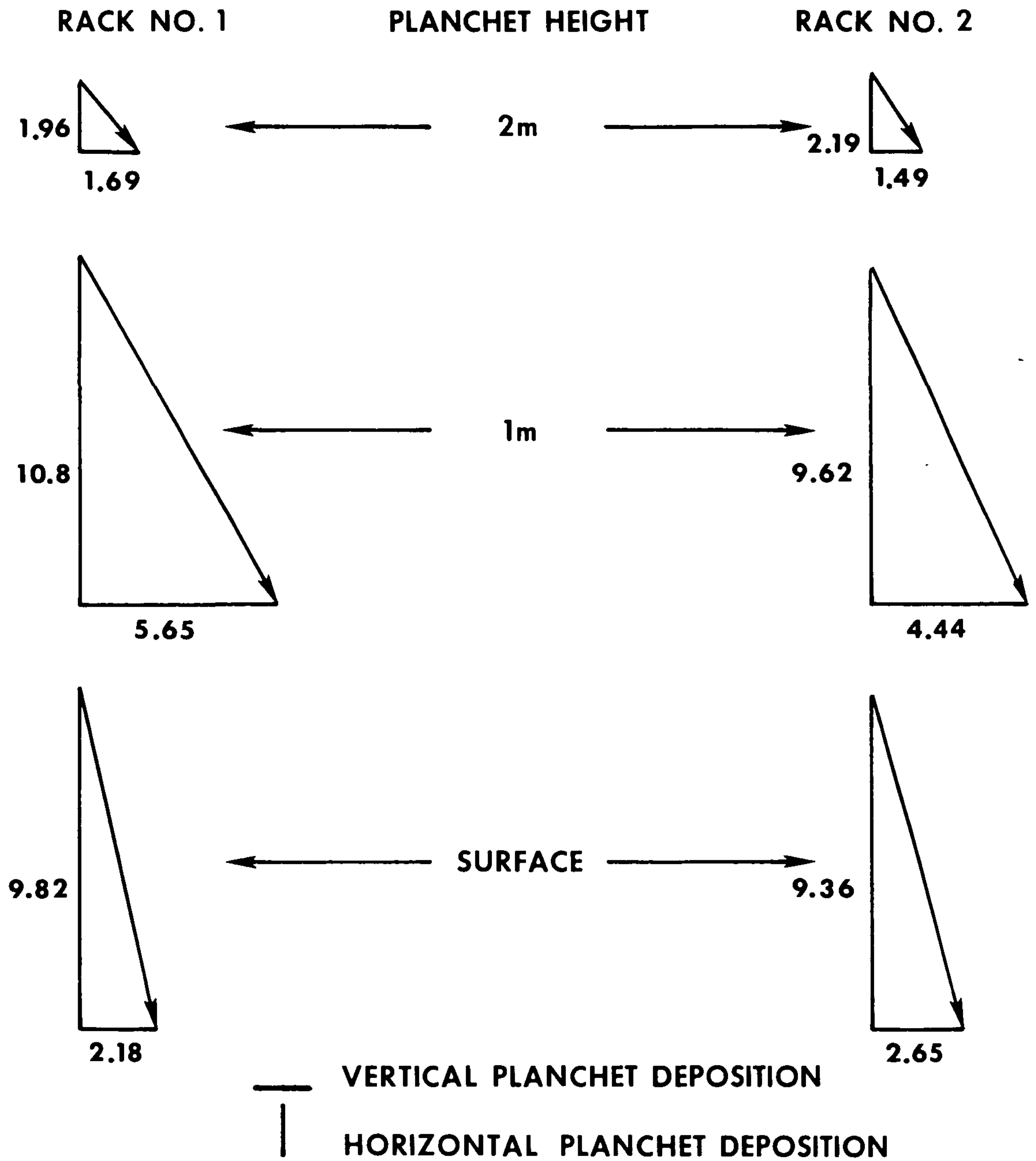


FIGURE 9 — PROJECT SIP PLANCHET RACK DATA ($\mu Ci/m^2$) AND DEPOSITION VECTORS

during transport across the experimental area. The activity on the two-meter elevation planchets dropped off markedly. The vector shows that the particles were being deposited in a nearly vertical mode close to the ground whereas at higher elevations the horizontal vector became stronger, probably due to the effect of the winds. The uniformity of the individual values between the two planchet racks separated by some 30 meters is remarkable.

The activity isopleths from Project Mice,[8] are shown in Figure 10 and indicate an average deposit of 0.66 $\mu Ci/m^2$. The three special air samplers were used on this study to confirm that $^{131}I_2$ was being generated.

The AC-1 charcoal impregnated filter paper has a high collection efficiency for elemental iodine but, at the flow rates used, would be inefficient for collecting CH_3I and HI because of the short residence times. An average of 86% of the activity collected by these samplers was found on the AC-1. An additional 12% was found on the Microsorban prefilter indicating the presence of some particulates. Microscopic examination of glass slides and electron microscope grids yielded a CMD of 0.01 μm for the particulates collected. The Unico Cascade Impactors, used on this study, yielded a mass median diameter (MMD) (1.6 μm) of the sampled aerosol. A CMD can be calculated from this value and also indicated a value of 0.01 μm.

The peak ^{131}I milk concentrations, from cows on fresh forage, obtained in these experiments are plotted on Figure 11 against the integrated air concentration divided by filter-to-charcoal ratios. In addition to the values for these studies, the results of two nuclear explosions are included, i.e., Pin Stripe and Palanquin. Although the levels are higher in the milk for the controlled releases, the data demonstrate that the controlled field releases yield data that correlate with actual fallout data. The values, one from cows fed green chopped alfalfa and one from cows fed baled hay, from Project Mice appear somewhat out of line, but that release, being primarily gaseous, was not representative of a true aerosol. The straight line plotted in the figure is best fit to the data and the

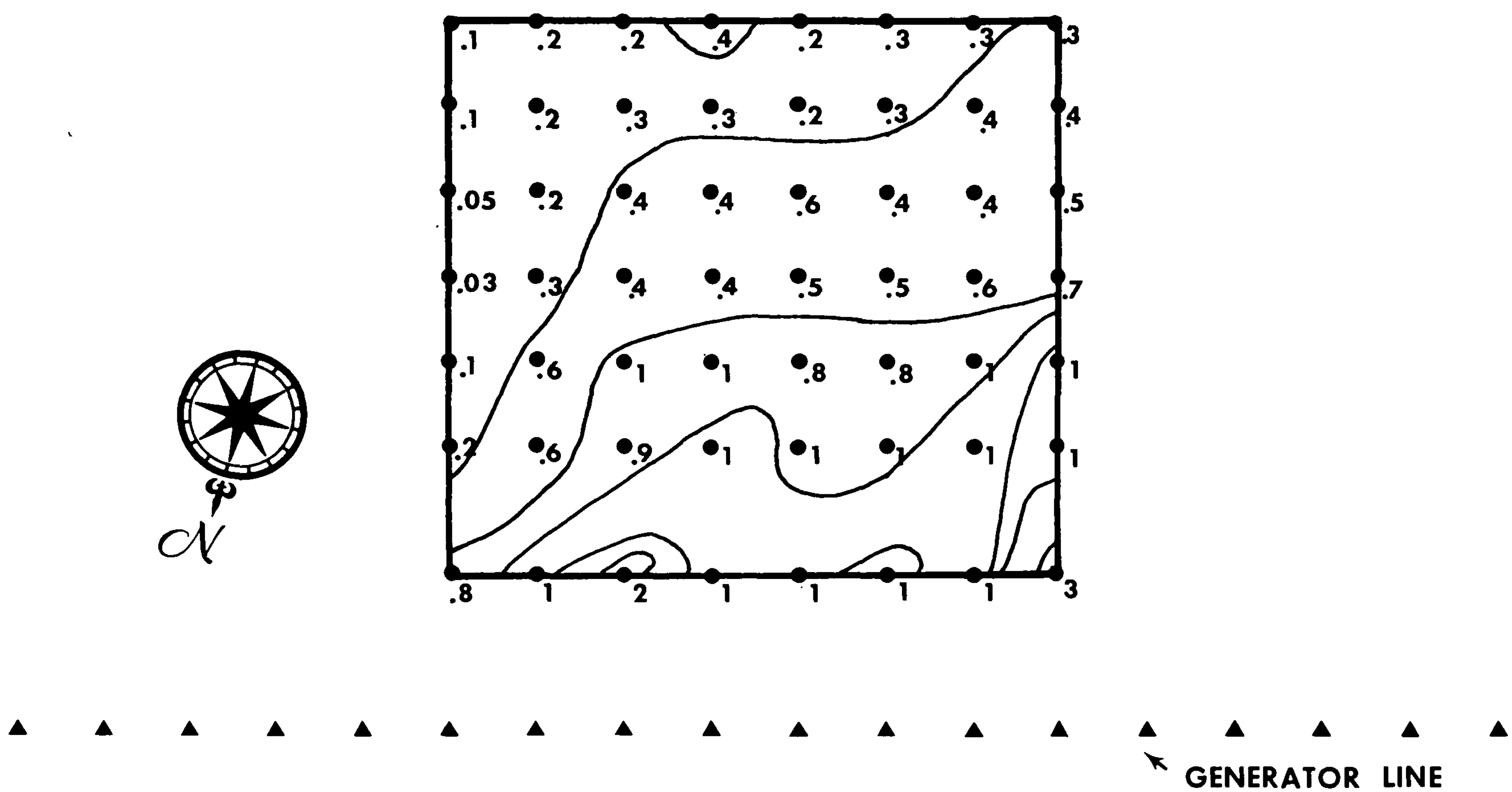
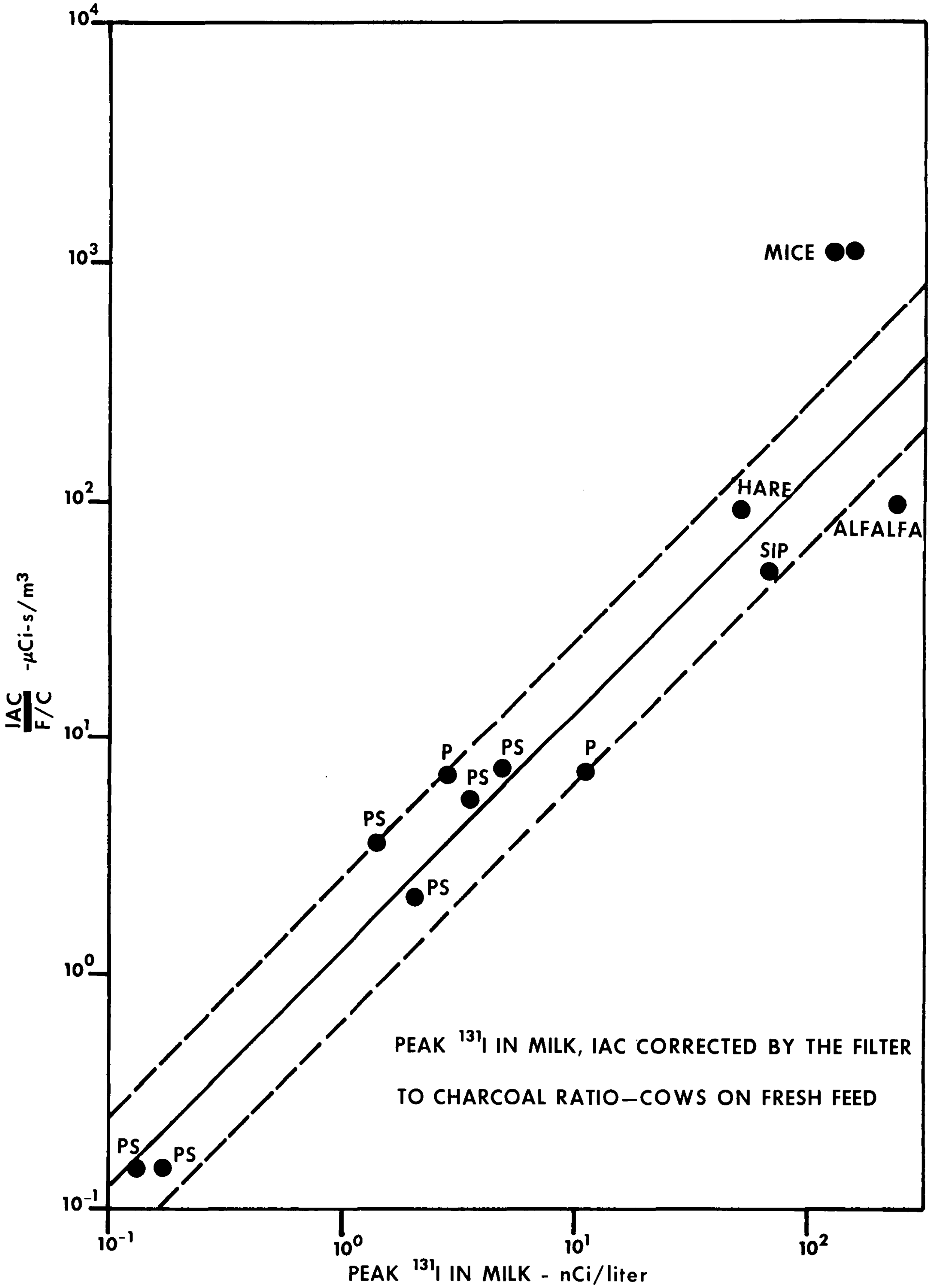


FIGURE 10 — PROJECT MICE ^{131}I ACTIVITY ($\mu Ci/m^2$) ISOPLETHS FROM PLANCHET DATA



MICE
HARE
ALFALFA
SIP
P
PS
PS
P
PS
PS
PS
PS
PS
PEAK ¹³¹I IN MILK, IAC CORRECTED BY THE FILTER
TO CHARCOAL RATIO—COWS ON FRESH FEED
IAC / F/C -μCi-s/m³
10⁴
10³
10²
10¹
10⁰
10⁻¹
10⁻¹
10⁰
10¹
10²
PEAK ¹³¹I IN MILK - nCi/liter

FIGURE 11

deviation plotted is within a factor of ±2. It can also be inferred from these data that the iodine is as available from the controlled releases as it is from the event-related aerosols.

Figure 12 is a plot of the particle size of the aerosol versus the deposition velocity. This latter value is calculated from the deposition on the planchets divided by the integrated air concentration at the same location. Two points are plotted for the Project Hare experiment, one for each type of forage used. The lower deposition velocity for the 23 μm aerosol (Hayseed) suggests that air bouyancy exerts some effect on large particles of these aerosols. The data for Project Mice, deposition velocity of 0.51, are in close agreement with the average value of 0.65 reported for the first two Controlled Environmental Radioiodine Tests conducted at the National Reactor Testing Station.[9] The elemental iodine was generated in the same manner during those studies.

Finally, on two studies, Projects Hayseed and Hare, the ratio of the deposition actually found on two types of forage to the value estimated from the planchet data was calculated. On Sudan grass, this ratio was 3.28 for Project Hayseed and 1.46 for Project Hare. On alfalfa, these values were 2.81 and 1.66 for the two studies, respectively. Since forage is a three dimensional sampler, the ratio would be expected to be greater than unity.

CONCLUSIONS

1. Aerosols of known particle size distribution can be generated in such a manner as to yield a known activity deposition level.
2. Lateral deposition was uniform for these studies and the downwind distribution was a function of the particulate size as expected.
3. The deposition velocity value can be used to estimate the aerosol particle size over a range of diameters.
4. Planchets can be used to estimate the activity per kilogram to be expected in forage. This estimation is more sensitive to particle size than it is to forage type.

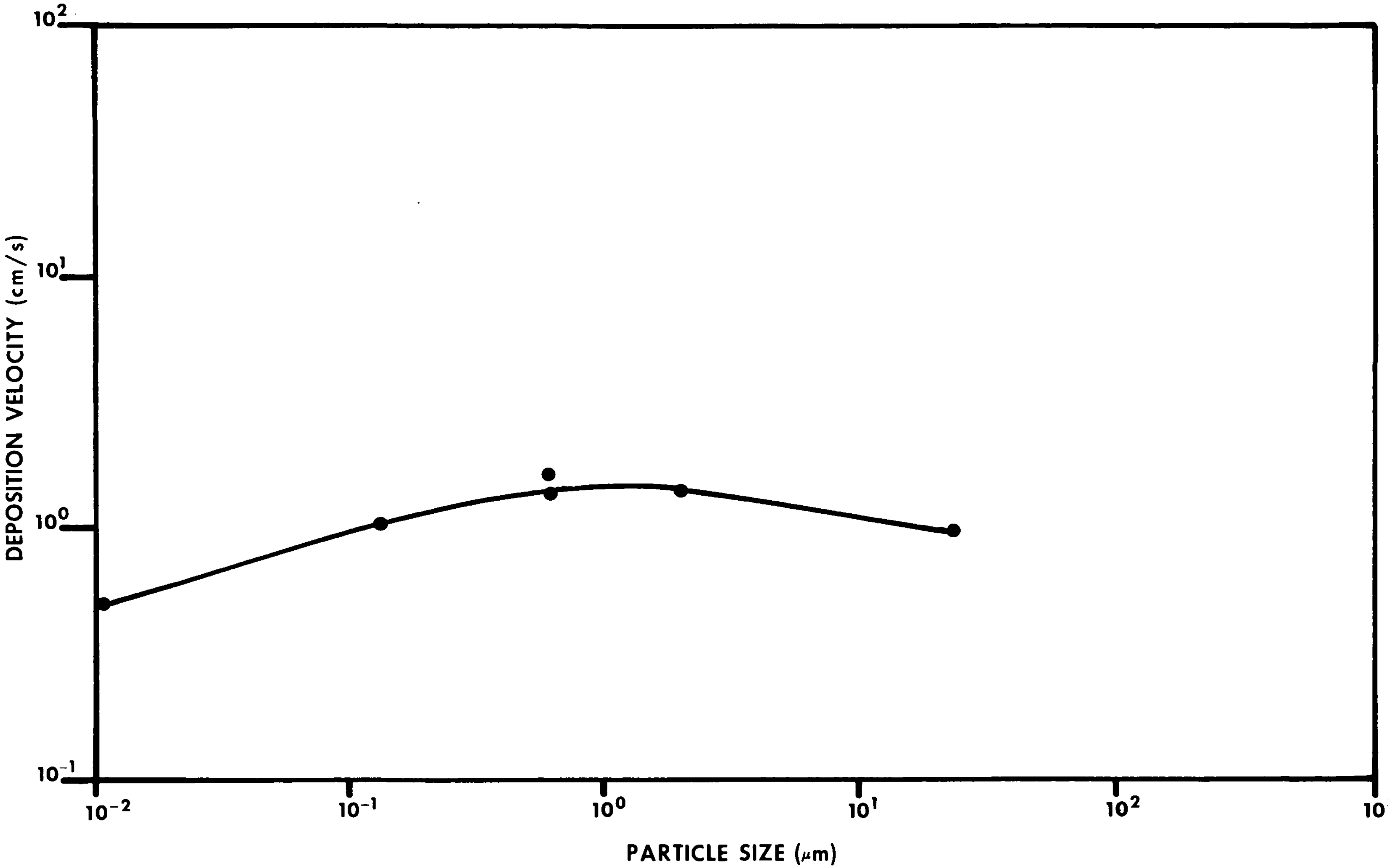


FIGURE 12 — PARTICLE SIZE VERSUS DEPOSITION VELOCITY FOR FIVE ^{131}I CONTROLLED RELEASES

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