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RESULTS OF A TEST OF SAMPLING IN I-131 PLUMES

Report Prepared by

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HANFORD LABORATORIES OPERATION
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RESULTS OF A TEST OF SAMPLING IN I¹³¹ PLUMES

TABLE OF CONTENTS

- I. Introduction
- II. Summary
- III. Emission Control
- IV. Sample Collections
- V. Meteorology Data
- VI. Analytical Method Used for Filter Samples
- VII. Results
 - A. Samples Collected with the Hanford AEC Beechcraft
 - 1. Equipment
 - 2. Results from Gross Gamma Monitoring
 - 3. Results from Filter Samples
 - B. Samples Collected
 - 1. Equipment Used
 - 2. Results from Samples Collected at 50 Miles
 - 3. Results of Close-in Sampling
 - C. Environmental Monitoring Samples
 - 1. Methods
 - 2. Results
 - D. Collection Efficiencies of Filters by Ram Sampling
- VIII. References

1083974

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LIST OF TABLES

PAGE NO.

I. Upper Winds at Hanford	18
II. Upper Level and Surface Winds at Selected Locations.	19
III. Hourly Wind Speeds and Directions	20
IV. I ¹³¹ in Air Sampled from the Hanford AEC Beechcraft.	29
V. I ¹³¹ in Background Filter Samples Taken September 13, 1962	33
VI. I ¹³¹ Results for Filter Samples Taken at Approximately 50 Miles on September 14, 1962	36
VII. Radioxenon and Kr ⁸⁵ in Air Samples	45
VIII. Results for Miscellaneous Samples Collected at Less than 50 Miles from the Source, September 14, 1962	50
IX. Summary of Air Sampler Locations	56, 57
X. Relative Collection Efficiency of the Filters in the Checkerboard Arrangement	76

LIST OF FIGURES

PAGE NO.

1. I ¹³¹ Emission Rate from Redox Stack	9
2. Plume Centerline Positions for Emissions from 0000 Z to 0800 Z, September 14, 1962.	13
3. Plume Centerline Positions for Emissions from 0900 Z to 1800 Z, September 14, 1962.	14
4. Gross Gamma Meter Readings on Sampling Runs Made by the Hanford AEC Beechcraft During I ¹³¹ Emission	28
5. Total Beta Activity on Filter Samples Taken in the 700 Area, Richland, Washington.	37
6. Particulate I ¹³¹ Activity on Filter Samples Taken in the 300 Area	37
7. I ¹³¹ Values on White Filters at 50 Miles Downwind	38
8. I ¹³¹ Values on Black Filters at 50 Miles Downwind	39
9. Sum of I ¹³¹ Values on White and Black Filter Pairs.	41
10. I ¹³¹ on the Filter Pairs.	42
11. Kr ⁸⁵ Concentrations in Air Samples Taken on September 14, 1962	46
12. Kr ⁸⁵ Concentrations in Air Samples Taken on September 13, 1962	47
13. Gamma Spectrum of a C-1 Filter Sample	53
14. Local Air Sampling Stations	54
15. Charcoal Absorber Tube Assembly	61
16. Milk and Pasture Grass Sampling Locations	64
17. I ¹³¹ in Air Measured Via Caustic Scrubbers.	66
18. I ¹³¹ in Air Measured Via Charcoal Cartridges.	68
19. I ¹³¹ in Sagebrush	70
20. I ¹³¹ in Pasture Grass	70

1083976

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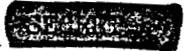
LIST OF FIGURES (Continued)

PAGE NO.

21. I ¹³¹ in Milk	70
22. I ¹³¹ in Local Pasture Grass and Milk	71
23. Filterable Gross Beta Activity in Air - Daily Samples. . .	73
24. Checkerboard Arrangement of the Test Filter Papers in the Ram Sampler.	75

1083977

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I. Introduction

On September 13 and 14, 1962, 8.3 curies of iodine-131 were emitted from the Hanford Redox Plant at a rate of from 0.35 to 0.65 curies per hour for a period of approximately 18 hours. During the emission, the plume trajectories were plotted from meteorological data and samples were collected across the predicted plume trajectories at several altitudes and at distances up to 50 miles from the Plant. Filter, charcoal trap, and air samples were collected during the emission using aircraft; and caustic scrubber, vegetation, and milk samples were collected during and after the emission at selected ground stations in the usual manner. Appropriate background samples were collected before the test began. The data and conclusions are given in this report.

II. Summary

A controlled iodine-131 emission from the Redox Plant was started at about 1700 local time on September 13, 1962, and continued for about 18 hours. The emission rate was controlled between 0.35 and 0.65 curies per hour. During the emission, filter and air samples were collected using aircraft and fixed ground collectors. Following the emission, sets of vegetation and milk samples were also collected. All samples were analyzed locally for I¹³¹ and some air samples were analyzed at ANL for krypton-85 and radioxenon.

The most important set of samples was collected on the morning of September 14, along an arc about 50 miles downwind from the Redox Plant. Filter and air samples were collected at regular intervals along the arc in three passes, at 1000, 2000, and 3000 feet above the terrain, respectively. Guidance in choosing the sampling time and altitudes and in selecting the arc to be flown was supplied by the Hanford meteorology staff, using plume trajectory predictions.

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As predictable meteorologically, no significant I^{131} concentrations were observed on the filters exposed at 3000 feet, but significantly high I^{131} results were obtained in the predicted plume positions at 1000 and 2000 feet. Background flights made at 1500 and 3000 feet over the same terrain on the previous day, before the emission was started, provided reference data.

Other filter samples were taken over the range of about 1 to 25 miles downwind of the Redox Plant using the Hanford Beechcraft plane [REDACTED]

The I^{131} plume was detected at several points out to about 10 miles by means of a gamma scintillation monitor carried in the Beechcraft. A Geiger counter-filter assembly [REDACTED] was not sufficiently sensitive to detect the plume. High I^{131} values were obtained on many filters collected downwind of the emission point out to about 10 miles, but the results at 20 to 25 miles were not clearly positive unless more than one pass was made through the plume. The sensitivity for detection of the plume by filter sampling was seriously hampered by the high concentration of fresh fallout in the air at the time of the test. Greater air flow rates through the filters would also have increased the sensitivity.

High Kr^{85} levels were found in some air samples collected at 50 miles, showing the presence of a plume from the separations plants in the vicinity expected. The Kr^{85} detected was virtually all emitted from the Purex Plant, however, rather than Redox, the concentrations being just as high on September 13 as on September 14.

A large number of air filter, vegetation, and milk samples were collected at ground stations before, during, and after the test emission. They were collected over a large area about the Hanford site, some as part of the normal regional monitoring program at Hanford, and others as special samples following the test. From the samples collected on September 14, I^{131} concentrations above the background due to fallout were found only in caustic scrubber air samples collected in the 200-E

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and 200-W areas and in sagebrush samples collected in the 200-E and 200-W areas and near the 100-F area. A high value obtained on one milk sample, collected near Moses Lake, Washington, on September 18, may be partly due to the emission. A pasture grass sample collected at the same location did not correlate with the milk result. Variations of I^{131} concentrations in other vegetation samples collected during the week following the test could not be considered significant because of an abrupt increase in fallout levels of I^{131} occurring within a day after completion of the test.

III. Emission Control

The I^{131} was generated by carrying out a dissolution of irradiated uranium in a Redox dissolver by a slow simmer process. The rate of dissolution was controlled by the temperature of the dissolving solution, as regulated by the steam supplied. Additional control of the emission rate was achieved by passing the dissolver off-gases through or around the silver nitrate absorbers, as necessary. A calibrated, continuous I^{131} monitor* on the plant exhaust stack provided the primary means of monitoring the emission rate. Dissolution rate was monitored by means of a continuous recording of the specific gravity of the dissolver solution, following its rate of increase. The records of the I^{131} emission rate from the Redox stack and the rate of change of the specific gravity of the dissolver solution are shown in Figure 1.

During the period of this test the Purex Plant was dissolving feed at a normal rate, but the cooling time of all of the material was greater than 190 days. I^{131} emissions from Purex during this period were negligible.

*The monitoring instrumentation has been described in reference 6.

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11

IV. Sample Collections

Approximately 11.5 hours after the radioiodine emission began, sampling flights were started. The twin-engine Beechcraft of the Richland Operations Office, AEC, was used for collecting particulate filter and charcoal filter samples and for measuring gross gamma radiation levels downwind of the Redox plant within and near the Hanford reservation. Sample collecting flights were made out to about 25 miles from the Redox plant. The airplane was operated by personnel of the Security Division, RLOO, and the sampling and counting equipment carried were operated by personnel of Hanford Laboratories. Further details about the sample collecting equipment and the results are given in Section VII.

In addition to the above, a large number of filter, caustic scrubber, vegetation and milk samples were collected during and after the test period through the air sampling station and regional monitoring network of the Hanford Environmental Studies and Evaluation Operation.

V. Meteorological Data

In order that the locations where air samples should be taken could be specified accurately, sufficient meteorological support to define the position of the plume at any time was necessary. Continual meteorological consultation to the control center to aid in the dispatching of the aircraft and in determining flight levels was required. This support was given by personnel of the Atmospheric Physics Operation, Hanford Laboratories. This force consisted of groups of specialists

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experienced in the vagaries of the local winds, and the diffusive properties of a plume released to the atmosphere and their relationship to the ambient meteorological conditions. The problem was similar to those with which the group is trained to cope in its normal responsibilities to the Hanford complex.

The predictions of the plume positions were based on measurements of the air properties at and about the source of the release (which is shown in Figure 2 and 3). Measurements of wind and temperatures which would initially affect the release were taken 2 miles northeast of the source at the Hanford Meteorology Tower. This 410-foot tower is instrumented at 50-foot levels for temperature, dew point, and wind direction and speed. The wind direction and speed through deeper levels of the atmosphere were determined at more or less regular intervals by the tracking of balloons near the Meteorology Tower. Mean wind vectors representative of 1200-foot layers were obtained. On a larger scale, wind measurements were available from other stations removed from the Hanford area. The most useful of these data, because of the trajectory of the cloud, were obtained from the Air Force Base at Moses Lake, 50 miles north-northeast of the source. These data were 5-minute averages of wind direction and speed centered on the hour. Significant deviations from these observations that occurred between reporting periods were also noted and factored into the analyses. Other wind and temperature measurements were received from Spokane, Seattle, The Dalles, Pendleton, Yakima, Ephrata, and Lewiston, Idaho, from routine Weather Bureau and aviation broadcast facilities and from the Richland and Pasco airports. Additional wind data which served to yield more detailed information for the intermediate scale were obtained from a telemetering network of several remote stations located throughout the Hanford area, outlined by a dashed line in Figures 2 and 3. These data were 1-hour averages from a 23-foot mast at each station. An important supplementary source of data was a roving

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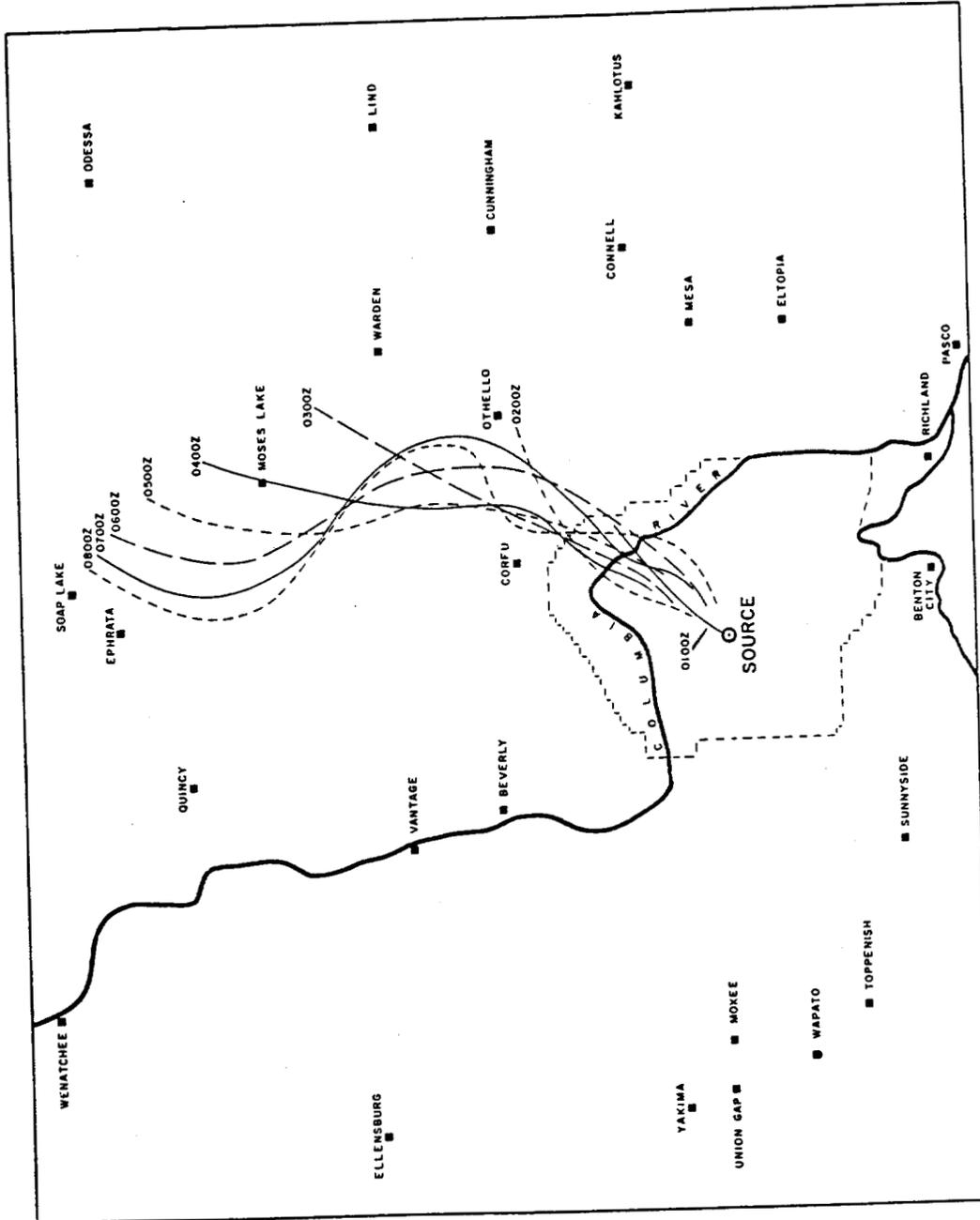


FIGURE 2

Plume Centerline Positions for Emissions From 0000Z to 0800Z Sept. 14, 1962. Centerlines Were Derived from Analyses of Trajectories Based on Meteorological Observations.

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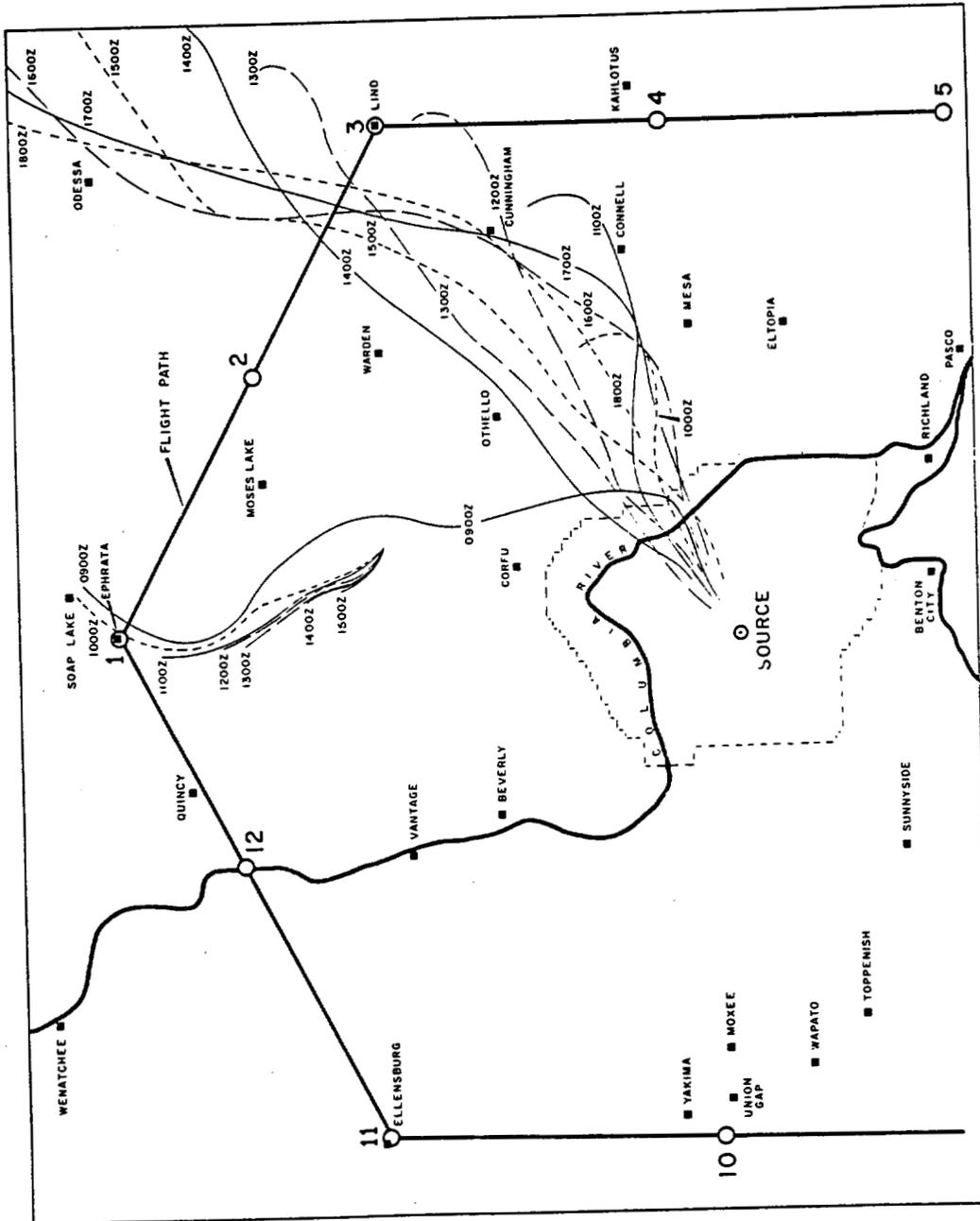


FIGURE 3

Plume Centerline Positions for Emissions From 0900Z to 1800Z Sept. 14, 1962. Centerlines Were Derived from Analyses of Trajectories Based on Meteorological Observations.

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team which could be dispatched to any location to obtain surface and upper level winds by tracking balloon releases.

Trajectories of material were obtained from the network of wind data that was available. The roving team was dispatched to areas of interest to obtain more detailed information than would otherwise be available, and would often obtain data between the regular reporting periods. These data analyzed in conjunction with the continuous records at the Hanford Meteorology Tower and the data-on-request from the local airports enabled a close surveillance on the plume. The upper winds were valuable in the qualitative assessment of the wind shearing effect on the emitted plume and were also used in the anticipation of wind changes in the lower levels.

Temperature profiles from the Hanford Meteorology Tower, Spokane, Salem, and Seattle were used to determine the stability of the atmosphere which directly controls the vertical dispersion of the plume and was thus a primary consideration in the determination of the flight level.

Meteorological support was given to the problem early on the morning of September 13 with a general analysis of the weather conditions at that time which led to the prediction of the trajectories from releases later in the day. This early analysis, although necessarily quite general because of the sparsity of observations in certain areas, proved to be valuable in that those areas which would be affected by the planned release later that day were correctly designated, and samples of the background were obtained on this basis. As it turned out, the variability in the background samples that were obtained gave rise to the consideration that some of the samples could, in addition to what is normally considered background, include materials which were stack products of other Hanford processes. This is discussed in detail in the section of this report which pertains to the

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analysis of the background samples. The meteorological problem here is to designate those areas which would be affected in the event that additional sources did affect the observed background observations.

From 0800Z to 2200Z September 13, the general circulation over the area of interest was such that any material released from Hanford into the atmosphere would be transported north to northwest of that area into a region in which the low level air flow is greatly influenced by rugged terrain. Wind measurements from the Hanford Tower, Richland Airport, the roving pibal team, Yakima, Ephrata, and Wenatchee show that a persistent southeast to northwest flow characterized the period, but the separation between these stations is too great to account for all of the local influences which would affect such a plume. The preferred path of such a release would follow the Columbia River Valley from Hanford northward toward Vantage, but it is also quite probable that portions of the release material would be influenced by many local small scale circulations that would carry it upslope into the valleys in and about Ellensburg to the west. From the data which were available, it is not possible to accurately position the plume at any particular time. However, it would be very likely that the segments 11 to 12 and 12 to 1 marked on Figure 3 would be affected by such a release. In the event that a release in this period is judged significant and is factored into the analysis, the most reasonable assumption is that the samples obtained in these segments do reflect a contribution from this plume and, therefore, are not true background data.

Meteorological support was intensified near the time of release at 2300Z, September 13 and continued from that time until the termination of the sampling at approximately 1900Z September 14. At the time of the release, the wind had shifted slightly, carrying the material to the northeast over a region in which the effect

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of topography on the wind variability was less than that which was previously observed. The plume trajectory could be more precisely predicted with the data which was available in this region. Table I summarizes the winds at the Hanford station as determined by the balloon ascensions. The time in the table is Z or Greenwich time. (To obtain local time, which was Pacific daylight savings time, 7 hours is subtracted from the values given.) The columns are headed by elevations at which the observations apply. Each entry in the table is the direction from which the wind is blowing in degrees and the wind speed in mph. Each entry is the mean wind vector in a layer 1200 feet deep centered about the given elevation. Table III summarizes the upper wind observations from the roving team. This table can be interpreted in the same manner as Table I, except that the surface wind was estimated at ground level at the location where the observation was taken. The location of the observation is also entered in the last column. Table III summarizes data obtained from the 410-foot tower. The wind measurements are entered in terms of the 16-point compass notations (i.e., V = variable, S = south, SW = southwest, etc.) and these indicate the direction from which the wind is blowing. The wind velocities are hourly averaged centered about the time entered.

The temperature difference between the 400 and 200-foot levels as measured on the tower ($T_{400} - T_{200}$) and the difference between the 300 and 100-foot levels ($T_{300} - T_{100}$) are indices of the degree of stability in the lower atmosphere. These temperature differences are listed in Table III and are 5-minute averages obtained one-half hour prior to the time designated in the table. These quantities are related to the vertical growth of the diffusing plume and are useful in estimating the altitude at which the plume can be detected. Values in the table greater than (-)0.6 are associated with gradual vertical plume growth and the atmosphere is described as being stable. Moderately stable conditions characterized the

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TABLE I

UPPER WINDS AT HANFORD

DATE TIME	ELEVATION (FEET)															
	680	1360	1995	2630	3235	3840	4430	5020	5610	6200	6790	7380	7970	8650	9150	9740
9-13 1643	115-12	125-9	140-7	175-7	210-9	215-12	220-16	240-20	250-27	250-26	240-26	240-27	240-28	240-28	240-29	
9-13 1755	110-7	150-3	180-4	210-6	215-12	210-14	215-16	225-23	230-25	235-25	245-25	255-27	260-29	260-26		
9-13 2011	160-7	190-6	200-9	210-10	230-17	250-23	250-24	240-25	235-26	235-29						
9-13 2100	150-4	195-4	200-9	210-8	225-14	235-18	240-25	245-29	245-28	240-28	240-27	235-27	230-28			
9-13 2200	265-1	220-2	215-5	225-8	235-14	235-19	230-23	230-23								
9-13 2300	240-14	250-16	255-17	255-14	245-16	235-18	230-16	225-11	230-37	230-43	230-36	235-36	230-39	225-41		
9-14 0005	250-25	250-27	240-25	240-31	240-34	235-16	230-16	220-20	225-38	230-43	240-41	250-41	260-27	255-25	250-23	250-27
9-14 0105	240-18	235-23	225-23	235-22	240-35	240-43	240-45	240-45	240-30	240-16	230-9	215-7	225-7	250-7	245-12	245-14
9-14 0202	225-16	225-21	225-18	230-16	235-20	245-21	245-26	240-27	240-28	240-29	235-29	230-25	230-21	230-18	230-20	235-25
9-14 0308	230-21	235-21	245-21	245-21	250-27	250-28	250-25	250-34	250-32	250-28	245-21	230-21	225-24	225-31	220-31	220-35
9-14 0401	235-24	240-29	240-29	235-21	235-23	240-18	245-18	250-20	250-24	245-27	235-24	225-34	205-24	175-16	185-18	
9-14 0555	240-25	250-39	250-35	250-22	255-21	255-18	245-14	240-12								
9-14 0642	245-14	245-15	255-16	260-13	260-14											
9-14 1000	250-20	240-24	250-24	260-21	250-25	240-26	235-28	230-27	230-28	230-28	230-28	235-28	245-24			
9-14 1100	245-23	245-23	255-23	255-23	250-21	240-20	230-24	225-28								
9-14 1606	255-14	250-12	260-9	265-9	255-12	255-18	250-23	240-26	245-30	230-32	220-30	225-20	225-30	225-33	230-35	235-35

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TABLE II
UPPER LEVEL AND SURFACE WINDS AT SELECTED LOCATIONS

DATE TIME	SFC WIND	ELEVATION (FEET)										LOCATION		
		680	1360	1995	2630	3235	3840	4430	5020	5610	6200		6790	
9-13 2155	--	130-7	150-9	160-9	190-10	210-16	210-23	210-23	215-20	220-18	220-18	220-18	220-18	Near Beverly
9-14 0845	--	215-18	250-20	260-23	245-18	230-17	220-18	215-20	215-20	220-18	220-18	220-18	220-18	2 mi. north of Othello
9-14 1033	225-11	230-24	230-32	225-34	230-28	235-24	235-26	235-28	230-30	225-33	225-33	225-34	220-37	5 mi. north of Othello
9-14 1130	210-9	225-16	230-24	230-24	230-24	225-25	220-24	215-22	205-24	210-23	220-28	220-28	230-30	5 mi. north of Othello
9-14 1234	205-7	225-17	235-20	230-21	220-24	215-26	215-23	220-23	225-26	225-30	225-33	225-29	230-28	5 mi. north of Othello
9-14 1332	220-12	230-14	240-17	245-17	235-18	225-19	220-24	220-23	220-30	220-35	225-29	225-29	220-40	5 mi. north of Othello
9-14 1445	195-16	215-16	240-17	245-18	240-19	230-23	230-23	225-20	230-25	220-34	215-40	215-40	220-40	5 mi. north of Othello
9-14 1603	185-16	200-16	220-18	235-17	245-18	240-28	240-28	230-30	220-18	215-43	225-33	225-33	235-24	10 mi. east of Othello
9-14 1647	180-12	215-20	220-17	230-15	240-15	240-19	240-24	240-29	235-37	230-38	220-39	220-39	210-37	10 mi. east of Othello

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TABLE III
HOURLY WIND SPEEDS AND DIRECTIONS

DATE	TIME Z	HANFORD METEOROLOGY TOWER							T ₃₀₀ -T ₁₀₀	T ₄₀₀ -T ₂₀₀	T ₃₀₀ -T ₁₀₀	MOSES LAKE SURFACE WIND
		7	50	100	200	300	400	T ₄₀₀ -T ₂₀₀				
9-13	2030	4	ESE4	4	ESE4	4	ESE4	4	ESE4	-0.9	-1.0	SW5
9-13	2130	2	V2	2	SSE2	2	SSE2	2	SSE2	-0.4	+0.3	SE4
9-13	2230	4	WSW5	5	SW6	6	SW6	6	SW6	-1.1	-0.3	SSE5
9-13	2330	7	SW10	11	SW12	12	SW13	12	SW13	-1.3	-1.2	SE4
9-14	0030	10	SW14	16	SW17	17	SW18	17	SW18	-0.4	-0.9	SE3
9-14	0130	7	SW10	11	SW13	13	SW13	13	SW13	-0.8	-0.1	SE6
9-14	0230	5	SSW10	13	SSW15	16	SSW18	16	SSW18	-0.3	+0.8	S8
9-14	0330	8	SSW14	17	SSW20	21	SSW23	21	SSW23	-0.6	+0.2	SE2
9-14	0430	7	SSW11	13	SSW17	18	SSW20	18	SSW20	-0.3	+0.5	Calm
9-14	0530	4	SW6	7	SW10	11	SW14	11	SW14	-0.9	-0.5	Calm
9-14	0630	7	SW11	15	SW18	19	SW21	19	SW21	+0.2	+0.8	Calm
9-14	0730	2	W5	8	W10	11	W12	11	W12	+0.1	+1.9	Calm
9-14	0830	2	W7	9	WSW12	14	WSW16	14	WSW16	+1.5	+2.3	Calm
9-14	0930	5	W10	12	WSW13	14	WSW16	14	WSW16	-0.3	+0.6	Calm
9-14	1030	6	W10	14	WSW16	15	WSW16	15	WSW16	-0.5	+1.0	Calm
9-14	1130	8	WSW14	17	WSW19	18	WSW20	18	WSW20	+0.0	+1.5	Calm
9-14	1230	6	WSW11	14	WSW17	18	WSW21	18	WSW21	+0.6	+1.7	Calm
9-14	1330	3	WSW6	8	WSW10	13	WSW16	13	WSW16	+1.1	+2.1	Calm
9-14	1430	2	WSW6	7	W10	11	W14	11	W14	-0.5	+1.2	Calm
9-14	1530	6	WSW7	9	WSW10	11	WSW12	11	WSW12	-0.4	-0.6	SW3
9-14	1630	4	WSW6	6	WSW6	6	WSW6	6	WSW6	-1.3	-0.8	SSE5
9-14	1730	7	WSW8	9	WSW10	10	WSW10	10	WSW10	-0.8	-1.2	
9-14	1830	8	SW11	12	SW13	12	SW13	12	SW13	-0.7	-0.3	

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lower atmosphere during most of the period of release and hence also during the period during which the samples were obtained. Estimates of the vertical extent of the plume at 50 miles downwind based on a reasonable extrapolation of experimental data obtained through previous Hanford research would be approximately 2000 feet. The reasonableness of this estimate has been supported further by the temperature profiles obtained by balloon releases at Salem, Oregon and Spokane, Washington. These data show that a stable layer of thickness 2000-3000 feet developed in the evening of September 13 and was sustained until 1600Z September 14. At 50 miles, the greatest concentrations could, therefore, be expected in the lower 1000 feet of the atmosphere. From 1000-2000 feet, the plume is still significant, but at 3000 feet the quantity of plume material is probably insignificant relative to background. At locations near the source, the peak could be expected between 300-800 feet.

Data from all locations were used to develop a description of the state of the atmosphere from which estimates of the dispersion and trajectories of the plume were made. Figures 2 and 3 show the trajectories that were obtained. Figure 2 shows the plume centerlines that were constructed using primarily the Hanford Tower, radiotelemetering, upper level, and Moses Lake surface winds. These are the centerlines which are associated with releases from the source from 0000Z September 14 to 0800Z September 14, 1962. The time is indicated with each trajectory so that direct evaluation of the sampling time relative to the plume centerline can be easily made. As can be seen from Table III, the conditions at Moses Lake became calm at the end of the period so that semistagnation occurred in this vicinity. The calm winds at Moses Lake should be interpreted as winds below the threshold of the anemometer used for the observation. There is little doubt that portions of the plume were still being transported, but at a much reduced rate. There is no basis for prognosis

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 beyond Soap Lake, but assuredly the width of the plume in this area will have a great lateral extent resulting from the direction variability associated with light winds and wind direction shear which was evident from the balloon runs. Figure 2 shows the plume centerlines as they existed from 0900-1800Z. It will be noted that the trajectory underwent an abrupt shift so that the centerlines are found northeast of the source. However, because of the semistagnant air to the west of Moses Lake, portions of the cloud which had been released during the night remained within that area throughout the period and sampling as late as 1830Z September 14 probably would detect it.

Figure 3 shows the plume centerlines relevant to the air sampling. The wind shift that occurred in the night, ~ 1900Z September 14, has brought about 2 prongs of dosage maximums. The first, which is directed west of Lind, is the preferred path of the material at the time the sampling commenced and the second is the result of stagnation in the Ephrata area of the release which had been made during the night. The flight path followed is indicated by the heavy line. Taking reference due east of the source and following the flight path counterclockwise, initial detection of the plume edge was expected 10 miles north of position 4. The highest concentrations, however, are to be found 5-10 miles northwest of position 3 and at position 1 (Ephrata) where stagnation had occurred. Detection is least likely in the area about position 2 where only fragments of the dispersed plume are likely. No detection was expected south of position 4 nor west of position 12.

Vegetation sample detection has much the same expectancies as those of the air sampling. However, because of the persistent nighttime trajectory, sampling near the line from Ephrata to Moses Lake to west of Othello to the source would have the highest probability of detection. The areas in which vegetation detection



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is likely are defined by the westward lobe in Figure 2 and the eastward lobe in Figure 3. It is significant that the location of the highest air dosage measurements may not correspond to those of the highest vegetation samples. Because of the stable nighttime conditions coupled with stagnation, the deposition on the ground in the westward lobe is expected to be greater than that found in the eastward lobe. Air samples, on the other hand, are expected to be a maximum in the eastward lobe where the plume dispersal has been much less than that in the westward lobe.

VI. Analytical Method Used for Filter Samples

All filter samples collected by the aircraft were initially analyzed by multi-channel gamma-ray scintillation spectrometry using NaI (Tl) detectors. The samples collected with the AEC twin-engine Beechcraft were initially counted in a five-inch well-type NaI (Tl) detector operated in anticoincidence with a large plastic scintillator shield (1). Detectable I-131 was observed on all samples collected by the Beechcraft during the emission. Zr-Nb-95 and Ru-Rh-103 activities were found on the paper particulate filters.

The filter samples were initially counted in sets of about eight or more between two opposing eight inch diameter by four inch thick NaI (Tl) detectors, or in the two inch diameter well of an eight inch diameter by eight inch thick NaI (Tl) detector. No radioactivity above background was detected on the charcoal impregnated filters collected at 50 miles. Zr-Nb-95 and lesser quantities of other "fall out" activities were observed on composites of the particulate filters (IPC paper). Above normal levels of I-131 were found on some of the composites containing filters exposed close to the source. Three five-inch diameter by five-inch thick well-type NaI (Tl) detectors were used to obtain gamma spectra on each of the "close-in" samples and on some of the IPC filters exposed at 50 miles. Estimates of the Ru-Rh-103, Ru-Rh-106, Ba-La-140, I-131, Cs-Ba-137, Ce-Pr-144, and Zr-Nb-95 activities were calculated by the GEM program (2) from the gamma-ray spectra. The two large IPC filters exposed during the entire flight were also analyzed by gamma-ray scintillation spectrometry. Above normal levels of I-131 were found on the filter.

The initial counts on composited samples indicated that the I-131 activity levels on most of the individual filter samples could not be measured by normal gamma-ray scintillation spectrometry. Consequently, these filters were analyzed by separating the iodine and counting by a multichannel beta-gamma coincidence

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method. The procedure used is as follows:

The filters were spiked with a known amount of I-125 and ashed in a quartz tube in a stream of oxygen. The off-gases from the ashing, containing the I-125 and I-131 activities were passed through a gas scrubber containing water saturated with SO₂ to collect the iodine. The iodine activity was collected by carrier-precipitation with a few milligrams of AgCl. The AgCl precipitate was dissolved with cyanide and then re-precipitated. The precipitate was finally transferred to the well of a small beta sensitive plastic scintillator and dried. The I-131 activity in the precipitate was measured using multichannel beta-gamma coincidence spectrometry. The background in the region of the I-131 photo peak was about 0.05 counts per minute. The counting efficiency as measured with a calibrated I-131 source was between 0.2 and 0.4 depending on the detector system. Each sample was counted for at least 1000 minutes. The disintegration rate of the I-131 at 2:00 p.m. on September 14, 1962, was calculated using the GEM program (2). The chemical yield of each sample was measured using a well type NaI (Tl) detector to count the I-125 X-ray and gamma-ray activity. The I-131 disintegration rates were then corrected for yields which averaged about 60 per cent.

The GEM computation program yields an estimate of the standard error based on counting statistics. An additional uncertainty was introduced in the beta-gamma coincidence measurements due to the presence of varying amounts of silver sulfate coprecipitated with the silver chloride, which reduced the beta counting efficiency and resulted in a bias. The procedure has been modified to eliminate the silver sulfate coprecipitation in the future. A few of the results may be biased on the high side due to traces of I-131 in the laboratory equipment. These results are indicated in the data presented later. It was found during these analyses that

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glassware which has never been used with I-131 tracer must be used for the chemistry.

VII. Results

A. Samples Collected With The Hanford AEC Beechcraft

The AEC twin-engine Beechcraft is periodically used for air sampling studies and for direct monitoring with a high sensitivity gamma-ray detector.

During the I¹³¹ emission, combined monitoring and sampling flights were made as planned at approximately 1, 3, 5, 10 and 25 miles from the source. Three additional flights were also made, one of which was in an "arc" at about 20 miles from the source and the other two were from points downwind to the source. The sampling paths, which were short, were traversed two or more times to give a total sampling time of about 10 minutes.

1. Equipment

The equipment used for gross gamma monitoring consisted of a 5-inch diameter by 3-inch thick sodium iodide crystal with its accompanying battery-powered photomultiplier and transistorized count rate meter and recorder⁽³⁾.

Air sampling was performed by drawing air into the plane through a 2-inch diameter port in the door of the plane at a direction perpendicular to the air flow. The air pump used was a Hurricane Air Sampler Model No. 16003 which was powered with nine 12-volt storage batteries. The air was pulled through a 9.6 square inch area of a standard IPC filter paper⁽⁴⁾ followed by a 1-inch thick bed of activated charcoal (cocoanut charcoal, 6-14 mesh, Cat. No. 5-685, Fisher Scientific Company) at a flow rate of 750 linear ft. per

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minute.

2. Results from Gross Gamma Monitoring

The general direction of wind from the separation plant stack was toward the northeast. The sampling was done by traversing the plume area at approximately 500 to 600 feet above ground level. The actual locations of the various sampling runs, plus the time at which they were flown, and the observed gross-gamma rate meter readings, are shown on the map in Figure 4. Most of the observed gamma monitor readings were rather low and probably near the normal background; however, there were a few points in the plume area which were about twice these lower values.

3. Results from Filter Samples

Measurements of the collection efficiency of particulate I^{131} and other radionuclides from fallout by IPC filter paper⁽⁵⁾ indicates that at flow rates of 750 linear feet per minute, about 50 per cent of the particulate iodine is collected by the filter. Most of the remaining particulate I^{131} is collected on the following one-inch thick activated charcoal bed. In Table IV the observed I^{131} content of the air which was sampled by the IPC filter and the activated charcoal bed are tabulated. The ratio of I^{131} on the IPC filter to that in the charcoal bed is also listed. Since the IPC filters were about 50 per cent efficient for particulate I^{131} , these ratios should be multiplied by two to determine the fraction of I^{131} actually associated with particulate material.

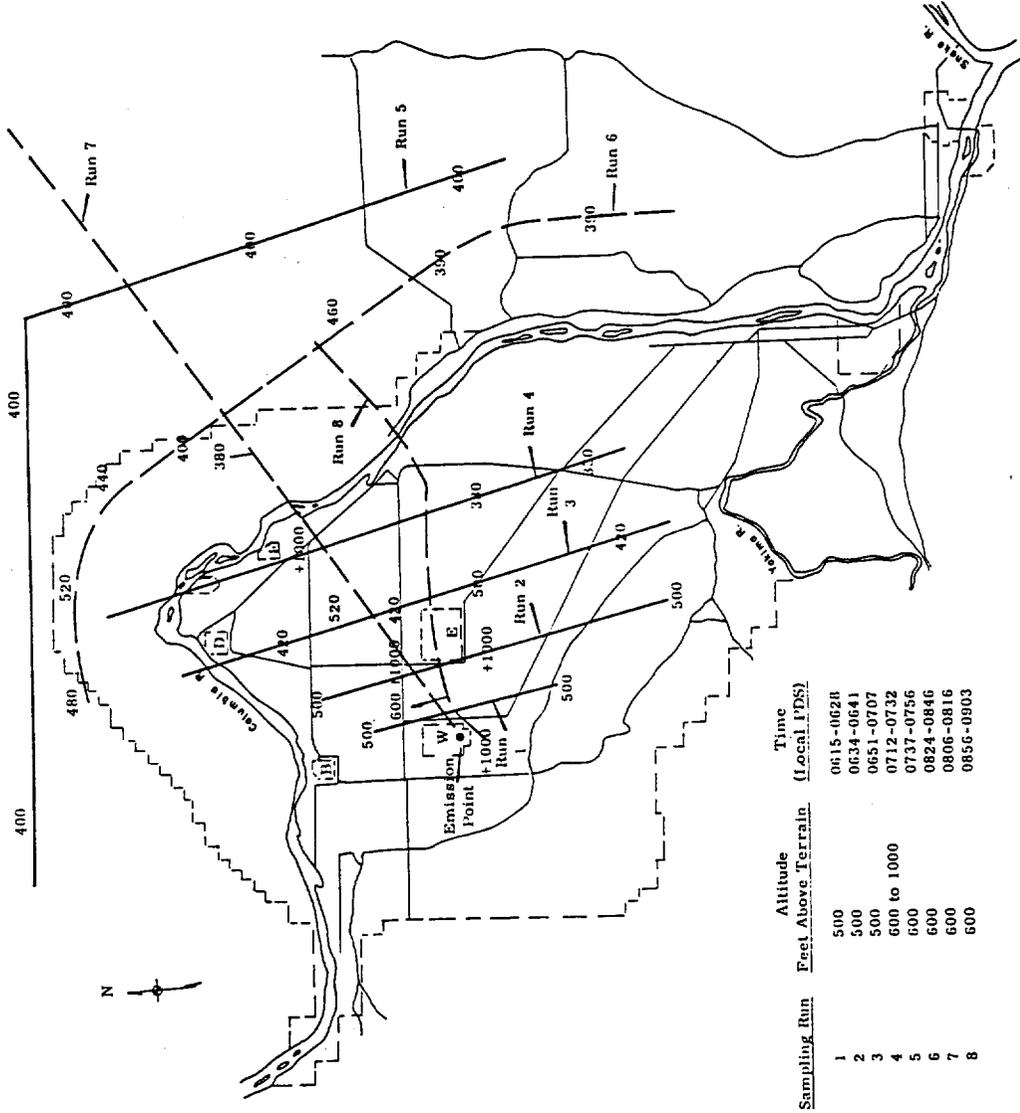


FIGURE 4

Gross Gamma Meter Readings on Sampling Runs made by the Hanford AEC Beechcraft During I¹³¹ Emission

TABLE IV

¹³¹I in Air Sampled from the Hanford AEC Beechcraft

¹³¹I in d/m/ft³

<u>Sample Number</u>	<u>Location*</u>	¹³¹ I on <u>IPC Filter</u>	¹³¹ I on <u>Charcoal Bed</u>	<u>Ratio**</u>
1	1 mile traverse	0.0101	0.156	0.065
2	3 mile traverse	0.0031	0.072	0.043
3	5 mile traverse	0.0047	0.043	0.11
4	10 mile traverse	0.0176	0.091	0.19
5	25 mile traverse	0.0074	0.0394	0.19
6	20 mile arc	0.0135	0.057	0.23
7	See map	0.052	0.143	0.36
8	See map	0.587	2.13	0.27

*See Map in Figure 4

** Ratio of ¹³¹I on the filter to that in the charcoal bed.

B. Samples Collected

1. Equipment Used

a. Filter samples

Most of the samples were collected with a five-inch filter holder in an air scoop device

The filter can be simply and quickly changed during flight and, in this experiment, the filters were generally changed at four minute or eight minute intervals.

A double filter consisting of a "white" filter backed by a "black" filter was used

The first filter was an IPC paper filter, Knowlton Paper Company type. This filter is a high strength filter having small pressure loss coefficients and high collection efficiency for the particulate matter typically produced in nuclear explosions⁽⁴⁾. The second filter was a porous fiber filter impregnated with activated carbon. It was used in this test in the hope that the black filter would absorb iodine very efficiently while the white filter ahead of it would collect most of the particulate matter; thus the ratio of iodine to the background of non-volatile fission products might be more favorable on the black filter. This proved to be of no advantage. The white filter was generally the more efficient collector of iodine (meaning, possibly, that most of the iodine was particulate) and the reduced air flow rate due to the additional filter was disadvantageous.

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b. Air samples

During all sampling runs at 50 miles, as described later, air samples were also collected for rare gas determinations. The samples were collected in spherical steel pressure bottles having a capacity of about 900 cubic inches using a positive displacement air pump operated from the aircraft power plant. Each sample was taken at a uniform rate during a specified portion of the sampling flight path.

2. Results from Samples Collected at 50 miles

a. Locations

The principal set of samples were collected on an arc about 50 miles downwind of the Redox Plant between 0730 and 0945, local time, on September 14, 1962, at which time the predicted plume location of the I^{131} from Redox was as shown on Fig. 3. The path taken by the sampling aircraft was from position 5 to position 11, as also shown on the map in Figure 3. A first pass was made at 3000 feet above the terrain starting at position 5 and ending at position 11. A second pass, made on the return flight, was at 2000 feet and a third pass was made at 1000 feet from position 5 back to position 12

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filter samples were taken continuously during these passes, with a filter change at either nominal 4-minute or nominal 8-minute intervals.

On the previous day, September 13, 1962, similar sampling runs were made between 2040 and 2200 local time to test equipment and to collect samples for establishing the I^{131} background. The flight path of the sampling aircraft on this background run was from position 4 to position 10. The first pass was made at about 1500 feet from point 4 to point 10 and the second pass was made at 3,000 feet on the return flight from point 10 to point 4. An additional background filter sample was collected during a flight from Richland Airport



b. Filter Samples for Background

The results on samples taken to establish the I^{131} background are tabulated in Table V. These samples were taken

on September 13, 1962, during which time the Redox Plant was not operating and the Purex Plant was and had been processing irradiated uranium which had been cooled about 190 days or longer. The average I^{131} emission rate during the 12 hours prior to the background sample collections was about 5×10^{-5} curies per hour. This is a factor of about 10^4 below the emission rate of the combined Redox and Purex Plants at the time of the test on September 14. The emission plume from the Purex Plant was moving Northward during this period, and the results of krypton-85 determinations show that the plane passed



TABLE V

¹³¹I in Background Filter Samples Taken September 13, 1962

Lab. Numbers	Sample Location	Exposure, Minutes	White Filters			Black Filters		
			¹³¹ I d/10 ⁻⁴ days	Std. Error	¹³¹ I on 4 min. basis **	¹³¹ I d/10 ⁻⁴ days	Std. Error	¹³¹ I on 4 min. basis **
3902-3	4 to 3, 1500'	6	0.051	0.023	0.034	0.117	0.023	0.078
3904-5	3 to 2, 1500'	7	0.162	0.032	0.092	0.080	0.013	0.046
3906-7	2 to 1, 1500'	7	0.301	0.026	0.17	0.202	0.016	0.115
3908-9	1 to 12, 1500'	7	0.140	0.050	0.08	0.193	0.016	0.11
3910-11	12 to 11, 1500'	7	0.188	0.023	0.108	0.253	0.058	0.145
3912-13	11 to 10, 1500'	8	0.047	0.021	0.0235	0.090	0.167	0.045
	Mean value:				0.085			0.090
3914-15	10 to 11, 3000'	7	0.070	0.055	0.04	0.090	0.023	0.051
3916-17	11 to 12, 3000'	8	0.272	0.034	0.136	0.120	0.031	0.060
3918-19	12 to 1, 3000'	5	0.214	0.041	0.171	0.003	0.067	0.00
3920-21	1 to 2, 3000'	7	0.172	0.033	0.098	0.010	0.027	0.006
3922-23	2 to 3, 3000'	7	0.132	0.033	0.075	0.042	0.020	0.024
3924-25	3 to 4, 3000'	8	0.109	0.023	0.055	0.099	0.038	0.0495
	Mean value:				0.096			0.032
3900-1		40	0.570	0.107	0.057	0.342	0.027	0.0342

* Std. error based on counting statistics, only
 ** Normalized to a 4-min. exposure

Over-all Average Bkgd:
 White Filters (4-min basis): 0.09 d/10⁻⁴ days
 Black Filters (4-min basis): 0.06 d/10⁻⁴ days

through the plume. These data will be discussed subsequently.

The mean values are shown for the I^{131} content of each type of filter, on the basis of a four minute exposure for each of two altitudes, 1,500 feet and 3,000 feet. The over-all mean for each type of filter was calculated and used subsequently. These mean values are not significantly different but they were not combined because it makes sense, scientifically, not to combine them. The four lowest results on the white filters occurred on the samples from the ends of the flight course, namely, at 3 to 4 and 10 to 11, suggesting a peak across the central part of the course. However, this was tested statistically and neither the differences between sample locations nor between altitudes were significant. The estimated precision of the results is the same for both types of filters. The standard error value given in Table V was based on the counting data only. The estimated over-all standard deviation of a single result, calculated from the 12 values on white filters, is about ± 0.04 d/ 10^{-4} day for a 4-minute sample, whereas the indicated precision of the counting was ± 0.016 d/ 10^{-4} day. The higher value includes variations due to sampling, laboratory processing prior to the counting, and any possible real variations in I^{131} content between samples.

As will be seen in the data presented in the next section, the presence of an appreciable I^{131} value due to fallout seriously limited the sensitivity for detection of the I^{131} release from the Redox Plant. An important variable in dealing with such an I^{131} background is that the fallout concentrations often change

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widely and abruptly with air mass changes. This effect is seen in the data in Figures 5 and 6. These data result from analyses of daily filter samples at Hanford for gross beta activity in micromicrocuries per cubic meter (Fig. 5) and I^{131} activity in disintegrations per minute per cubic foot (Fig. 6). The period during which the aircraft sampling was done is indicated. A sample for I^{131} determination was not taken at the 300 Area on September 14, because the filtering equipment was being used in the airplane that day. The weather front which passed over this area on September 14 evidently brought in an air mass with about a 5 to 7 fold higher fallout concentration. Fortunately, this occurred after the experiment was completed.

c. Filter Samples Taken During Emission

The results for the white and black filter samples collected at about 50 miles downwind of the Redox Plant during the I^{131} emission on September 14, 1962 are given in Table VI. Certain values in column seven are marked by an asterisk to indicate possible contamination, and are mentioned earlier in Section VI.

The sample position numbers identify the flight course (see Figure 3) during the interval the filter was exposed. The standard error values are based only on the counting data. The I^{131} values in columns 6 and 9 are normalized to a 4-minute exposure period and these values are plotted on Figures 7 and 8 for each of the three altitudes. The mean background value is indicated by a heavy dashed line on each graph.

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TABLE VI

I-131 Results for Filter Samples Taken at Approximately 50 Miles
 September 14, 1962

Lab. Numbers	Sample Location	Exposure, Minutes	WHITE FILTERS			BLACK FILTERS			Feet above Terrain
			I-131 d/10 ⁻⁴ days	Std. Error	I-131 on 4 min. basis	I-131 d/10 ⁻⁴ days	Std. Error	I-131 on 4 min. basis	
3886-7	5 to 4	6	0.084	0.033	0.056	0.123*	0.011	0.082	1000
3888-9	4 to 3	8	0.099	0.030	0.05	0.248*	0.012	0.124	"
3890-1	3 to 2A	3.5	0.083	0.030	0.095	0.251*	0.020	0.29	"
3892-3	2A to 2	3.5	0.187	0.045	0.21	0.169*	0.017	.19	"
3894-5	2 to 1A	3.5	0.075	0.021	0.086	0.047*	0.025	0.05	"
3896-7	1A to 1	3.5	0.109	0.021	0.13	0.169*	0.030	.19	"
3898-9	1 to 12	7	0.038	0.016	0.022	0.232*	0.040	.13	"
3926-7	11 to 12	7	0.576	0.072	0.33	0.055	0.032	.032	2000
3928-9	12 to 1	7	0.401	0.058	0.23	0.063	0.015	.036	"
3930-1	1 to 1A	3.5	0.037	0.042	0.042	0.051	0.022	.058	"
3932-3	1A to 2	3.5	0.070	0.025	0.08	0.017	0.024	.019	"
3934-5	2 to 2A	3	-0.139	0.109	0.0	-0.021	0.045	0.0	"
3936-7	2A to 3	4	0.184	0.30	0.18	0.001	0.096	0.00	"
3938-9	3 to 4	8	0.477	0.084	0.24	0.260	0.024	0.13	"
3940-1	4 to 5	7	0.052	0.053	0.03	0.089	0.021	.051	"
3942-3	5 - 4	7	0.229	0.020	0.13	0.059	0.035	.034	3000
3944-5	4 - 3	7	0.066	0.056	0.04	0.073	0.043	.042	"
3946-7	3 - 2A	3.5	0.034	0.039	0.04	0.076	0.022	.087	"
3948-9	2A - 2	3.5	0.088	0.025	0.10	0.036	0.019	.041	"
3950-1	2 - 1A	3.5	0.111	0.023	0.13	-0.079	0.048	.0	"
3952-3	1A - 1	4.5	0.053	0.011	0.05	0.044	0.022	.04	"
3954-5	1 - 12	7	0.22	0.031	0.13	0.108	0.024	.062	"
3956-7	12 - 11	8	0.292	0.061	0.15	0.167	0.082	0.084	"

* Possible positive bias due to contamination

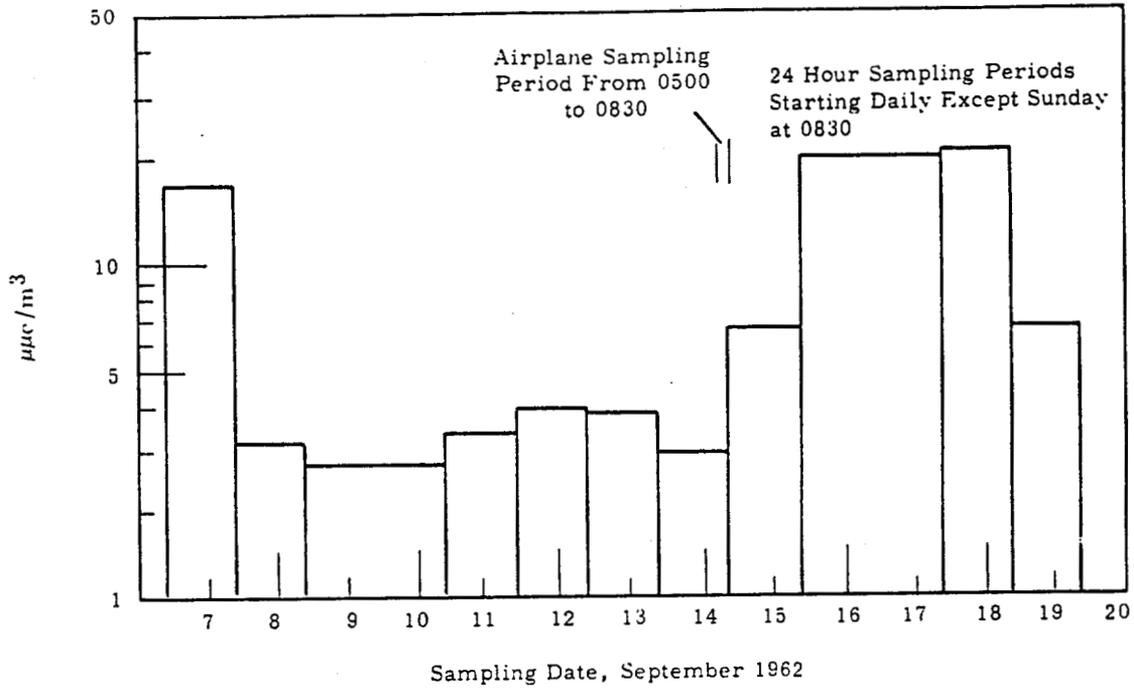


FIGURE 5

Total Beta Activity on Filter Samples Taken in the 700 Area, Richland, Wn.

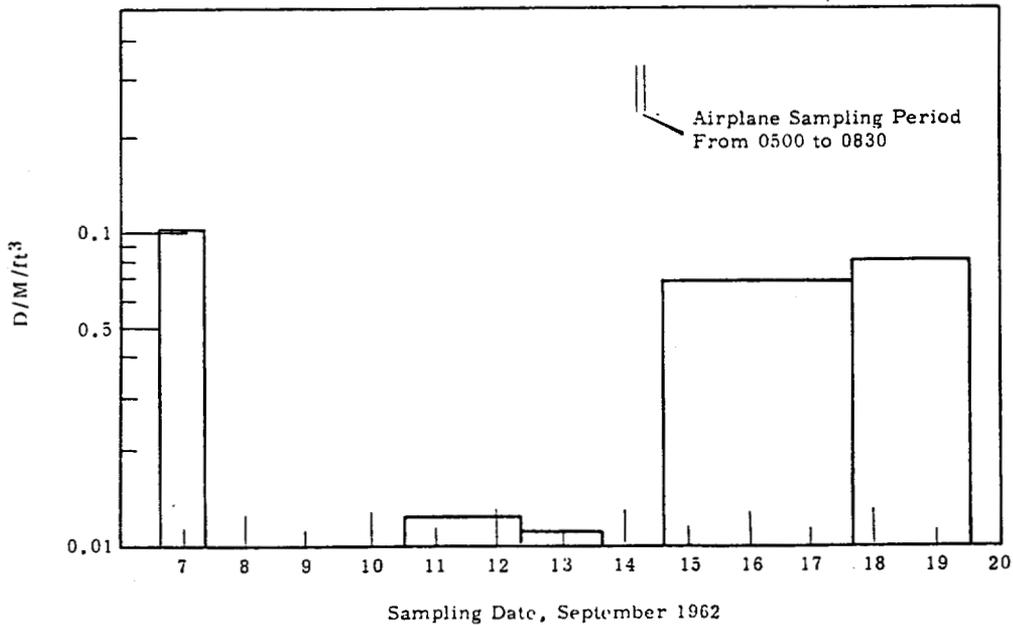


FIGURE 6

Particulate I^{131} Activity on Filter Samples Taken in the 300 Area

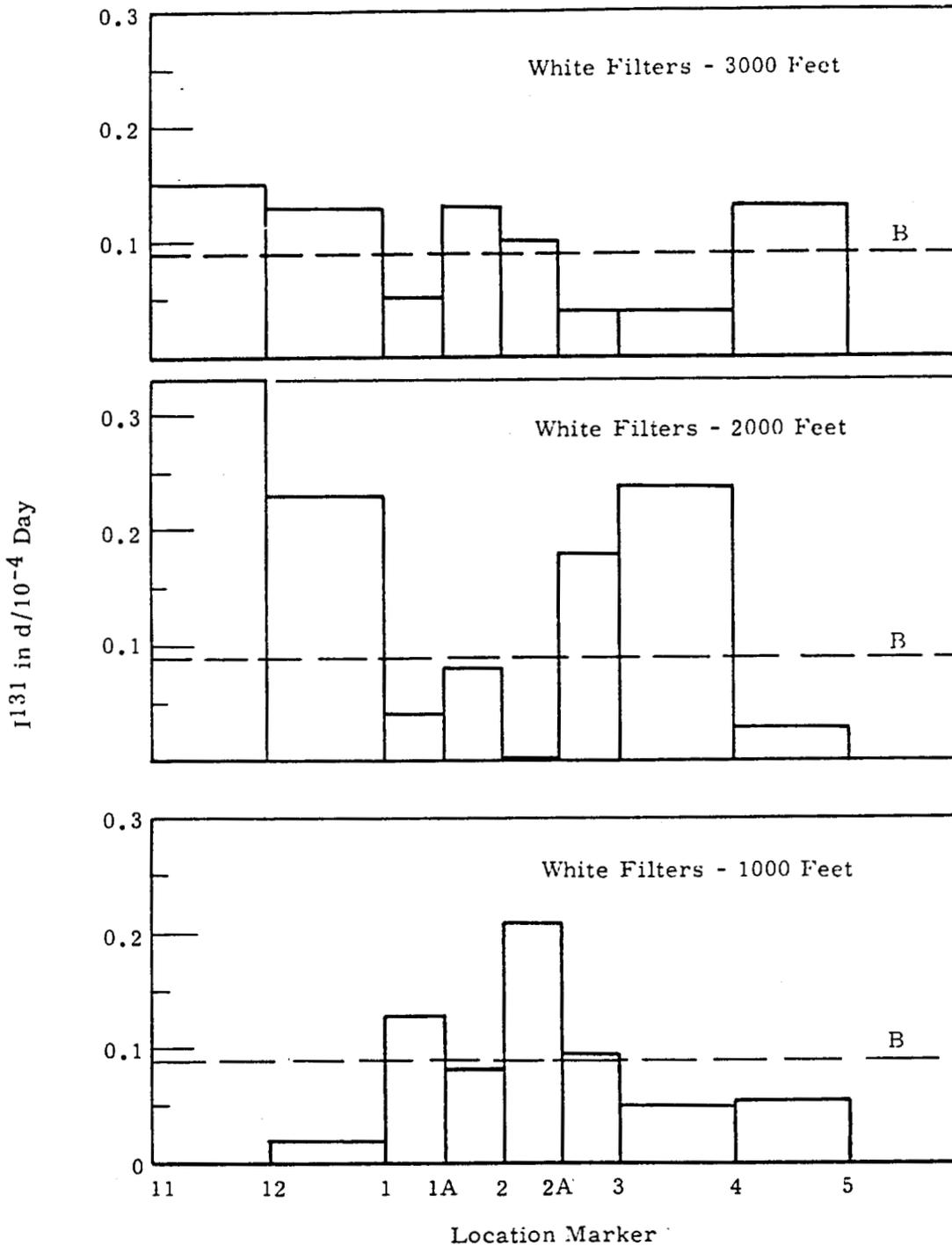


FIGURE 7

I¹³¹ Values on White Filters at 50 Miles Downwind

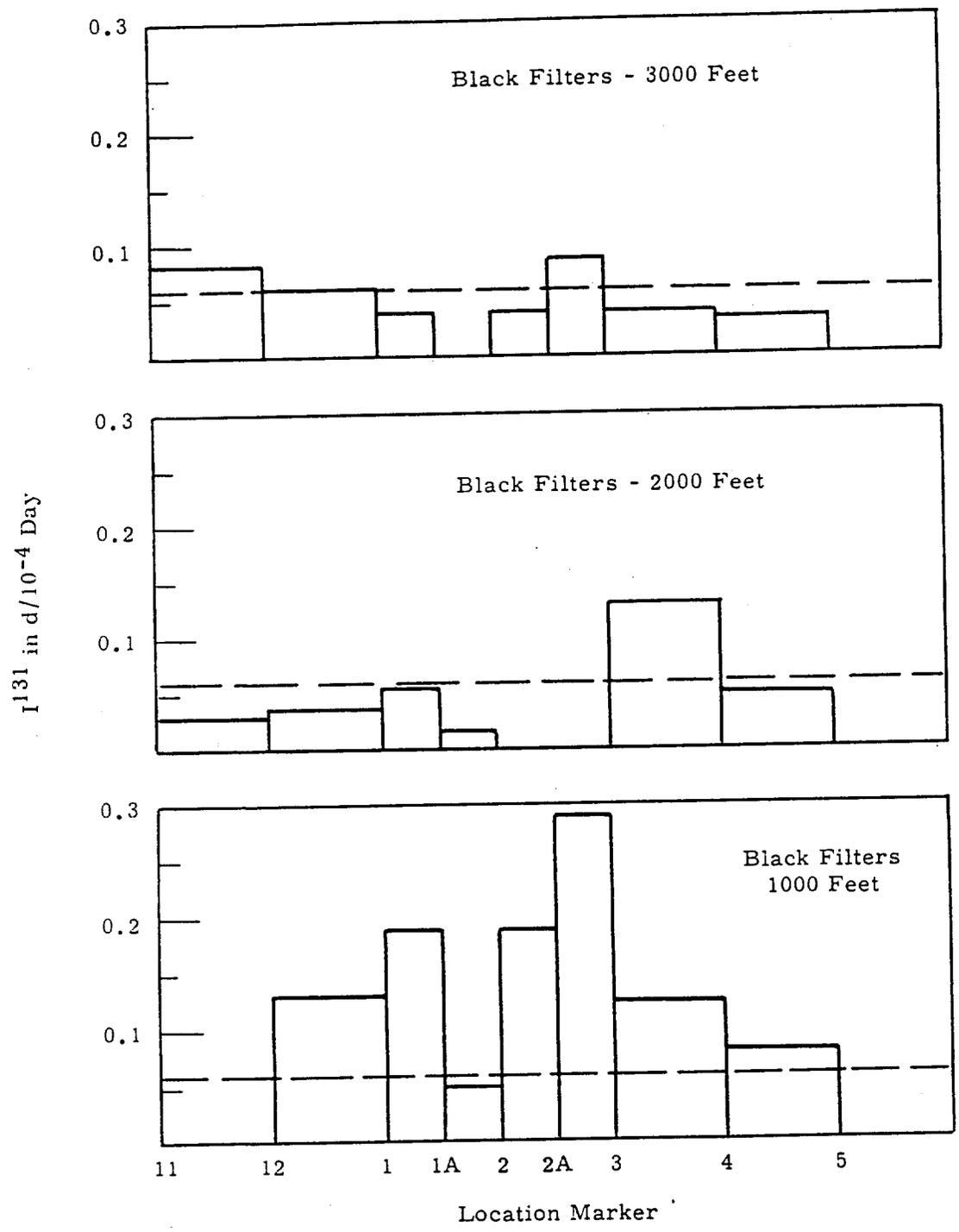


FIGURE 8

I^{131} Values on Black Filters at 50 Miles Downwind

The graphs show that I^{131} was not detectable above background at 3000 feet but the peaks occurring in the 1000 and 2000 foot data appear to be significant. It is particularly interesting that two peaks appear at 2000 feet, namely at 11 to 1 and at 2A to 4 respectively. These correspond reasonably well with the predicted plume trajectories given previously. The absence of a peak at 12 to 1 at 1000 feet is mysterious. However, the apparent displacement of the central peak from 2-2A to 2A-4 at the higher altitude suggests a shearing effect as discussed in Section V. Thus, the second peak at 1000 feet may be in the 11 to 12 position, where a sample was not taken.

The results from the black filters, shown on Figure 8, tend to support those from the white filters at 1000 and at 3000 feet. However, the results for 2000 feet were too low to be conclusive.

The total I^{131} collected on both filters is plotted in Figure 9. The results show the patterns at 1000 and 2000 feet clearly. The data are plotted in a paired manner in Figure 10, the I^{131} values on white filters being plotted against the corresponding values on the black filters. The area blocked out by a dashed line encloses what appears to be the background area.

Using the average background value for white filters of 0.09 and the standard deviation of ± 0.04 , one can compare the white filter values with the $2\hat{\sigma}$ upper limit of the background. Doing this, we observe that one result at 1000

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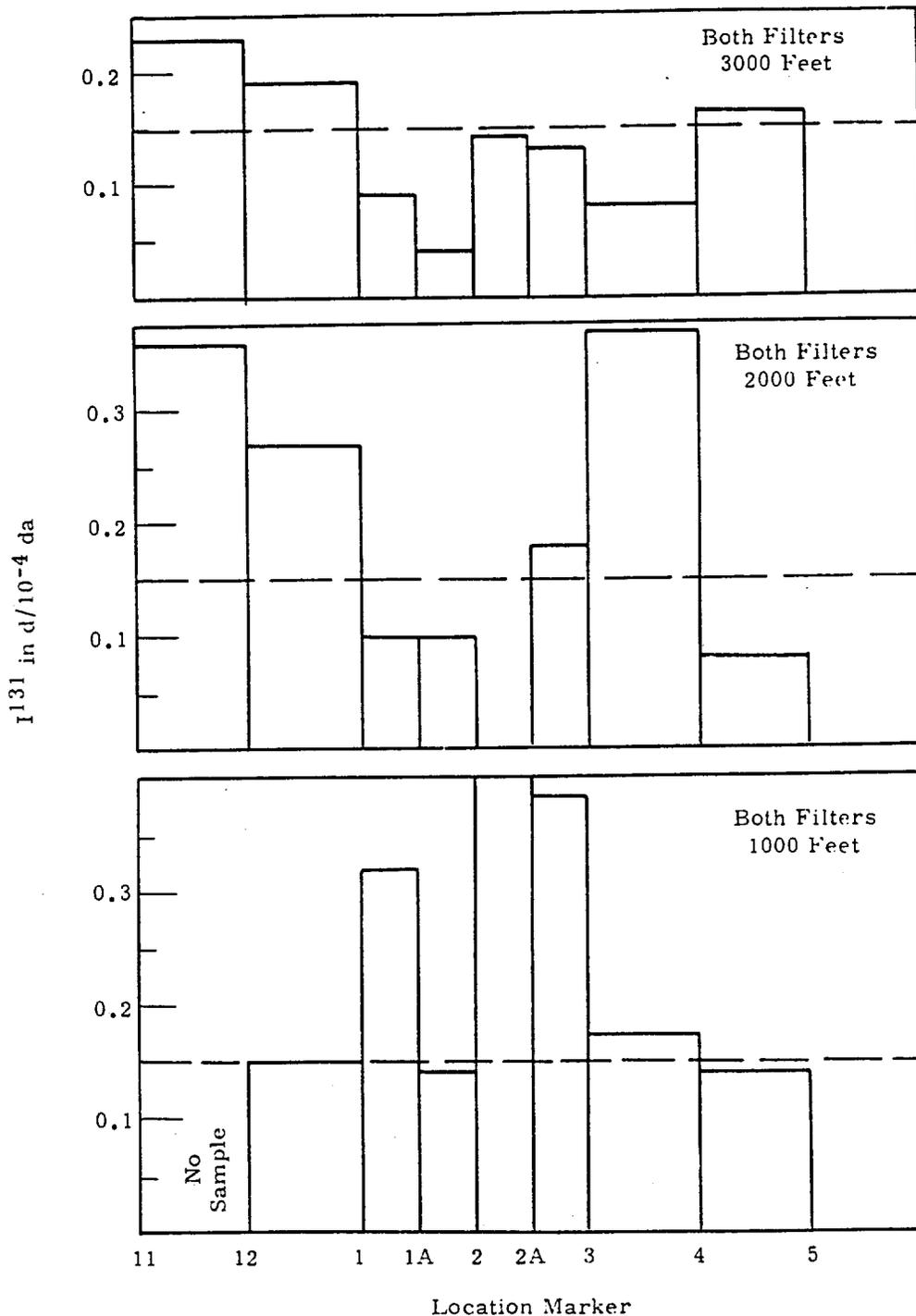
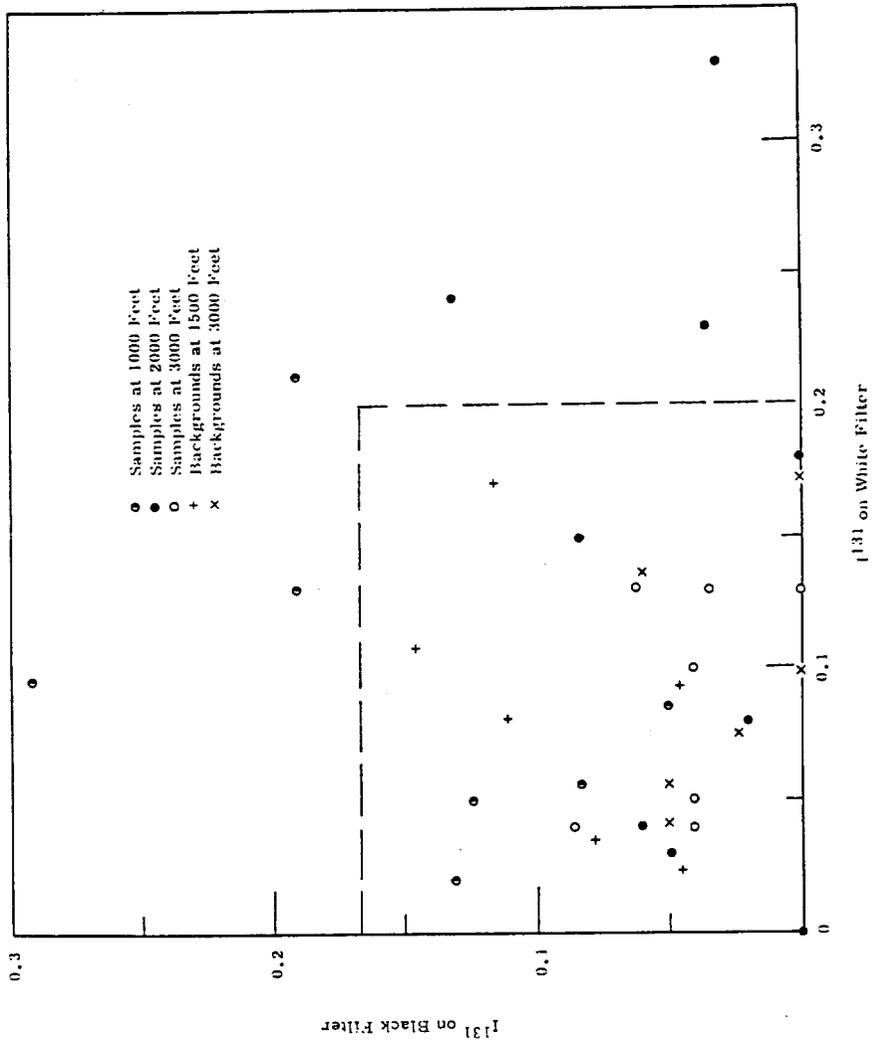


FIGURE 9

Sum of I^{131} Values on White and Black Filter Pairs



feet, three at 2000 feet, and none at 3000 feet are appreciably above the 2σ limit. However, the significance of the 2σ limit is uncertain because the statistical distribution of the results is not known and it may not be a normal distribution.

The significance of the high I^{131} values on certain filters was therefore tested by a ranking method. This was done with the combined results, that is, the sums of the paired values, as they are plotted on Figure 9. When all 23 samples and 11 background values are ranked, it is found that the highest five values are samples from 1000 and 2000 feet. The probability of a chance occurrence of five results from the set of 1000 and 2000 foot data being higher than any of the background results is only 0.037. Similarly, six values in the 1000 and 2000 foot set are higher than any value in the 3000 foot sample set. The probability that this could occur by chance is 0.05. Thus, the set of 1000 and 2000 foot data is significantly different from the background data, and it is also significantly different from the 3000 foot data, as predicted meteorologically. The 3000 foot sample data is probably representative of background; hence, it could be included with the background data set of September 13. When this is done, the significance of the occurrence of five 1000 and 2000 foot results above all background and 3000 foot data combined can be tested. The probability of this occurrence is 0.0092. These results indicate that the I^{131} plume was

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detected at 1000 and 2000 feet above the terrain by the filter technique used.

Two conditions seriously limited the detection sensitivity in this experiment. These were the presence of a substantial I¹³¹ background because of the nuclear weapons testing in progress at the time, and the low air sampling rate attained.

d. Air Samples

Samples of air were collected for krypton and xenon determinations during the flights at 50 miles downwind of the Redox Plant. The samples were analyzed at the Argonne National Laboratories and the results are given in Table VII.

The krypton results were no higher at these locations than those obtained on the background samples of September 13,

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which are shown in Figure 12. This indicates that the rare gas releases from the Purex Plant, which was operating without interruption throughout this period, dominated those from Redox during the experiment. The plumes were evidently located between positions 11 and 3 on both days. The xenon values are reported in Table VII. The xenon and krypton results do not correlate and the positions of the xenon peaks do not agree with either the Kr^{85} and I^{131} data. Evidently, the xenon activity is predominately from weapons tests. The xenon isotope ratios were not measured.

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FIGURE 11

Kr⁸⁵ Concentrations in Air Samples Taken September 14, 1962

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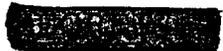


FIGURE 12

Kr⁸⁵ Concentrations in Air Samples Taken on September 13, 1962
(Altitude 1500 Feet)

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3. Results from Close-in Sampling

a. Locations

In addition to the sampling runs made at 50 miles, a large number of sampling runs were made in the range of 1 to 25 miles downwind of the Redox Plant. These runs were made in an attempt to detect the I^{131} plumes by means of a Geiger tube and count rate meter device

The Geiger tube in this device was an end-window type mounted directly behind the filter paper. With this detector, the presence of radioactivity in the sample being collected, as in a cloud from an atomic bomb test, can often be monitored visually by watching the recorder trace of the count rate meter. In these tests at Hanford, however, the I^{131} plumes were not detectable by this device, although they were detected by the more sensitive scintillation counter used in the Beechcraft sampling runs (See Section VII A). During these close-in sample runs filter samples were taken regularly at four minute intervals, which corresponded to approximately 10 to 12 miles each. The plane was flown in arcs across the Northeast quadrant centering at the Redox Plant. The area covered is roughly defined by the Columbia River between the Hanford Reservation boundary and Vantage, Washington, on one side and a line between Richland and Kahlotus, Washington, on the other side.

b. Filter Samples Taken During Emission

Approximately 50 double filter samples were collected during several hours of flying at low altitude downwind of

the Redox Plant up to within approximately one mile from the Redox Plant. Most of the time, the filters were changed at exactly 4 minute intervals, regardless of the location of the aircraft at the time. The flying altitude was in the range of 500 to 2000 feet above the terrain throughout the sampling period. In many cases, the exact location of the sample could not be determined, after the fact, since the plane was often out of the tracking range of the radar (Othello Radar Station) because of the low altitude and the hilly terrain. The longitude and latitude information on the sample data log was often not helpful, either, because of the frequent circling of the plane. Consequently, many of these samples were not analyzed by the more sensitive β - γ coincidence method, although every filter was analyzed by multichannel gamma spectrometry. Those samples which gave a pronounced I^{131} gamma peak over the background of other radioactive matter on the filters were processed through the β - γ method. To these were added several additional samples which were of interest because the location of the sample was known and was of significance. Sometimes the location was of interest simply because the sample was taken at a place known to be out of the plume.

The I^{131} results obtained on these samples are tabulated in Table VIII. The net I^{131} values (6th and 9th columns) have been corrected for the background average. The approximate distances and directions from the source are given in the last

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TABLE VIII

RESULTS FOR MISCELLANEOUS SAMPLES COLLECTED AT LESS THAN 50 MILES FROM THE SOURCE, SEPTEMBER 14, 1962

Sample Number	Lab. Exposure, Minutes	WHITE FILTERS			BLACK FILTERS			Approx. distance from Redox Plant, Miles	Approx. Direction	
		Net I ¹³¹ per 4 min, d/10 ⁻⁴ da	Std Error d/10 ⁻⁴ da	I ¹³¹ d/10 ⁻⁴ da	Std Error d/10 ⁻⁴ da	Net I ¹³¹ per 4 min, d/10 ⁻⁴ da				
2	3962-3	155	4.772	0.389	0.05	3.729	0.181	0.039	10 to 50	Varied from NNW to ENE
3	3964-5	4	0.028	0.047	nil	0.014	0.098	nil	15	ESE
9	3976-7	4	0.0117	0.064	nil	0.00	nil	nil	13	N
11	3980-1	4	1.071	0.140	0.99	6.543	0.159	6.49	10	NNE
12	3982-3	4	2.881	0.330	2.8	8.518	0.181	8.46		Within Hanford reservation
16	3990-1	4	8.525	0.245	8.45	16.714	0.331	16.65	"	"
20	3998-9	4	5.430	0.469	5.35	8.765	0.236	8.71	1 to 10	N to E
31	4293-4	4	1.453	0.118	1.38	2.904	0.248	2.85	1 to 10	"
34	4299-0	4	0.869	0.057	0.79	1.236	0.103	1.18	1 to 10	"
35	4301-2	4	0.206	0.066	0.13	0.160	0.241	0.10	1 to 10	" (?)
45	4321-2	4	0.204	0.025	0.13	0.214	0.020	0.16	25	NNW
46	4323-4	4	0.015	0.031	nil	0.027	0.020	nil	30	NNW
47	4325-6	4	0.143	0.027	0.065	0.026	0.020	nil	35	NNW
49	4329-0	4	0.290	0.042	0.21	0.292	0.089	0.24	28	NE
50	4331-2	38	2.070	0.104	0.14	1.061	0.192	0.05	30 to 45	ENE to NNE

*Corrected for background and normalized to 4 min. sample period.

two columns. The data show that very high I^{131} values per 4 minute filter were obtained when the plane passed through the plume at distances out to about 10 miles from the source. At greater distances, detection is less positive. All large I^{131} values were obtained on samples within the NE quadrant. Samples 3, 45, 46 and 47 were taken well outside the expected plume trajectory. Generally, good correlation between the white and black filter pairs was obtained.

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The data obtained by gamma spectrum analysis of all filters, including those taken on the 50 mile flight course, were analyzed by a general spectral analysis computer program for the seven predominant fission products, Ru-Rh-103, Ru-Rh-106, Ba-La-140, I-131, Cs-Ba-137, Ce-Pr-144, and Zr-Nb-95. With the exception of _____ filters exposed for long periods, the low level of activity and the complexity of the spectrum prevented useful spectral analysis, the error limits generally being larger than the value. In the cases where I^{131} was high, already noted in Table VIII, a reasonably accurate I^{131} value was obtained from the gamma data but the precision was much inferior to that obtained by β - γ coincidence counting after separation.

The long exposure samples, which had high gamma count rates, yielded a typical current fallout spectrum with varying amounts of additional I^{131} imposed upon it. One of the spectra is shown in Figure 13. The I^{131} peak is very prominent, being considerably higher than fallout levels, as monitored by the La-140 peaks (note the high sensitivity range used).

It was mentioned earlier that the Geiger tube counting results _____ were all negative.

C. Environmental Monitoring Samples

Samples of air, water, vegetation, milk, and produce, and measurement of ambient dose rates are obtained routinely within the HAPC environs by the Environmental Studies and Evaluation Operation. Certain portions of this environmental surveillance

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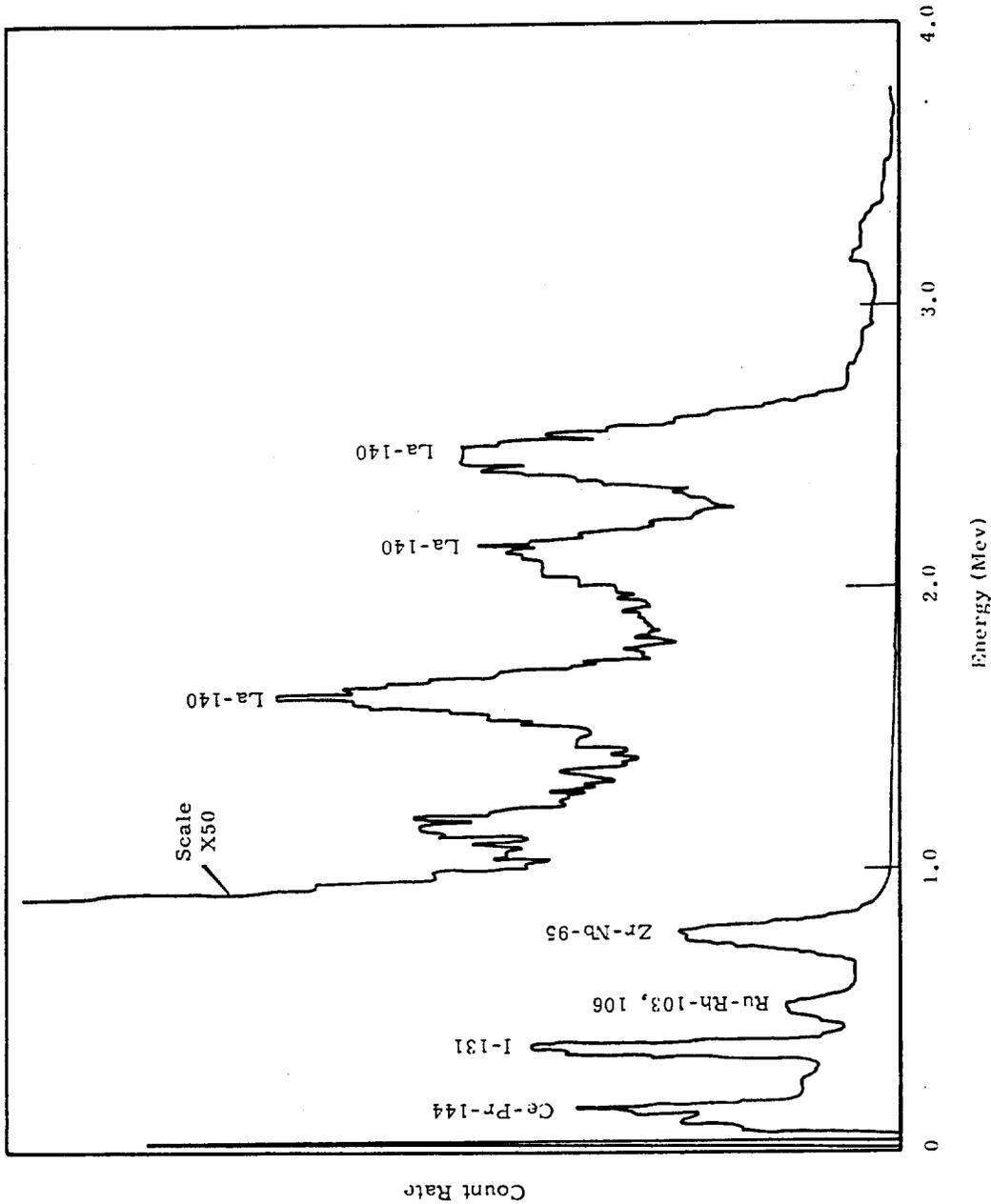


FIGURE 13

Gamma Spectrum of a C-1 Filter
(Counted September 15, 1962)

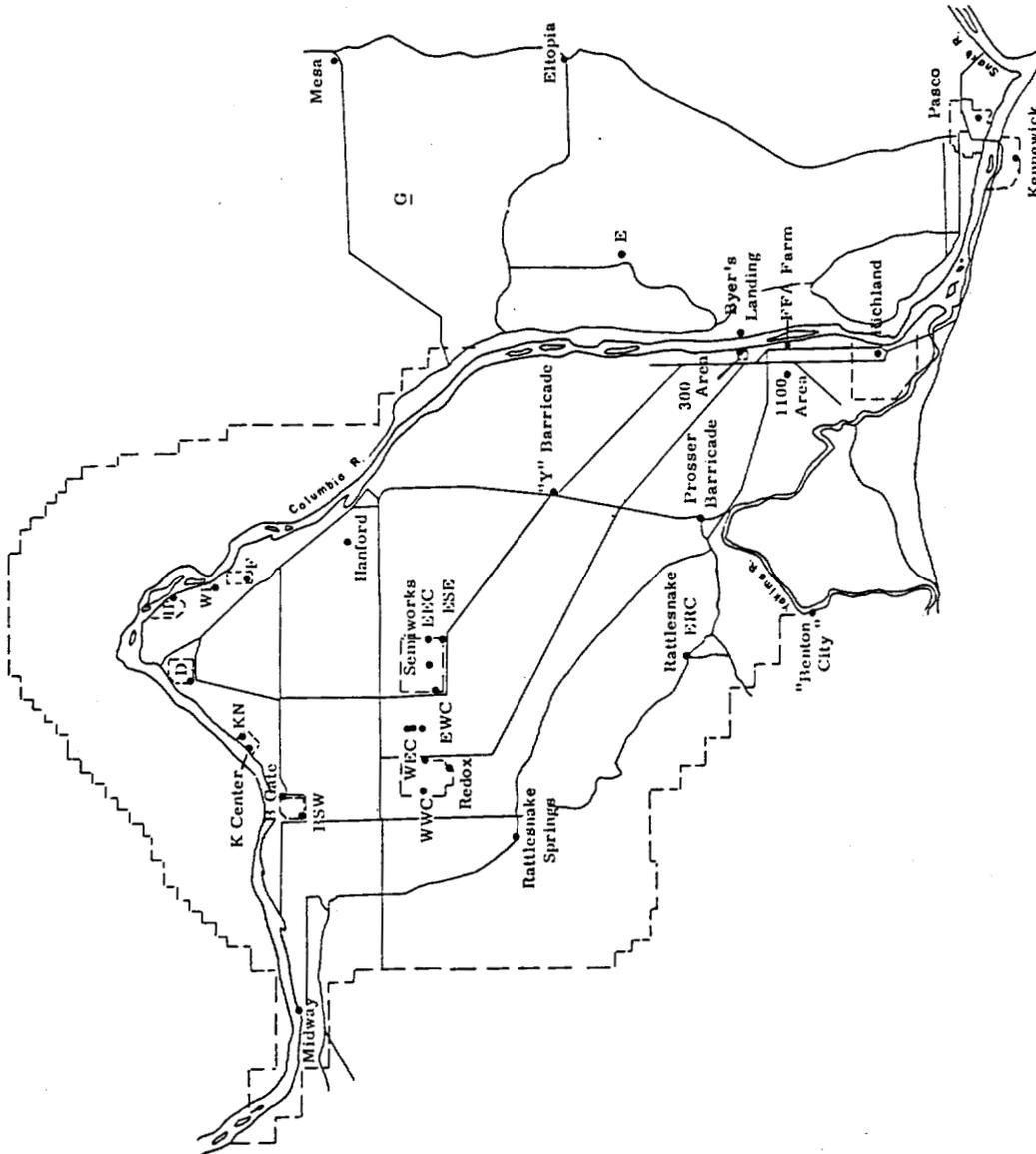


FIGURE 14

Local Air Sampling Stations - September 1962



program are directed towards the measurement of I-131. Additional sampling locations and increased sampling frequencies were established for the test period to aid in evaluating the effects of the additional I-131 release on the environs.

1. Methods

A. Air Samples

Two types of air samples are routinely collected - filter paper samples and caustic scrubber samples. In addition to these, several temporary samplers, consisting of an air filter followed by a charcoal cartridge I-131 adsorber, were established for the test period. All locations where air samplers were operated during the test are shown in Figure 14 and are summarized in Table IX.



TABLE IX

SUMMARY OF AIR SAMPLER LOCATIONS

September 1962

<u>Location</u>	<u>Normal</u>		<u>Special</u>
	<u>Air Filter</u>	<u>Caustic Scrubber</u>	<u>Pre-Filtered Charcoal Adsorber</u>
<u>200 Areas and Vicinity</u>			
200 West, Redox *	D**	W**	-
200 West, East Center *	W	W	-
200 West, West Center *	W	-	-
200 East, West Center *	W	W	-
200 East, East Center *	W	W	-
200 East, Southeast *	W	W	-
200 East, Semiworks *	W	W	-
Meteorology Tower - 3'	W	-	-
Meteorology Tower - 200'	W	-	-
Meteorology Tower - 400'	W	-	-
<u>100 Areas and Vicinity</u>			
Midway Power *	-	-	W
100-B Gate *	W	-	-
100-B Southwest *	-	-	W
100-K Center *	W	-	-
100-K North *	-	W	-
100-D Southwest *	-	-	W
100-H East *	-	W	-
White Bluffs *	W	W	-
100-F Southwest *	-	-	W
Hanford *	-	-	W
<u>Intermediate Locations</u>			
'Y' Barricade	-	-	W
Prosser Barricade	-	-	W
Rattlesnake Mtn. ERC	-	-	W
Rattlesnake Springs	-	-	W
<u>300 Area and Vicinity</u>			
300 Area *	D	W	-
Byers Landing *	W	W	-
1100 Area *	W	W	-
FFA School Farm	-	-	W

*Permanent Atmospheric Monitoring Station

**D = Daily, W = Weekly

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TABLE IX (Continued)

SUMMARY OF AIR SAMPLER LOCATIONS

<u>Location</u>	<u>September 1962</u>		<u>Special Pre-Filtered Charcoal Adsorber</u>
	<u>Air Filter</u>	<u>Normal Caustic Scrubber</u>	
<u>Tri-Cities and Benton City</u>			
Richland *	D	W	-
Kennewick *	W	-	-
Pasco *	W	W	-
Benton City *	W	W	-
<u>Dairy Farms</u>			
"E"	-	-	W
"G"	-	-	W
<u>Remote Locations</u>			
Seattle, Wash.	W	-	-
Spokane, Wash.	W	-	-
Walla Walla, Wash.	W	-	-
Yakima, Wash.	W	-	-
Meachum, Oregon	W	-	-
Klamath Falls, Oregon	W	-	-
Boise, Idaho	W	-	-
Lewiston, Idaho	W	-	-
Great Falls, Montana	W	-	-

*Permanent Atmospheric Monitoring Station

*D = Daily, W = Weekly

(1) Caustic Scrubbers

The concentration of I-131 in the atmosphere is measured by caustic scrubber samplers located in fifteen monitoring stations in the plant environs. Four of these stations are located off the project, and the remainder are located within the various plant operating areas. Each station is a 6' x 6' x 8' high building which houses the necessary monitoring equipment⁽⁶⁾.

The samplers consist of a calibrated, electrically-driven vacuum pump which draws 2.0 cfm of air through one liter of 0.1 normal NaOH solution. A balancing platform and siphon arrangement permits introduction of distilled water into the scrubber at a rate equal to the rate of evaporation. This water feeder helps maintain constant liquid head, air flow rate, and scrubber efficiency.

After one week of operation, the scrubber bottle is replaced and taken to the radiochemical analysis laboratory for determination of the I-131 content. The analytical procedure used provides for the addition of an iodine carrier and AgNO_3 to the scrubber solution, followed by filtration of the resulting silver iodide precipitate. The radiation from the I-131 on the filter is measured by an end-window G.M. tube connected to a scale-of-64 scaler.

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Atmospheric concentrations of I-131 are then calculated from these counting rates by applying factors for counter calibration, chemical recovery of the I-131, scrubber efficiency and the volume of air sampled. The detection limit for I-131 in the atmosphere using the above method is 10^{-14} μc per cc of air. In addition to the regular sampling frequency all of the scrubbers were changed on September 13 and again on September 14, 1962.

(2) Filter Paper Samples

Airborne particulate materials are sampled at several of the Atmospheric Monitoring Stations and at off-site locations (Table IX). Fifteen of these samplers are operated within the project, five are operated in nearby off-project locations, and nine others are operated in more distant Pacific Northwest communities. Two and one-half CFM of air are drawn through 2" x 4" H-70 paper, 18 mils thick, by an electrically driven vacuum pump. The filters are changed daily at four locations and weekly at the others. Gross beta radioactivity collected by the filters is measured with a thin window, propane gas, proportional counter calibrated to give 40 per cent over-all counting efficiency for a RaDEF source distributed on filter paper. A delay of 48 hours is used between sample collection and counting to permit decay of the short life naturally radioactive substances present. One

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of the purposes for operating these filters is to signal and record influxes of radioactive fallout, which can influence the concentrations of I-131 and mixed fission products found in other environmental samples.

(3) Charcoal Cartridge Adsorber Samples

During the test period charcoal cartridge adsorbers were operated at several locations where no permanent atmospheric monitoring stations existed (see Table IX and Figure 14). One additional portable unit was operated in the vicinity of the 100 Areas from 0800 to 1000 on September 14, 1962. The air samples were first drawn through a 2" x 4" H-70 filter paper and then through the charcoal adsorber at 1 CFM. The sampling unit is shown in Figure 15. The outer tube is a 1/2" o.d. by 1/32" wall, transparent acetate cylinder, 2" long. Activated charcoal, 12 x 30 mesh, is packed in the center inch and one-quarter of the tube to a density of 34 pounds per cubic foot. Wire mesh screens retain the charcoal in place while snap rings prevent channeling of the gas between the tube wall and the charcoal.

Used cartridges were analyzed by placing them on top of a 3" NaI scintillation crystal and performing a quantitative gamma energy analysis. Atmospheric

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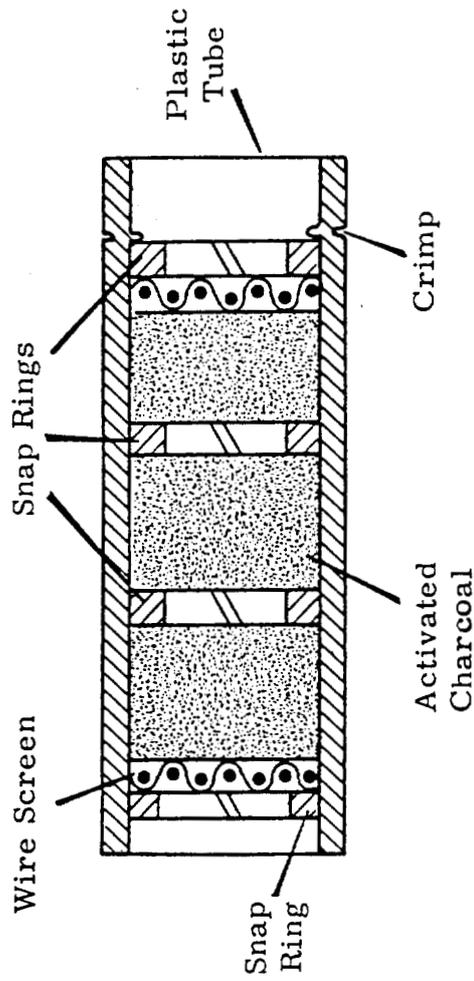


FIGURE 15

Charcoal Adsorber Tube Assembly

concentrations are calculated from the counting rate obtained at the characteristic 0.36 Mev gamma energy by applying corrections for counter calibration, collection efficiency, and volume of gas sampled. The detection limit for I-131 in the atmosphere using the charcoal adsorbers and a weekly sampling schedule is about 2×10^{-13} μc I-131 per cc of air. Gross beta radioactivity collected on the pre-filters was measured on the propane gas proportional counter.

b. Vegetation

(1) On-Project

Samples of sagebrush and native grasses are collected weekly from the 200 West and 200 East Areas. The sample from each area is a 150 g composite of smaller samples collected at ten specific locations within the area. The vegetation is placed in a 250 ml jar and counted on top of a three inch NaI well crystal. Radionuclide concentrations are calculated from counting rates obtained at specific gamma energies, after applying corrections for Compton scatter and background radiation. The detection limit for I-131 in these samples varies with the amounts of interfering fallout nuclides present, but it is normally about 10^{-6} μc per gram.

During the morning of the test additional sagebrush

[REDACTED]

samples were collected at 100-D, 100-H, White Bluffs, and Hanford, during operation of a portable charcoal adsorber unit.

(2) Off-Project

In conjunction with the milk sampling program discussed below, two pound samples of pasture grass are collected routinely whenever a milk sample is obtained from one of the local dairy farms. Pasture grass was also sampled when the special milk samples were collected on September 18 and 21, 1962.

The pasturage is chopped, compacted into a 500 ml jar and counted inside of a nine inch NaI well crystal. Radionuclide concentrations are calculated in a manner similar to that used for the sagebrush samples and the detection limit for I-131 is about 5×10^{-8} μc per gram of pasture grass. Figure 16 shows the pasture grass sampling locations.

c. Milk

Milk samples are collected from five local farms (four weekly and one daily) and composite samples representing each of two milk sheds supplying a local creamery are collected twice a month. In addition, three commercial brands of milk are purchased monthly from a local grocery store. A few days after

[REDACTED]

the test period, milk samples were also collected from several dairy farms located near Highway 11-G between Othello and Moses Lake, Washington. All milk sampling locations are shown on Figure 16.

Several gallons of milk are required to permit detection of the vanishingly small quantities of I-131 present as a result of normal Hanford operations. The milk is passed through an ion-exchange column where the radioiodine is trapped and concentrated. The I-131 in the resin is then measured with gamma scintillation counting equipment. The detection level for I-131 in milk using this procedure and a three gallon sample is about two $\mu\mu\text{c}$ per liter.

2. Results

The analytical results obtained from the various environmental samples collected prior to, during, and after the test period are illustrated in Figures 17 through 22.

a. I-131 in Air

I-131 concentrations in air, as measured by caustic scrubbers (Figure 17) increased significantly during the test period (September 13-14, 1962) at all 200 Area locations. The highest I-131 concentration obtained was $9.7 \mu\mu\text{c}/\text{m}^3$ at the Redox atmospheric monitoring station, during the period from 1400 on September 13 to 1140 on September 14, 1962.

Caustic scrubber samplers operated in the vicinity of the 100

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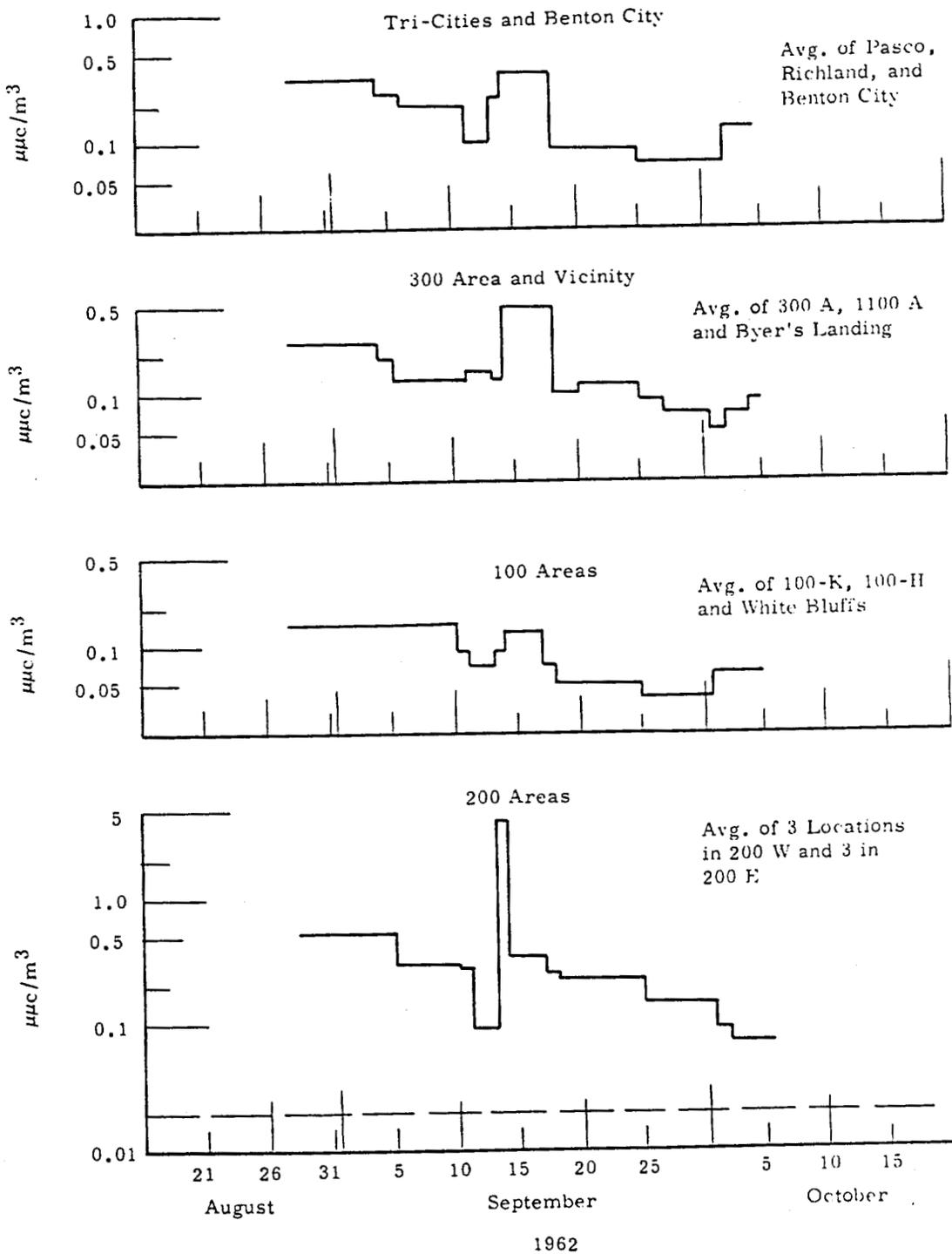


FIGURE 17

I^{131} in Air - Measured via Caustic Scrubbers

Areas indicated a small, non-significant increase in I-131 concentration the day of the test, and another small increase during the period from noon on September 14 through noon on September 17, 1962. A similar trend in air concentrations was noted at the cities of Richland, Pasco, and Benton City.

A somewhat larger increase than that found in the 100 Areas, was noted by caustic scrubber samplers operated in the 300 Area and vicinity during the four day period starting about 1300 on September 14, 1962. The largest increase and the maximum concentration for the three samples in this group occurred at the 300 Area itself, where the concentration increased from $0.2 \mu\mu\text{c}/\text{m}^3$ on September 13, to an average of $0.8 \mu\mu\text{c}/\text{m}^3$ over the following four day period.

The charcoal cartridge absorber units operated during the test period yielded values of I-131 concentrations which were generally at or below the detection limit (Figure 18). This detection limit varied with the total amount of air sampled and the amounts of interfering nuclides present. The one day samples obtained at Hanford and 100-F Area on September 13-14, were significantly higher than those operated at other locations, but were just above the detection limit for a one day sample (0.5 to $1.0 \mu\mu\text{c}/\text{m}^3$). The portable unit operated consecutively at 100-D, 100-H, White Bluffs and Hanford for a total of two hours gave a value below the detection limit of $9 \mu\mu\text{c}/\text{m}^3$ upon laboratory analysis.

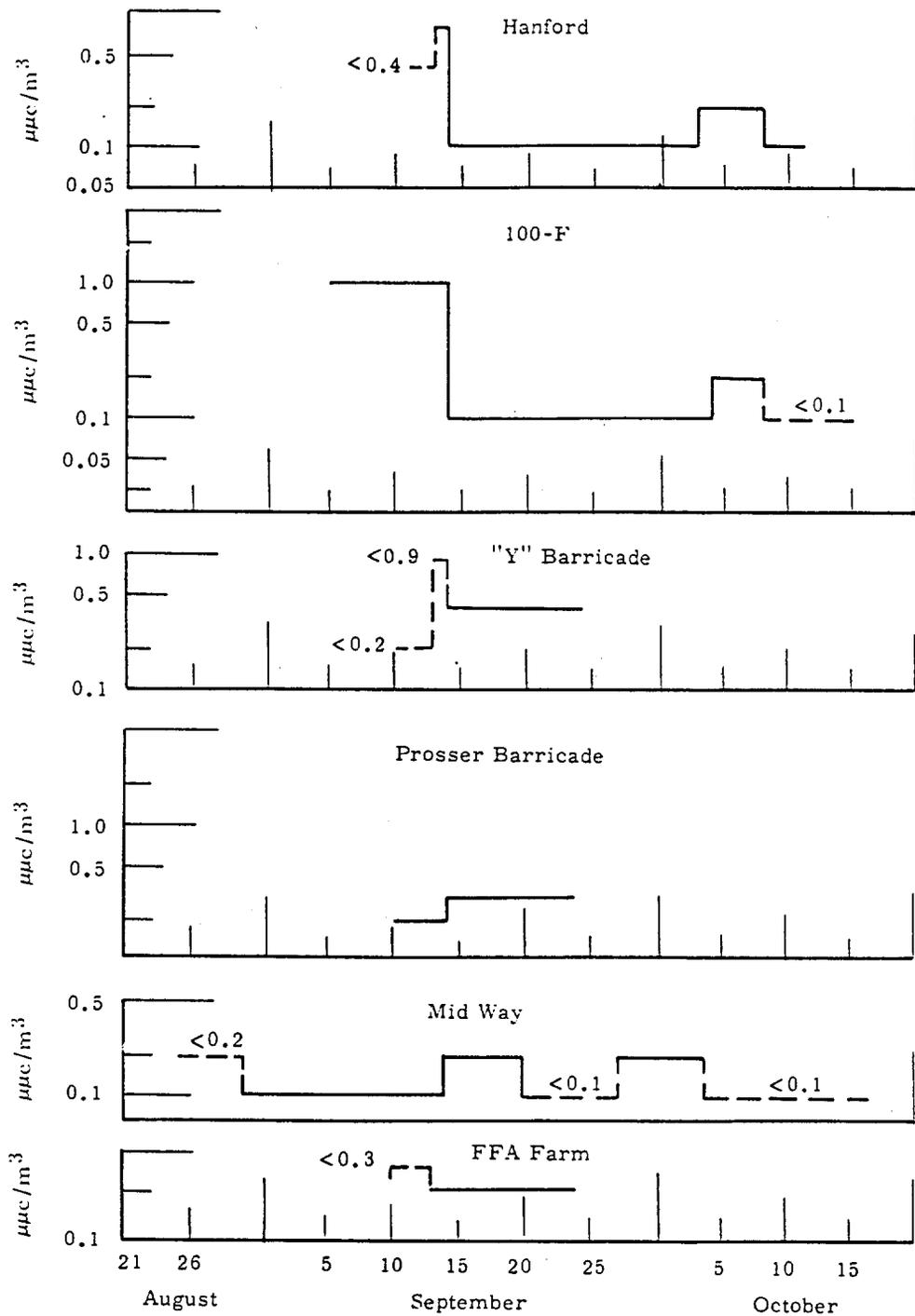


FIGURE 18

I^{131} in Air - Measured via Charcoal Cartridges

b. I-131 on Project Vegetation

The increased I-131 concentration at the 200 Areas was reflected in an increased I-131 content of sagebrush samples collected at 200 West and 200 East Areas (Figure 19). Sagebrush samples collected in the vicinity of the 100 Areas, during the operation of the portable charcoal unit, gave increasing results with time and with closeness of approach to 100-F. These four results are also shown on Figure 19.

c. I-131 in Milk and Pasture Grass

No significant increase was noted in the I-131 content of the daily milk and pasture grass samples collected immediately after the test (Figure 22). Increased concentrations were noted in milk samples collected from all routine locations four to five days after the test, but these increases were caused in part by the influx of fallout materials which began on the 15th of the month (see air filter results below).

The special milk and pasture grass samples from dairies along Highway 11-G (Figures 20 and 21) were not collected until September 18, in an attempt to sample during the probable time of the peak I-131 concentration. The influx of fallout materials had already started by then, but was not signaled until September 19 when several filter samples from the weekend of September 14 to September 17 had been processed.

The I-131 concentration in the special milk sample collected near Moses Lake (K) was significantly higher (780 $\mu\mu\text{c}/\text{l}$) than

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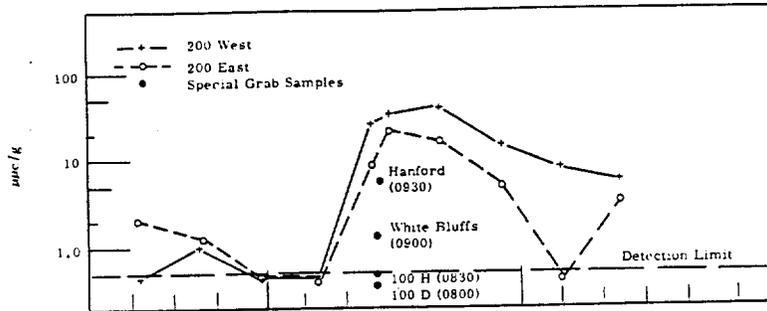


FIGURE 19

I¹³¹ on Sage Brush

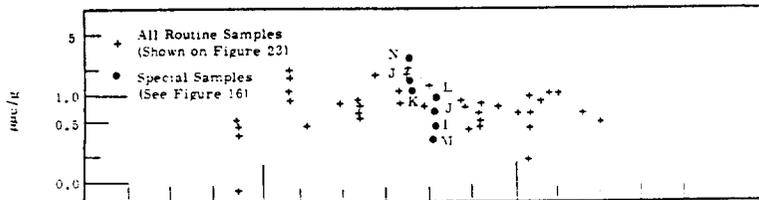


FIGURE 20

I¹³¹ on Pasture Grass

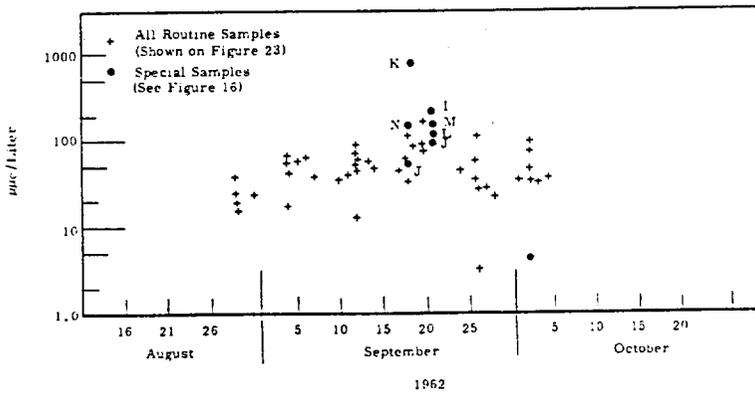


FIGURE 21

I¹³¹ in Milk

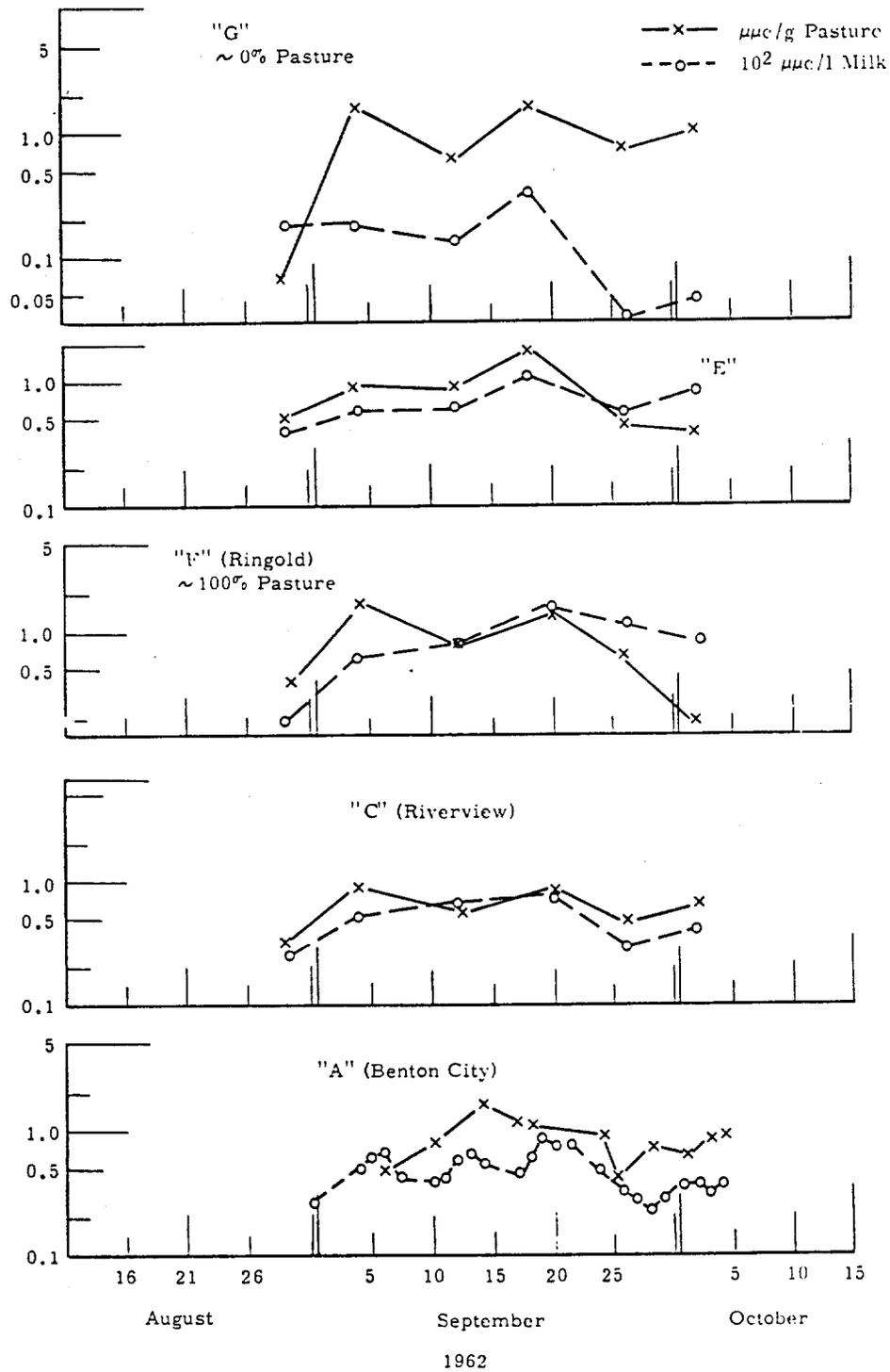


FIGURE 22

I^{131} in Local Pasture Grass and Milk

the concentration found in any of the other routine or special milk samples. It was not possible, however, to determine what fraction of the measured I-131 came from the test and what fraction was due to fallout. If milk samples had been collected from this region prior to and immediately after the test, a better definition of the influence of the test might have been possible.

Four dairies were sampled on September 21, one being a resample of the "J" dairy. Samples from "K" and "N" were not obtainable on that day. The average of the results obtained on the samples collected on September 18, 1962 were 1.8 $\mu\mu\text{c}$ I-131 per gram of pasture grass and 330 $\mu\mu\text{c}$ I-131 per liter of milk. Average values obtained on September 21, 1962 were 0.58 $\mu\mu\text{c/g}$ of grass and 140 $\mu\mu\text{c/l}$ of milk.

None of the results of the special pasture grass samples deviated significantly from general population of results obtained from the routine locations sampled after the test (Figure 20).

d. Gross Beta Radioactivity Collected on Air Filters

Filter sample results can be used to signal the influx of fallout materials. Usually when sudden general increases in concentrations of radioactive particulate materials are found, relatively fresh fallout materials containing some I-131 are present. It appears from the air filter data plotted in Figure 23, that an influx of fallout materials occurred throughout the Pacific Northwest beginning on September 15, the day after the test was completed at Redox. This influx undoubtedly affected the I-131

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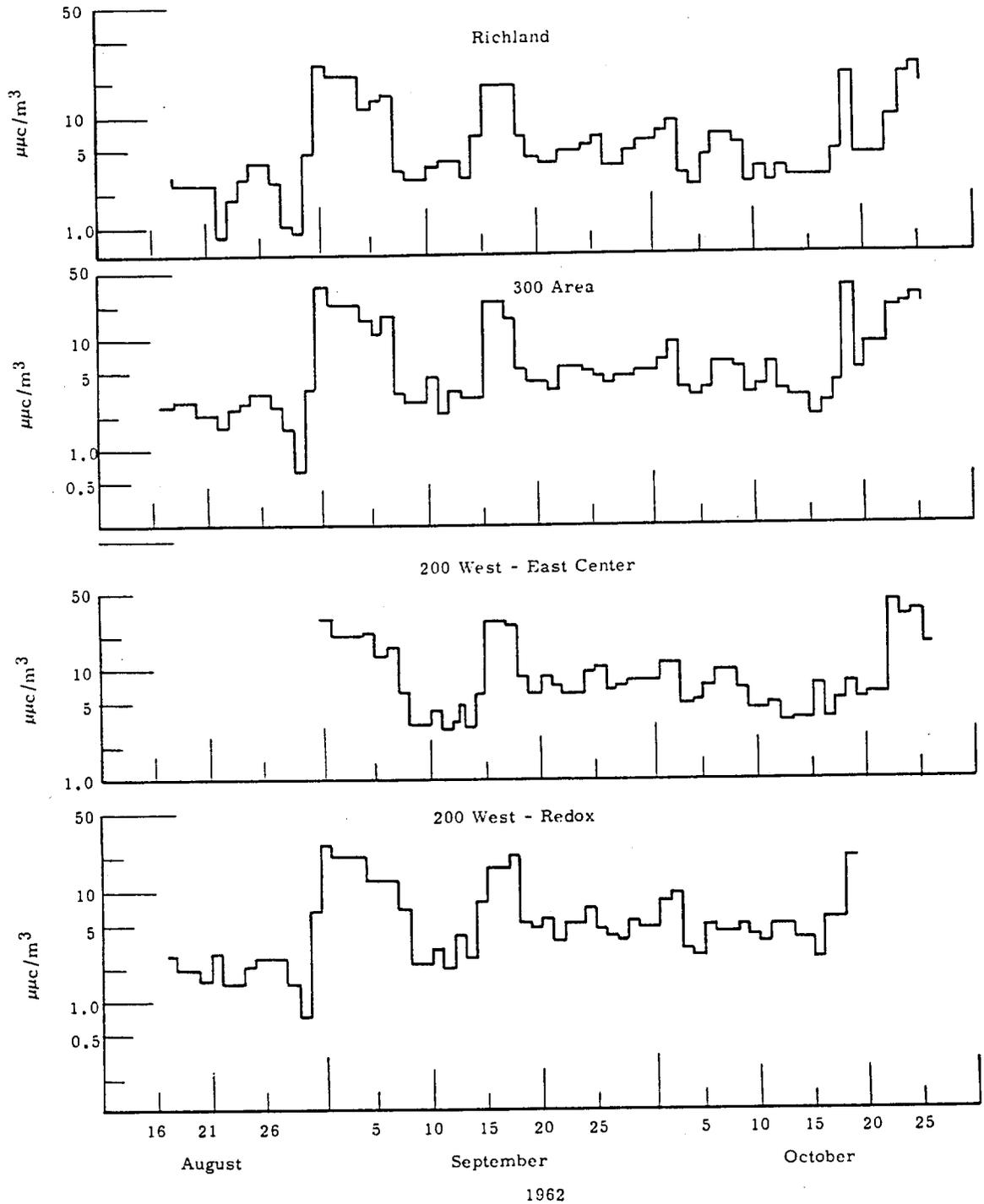


FIGURE 23

Filterable Gross Beta Activity in Air - Daily Samples

concentrations found in environmental samples collected after the test, and the results of air filter samples are presented here to aid in evaluating the possible effects of fallout I-131 on the test results.

D. Collection Efficiencies of Filters by Ram Sampling

Two types of filter papers were used during these tests. These were IPC Paper and an extremely thin charcoal impregnated paper. For their collection of ram samples, an IPC filter was placed in front of a charcoal impregnated filter with the object of obtaining some degree of separation of particulate and gaseous I¹³¹. To aid in the interpretation of their results and obtain data for future sampling, it was desirable to compare the efficiencies for total I¹³¹ collection by ram sampling using IPC paper alone, charcoal impregnated paper alone, and an IPC filter followed by a charcoal impregnated filter. To determine these relative efficiencies, a ram sample filter mount consisting of a "checkerboard" arrangement containing three single IPC filters, three single charcoal impregnated filters, and three IPC filters backed with charcoal impregnated filter was prepared (See Figure 24). The "checkerboard" filter was placed in the center of its ram sleeve (a rectangular tube 2 ft. long mounted on the door of the Beechcraft) and the plane was flown at 120 mph for two hours in the plume from a Hanford Separations Plant stack.

The observed I¹³¹, BaLa¹⁴⁰, ZrNb⁹⁵, and Ru¹⁰³ contents of these filters are summarized in Table X. It is evident that for the same exposure time the IPC filter collects 5 to 6 times as much I¹³¹ as

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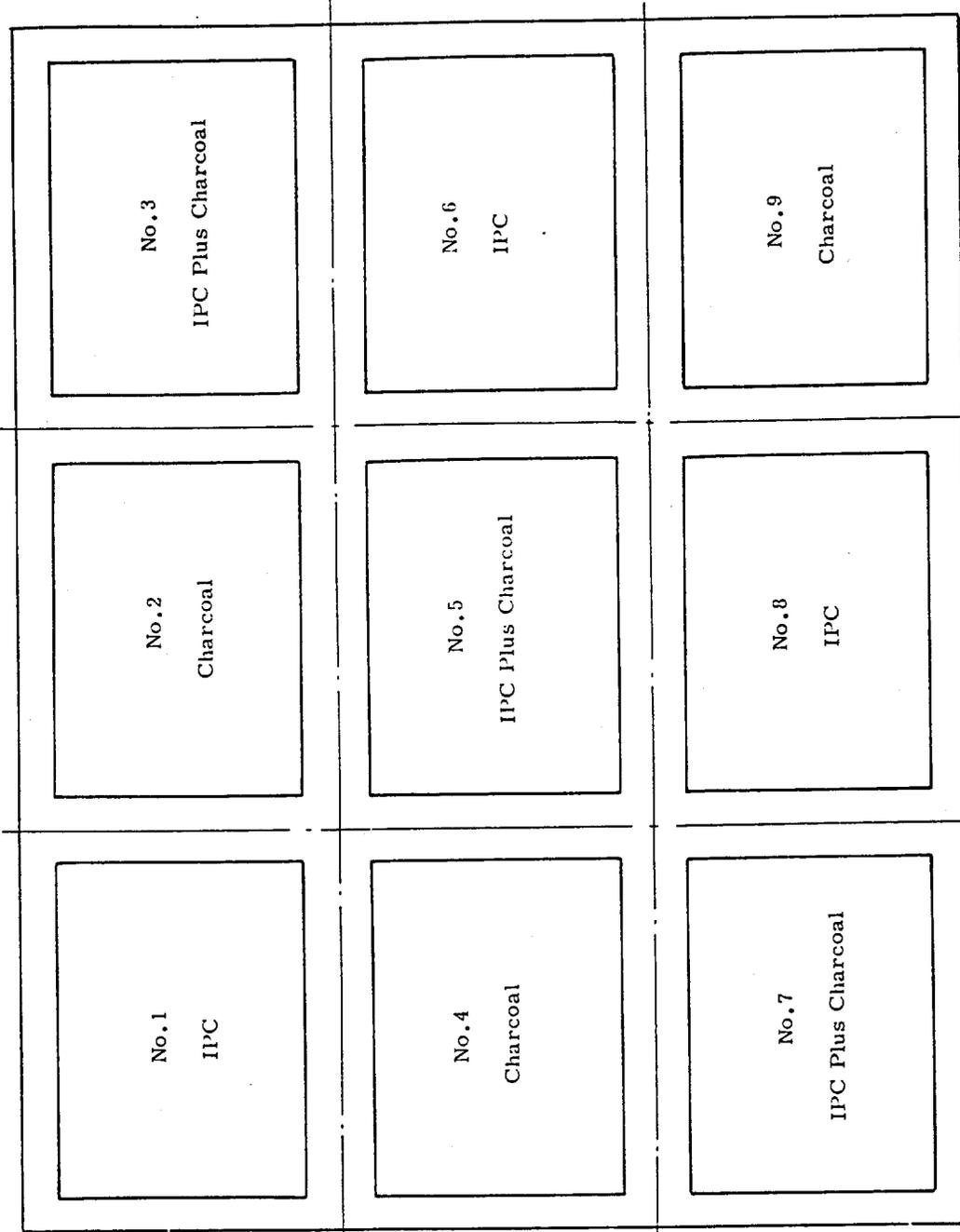


FIGURE 24

Checkerboard Arrangement of the Test Filter Papers in the Ram Sampler

TABLE X

RELATIVE COLLECTION EFFICIENCY OF THE FILTERS IN THE CHECKERBOARD ARRANGEMENT

Filter No.*	Filter Type	d/m/filter			
		Ba-La ¹⁴⁰	ZrNb ⁹⁵	Ru ¹⁰³	I ¹³¹
1	IPC	450	789	437	140
6	IPC	553	706	421	147
8	IPC	453	802	347	100
	Avg. IPC Values	485	766	402	129
2	Charcoal Impregnated	58	23	22	21
4	Charcoal Impregnated	53	21	24	24
9	Charcoal Impregnated	40	31	37	22
	Avg. Charcoal Impregnated	50	25	28	22
3	IPC plus Charcoal Impregnated	128	110	95	40
5	IPC plus Charcoal Impregnated	146	106	75	47
7	IPC plus Charcoal Impregnated	87	48	71	30
	Avg IPC plus Charcoal Impregnated	120	88	80	39

*See Figure 24

the charcoal impregnated filter and 3 to 4 times as much as both filters in series. However, it is also evident that charcoal filters collect a much smaller fraction of the other radionuclides relative to I¹³¹ than the IPC paper.

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28

VIII.

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