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FALL-OUT IN SOUTHEASTERN UNITED STATES DURING JANUARY AND FEBRUARY 1951 FROM THE NEVADA ATOMIC TESTS

OAK RIDGE NATIONAL LABORATORY
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Contract No. W-7405-Eng-26

HEALTH PHYSICS DIVISION

FALL-OUT IN SOUTHEASTERN UNITED STATES DURING JANUARY AND FEBRUARY 1951 FROM THE NEVADA ATOMIC TESTS

F.J. Davis

Date Issued

NOV 8 1951

OAK RIDGE NATIONAL LABORATORY
operated by
CARBIDE AND CARBON CHEMICALS COMPANY
A Division of Union Carbide and Carbon Corporation
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Oak Ridge, Tennessee

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ABSTRACT

Air filter and rainwater collections of atmospheric fall-out were made at Atlanta, Georgia; Urbana, Illinois; Topeka, Kansas; Fort Worth, Texas; and Nashville, Tennessee. The air filters showed activity as high as 1400 counts per minute compared to an average thorium background of 7 counts per minute; the precipitate from rainwater collections showed activity increases as high as 400 times normal. Radioautographs proved the particulate nature of the activity, and radiochemical analyses identified the activity as recent fission products. Observed activity peaks correlate well with meteorological positioning of the radioactive cloud from the Nevada tests.

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INTRODUCTION

On January 17, this Laboratory was asked by the Division of Biology and Medicine of the Atomic Energy Commission to participate in a program of measuring the fall-out from the Nevada atomic tests. It was decided to establish stations at the following places: Emory University, Atlanta, Georgia; University of Illinois, Urbana, Illinois; Forbes Air Force Base, Topeka, Kansas; Carswell Air Force Base, Fort Worth, Texas; and Vanderbilt University, Nashville, Tennessee.

APPARATUS

Since time for preparation was so limited it was necessary to use readily available equipment. All stations were equipped with the following: a filtron, a constant air monitor, auxiliary counting apparatus, and equipment for rainwater or snow collection. In addition Urbana, Topeka, and Nashville were equipped with moving filter monitrons.

The filtron is an air filter apparatus which pulls air at a rate of about 5 CFM through a filter paper 4-1/2 x 8 inches. The paper is removed after a specified time interval and the activity counted. The constant air monitor is an air filtering device which draws air through a filter paper 3 x 5 inches at a rate of about 4 CFM. The paper surrounds a beta counter tube and the accumulated activity in the filter paper records continuously on an Esterline-Angus recorder. The moving filter monitrons use 4-inch strip filter paper which travels at the rate of 6 inches per hour around a perforated cylindrical suction tube and under a beta counter tube. The distance between the suction tube and the counter tube can be adjusted so that the time interval between collection and counting of activity can be varied. This time interval was set to the maximum of about 4-1/2 hours. The recording apparatus consisted of a scaler, a Berkeley counting rate computer and an Esterline-Angus recorder. The auxiliary apparatus for the counting of the filters consisted of a beta counting tube approximately 3/4 inch diameter and 11 inches long with a sensitive area about 7 inches long and a wall thickness of 30 milligrams per square centimeter. The beta tube was mounted in a lead pig and a holder rigged to hold the filter paper in the same geometry as when the activity was collected. The beta count was obtained by use of a scaler and stopwatch. Containers for snow or water collections were coated on the inside with rubber-base paint. Urbana was the only station which did not collect rainwater or snow. The filter paper used in the filtrons and constant air monitors was type CW-6. In the moving filter monitrons Hollingsworth and Vose type H-70 was used.

The counting instruments were calibrated by use of a known source of uranium deposited on aluminum foil of the same size as the filter paper. The air flow of each filtering device was measured by a gas meter so that the activity could be measured in microcuries of collected activity per cc of air.

PROCEDURE

Two men were sent to each station to set up the apparatus and start the collection of data. After operations were progressing smoothly only one person was required to maintain the equipment and collect data. As long as two people were available, the routine instructions given to each station were as follows:

1. Change filtron paper every 8 hours.
2. Change constant air monitor filter every 24 hours.
3. Count all samples 8 hours after removal.
   Count samples and background for 20 minutes each.
4. If any activity is noticed, count samples again at 16 and 24 hours after removal.
5. If activity is all ThB (half-life 10.6 hours) the activity at 24 hours will be 35 per cent of that at 8 hours.

   If activity at 24 hours is more than 35 per cent of that at 8 hours, keep counting samples at 8 hour intervals until 48 hours after removal.
6. Plot activity above background for each series of counts on a sample on two cycle semi-log paper.
7. Send to Oak Ridge by air mail any samples which show activity other than ThB, to be further counted and radioautographed.
8. Treat any rainwater and send in floc by air mail.

When only one person was maintaining the equipment, the rountine was changed to 12 hour intervals. The procedure on handling rainwater will be discussed under Rainwater Treatment.

OBSERVATIONS AND COMPUTATIONS

In the computations of the activity shown in Figures 1-5 it was assumed that (1) all radium decay products had decayed by the first count, (2) the thorium B + C count decayed with a half-life of 10.6 hours and (3) the fission products collected decayed as $T^{-1.2}$ where T is the age of the fission products.

The formula for computing the fission product activity F_x at the time of the first count is

$$F_x = \frac{Y - X e^{-\lambda t}}{\left(\frac{T}{T + t}\right)^{1.2} - e^{-\lambda t}}$$

where X is the net counting rate during the first count

Y is the net counting rate during the second count

λ is the decay constant for thorium B

t is the time interval between counts

T is the age of the fission products at the time of the first count.

This formula should be corrected, however, to a time at the midpoint of the collection period so that it approximates the average activity when collected. The complete formula used, including the correction factor is

$$F = \left(\frac{T}{T - t_1}\right)^{1.2} \left[\frac{Y - X e^{-\lambda t}}{\left(\frac{T}{T + t}\right)^{1.2} - e^{-\lambda t}}\right]$$

[redacted]

where F is the fission product activity at the midpoint of the collection period

t_1 is the time interval from the midpoint of the collection period to the time of the first count

In Figures 1-5 the widths of the rectangular ordinates represent the collection times. The activity peaks are attributed to the particular shots as indicated on the figures. These were arrived at by a study of meteorological maps furnished by AFOAT-1 showing the trajectories of the activity and by surface winds shown on daily weather maps. The times of firing the shots were early morning on the dates of January 27, January 28, February 1, February 2, and February 6, 1951.

Figure 1 data from Urbana shows the highest activity recorded. The time delay for the activity to reach Urbana appears to be approximately 2 days for the first shot and 3 days for the second.

Figure 2 shows the activity collected at Topeka. The activity peak shown is thought to be from shot number 2 but some of the early activity on the constant air monitor filter may be due to shot number 1. The delay in arrival as shown by the filtron peak appears to be approximately two days.

Figure 3 shows activity collected at Nashville. This peak is probably also from shot number 2 and again a two-day delay is shown. The activity collected is considerably more than that collected at Topeka.

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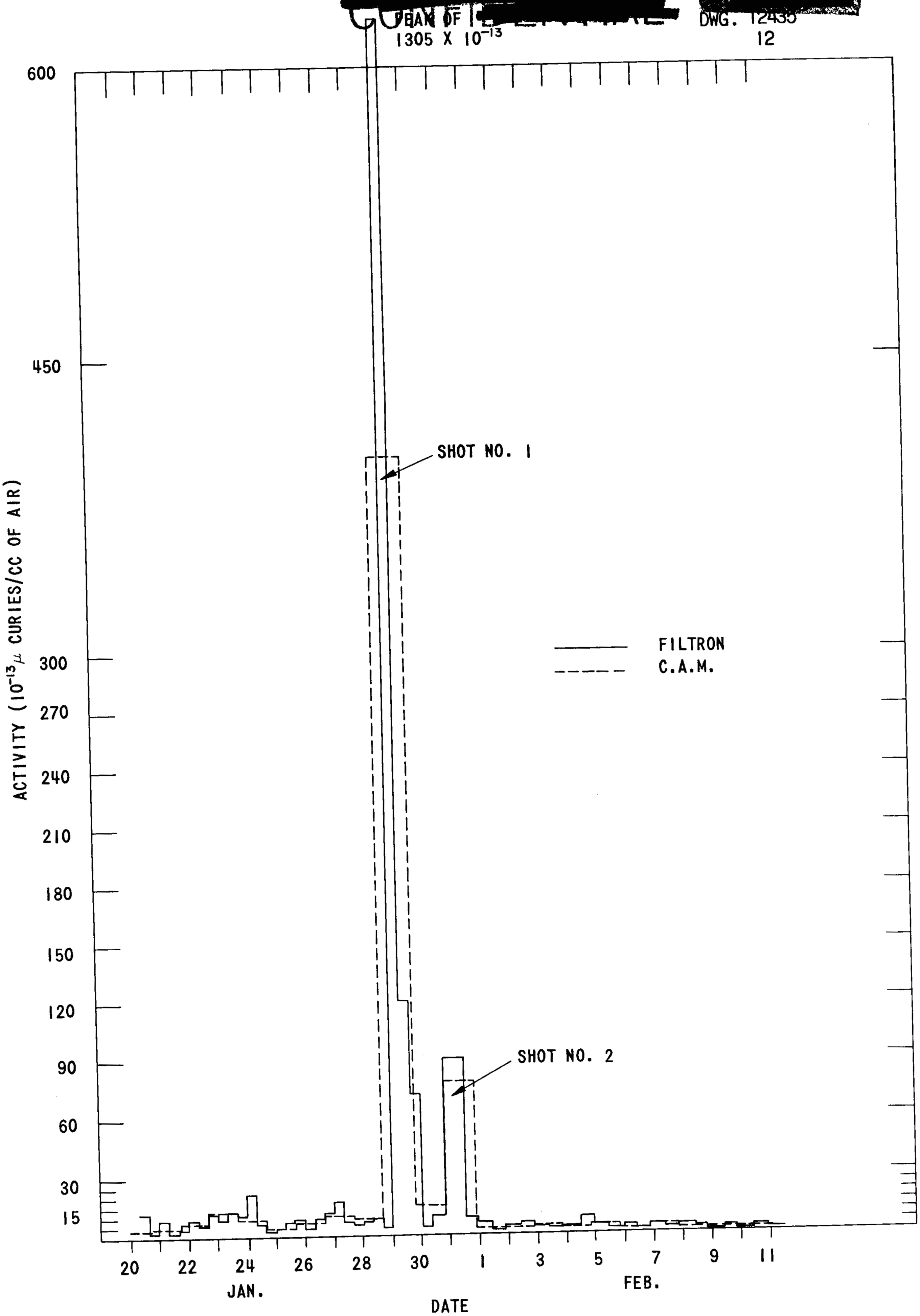

Fig. 1— URBANA, ILLINOIS

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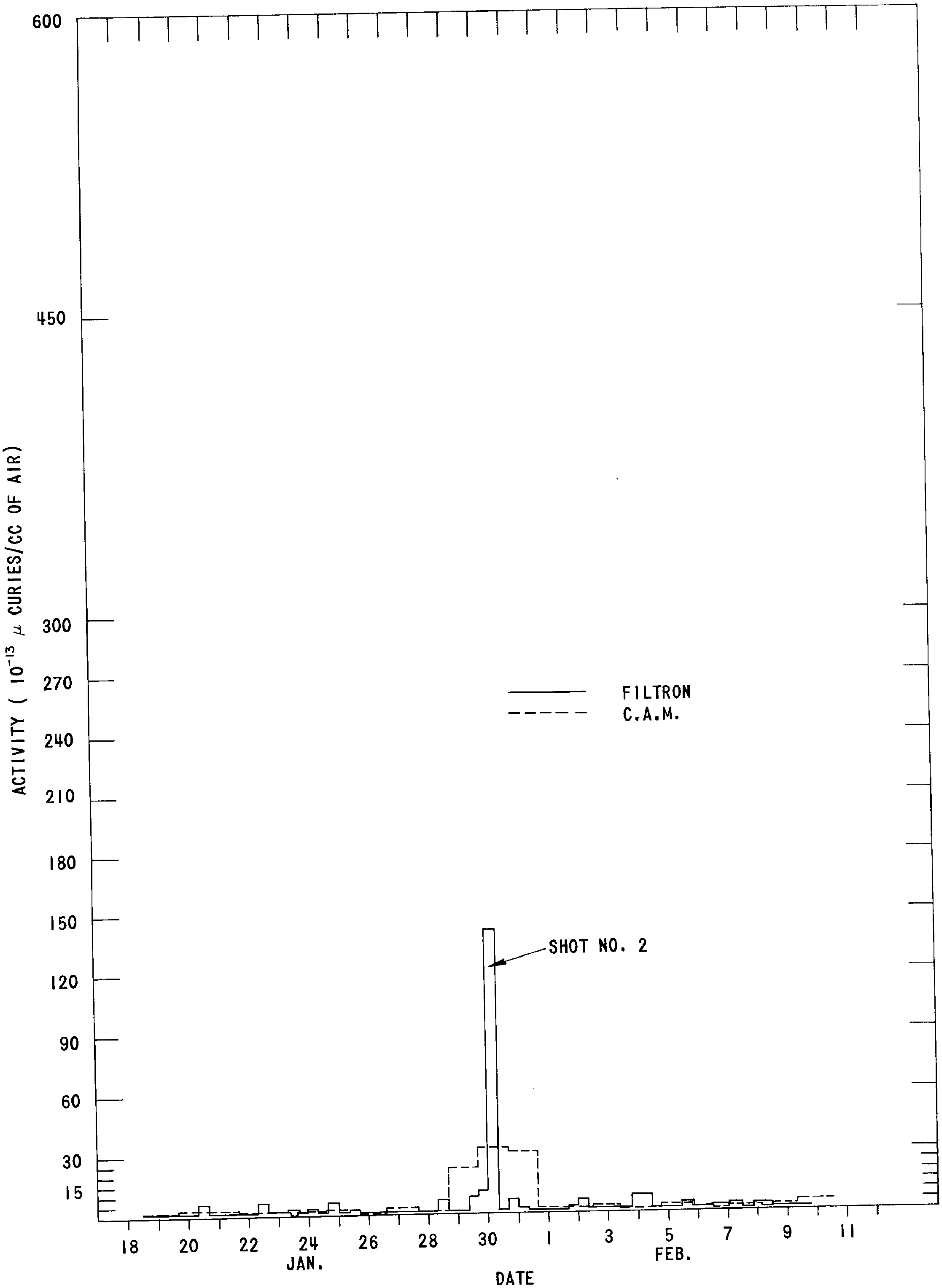

Fig. 2— TOPEKA, KANSAS

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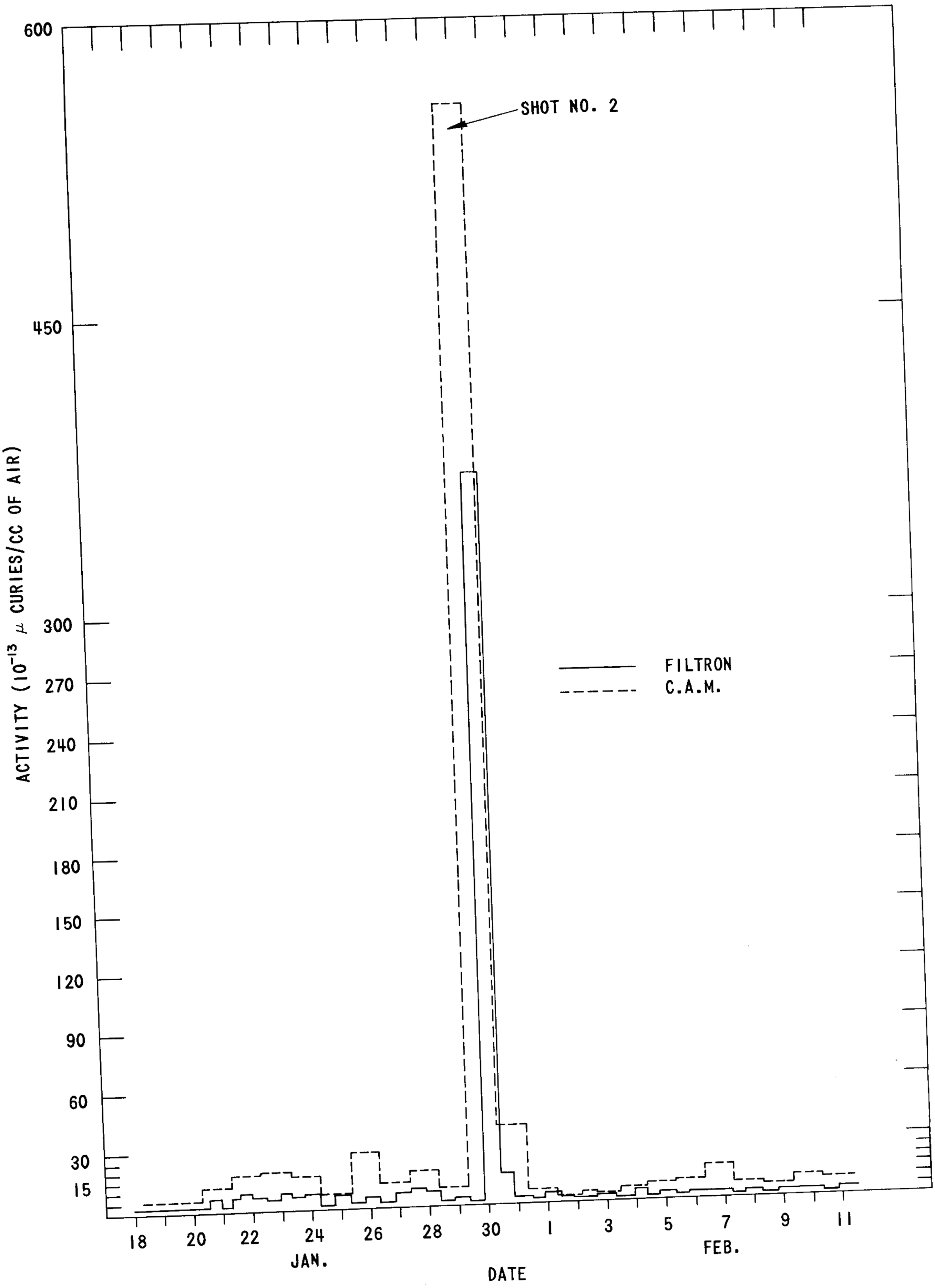

Fig. 3— NASHVILLE, TENNESSEE

Figure 4 from Atlanta shows the smallest amount of activity collected at any of the five stations. The delay shown here is approximately five days after the fourth shot. While the upper air carried the activity into the region south of Atlanta in two days, surface winds apparently carried the activity still further south so that none was observed in Atlanta until the fifth day.

Figure 5 from Fort Worth shows activity collected from three shots, numbers 2, 4, and 5. The activity collected from number 5 is quite small but shows up definitely on the radioautograph shown in Figure 12. The delay on shot 2 is about 2 days, shot 4 four days and shot 5 five days.

The two records shown in Figure 6 are from Urbana on the morning of January 29. Figure 6b is the record from the constant air monitor giving the accumulated activity collected versus time. The activity is counted from the time it is collected until the filter is changed so that the radium and thorium products are both included. Figure 6a is the record from the moving filter monitron showing activity versus time. Since the moving filter monitron counts the activity 4-1/2 hours after it is collected to allow radium products to decay out, the record was shifted so that the peaks correspond with the time of collection as shown on the constant air monitor record. The peaks in Figure 6a should correspond to changes of slope in Figure 6b. The records read in opposite directions, the higher activity in

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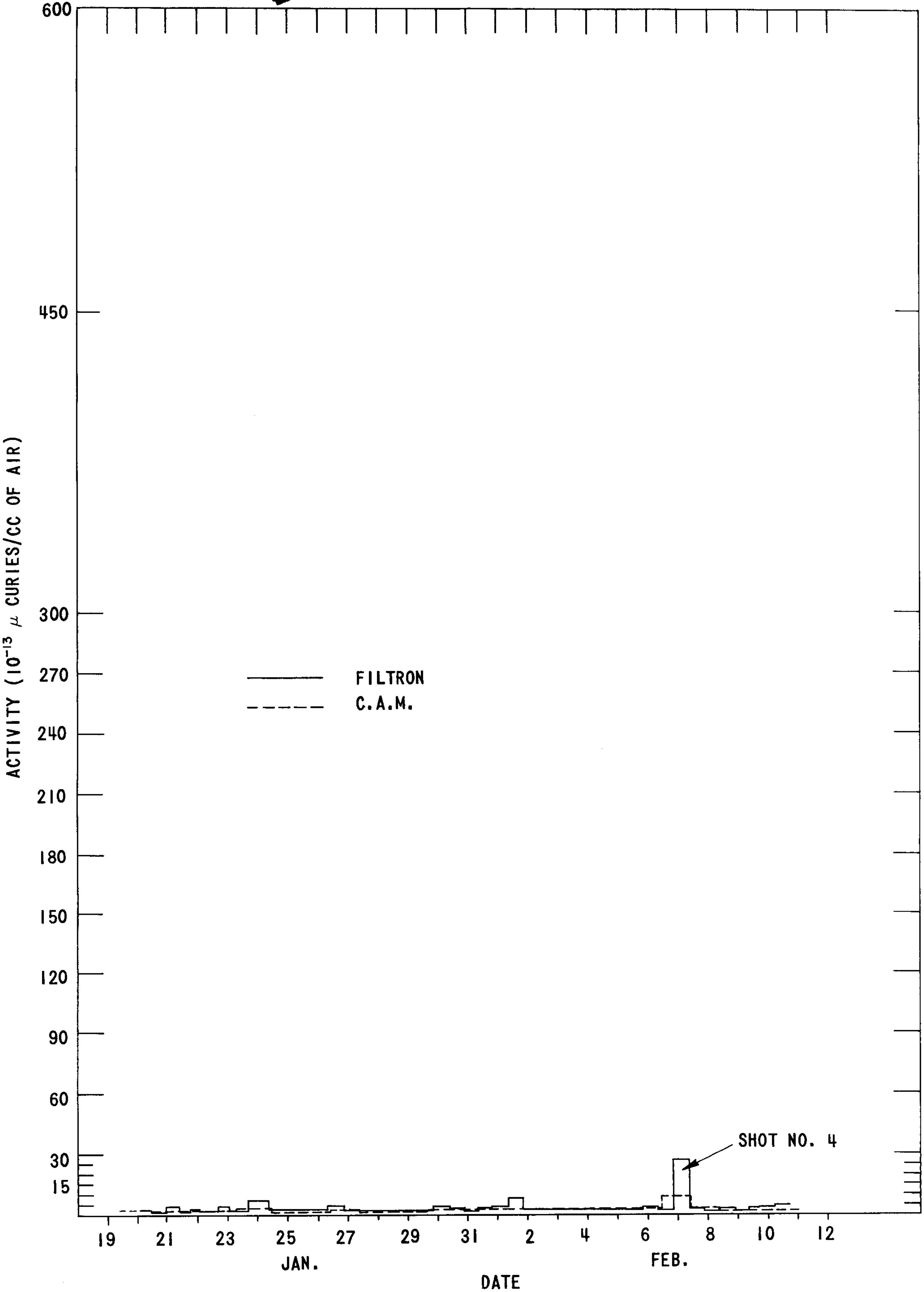

Fig. 4— ATLANTA, GEORGIA

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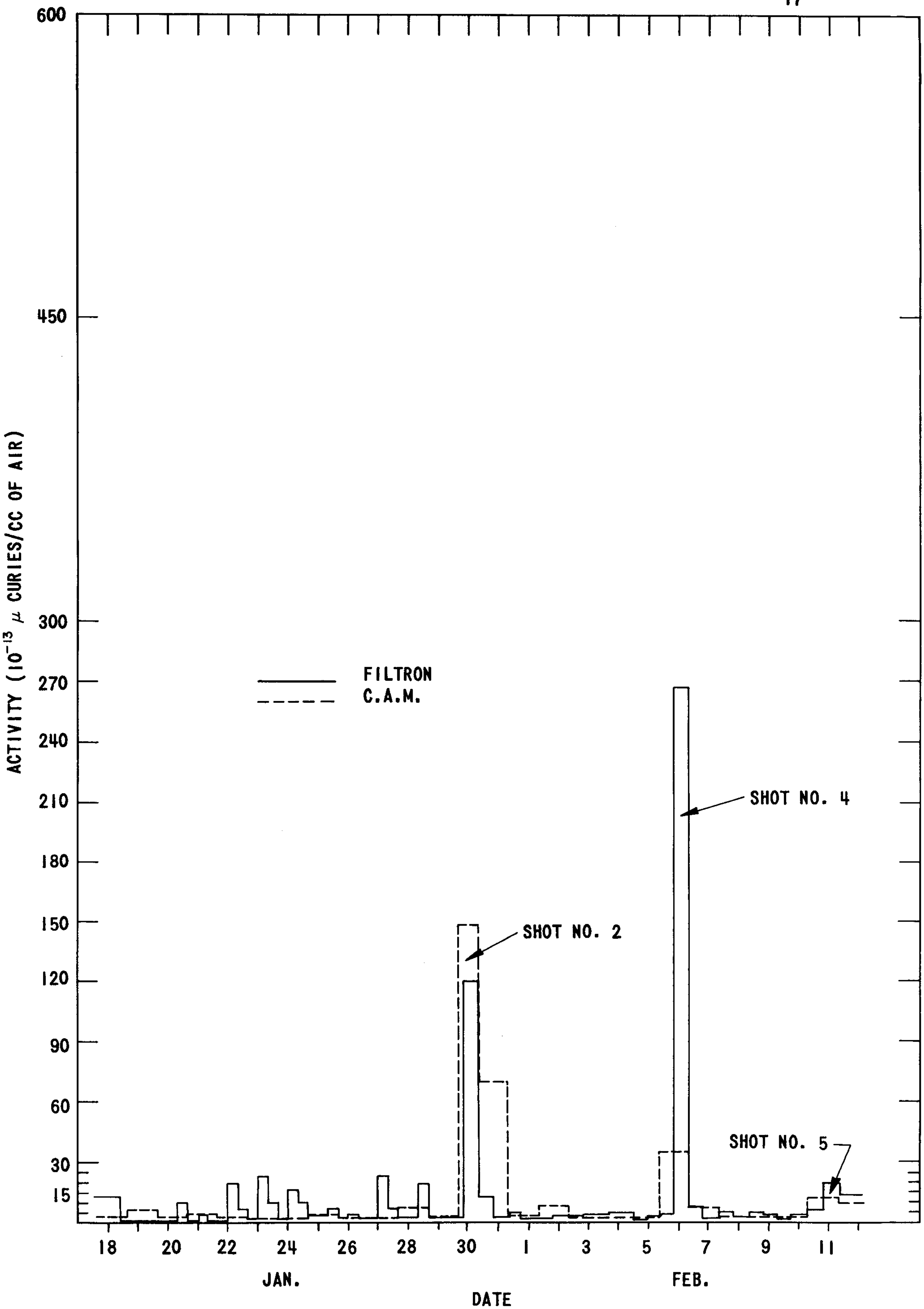
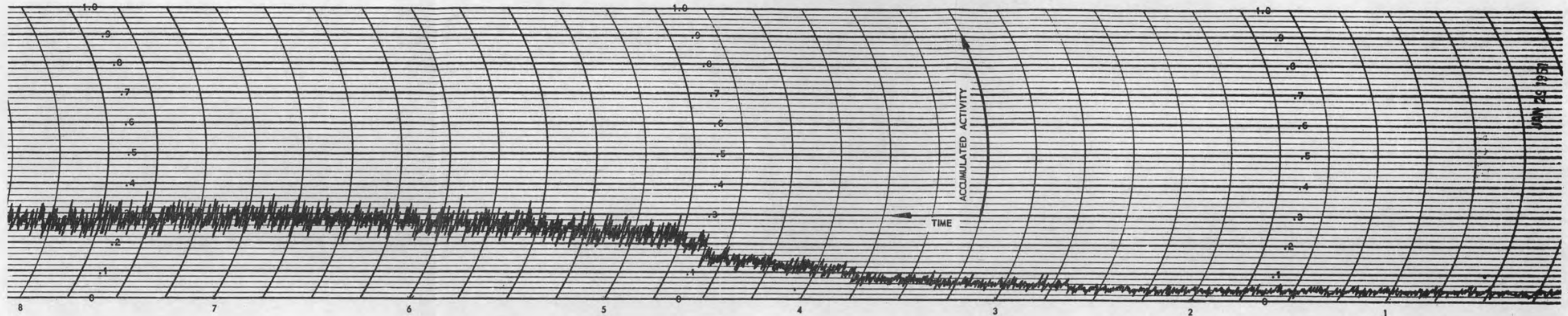


Fig. 5 - FORT WORTH, TEXAS

Fig. 6 b – CONSTANT AIR MONITOR RECORD FROM URBANA ON JANUARY 29, WHICH SHOWS ACCUMULATED ACTIVITY COLLECTED VERSUS TIME. ACTIVITY INCREASES TOWARD THE RIGHT. CHANGES OF SLOPE CORRESPOND TO ACTIVITY PEAKS IN FIGURE 6a.



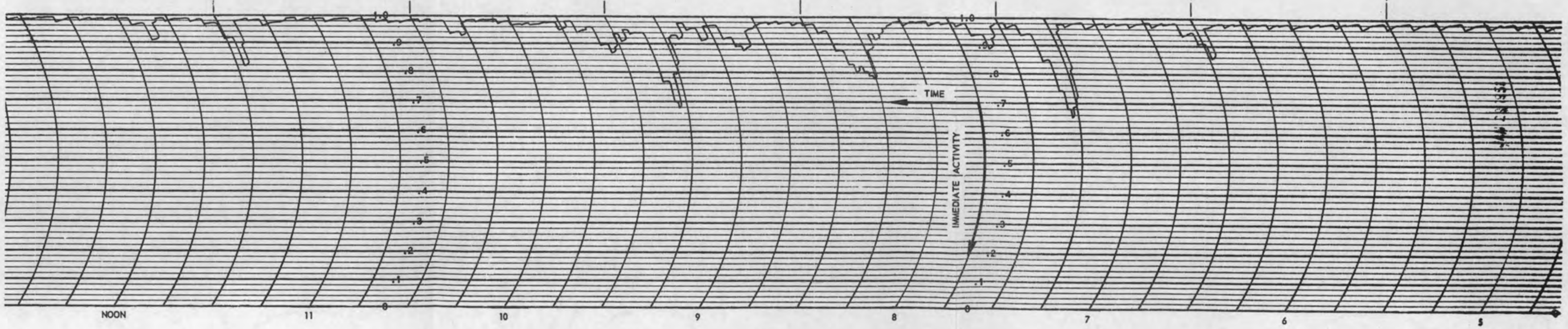


Fig. 6a – MOVING FILTER MONITRON RECORD, DECAY VERSUS TIME, FROM URBANA ON JANUARY 29, WHICH SHOWS ACTIVITY AFTER 4½ HOURS. ACTIVITY INCREASES TOWARD LEFT. RECORD HAS BEEN SHIFTED 4½ HOURS SO TIME OF COLLECTION WILL CORRESPOND WITH TIME SHOWN IN FIGURE 6b.

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Figure 6a is shown by greater displacement to the left while in Figure 6b higher activity displaces the trace to the right.

Since the counter on the moving filter sees a point on the filter for about ten minutes some of the peaks may be due to only a single particle. Since the activity on both records shows good correlation it is felt that the peaks are not due to single particles. This is also borne out on the radioautograph shown in Figure 9 which shows a collection of a large number of particles. It is interesting to note the large variations of the activity shown on the record. This may be due to shifting winds or other meteorological changes.

BACKGROUND

The average count rate due to thorium products collected by the filtrons is shown in the second column of Table I. The standard deviations of the thorium collections are shown in the third column; this gives an indication of the variation of the thorium content in the air from day to day. For comparison the peak values for the first count (X) are given in cts/min in the fourth column. In the fifth column are given the values of $Y - Xe^{-\lambda t}$ for the peaks, i.e. the difference between the second count (Y) and the first count (X) multiplied by the decay of thorium B; this gives values which are proportional to the fission product activity with the thorium activity taken out. The sixth column gives the standard deviations

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of this difference when no fission products are present which is an indication of the reliability of the numbers given in column 5.

The advantage of making two counts on a sample, so that the thorium activity can be calculated and subtracted, is shown using data from Topeka by comparing the ratio of the fifth and sixth columns ($\frac{152}{6} = 25.3$) to the ratio of the fourth and third column ($\frac{330}{49} = 6.7$). This is analogous to signal-to-noise calculations. If there are few thorium products present in the air such as shown for Urbana and Nashville there is no advantage in making two counts to subtract out the thorium activity since one introduces more error from making the two counts than there is variation of the thorium content. For this reason the fission product activity for Urbana and Nashville (Figures 1 and 3) was calculated from its first count only. During most of the period of observation at Urbana and Nashville, snow covered the ground so that one would not expect large amounts of thorium products. The other three stations had little or no snow during the period of observations.

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Table I

Station	Average Thorium Collections (cts/min)	Standard Deviations of Thorium Collections (cts/min)	Peak ct/min First Count (X)	$Y - Xe^{-\lambda t}$ (cts/min)	Standard Deviations
Urbana	7	5	1410 144	392 66	2
Topeka	72	49	330	152	6
Nashville	5	4	520	---	-
Atlanta	20	18	99	36	3
Ft. Worth	51	33	270 495 82	68 197 11	4
1	2	3	4	5	6

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RADIOAUTOGRAPHS

In an effort to determine the nature of the activity on the filters from the Constant Air Monitors and the Filtrons, radioautographs were made on some of the filters. Inasmuch as the activity on the filters had decayed due to counting time at the sampling site, travel time of filters to ORNL, and then another 24 hours of counting time at ORNL, it was decided to radioautograph only the two highest activity filters from each sampling station. Table II shows the data on filters that were radioautographed.

Figures 7 through 12 show the negatives of the radioautographs with white spots corresponding to dark radiation spots on the films. Eastman's X-ray film (No-screen) was used from which non-gloss contact print was made. For this report two additional steps in reproduction are required thus decreasing to a considerable extent the resolution inherent in the original radioautographs. The radiation spots in Figures 7 and 8 have been circled so they can be located more easily. Any other white spots on these two figures appearing in the final reproduced report do not represent radiation but specks of dirt, etc., involved in processing for reproduction. Normal radiation spots will usually have a white center with a gradual graying to the background of the film. Non-radiation spots are usually sharp white spots in the normal background with very little graying edges. In Figures 9, 10, 11, and 12 there were so many radiation spots that circling them was not practical.

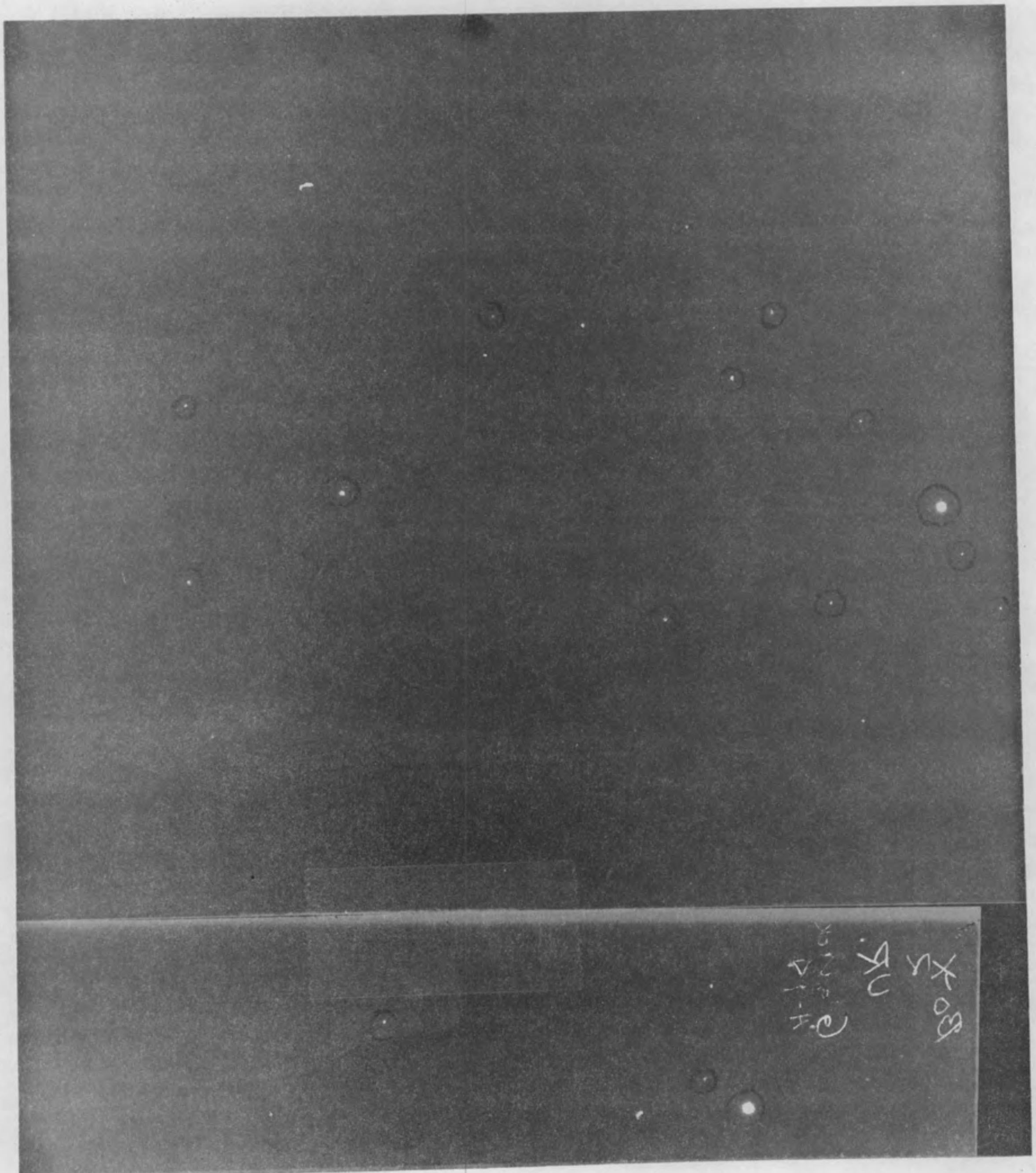

Figure 7

Upper Photo: Ft. Worth, Texas (F-37) Sampling Time 1-29:1621 to 1-30:0821 Counts/min 30 mg/cm^2 [2-5-51(107.4)] [2-6-51(107.2)] Radioautographed 70 hrs 2-9 to 2-12

Lower Photo: Ft. Worth, Texas (CAM-14) Sampling Time 1-30:0831 to 1-31:0916 Counts/min 30 mg/cm^2 [2-5-51(76.5)] [2-6-51 (71.3)] Radioautographed 70 hrs 2-9 to 2-12

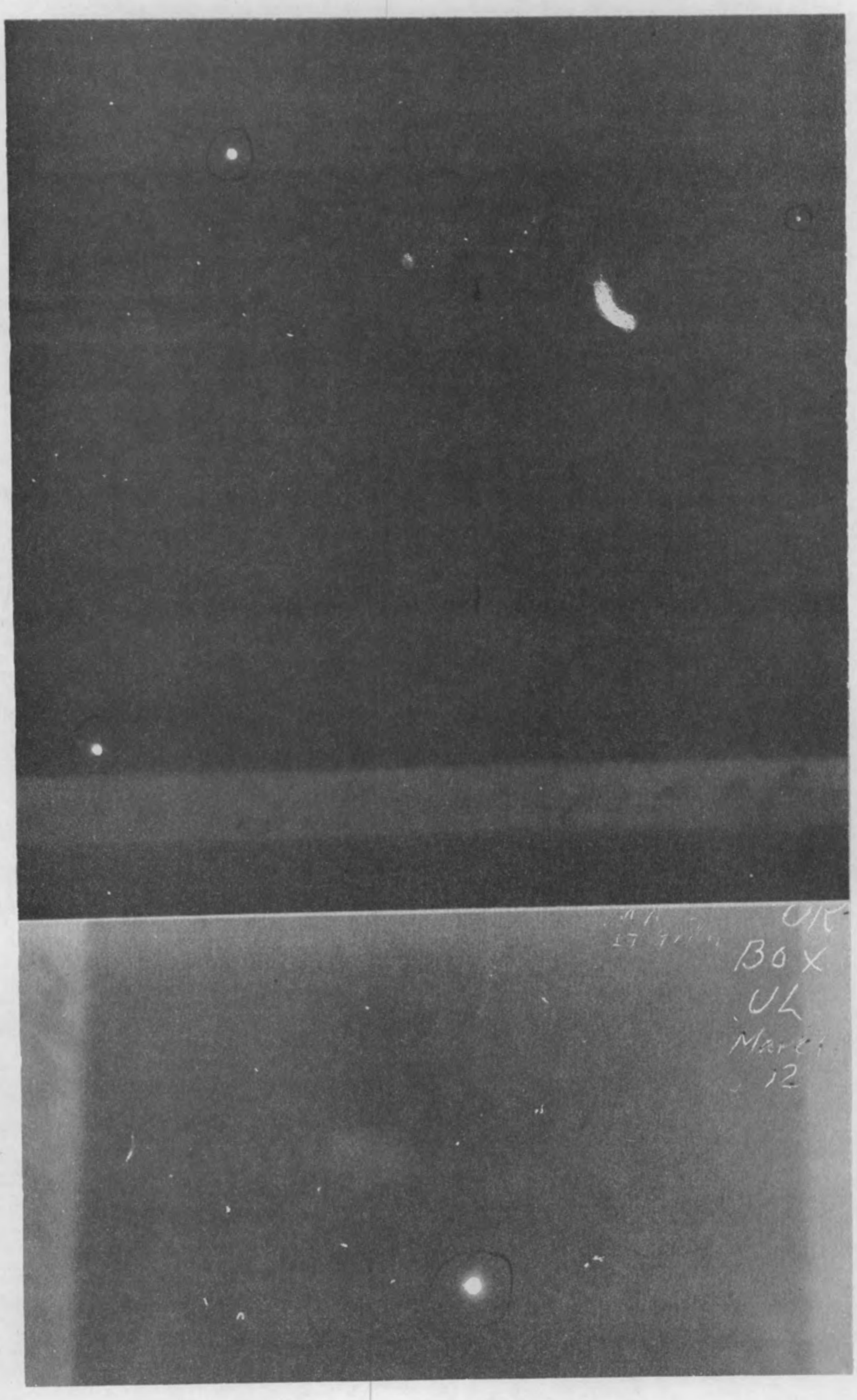

Figure 8

Upper Photo: Topeka, Kansas (F-35) Sampling Time 1-29:1201 to 1-30:0830 Counts/min 30 mg/cm^2 [2-8-51(13.4)] [2-9-51(12.0)] Radioautographed 70 hrs 2-9 to 2-12

Lower Photo: Topeka, Kansas (CAM-12) Sampling Time 1-29:1617 to 1-30:1620 Counts/min 30 mg/cm^2 [2-8-51(35.0)] [2-9-51(27.9)] Radioautographed 70 hrs 2-9 to 2-12

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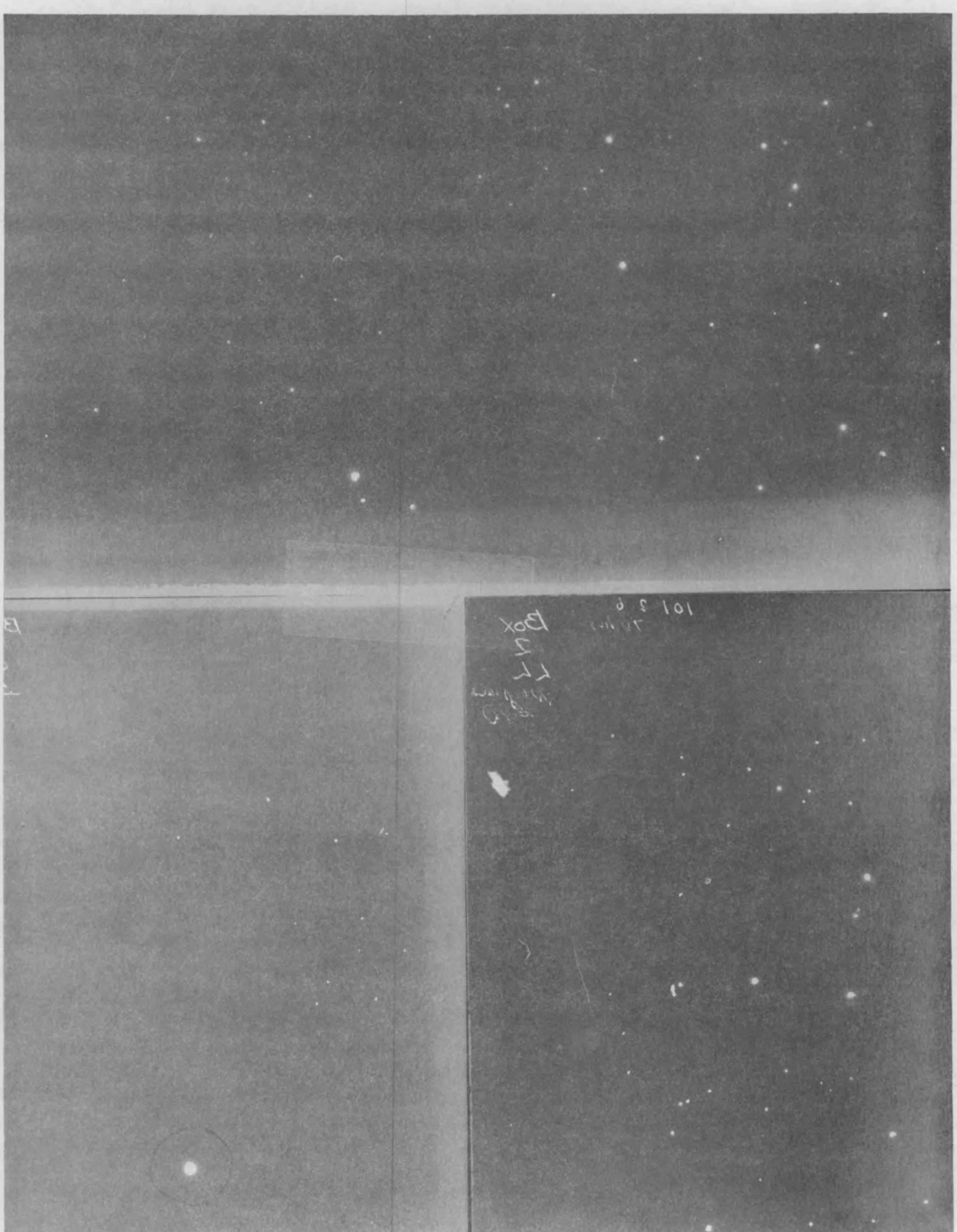

Figure 9

Upper Photo: Urbana, Illinois (F-27) Sampling Time 1-28:2400 to 1-29:0800 Counts/min 30 mg/cm^2 [2-5-51(236.2)] [2-6-51(210.1)] Radioautographed 70 hrs 2-9 to 2-12

Lower L Photo: Urbana, Illinois (CAM-12) Sampling Time 1-30:2100 to 1-31:2100 Counts/min 30 mg/cm^2 [2-5-51(74 c/m)] [2-6-51 (70.4)] Radioautographed 70 hrs 2-9 to 2-12

Lower R Photo: Urbana, Illinois (CAM-10) Sampling Time 1-28:1600 to 1-29:1600 Counts/min 30 mg/cm^2 [2-5-51(136.1)] [2-6-51 (101.0)] Radioautographed 70 hrs 2-9 to 2-12

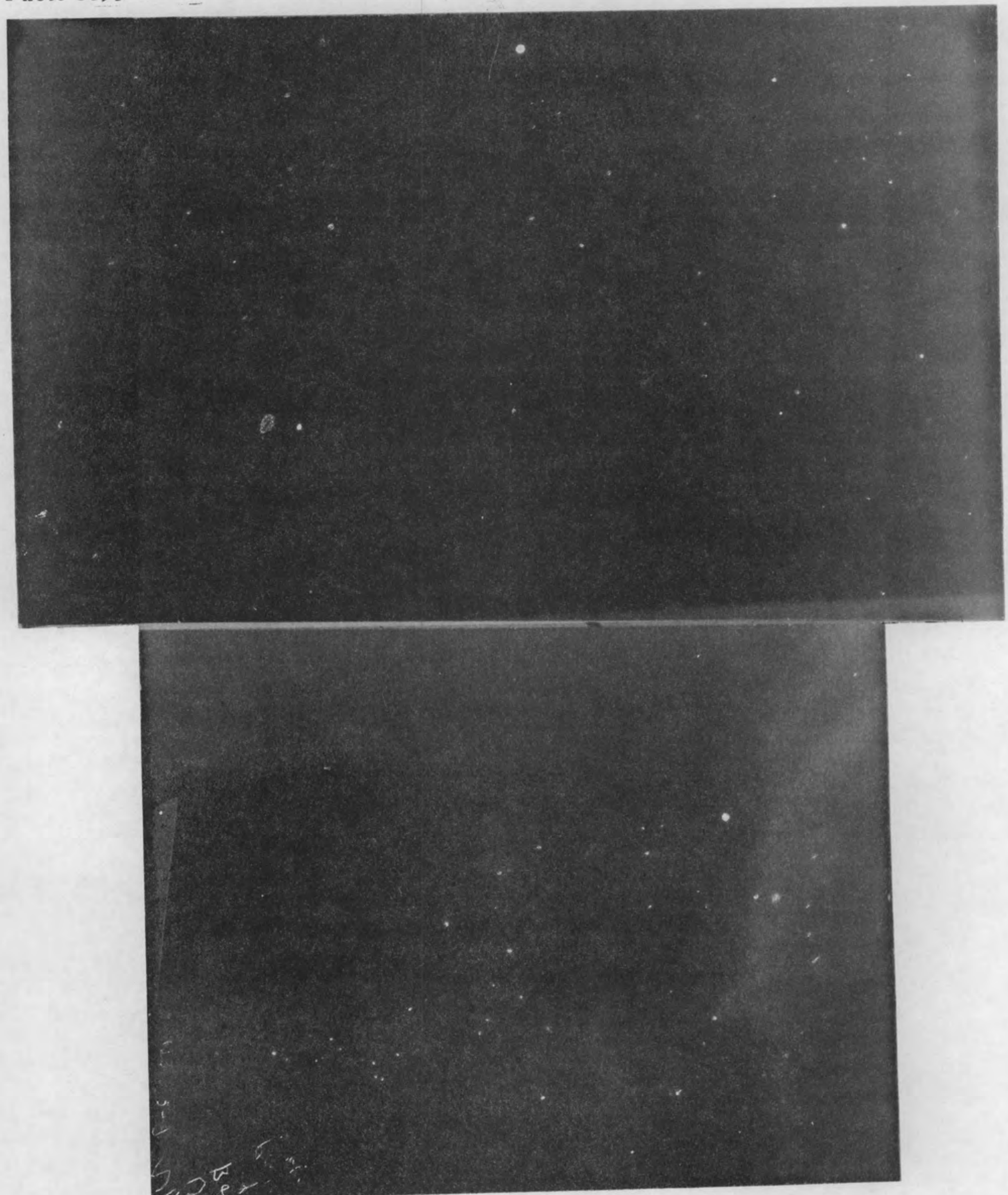

Figure 10

Upper Photo: Nashville, Tennessee (F-27) Sampling Time 1-29:0700 to 1-30:0930 Counts/min 30 mg/cm^2 [2-8-51(157.3)] [2-9-51 (139.9)] Radioautographed 70 hrs 2-9 to 2-12

Lower Photo: Nashville, Tennessee (CAM-12) Sampling Time 1-29:0930 to 1-30:0930 Counts/min 30 mg/cm^2 [2-8-51(192.4)] [2-9-51 (173.3)] Radioautographed 70 hrs 2-9 to 2-12

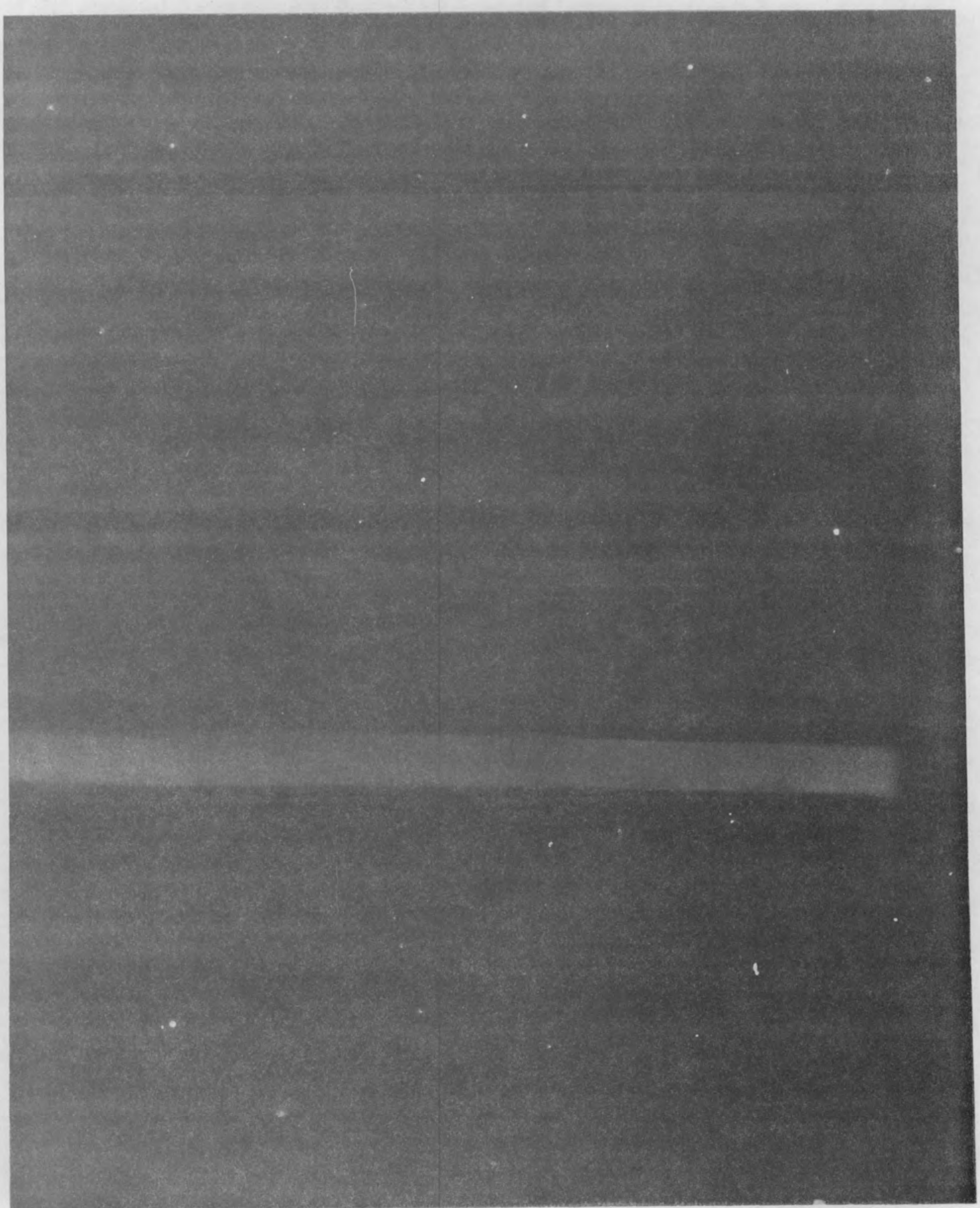

Figure 11

Upper Photo: Atlanta, Georgia (F-43) Sampling Time 2-6:0830 to 2-7:0830 Counts/min 30 mg/cm^2 [2-12-51(44.4)] [2-13-51(33.3)] Radioautographed 70 hrs 2-16 to 2-19

Lower Photo: Atlanta, Georgia (CAM-20) Sampling Time 2-6:0830 to 2-7:0830 Counts/min 30 mg/cm^2 [2-12-51(22.3)] [2-13-51 (22.9)] Radioautographed 70 hrs 2-16 to 2-19

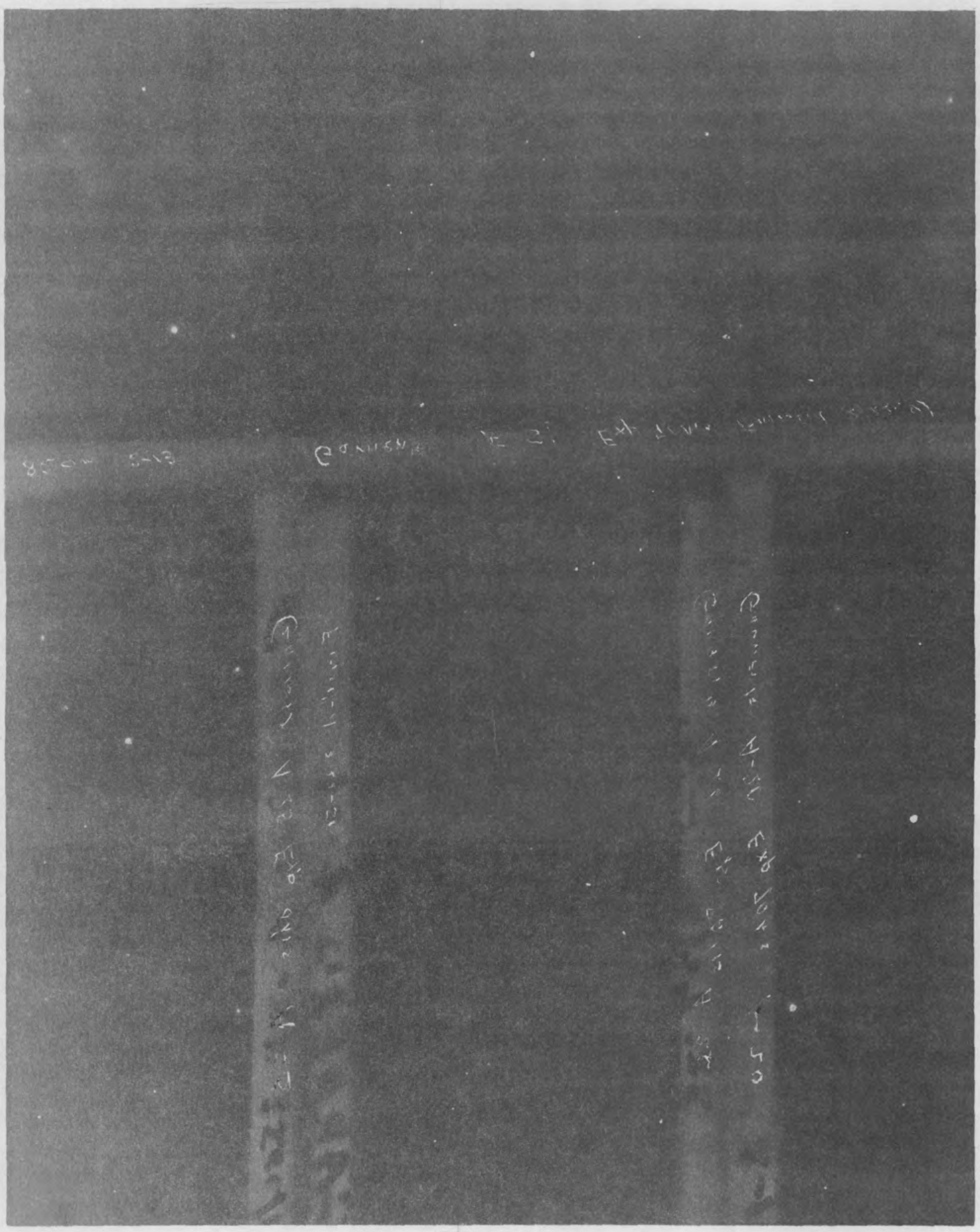

Figure 12

Upper Photo: Ft. Worth, Texas (F-51) Sampling Time 2-5:2021 to 2-6:0820 Counts/min 30 mg/cm^2 [2-12-51(24.3)] [2-13-51(8.2)] Radioautographed 70 hrs 2-19 to 2-22

Lower L Photo: Ft. Worth, Texas (CAM-25) Sampling Time 2-10:0835 to 2-11:0830 Counts/min 30 mg/cm^2 [2-15-51(35.3)] [2-16-51 (35.0)] Radioautographed 70 hrs 2-19 to 2-22

Lower C Photo: Ft. Worth, Texas (CAM-26) Sampling Time 2-11:0825 to 2-12:0830 Counts/min 30 mg/cm^2 [2-15-51(34.1)] [2-16-51 (32.2)] Radioautographed 70 hrs 2-19 to 2-22

Lower R Photo: Ft. Worth, Texas (CAM-20) Sampling Time 2-5:0835 to 2-6:0830 Counts/min 30 mg/cm^2 [2-7-51(149.9)] [2-13-51 (36.1)] Radioautographed 70 hrs 2-19 to 2-22

Table II

Filter Location	Filter Number	Sampling Time at Location: Start	Sampling Time at Location: Stop	Counting Date at ORNL and Counts/min thru 33 mg/cm²		Radioautographed 70 Hours Between
Fort Worth	F-37[1]	1-29:1621	1-30:0821	2-5-51 - 107.4	2-6-51 - 107.2	2-9 to 2-12
	CAM-14[2]	1-30:0831	1-31:0916	2-5-51 - 76.5	2-6-51 - 71.3	2-9 to 2-12
Topeka	F-35	1-29:1201	1-30:0830	2-8-51 - 13.4	2-9-51 - 12.0	2-9 to 2-12
	CAM-12	1-29:1617	1-30:1620	2-8-51 - 35.0	2-9-51 - 27.9	2-9 to 2-12
Urbana	F-27	1-28:2400	1-29:0800	2-5-51 - 236.2	2-6-51 - 210.1	2-9 to 2-12
	CAM-10	1-28:1600	1-29:1600	2-5-51 - 136.1	2-6-51 - 101.0	2-9 to 2-12
	CAM-12	1-30:2100	1-31:2100	2-5-51 - 74	2-6-51 - 70.2	2-9 to 2-12
Nashville	F-27	1-29:0700	1-30:0930	2-8-51 - 157.3	2-9-51 - 139.9	2-9 to 2-12
	CAM-12	1-29:0930	1-30:0930	2-8-51 - 192.4	2-9-51 - 173.3	2-9 to 2-12
Atlanta	F-43	2-6 :0830	2-7 :0830	2-12-51 - 44.4	2-13-51 - 33.3	2-16 to 2-19
	CAM-20	2-6 :0830	2-7 :0830	2-12-51 - 22.3	2-13-51 - 22.9	2-16 to 2-19
Fort Worth	F-51	2-5 :2021	2-6 :0820	2-12-51 - 24.3	2-13-51 - 8.2	2-19 to 2-22
	CAM-20	2-5 :0835	2-6 :0830	2-7-51 - 149.9*	2-13-51 - 36.1	2-19 to 2-22
	CAM-25	2-10:0835	2-11:0830	2-15-51 - 35.3	2-16-51 - 35.0	2-19 to 2-22
	CAM-26	2-11:0825	2-12:0830	2-15-51 - 34.1	2-16-51 - 32.2	2-19 to 2-22

*Counted at Fort Worth, Texas.

[1] F refers to a filter from a filtron.

[2] CAM refers to a filter from a constant air monitor.

On the basis of the above photographs, Urbana had the most radioactive particulates with Nashville second. Urbana was on the predicted central path for shots one and two. Fort Worth and Topeka showed fewer radioactive particles, but a greater proportion of the particles produced large spots. Assuming constant specific activity in all particles, one would state that fewer small particles were deposited at these two stations. It is presumed that the larger particles at Fort Worth and Topeka fell quickly and that the finely divided material did not appear at ground levels until Urbana and Nashville were reached. It is to be emphasized that due to the rapid succession of detonations, it is quite difficult to state that a particle came from one shot or the following one, when meterological conditions make the carrying wind stratas overlap.

The circled radiation spot in Figure 9 was the only radiation effect produced by the filter and it was assumed that all counts obtained on the filter were due to this particle.

An isolation of this particle was attempted. It was finally separated on a microscope slide which contained several other non-active particles. The largest particle on the slide measured 17 microns, therefore, the radioactive particle was 17 microns or less. Approximately six months after collection this particle gave 62 disintegrations per minute.

RAINWATER TREATMENT

The rainwater samples at the stations were collected by tapping into drain lines from roofs. Where snow was collected it was either shoveled off the ground or collected on a tarpaulin. These samples, ranging in volume from 25 to 52 gallons, were treated with aluminum sulfate and sodium carbonate. The resulting aluminum hydroxide precipitate was decanted and the floc shipped to ORNL. Here it was dissolved in nitric acid, and the gross beta activity levels were determined by counting suitable aliquots with an end-window GM counter.

In Table III is shown the gross activity of the rain collections. The dates of the high values obtained for certain of the collections are in excellent agreement with the air filter observations. The rainwater activity level at Atlanta, Georgia, on February 6 and 7 was higher than the activity level on January 24 by a factor of at least 425. In Table IV are given the radiochemical analyses of two of the highest samples which shows the activity to be that of fission products.

The presence of a number of short-lived fission products (i.e., Ba^{140}, I^{131}, and Te^{132}) indicates that the fission product contaminant is not old.

The decay curves for four of the radiochemical separations are shown in Figures 13 through 16.

Table III

Rainwater Activity Levels* by Date and Location

Collection Date	Location	Volume Collected	d/m per Volume	d/m per 100 Gallons	10^{-7} μc/cc
1-24-51 (Rain)	Nashville	50 gal.	470	940	0.011
1-24-51 (Rain)	Atlanta	25 gal.	290	1180	0.014
1-31-51 (Snow)	Ft. Worth	52 gal.	1.21×10^5	2.33×10^5	2.77
2-6-51 (Rain)	Ft. Worth	52 gal.	1.22×10^5	2.35×10^5	2.79
2-6 and 2-7-51 (Rain)	Atlanta	43 gal.	2.15×10^5	5.00×10^5	5.95
2-6-51 (Rain)	Topeka	25 gal.	1.41×10^4	5.64×10^4	0.67

*The quoted activity levels are not corrected for the carrying efficiency of the aluminum hydroxide floc.

Table IV

Identity of Contaminants by Radiochemical Analyses of Two Samples

(activity in terms of per cent of gross beta count)

	Total Rare Earths*	Ce	Ba	I	Ru	Te	Zr	Nb	Cs	Sr
1-31-51 Ft. Worth	52.5	12.8	7.9	3.0	3.0	- - - Not analyzed for - - -				2.0
2-6 and 2-7-51 Atlanta	51.5	12.7	4.2	3.9	4.7	2.7	1.2	0.2	0.2	0.7

* Total rare earths including cerium.

[redacted] [redacted]

DWG. 12431

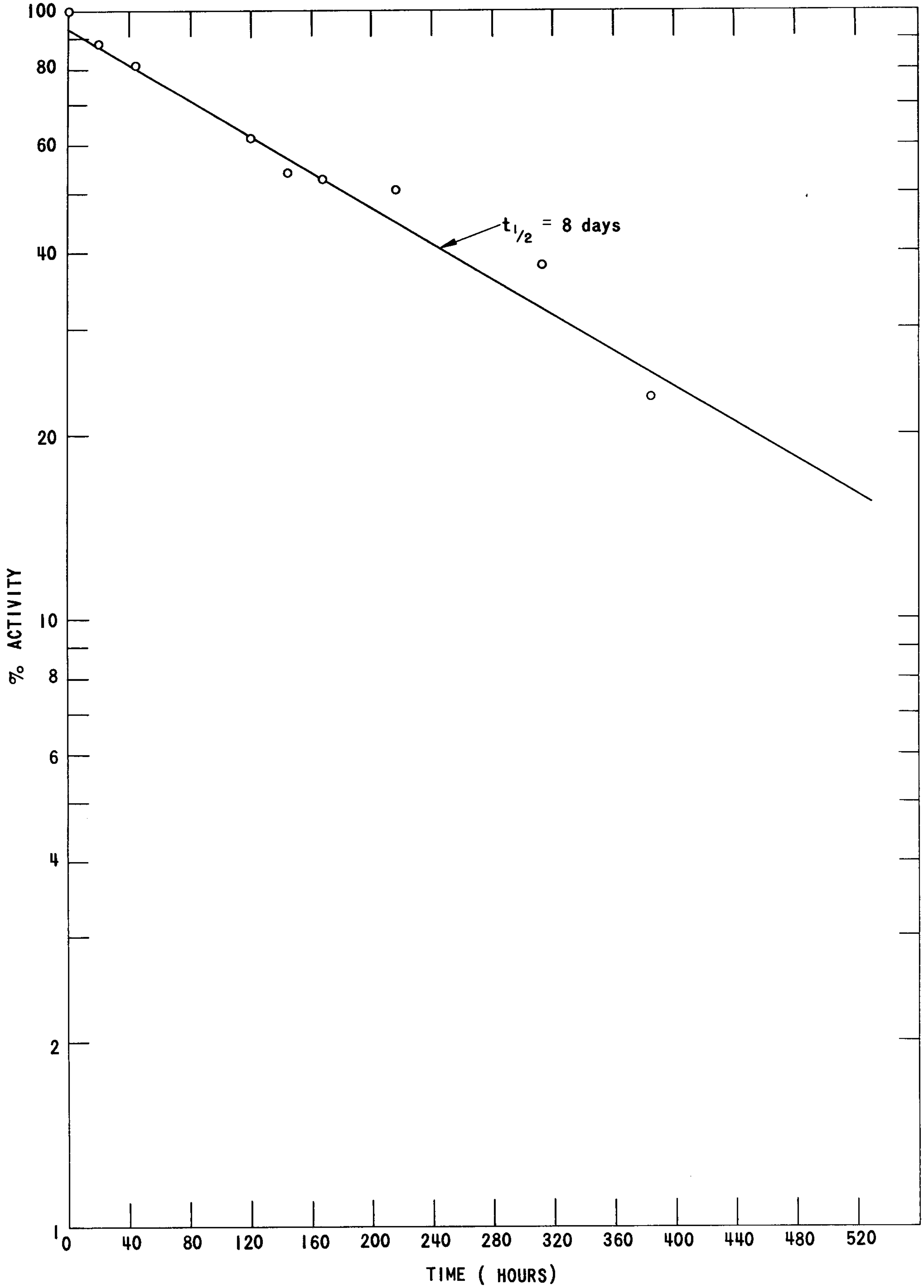

Fig. 13 — IODINE[131] DECAY COLLECTED 2-6-51, ATLANTA

[redacted] [redacted]

DWG. 12432
35

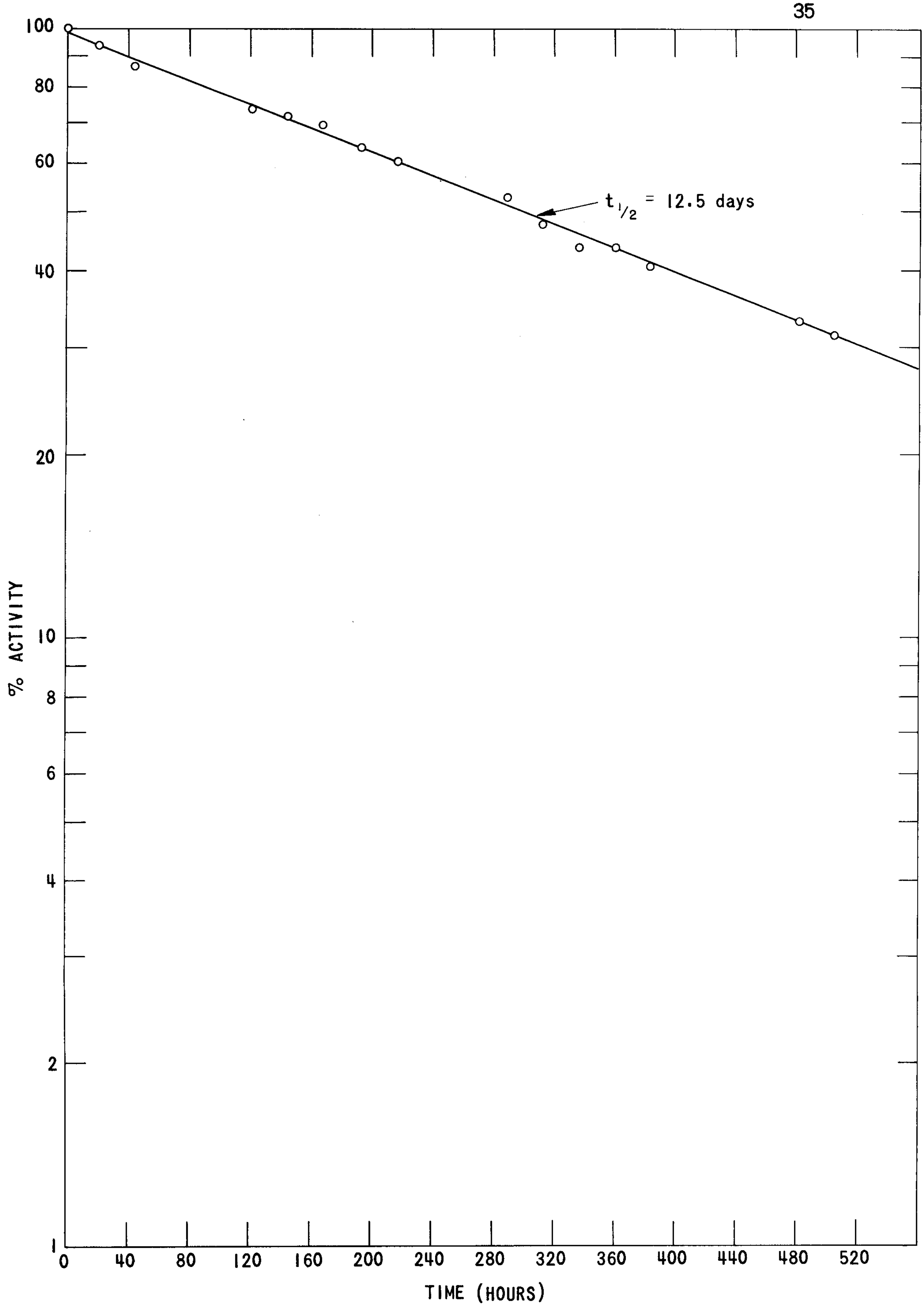

Fig. 14 — BARIUM[140] DECAY COLLECTED 2-6-51, ATLANTA

[redacted] [redacted]

DWG. 12440
36

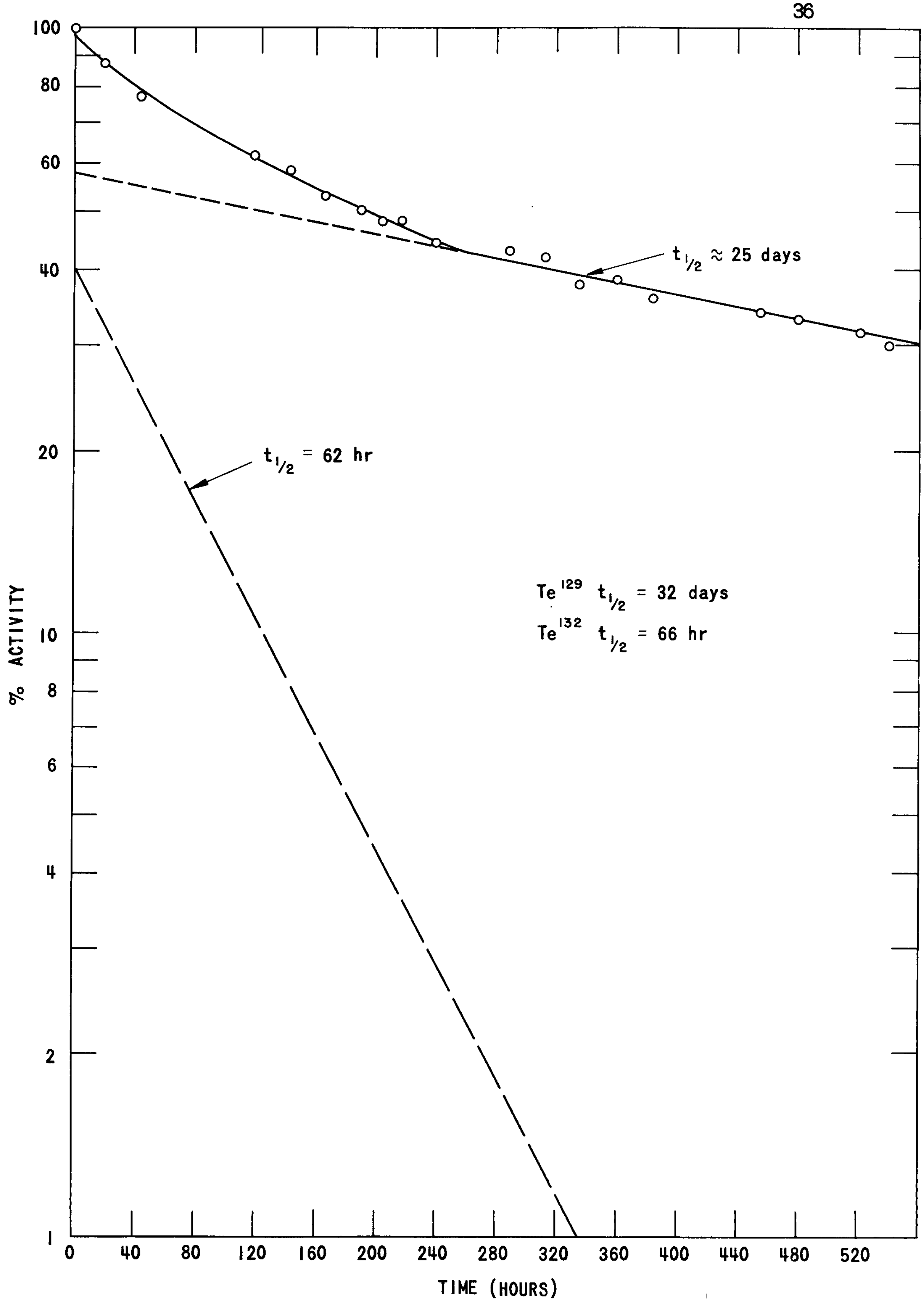

Fig. 15— TELLURIUM DECAY COLLECTED 2-6-51, ATLANTA

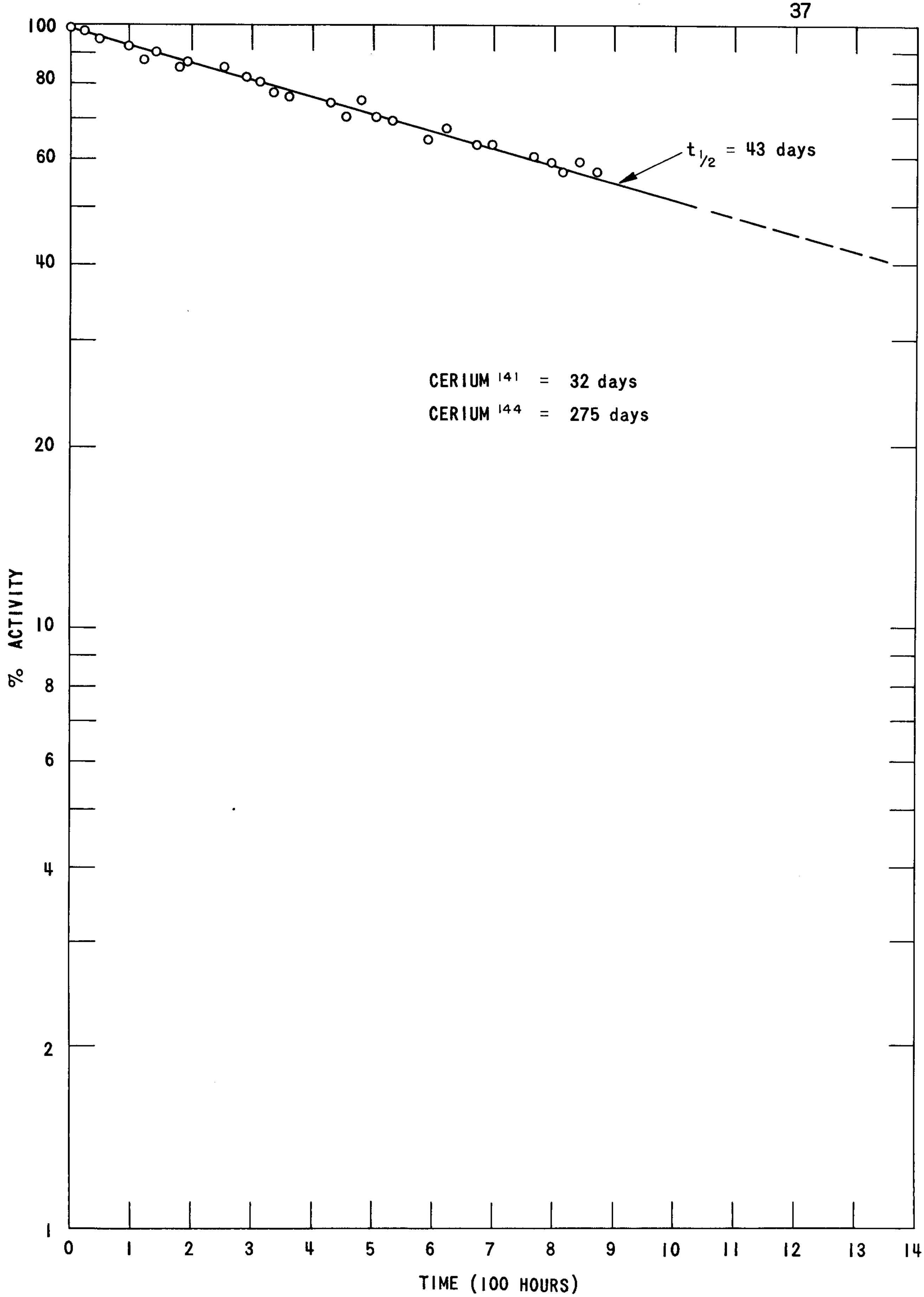

Fig. 16 — CERIUM DECAY COLLECTED 2-6-51, ATLANTA

[redacted]

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ACKNOWLEDGEMENTS

The author thanks the following institutions and people for the use of their facilities and their cooperation:

Emory University, Atlanta, Georgia

Dr. W. H. Jones - Chemistry Department

[redacted]

University of Illinois, Urbana, Illinois

Dr. A. O. Hanson - Physics Department

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