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ENVIRONMENTAL EVALUATION OF AN ACUTE RELEASE OF I^{131} TO THE ATMOSPHERE

AUTHOR

J.K.Soldat

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ENVIRONMENTAL EVALUATION OF AN ACUTE RELEASE OF I-131 TO THE ATMOSPHERE

by

J. K. Soldat

Environmental Studies and Evaluation

For Presentation to the
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RADIATION PROTECTION OPERATION
HANFORD LABORATORIES

HANFORD ATOMIC PRODUCTS OPERATION
Richland, Washington

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ABSTRACT

The inadvertent release of 60 curies of I^{131} from a Hanford Separations plant stack occurred in September, 1963. Plant operations were shutdown as soon as the abnormal release was detected, and a comprehensive program of environmental surveillance was undertaken to define the extent and magnitude of the I^{131} deposition.

Initial sampling efforts were guided by rapid calculations of the probable region of maximum deposition made by plant meteorologists. The results of the initial data collected allowed a more precise definition of the area of maximum I^{131} deposition and the location of the individual who probably received the maximum thyroid radiation dose resulting from the accidental I^{131} release. This individual was a child residing at the small farm where the maximum I^{131} concentrations in milk were measured. The estimated maximum total thyroid dose to this child of 0.035 rem was supported by direct measurement of the thyroid burden in the Hanford Whole Body Monitor. For comparison, the Radiation Protection Guide published by the Federal Radiation Council for individuals is 1.5 rem per year to the thyroid.

The results of the surveillance program also supported previously derived parameters for the behavior of I^{131} in various media in the Hanford environs and provided guidance for rapid evaluation of future accidental releases.

I. INTRODUCTION

A study of the behavior in the Hanford environs of I^{131} arising from the influx of debris from nuclear tests during 1961 and 1962 was previously published.(1) The present paper describes the environmental evaluation of an acute release of 60 curies of I^{131} from the Hanford Purex Chemical Separations Plant stack on September 2 and 3, 1963 and contrasts the I^{131} behavior found to that observed in the earlier study. Figure 1 illustrates the Hanford plant environs and various sampling sites for air, grass and milk. The Purex stack is approximately centrally located within the project boundary.

II. METEOROLOGICAL PREDICTIONS

Measurements of wind speed and direction made at the plant meteorology tower during the I^{131} release guided initial environmental surveillance efforts on September 2 and Setpember 3. On September 3 meteorological personnel calculated the I^{131} deposition pattern shown in Figure 2, based on the laboratory analyses of stack effluent samples operated up through the morning of September 3, 1963. About 10% of the I^{131} was released in the afternoon of September 2, 1963 during unstable meteorological conditions, while most of the remainder of the I^{131} was released during the night of September 2 to September 3 under very stable meteorological conditions. The mathematical models used by the meteorologiststo calculate the pattern shown in Figure 2 tends to maximize deposition estimated for off-project locations.(2)(3)(4)

III. ENVIRONMENTAL MEASUREMENTS

A. Vegetation Samples

Extensive vegetation sampling was performed beginning on September 2 and continuing for the next week to define the pattern of I^{131} deposition. Leafy sagebrush was collected whenever possible at on-site locations. A few samples consisted of leafy weeds, cheat grass and in one case, bare sage stems where a fire had previously destroyed the normal vegetation growth. Off-site vegetation samples consisted of pasture grass samples from local dairy farms or leafy weeds and native grasses along the highways and at the permanent atmospheric monitoring stations.

Guided by the predicted deposition pattern, environmental surveillance was intensified east and southeast of the project on September 3 and September 4, 1963. Although the vegetation contamination found south of the project was about as predicted by meteorology personnel, the large area of 10 to 100 pc I^{131}/gram of vegetation north of Pasco was not found. Sampling of pasture grass and milk was extended as far as 60 miles southeast in an unsuccessful effort to locate the predicted maximum deposition.

As sample results were received from the laboratory, it became evident that the position of maximum deposition was probably in the Benton City-West Richland area. Vegetation samples from this latter region ranged from 1 to 10 pc I^{131}/gram (wet weight). The vegetation pattern actually found is shown in Figure 3. Maximum off-site vegetation contamination of 13 pc I^{131}/gram was measured on a sample of green hay from a farm 20 miles SSE of the Purex stack where no cattle were being grazed. Maximum on-site vegetation contamination was found within two miles of

the Purex stack where several samples contained 50 to 250 pc I^{131}/gram (Table I). Maximum I^{131} concentration detected on pasture grass was 2.7 pc I^{131}/gram at a farm 22 miles SE of the point of release (Table II).

B. Air Sampling

Twenty-two permanent atmospheric monitoring stations are maintained within the Hanford site and the immediate environs (Figure 1). Equipment installed in these stations includes air samplers consisting of an HV-70 asbestos film particulate filter and a caustic scrubber I^{131} collector in series. These permanent air sampling stations were supplemented by several temporary caustic scrubber and charcoal cartridge I^{131} samplers during September, 1963.

The maximum off-site air concentration of 0.24 pc I^{131}/m^3 was measured at the city of Richland over the period September 3 to 7, 1963. Maximum on-site air concentration detected was 73 pc I^{131}/m^3 at one-fourth of a mile NE of the Purex stack during the period from the morning of September 2 to the morning of September 3. Table I summarizes the maximum I^{131} concentrations measured in air at 14 of the permanent air monitoring stations. Table II summarizes air measurements made in the vicinity of local dairy farms.

C. Milk Samples

Routine milk samples collected from the Hanford environs in 1963 included one daily and six weekly samples from dairy farms, two milk shed composites obtained twice per month from the collection truck of a local dairy plant, and three commercial brands of milk purchased twice per month from a local grocery store. On September 4, two of the weekly milk collection schedules were increased to a daily frequency

for the remainder of the month. In addition, a new location (Farm B) was added at a daily collection frequency starting on September 12, 1963. Spot sampling at several other dairy farms brought the total number of farms where milk and grass were sampled up to 15 during the month of September.

Maximum milk concentrations in the first few days after the emission were detected at Benton City, 18 miles south of the Purex stack, where a peak value of 120 pc I^{131}/liter was obtained on September 5, 1963. Sample results obtained in the next few days (shown in Figure 4 and Table II) indicated the possibility of a maximum milk concentration at West Richland. A successful search was made in that area, and a sample collected on September 12, 1963 from a farm 16 miles SSE of the point of release (Farm B) was found to contain 140 pc I^{131}/liter of milk. The subject milk was obtained from a single cow maintained for the sole use of the owner's family.

IV. EVALUATION OF ENVIRONMENTAL RADIATION EXPOSURES

The maximum off-project radiation exposure was estimated to be that received by a four year old child residing at the farm where the maximum I^{131} concentration in milk were measured. Since milk sampling at this farm (called Farm B) was not initiated until September 12, 1963, some estimate of the probable peak milk concentration had to be obtained. Daily milk samples had been collected at a farm (called Farm A) ~7.5 miles SE of Farm B during the period September 4 to October 1, 1963 and weekly samples were collected during the other 11 months of 1963. Figure 5 illustrates the consistent ratio of the I^{131} concentrations measured in milk samples from the two farms during September and October, 1963. Using this ratio, back-extrapolation of the I^{131} content in milk at Farm B to a probably peak of 450 pc I^{131}/liter on September 5, 1963 was possible.

Using the extrapolated and measured daily milk concentration values shown in Figure 5 for Farm B, a probable thyroid burden versus time and a probable total thyroid radiation dose was estimated for the four year old resident of Farm B. Assumptions used for the estimate were; a) one liter per day of milk was consumed, b) 30% of the ingested I^{131} reached the thyroid where it remained with an effective half-life of 7.6 days, c) the child's thyroid weighed 4 grams, and d) the amount of I^{131} contributed by inhalation and ingestion of fresh leafy vegetables was insignificant compared to that contributed by the milk.

Applying the above assumptions, the total estimated I^{131} ingested during September and October, 1963 of 3600 picocuries was estimated to have yielded a radiation dose of 0.035 rem to a 4-gram thyroid. For comparison, the Radiation Protection Guide published by the Federal Radiation Council for individuals is 1.5 rem per year.(5)

Several unsuccessful attempts were made to arrange for thyroid counts of the residents of Farm B. Since the actual radiation doses involved were insignificant and since it was desirable not to arouse undue concern on the part of the family, no urgency was implied in the requests for thyroid counts. Finally on October 19, thyroid counts were obtained on the four year old boy and his eight year old sister. The measured thyroid burden of the boy was 73 picocuries and that of his sister was below the detection level of 30 picocuries.

In the meantime, a running estimate of the boy's probable daily thyroid burden had been made assuming the previously mentioned parameters for consumption, uptake, and retention. This estimated burden for October 19, 1963 was 75 picocuries, in excellent agreement with the measured value of 73 picocuries. This correspondence between measurement and calculation was

probably a fortuitous combination of compensating errors in the assumed factors, and it is unfortunate that more thyroid counts were not available to allow determination of the exact factors involved.

Milk consumption estimated by the parents was one gallon per day for the boy and one quart per day for the girl. These consumptions seem to be somewhat high, but the ratio of the measured thyroid burdens for the two children is about right considering the consumption ratio of four to one quoted by the parents.

V. ENVIRONMENTAL BEHAVIOR OF THE I^{131}

The individual results obtained for concentrations of I^{131} in air, pasture grass, and milk versus time were used to estimate the environmental behavior of the I^{131}. This behavior was then contrasted to that previously reported for the fall of 1961 and 1962 during the influx of I^{131} from fallout of nuclear test debris.(1)

The ratios observed in September, 1963 are similar to those found in 1961 and 1962, with the exception that the concentrations of I^{131} found on pasture grass samples seemed to be high by about a factor of 2 when compared to that found in either the air or the milk. The ratios of dose to a 2-gram thyroid received as a result of milk consumption to that received from breathing air remained about the same in 1963 as was found in the 1961 to 1962 study, i.e., 400 to 1. These ratios are summarized in Table III.

The average delay period found between grass and milk was approximately three days in September, 1963 compared to a value of approximately six days found in the 1961 to 1962 study. The effective half-times for I^{131} in grass and milk were found to be approximately four days and approximately five days, respectively during September, 1963. Corresponding values determined

during the earlier study were approximately six days and approximately nine days, respectively. The acute nature of the 1963 release was undoubtedly responsible for the decreased parameters observed.

VI. CONCLUSIONS

The inadvertent release of 60 curies of I^{131} from the Hanford Purex facility in September, 1963 did not result in any significant radiation exposure to persons residing in the Hanford environs. The maximum off-site exposure was deemed to be that received by a small child residing at the farm where maximum I^{131} concentrations in milk were measured. The radiation dose to the 4-gram thyroid of this child was calculated to be ∿0.035 rem compared to the FRC Radiation Protection Guide of 1.5 rem per year for an individual member of the public. Iodine-131 deposition on vegetation was about as predicted by meteorology for close distances, but was much less than predicted at distances of 20-30 miles. On the other hand, I^{131} deposition on pasture grass seemed to be high when compared to concentrations measured in air and milk and to ratios of concentrations expected on the basis of earlier studies. The ratio of thyroid radiation dose received via milk consumption to that received from inhalation of air was the same for both the 1963 and the 1961 to 1962 studies (or 400 to 1 for a 2-gram thyroid).

The delay period between the appearance of I^{131} in grass and its subsequent peak concentration in milk was approximately three days for the acute release of September, 1963. The effective half-time for clearance of the I^{131} from pasture grass and milk was approximately three days and approximately five days, respectively.

It was demonstrated that meteorological guidance, plus rapid collection and analysis of grass samples can lead to prompt definition of the probable

position of maximum deposition. It was also shown that use of a combination of standard assumptions for milk consumption, I^{131} uptake and thyroid retention appeared to lead to a satisfactory estimation of thyroid burden versus time and of total thyroid radiation dose, in spite of the probable inaccuracy of each of the assumptions taken alone.

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(3) G. R. Hilst, "Diffusion of Stack Gases in Stable Atmospheres", Journal of the Air Pollution Control Association, (November, 1957).

(4) G. R. Hilst, and C. L. Simpson, "Observation of Vertical Diffusion in Stable Atmospheres", Journal of Meteorology 15.1, pp 125-6, (February, 1958).

(5) Federal Radiation Council, "Background Material for the Development of Radiation Protection Standards", Staff Report No. 2, (September, 1961).

TABLE I

MAXIMUM I^{131} CONCENTRATIONS MEASURED IN AIR AND ON NATIVE VEGETATION COLLECTED AT AIR SAMPLING STATIONS

September 1963

Distance from Purex Stack	Vegetation		Air	
	Date	pc I^{131}/g (wet weight)	Date	pc I^{131} per m^3
On-Project				
8 miles NW	9/4/63	6.5*	8/26-9/4	0.85
8 miles NNW	9/4/63	0.70	8/26-9/4	0.70
9 miles North	9/4/63	0.09	9/2-9/4	0.35
10 miles North	9/4/63	0.38	9/2-9/4	0.28
7 1/2 miles NNE	9/4/63	0.10	9/2-9/4	0.32
1/2 mile SSW	9/3/63	91	9/2-9/3	52.6
3 1/2 miles West	9/4/63	14	9/2-9/4	2.3
1/4 mile NE	9/4/63	125	9/2-9/3	72.7
1 1/2 miles NW	9/4/63	4	9/2-9/3	4.7
2 1/2 miles WSW	9/4/63	250	9/3-9/6	3.0
Off-Project				
17 miles SE	9/7/63	0.81**	8/29-9/7	0.10 ≠
28 miles SE	9/7/63	0.51	9/3-9/7	0.08
28 miles SE	9/7/63	0.60	9/3-9/7	0.10
15 miles South	9/7/63	1.53	9/3-9/7	0.14

* All on-project vegetation samples listed were leafy sagebrush except this one which was bare sage stems.

** All off-project vegetation samples were leafy weeds.

≠ Average of three samples, the highest of which was 0.14 pc/m^3 on 8/29-9/3.

TABLE II

MAXIMUM CONCENTRATIONS OF I^{131} MEASURED IN SAMPLES OF AIR, PASTURE GRASS, AND MILK FROM THE HANFORD ENVIRONS

September 1963

Distance from Purex Stack	Pasture Grass		Milk		Air		Sampling Frequency Grass and Milk
	Date	pc/gram	Date	pc/liter	Date	pc/m^3	
Individual Farms							
11 miles East	9/3	1.25	9/9	89			Daily, 9/4-10/1
17 miles East	9/3	0.26	9/11	34			Weekly
17 miles ESE	9/4	0.17	9/11	10 ≠	9/2-9/4	0.15	Weekly
16 miles SSE	9/14	1.15	9/12	140	9/3-9/7	0.24*	Daily, 9/12-11/27
16 miles South	9/7	1.60	9/7	91	9/3-9/7	0.14	Single Sample
17 miles South	9/7	1.36	9/7	57	9/3-9/7	0.14	Single Sample
18 miles South	9/7	2.61	9/5	120	9/3-9/7	0.14	Daily, 9/4-10/1
18 miles SE	9/7	0.81	9/7	37	8/29-9/3	0.14	Weekly
22 miles SE	9/5	2.68	9/5	40	9/3-9/7	0.16	Weekly
26 miles SE	9/3 & 9/8	0.61	9/9	37	9/3-9/7	0.08	Daily
29 miles SE	9/6	0.93	9/6	75	9/3-9/7	0.10	4 in 1 week
52 miles SE	9/12	0.11	9/12	10 ≠			Single Sample
54 miles SE	9/12	0.46	9/12	68			Single Sample
58 miles SE	9/12	0.24	9/12	14 ≠			Single Sample
60 miles SE	9/12	0.14	9/12	25 ≠			Single Sample
Milk Truck Composites							
East of Project			9/8	18			3 per month
South of Project			9/7	59			3 per month
Commercial Milk from Stores							
Brand A			9/16	12			2 per month
Brand F			9/16	8			2 per month
Brand H			9/26	4			2 per month

≠ These four farms were known to have their cows on all or nearly all dry feed.

* This air station was ∿5 miles down wind of the farm. All other air stations were only ∿2 to 3 miles from the farms.

TABLE III

RATIO OF I CONCENTRATIONS
MEASURED IN VARIOUS ENVIRONMENTAL MEDIA

Ratio	September 1963	1961 and 1962
$\frac{\text{pc/kg Sagebrush (on-project)}}{\text{pc/m}^3 \text{ air}}$	∿ 2,300	--
$\frac{\text{pc/kg Weeds (off-project)}}{\text{pc/m}^3 \text{ air}}$	∿ 6,700	--
$\frac{\text{pc/kg Pasture Grass (Dairy Farms)}}{\text{pc/m}^3 \text{ air}}$	∿ 10,000	∿ 4,200
$\frac{\text{pc/liter of Milk}}{\text{pc/kg grass}}$	∿ 0.07	∿ 0.15
$\frac{\text{pc/liter Milk}}{\text{pc/m}^3 \text{ air}}$	∿ 700	∿ 600
$\frac{\text{Dose to 20-gram Thyroid from Milk}}{\text{Dose to 20-gram Thyroid from Air}}$	∿ 40	∿ 40
$\frac{\text{Dose to 2-gram Thyroid from Milk}}{\text{Dose to 2-gram Thyroid from Air}}$	∿ 400	∿ 400

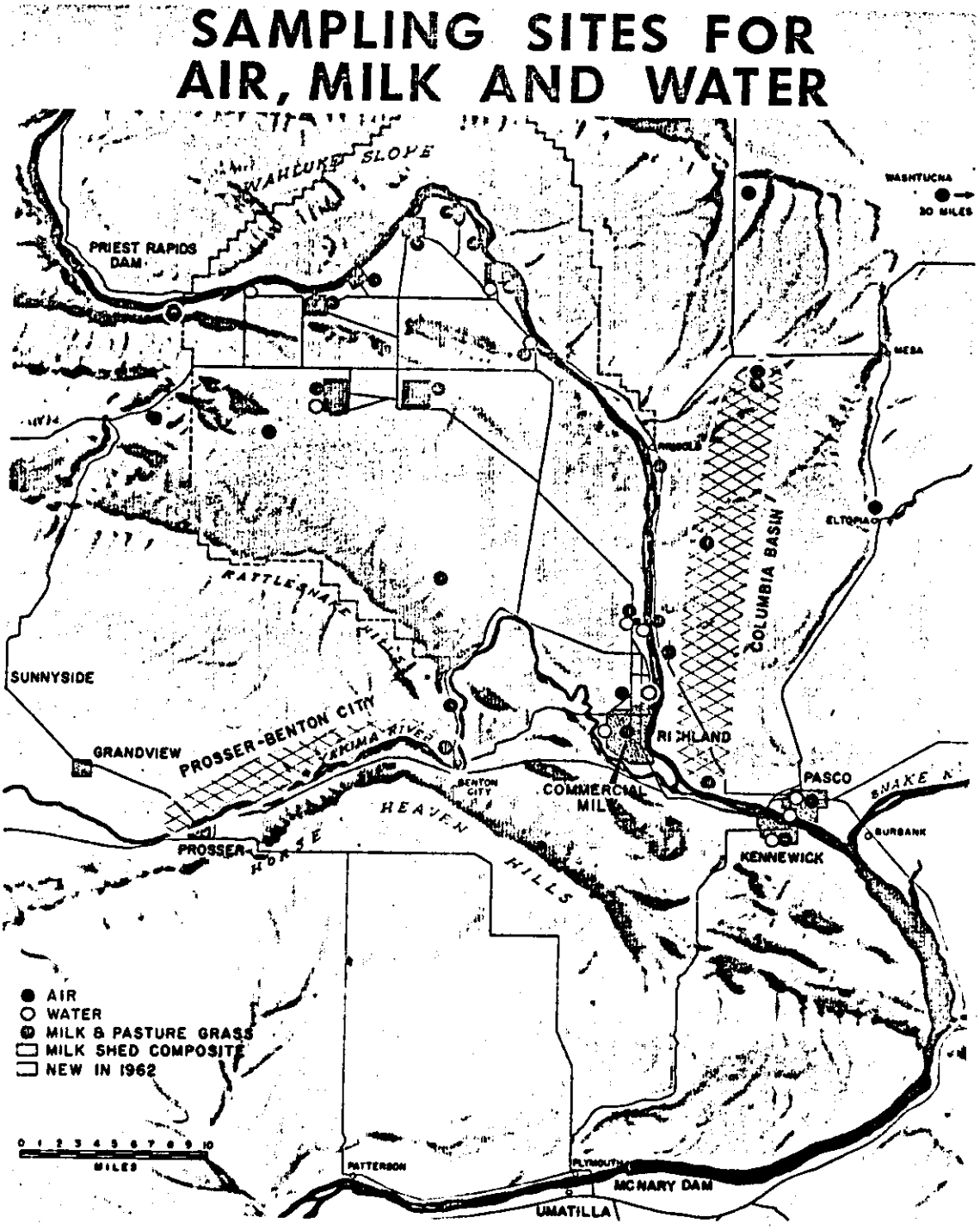

FIGURE 1

Hanford Plant Environs and Sampling Sites

AEC-GE RICHLAND, WASH.

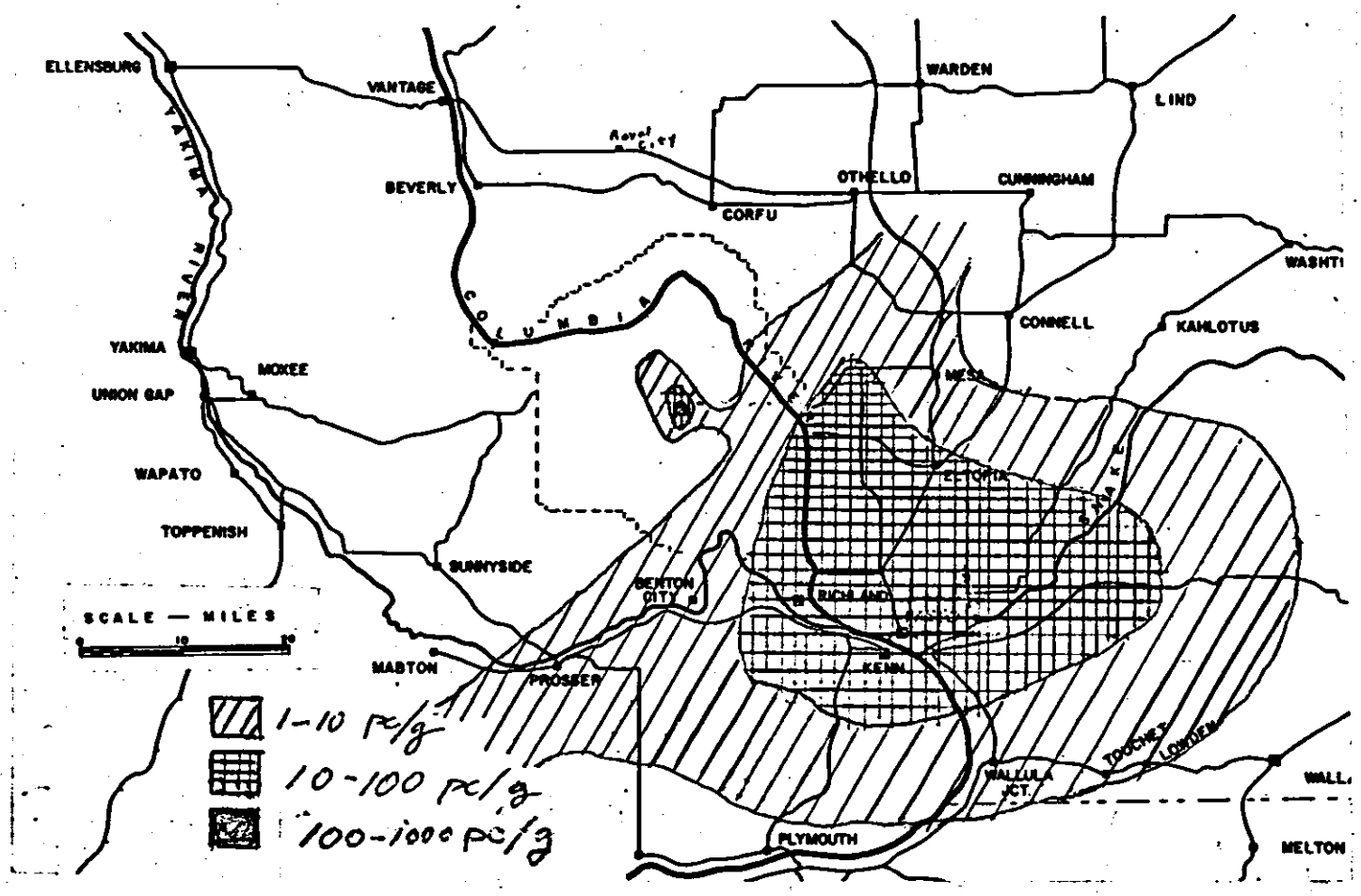

FIGURE 2

PREDICTED MAXIMUM CONCENTRATIONS
OF I^{131} ON VEGETATION

September, 1963

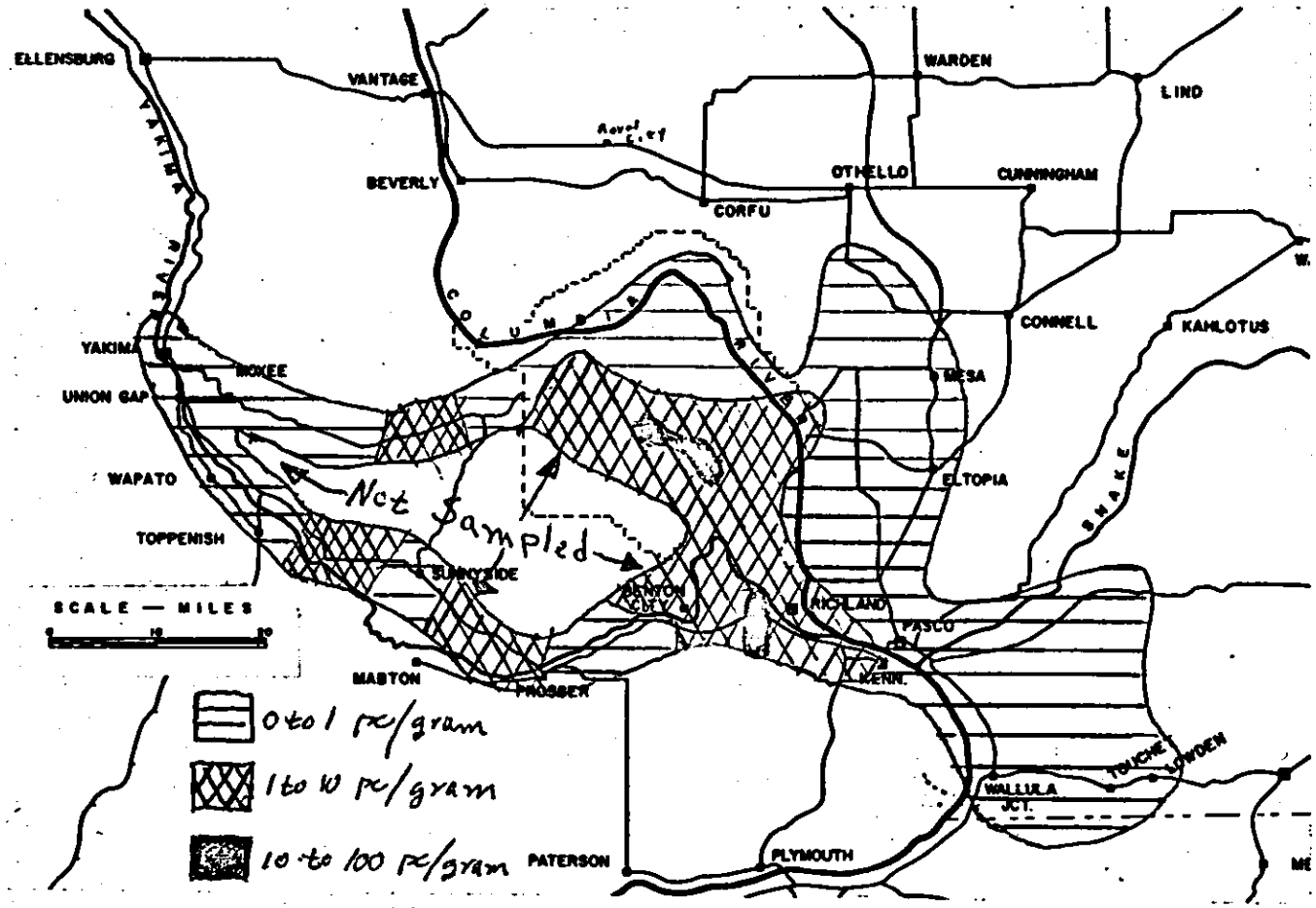

FIGURE 3

MAXIMUM CONCENTRATIONS
OF I^{131} MEASURED ON VEGETATION

September, 1963

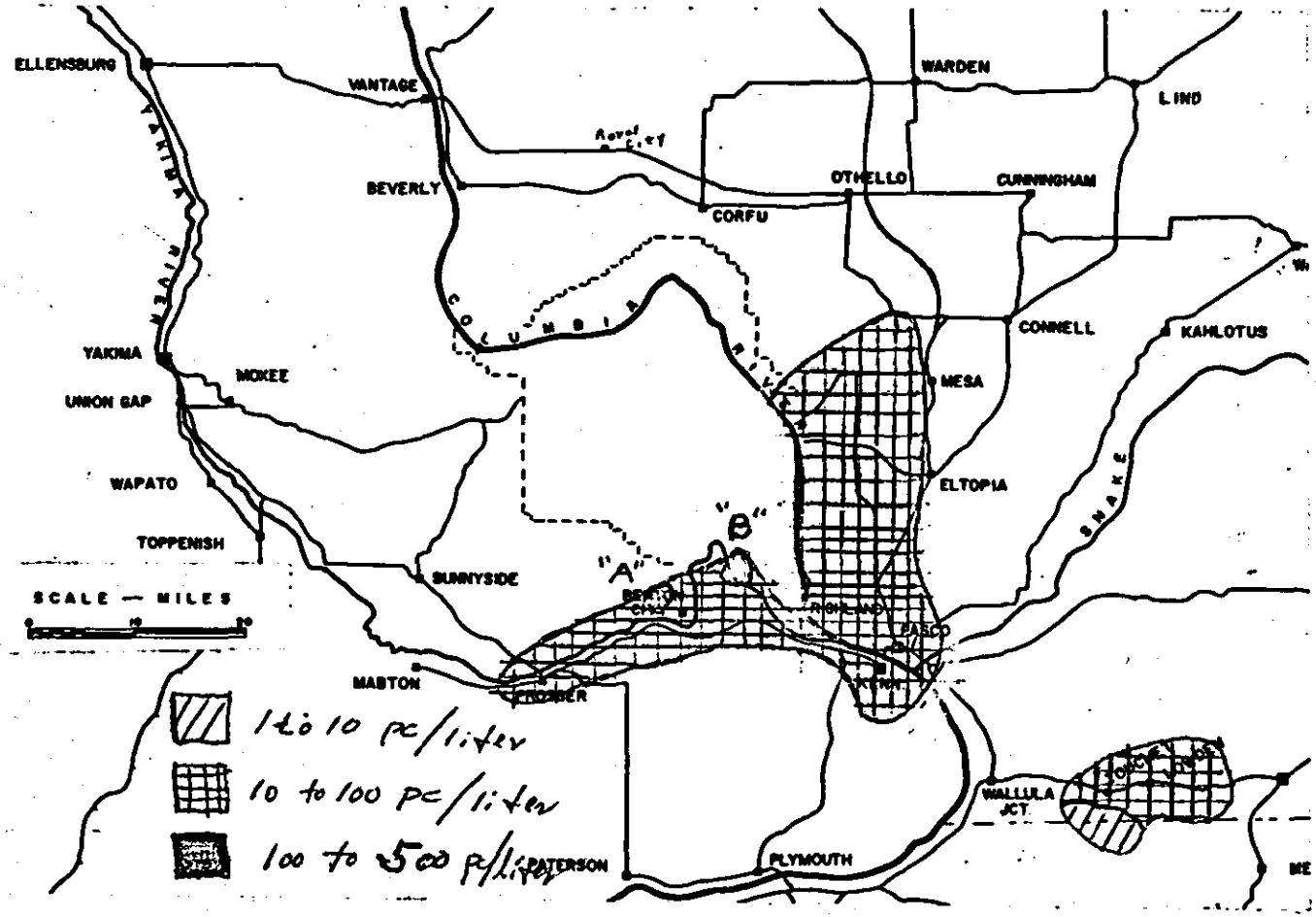

FIGURE 4

MAXIMUM CONCENTRATIONS
OF I^{131} MEASURED IN FARM MILK

September, 1963

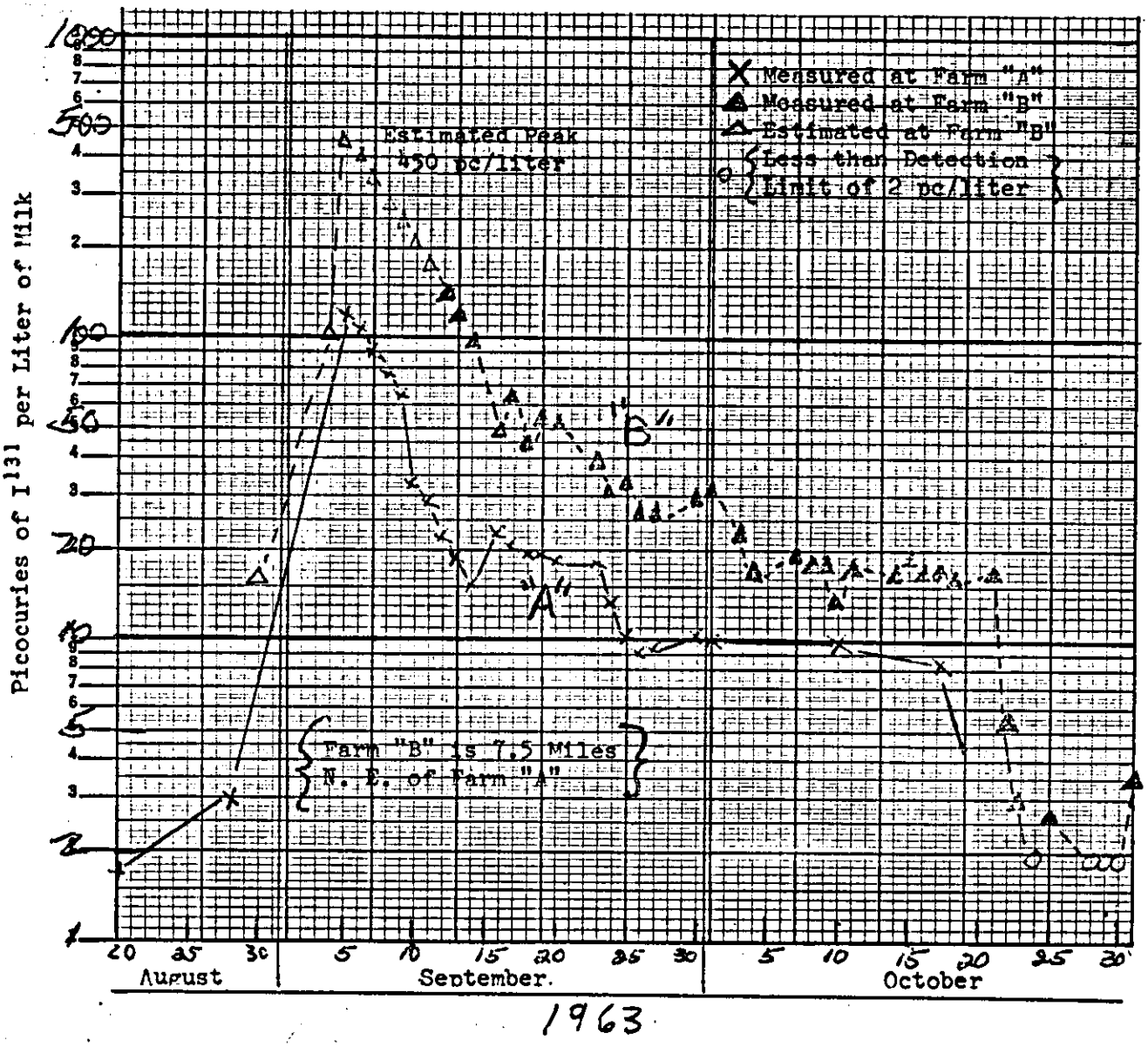

FIGURE 5

CONCENTRATIONS OF I^{131} IN MILK SAMPLES
COLLECTED DAILY FROM TWO FARMS
IN THE REGION OF MAXIMUM DEPOSITION