

SPECIAL RE-REVIEW

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HW-10758

FINAL DETERMINATION

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BY *JP Drown* DATE 2-19-81BY *JN Jordan* DATE 2-19-81

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August 17, 1948

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LONG-LIVED FISSION ACTIVITIES IN THE STACK GASES AND VEGETATION
 AT THE HANFORD WORKS

The deposition of radio-iodine (I^{131}) from the stacks in the 200 Areas has long been recognized as a possible hazard, especially to live-stock grazing on the contaminated vegetation. (1)(2) The presence of other fission products with long half-lives in the stack gases has presented the possibility of a similar hazard with these materials. (3) During the early days of plant operation, the major part of the work by the H.I. Group on the stack was devoted to evaluating the hazard due to the iodine. (4) Some effort was expended, however, in making a few spot measurements on the other fission products in the stack effluent and on the vegetation. In general, the work was uncorrelated and somewhat sketchy because of the lack of personnel to assign to this one problem.

This report presents the data available in the Health Instrument Divisions on the stack problem for those interested in the problem. More detailed work is at present being done by both the Technical and Health Instrument Divisions.

Summary

A series of decay curves on filter papers through which air from the stack and from locations on the reservation was pulled indicate that the long-lived activity is composed of materials with a half-life of 30-60 days (60-80%), 275-300 days (30-40), and a small amount (1-2%) with very long half-life. These curves are confirmed by a series of radiochemical analyses. The total amount of long-lived activity discharged from each stack is estimated at 0.1 to 180 millicuries per day. There is some indication that this value has increased in the past year. The long-lived activity in vegetation is shown to have a composition similar to that which would be expected after three years exposure to the stack effluent. The long-lived activity level ranges from < 0.02 $\mu\text{c/kg}$ at Richland to 0.40 $\mu\text{c/kg}$ in the 200 Area.

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Procedures

A. Stack Sampling

Early samples from 1945 and 1947 were obtained by pulling air with a Motocaire pump at approximately 2 CFM, from the 50 foot level in the stack through a piece of asbestos filter paper about $1\frac{1}{2}$ inches in diameter. The time of sampling varied depending upon the particular conditions at the time. Since the primary reason for sampling was investigation of the plutonium content of the air, only a few of the filters were saved for decay measurements.

The results from 1948 were obtained by pulling the air from the tunnel between the Canyon Building and the Fan Building, from the breach between the fans and the stack, from the dissolver off-gas line, and from the 50 foot level of the stack. These samples were obtained by the 200 Area Plant Assistance Group of the Technical Division. The filters were analyzed for total beta activity by extracting the activity with hot nitric acid or with nitric acid and potassium perchlorate followed with a beta count on an aliquot. The quantities of individual fission products were found by analyzing an aliquot of the nitric acid extract according to the procedures listed in the literature. (5)

B. Air Sampling

The Site Survey Group has filter units, similar to the one described for stack sampling, at various locations to monitor the quantity of fission products in the air. Some of the filter papers from such monitors have been saved for decay studies in order to obtain a measure of the long-lived activity concentration. In addition, some samples were taken from the pads in the desert coolers of the 222 and 224 Buildings after a downdraft in May 1945, had contaminated these buildings with radio-iodine.

C. Vegetation Sampling

A routine program for measuring the activity deposited on vegetation was started late in 1945. The vegetation most prevalent in the particular region was sampled and one gram aliquots mounted in the form of a small pellet about $1\frac{1}{2}$ inches in diameter by using a hand operated press. A series of tests of adding a known amount of active iodine to blank samples indicated that this type of sample must, on the average, be corrected by a factor of three for self-absorption and spread of the sample. The applicability of such a correction factor to the long-lived activity is doubtful.

A few of the samples obtained on this routine program were saved for decay measurements. In general, only the more active samples were saved and counted.

A more recent program instituted in early 1948 was designed to measure directly the long-lived activity by taking advantage of the volatility of iodine in hot nitric acid. A 50 gram sample of the vegetation was wet ashed with nitric acid until only the mineral salts were left. A 1/50th aliquot of this residue was placed directly on a stainless steel plate for beta counting.

D. Counting

All samples were counted on standard 1 inch diameter thin mica window counters contained in a lead pig with $1\frac{1}{2}$ inch thick walls. The shelves were arranged so that the sample was about 1/8 inch below the counter face on the first shelf and about 3/4 inch on the second shelf. Stack samples and air filters were mounted directly on $3\frac{1}{4} \times 2\frac{1}{2}$ cards, covered with cellophane, and counted on the first shelf. The vegetation samples formed by the press were counted on the second shelf because of the bulky pellet.

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Counting rates were measured on all decay samples at intervals of one day to several weeks depending on the type of sample and the length of time that it had been decaying. All measurements for a given sample were made on the same counting equipment, although GM tubes were changed during the course of the experiment. These counting rates were not corrected for absorption in the counter window or back scatter. The analytical results for total amounts of activity on the filters were corrected for back scatter from the counting plate, absorption in the counter window, and geometry, but not for self-absorption or self-scatter. The results expressed in microcuries have not been corrected for disintegration scheme; for this data one microcurie is considered as 3.7×10^4 beta particles emitted from the sample per second.

E. Decay Curve Analysis

In order to obtain the most accurate analysis of the curves, advantage was taken of the analytical results available from 1948 (Table IIX). These results indicated a given percentage of individual elements in the stack samples. An estimate of the isotopic ratios in those cases where two isotopes are formed may be calculated by assuming 200 days of irradiation and 110 days cooling time. The final percentages taken for a comparison curve are given in Table I.

TABLE I
COMPONENTS OF COMPARISON CURVE

<u>Isotope</u>	<u>Percentage of Total Activity</u>	<u>Half-Life</u>
Ce ¹⁴¹	8	30 d
Sr ⁸⁹	17	55 d
Y ⁹¹	36	57 d
Zr ⁹⁵	10	65 d
Ce ¹⁴⁴	28	275 d
Sr ⁹⁰ and others	1	>10 y

A decay curve was made for a mixture having this composition at the start and all measured curves were compared with this one by overlaying the two sheets. It was found that the curves could be made to match by displacing the time scale so that the start of the measured curve was some time before or after the calculated one. The percentages of the components were then corrected for the amount of decay which would have occurred for each isotope in the time of this displacement.

The results are expressed in terms of the half-lives since it was felt that it would be impossible to separate the 55 day half-life due to Sr⁸⁹ from the 65 day due to Zr⁹⁵ by this means. Thus, the fifty-five day group would include Ce¹⁴¹ (30d), Sr⁸⁹ (55d), Y⁹¹ (57d), Zr⁹⁵ (65d), and Ru¹⁰³ (45d). The 275 day group would include Ce¹⁴⁴ (275d) and Ru^{104,105} (1 year).

Results

A. Stack Samples

A series of decay curves from samples taken in 1945 and 1947 from the "T" Plant stack are presented in Figures 1, 2, and 3. The solid line represents the decay curve obtained from the procedure described in the preceding section. Some values obtained from these curves and from several other unpublished curves are given in Table II.

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TABLE II

RESULTS FROM DECAY CURVES ON STACK SAMPLES

Sample Date	Time Sample	Long-Lived Activity	Apparent Composition		
			55 Day	275 Day	Long
~ 5/11/45	-	2.4×10^{-2}	59	40	1
~ 5/11/45	-	1.0×10^{-2}	55	44	1
4/8/47	24 Hr.	9.5×10^{-4}	--	--	-
4/9/47	24 Hr.	2.3×10^{-3}	--	--	-
4/9/47	5 Hr.	1.6×10^{-3}	71	28	1
4/16/47	7 Da.	1.6×10^{-2}	71	28	1
4/18/47	22 Da.	1.9×10^{-3}	71	28	1

A few other curves are available, but unfortunately, the sampling date cannot be set within a two or three month period so that the composition at the time of sampling is unobtainable.

Several analyses were run by R. C. Thorburn for fission products on filters obtained by the Technical Division from the stack system of the "B" Plant during investigation of the active particle problem. The results of some of the analyses are given in Table III.

TABLE III

F.P. ANALYSES ON STACK SAMPLES FROM THE "B" PLANT

Element	Dissolver Off-Gases	Dissolver Off-Gases %	Tunnel %	50' Level %	50' Level %
Ce	35.6	36.3	12.7	37.6	45.2
Rare Earths*	6.4	4.2	0.7	0.1	- -
Y	36.8	36.8	51.3	34.6	- -
Sr	17.7	15.1	4.8	30.7	26.6
Eu	4.8	3.9	40.6	- -	8.7
Cs	0.2	0.3	- -	- -	- -
Zr	7.5	9.3	- -	- -	9.2
Cb	1.4	1.5	- -	- -	- -
Te	1.1	1.5	- -	- -	- -
I	9.8	4.2	- -	8.9	1.3

*Other than those listed

These values represent percentages of total activity and are not corrected for self-absorption or self-scatter occasioned by use of carrier. The cerium value represents the equilibrium counting rate for the parent-daughter relation. The strontium value represents about fifty percent of the equilibrium counting for the buildup of the Y⁹⁰ daughter from Sr⁹⁰.

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Several additional values giving total activity on the filter paper were obtained by L. C. Schwendiman in the course of an experiment designed to calibrate a C. P. meter for total activity in microcuries. These values are listed in Table IV.

TABLE IV
ACTIVITY OF FILTERS

Sampling Location	Sampling Date	Time Sample Hours	Sampling Rate CFM	MC per Filter
Stack	3/8/48	24	6.6	18.1
Stack	3/10/48	24	6.6	37.2
Stack	3/11/48	24	7.8	21.1
Tunnel	3/30/48	22.3	10	13.4
Stack	3/30/48	23.8	8	30.7
Breesh	3/30/48	23.1	9.8	24.5
Tunnel	3/31/48	24.9	11	31.6
Breesh	3/31/48	24.8	10	22.4
Stack	3/31/48	24.7	10	60.5
Stack	4/1/48	22.4	10	32.1
Tunnel	3/30/48	23.3	10	46.2
Tunnel	4/1/48	19.5	9.8	36.1

Some values on the amounts of alpha emitters in the stack effluent were obtained by a series of alpha counts on filter papers taken in 1946 from the 60' level of both stacks. A summary of these results is listed in Table V.

TABLE V
ALPHA ACTIVITY ON FILTERS

T Plant			B Plant		
Date	Sampling Time	Estimated Alpha Activity	Date	Sampling Time	Estimated Alpha Activity
	Hours	d/m		Hours	d/m
8/1/46	22.25	7600	3/27/46	9.8	28,000
8/2/46	22.8	43	4/2/46	17	3400
8/5/46	71.8	32,000	4/4/46	24	6900
8/8/46	21.8	8350	4/25/46	25	1290
8/7/46	27.3	6700	4/26/46	16.7	2400
8/9/46	50.7	2400	5/7/46	15.5	1630
8/10/46	26.3	4850	5/8/46	25.5	57
8/11/46	6.8	640	5/9/46	24	6000
8/11/46	17.1	630	5/10/46	23.5	5150
8/12/46	6.8	210	5/11/46	24.5	6250
8/12/46	17.3	1450	5/12/46	22.5	6150
8/13/46	8.1	300	5/14/46	52.25	8400
8/22/46	220	48,000	5/29/46	5.3	1100

These values have been increased by 30% to compensate for self-absorption in the filter paper. Some values are available on the beta activity but are, in general, too inaccurate to include without further study.

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B. Air Filters - Decay curves from filter papers taken on the reservation are presented in Figures 4, 5, 6, and 7. Some values on the composition of the activity as measured by this means are given in Table VI.

TABLE VI

RESULTS FROM DECAY CURVES ON AIR FILTERS

Sampling Date	Location	Composition		
		55 Day	275 Day	Long
		% Activity	% Activity	% Activity
5/7/45	200 Areas	83	17	~ 0.5
5/9/45	200 Areas	83	17	~ 0.5
5/14/45	200 Areas	69	30	1
5/15/45	Richland	71	28	1
5/15/45	222-T	69	30	1
3/11/46	200-East	52	46	2
3/19/46	200-East	65	34	1
11/12/46	200-East	65	34	1
8/5/47	200-East	71	28	1

These values are presented as a supplement to the data from the stack samples. The composition should be the same for these as for the stack samples since the material is undoubtedly collected shortly after discharge from the stack.

C. Vegetation - The decay curves measured on vegetation are, in general, more inaccurate and of lower reproducibility because of the smaller counting rate and the more uncertain factors of deposition on various parts of the plant. Several curves from samples taken in 1946 and 1947 are given in Figures 8, 9, and 10, for comparison with the curves measured for stack samples. A tabulation of results from some of these curves is given in Table VII.

TABLE VII

DECAY MEASUREMENTS ON VEGETATION

Sampling Date	Sampling Location	$\mu\text{C/kg}$ I-131	$\mu\text{C/kg}$ Others	Composition		
				55 Day	275 Day	Long
				% Activity	% Activity	% Activity
11/28/45	R3M2	13	1.9	62%	37%	~ 1%
11/4/46	4S M4	0.6	0.1	49%	49%	~ 2%
11/25/46	4S M7	14	0.7	41%	56%	~ 3%
4/29/47	Meteorology	0.6	0.3	71%	28%	~ 1%
6/20/47	200-East	2.6	2.2	49%	49%	~ 2%
7/22/47	200-West	1.1	0.7	41%	56%	~ 3%
7/14/47	200-West	3.3	5	49%	49%	~ 2%

The values listed for $\mu\text{C/kg}$ have been corrected by a factor of three for self-absorption. The use of this factor for beta particles from the other fission products may cause the result as listed to be high.

A second series of values are available from the period of February - May, 1948. These samples were analyzed for long-lived activities by digestion of the vegetation and counting of the residue. The results of these tests are presented in Figure 11. Each individual value on the map represents one determination of the long-lived activity.

A single complete fission product analysis has been run on a sample of sage from the 200-West Gate. This sample indicated 49% cerium, 19% yttrium, 2% rare earth group, 8% ruthenium, 8% strontium, 3% cesium, and 7% zirconium.

IV. DISCUSSION:

The fission product analyses on filter papers through which stack gases were pulled indicate that the main activities discharged are cerium, yttrium, and strontium, with only 3 - 8 percent ruthenium. The decay curve data indicates that the activity is 60 - 80 percent of approximately 50 day half-life and 20 - 50 percent approximately 300 day half-life. Only 1 - 2% shows negligible decay over the period of measurement.

An estimate of the amounts of activities other than I^{131} and Xe^{133} discharged from the stack may be made from the data in Tables II and IV. Although the sampling data is not available for the first two samples in Table VIII, the best estimate available as to conditions are as given in the following table. The conditions should be within at least a factor of ten of the true values.

TABLE VIII

AMOUNT OF ACTIVITY DISCHARGED FROM STACK

Date	Time Sample Hrs.	Estimated Flow Rate CFM	μo Long on Filter	Estimated Air Up-Stack CPM	mo/da Discharged
5/11/48	2	1	2.4×10^{-2}	60,000	17
5/11/48	2	1	1.0×10^{-2}	60,000	7
4/8/47	24	1	9.5×10^{-4}	30,000	0.029
4/9/47	24	1	2.3×10^{-3}	30,000	0.069
4/9/47	6	1	1.6×10^{-3}	30,000	0.230
4/18/47	168	1	1.6×10^{-2}	30,000	0.069
4/18/47	24	1	1.9×10^{-3}	30,000	0.028
3/8/48	24	6.6	18.1	30,000	82
3/10/48	24	6.6	37.2	30,000	170
3/11/48	24	7.8	21.1	30,000	81
3/30/48	23.8	8	30.7	30,000	116
3/31/48	24.7	10	60.5	30,000	176
4/1/48	22.4	10	32.1	30,000	103

All of these values were obtained from the sampling line leading from the 50' level in the stack to the 292 Building.

A further estimate of the activity at several other points in the stack system may be obtained from Table IV. These values are given in Table IX.

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TABLE IX
ACTIVITY FROM VENTILATION SYSTEM

Date	Sampling Point	Time Sample	Sampling Rate	μ o Long-Lived Activity	Mo/da
		Hrs.	CFM		
3/30/48	Tunnel	23.3	10	13.4	43
3/31/48	Tunnel	24.9	11	31.6	83
3/30/48	Tunnel	23.3	10	46.2	143
4/1/48	Tunnel	19.5	9.8	36.1	133
3/30/48	Breach	23.1	99.8	24.5	78
3/31/48	Breach	24.8	10	22.4	65

From these data, an average of 80 - 150 millicuries of fission products are at present being discharged to the atmosphere each day. The meager data available from 1945 and 1948 indicates that the amount of activity discharged may have been considerably less at this time.

An estimate of the total alpha activity disposed of by the stack may be obtained from Table V. These samples were taken with a flow through the filter of 1.8 CFM and an air flow up the stack of 30,000 CFM. If all of the alpha activity on the paper is assumed to be plutonium, the amount discharged is given in Table X.

TABLE X
PLUTONIUM IN STACK GASES IN 1946

T PLANT		B PLANT	
Date	Estimated μ g/day	Date	Estimated μ g/day
8/1/48	980	3/27/48	8150
8/2/48	50	4/2/48	570
8/5/48	1290	4/4/48	820
8/6/48	1090	4/25/48	150
8/7/48	700	4/26/48	410
8/9/48	134	5/7/48	300
8/10/48	530	5/8/48	7
8/11/48	2700	5/9/48	710
8/11/48	106	4/10/48	630
8/12/48	90	5/11/48	730
8/12/48	240	5/12/48	740
8/13/48	1060	5/14/48	350
8/22/48	625	5/29/48	600

The extrapolation from short periods to 24 hours is seen to introduce considerable error. Thus, the seven hour run on 8/11/48 gives a value of 2700 μ g/day and on 8/12/48 it gives a value of 90 μ g/day. The true values for these days are 150

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$\mu\text{g/day}$ and $200 \mu\text{g/day}$. The variation is rather wide but most of the longer runs appear to give from $0.3 \mu\text{g/day}$ to $1.2 \mu\text{g/day}$ with occasional very low values.

The total amount of activity in existence at any time is dependent upon the rate at which the activity is released to the atmosphere and on the decay rate. If the average rate of evolution for the past three years has been ten millicuries per day from each stack, then the total distributed over the surrounding countryside is about 3.5 curies with about 1.1 curies of 50 - 60 day half-life, 2.2 curies of ~ 300 day half-life and 0.2 curies of very long half-life. If the later values on evolution are accepted for the average rate, then these figures may be multiplied by a factor of 10.

The various analytical results all confirm cerium, yttrium, and strontium as the major fission products from the stack. From the decay curve data on air filters and stack samples, the activity in the air averages about 70% of the 50 - 60 day, about 29% of the 300 day, and about 1% of the long half-life material. If all of this activity were deposited uniformly on the vegetation, then the composition of the activity present would be as given in Table XI.

TABLE XI
COMPOSITION OF VEGETATION ACTIVITY AS CALCULATED
FROM THE STACK DATA

Time Since Startup	% 55 Day	% 275 Day	% Long
1 yr.	42%	55%	3
2 yr.	35%	61%	4
3 yr.	32%	62%	6
5 yr.	30%	62%	8

These values compare favorably with the one complete analysis for fission products in vegetation if the 55 day group is said to include the Y, Re, Cs, and Zr, which total 30%, and the 275 day group includes the cerium and ruthenium which total 57%. The 8% strontium is then regarded as entirely the 25 year isotope. Actually, some of the ruthenium, cerium, and strontium are in the 55 day group, but the majority of the activity after this time should be long-lived.

The 55 day and 275 day components in the vegetation have nearly reached radioactive equilibrium at the moment. If the rate of discharge of these materials from the stack does not change, the activity in vegetation should remain about the same as is given in Figure 11 until the buildup of the very long-lived activity becomes significant. From the data presented in Table IX, it would seem that an increase of 1 - 2% per year in the long-lived component may be expected for the next few years.

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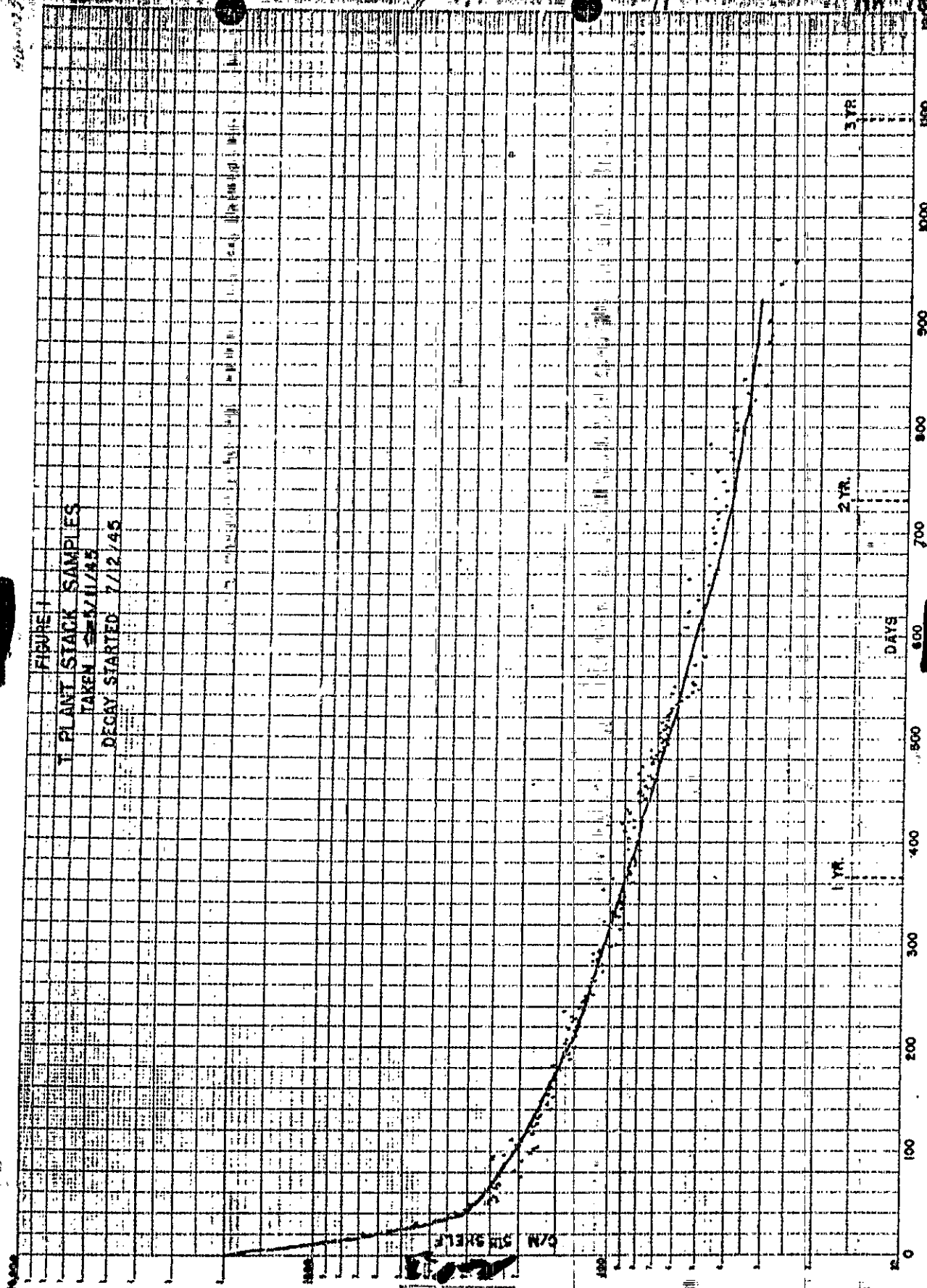
REFERENCES:

1. 7-3217 "Tolerable Concentrations of Radio-Iodine on Edible Plants"
H. M. Parker
2. 8-3455 "¹³¹I Accumulation in the Thyroid of Sheep Grazing Near HEW"
K. E. Herde
3. 7-5520 "Disposal of Separation Plant Off-Gases"
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4. 3-5402 "Trend of Contamination Observed in the Air, The Columbia River, and
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L. D. Turner
5. CL-697 "Project Handbook, Chapter III"

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FIGURE 1
T PLANT STACK SAMPLES
TAKEN 5-11-45
DECAY STARTED 7/12/45



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TAXES: 2.000000

DECAY STAGE 1 7/12/81

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2-yr.

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1 DAY

3

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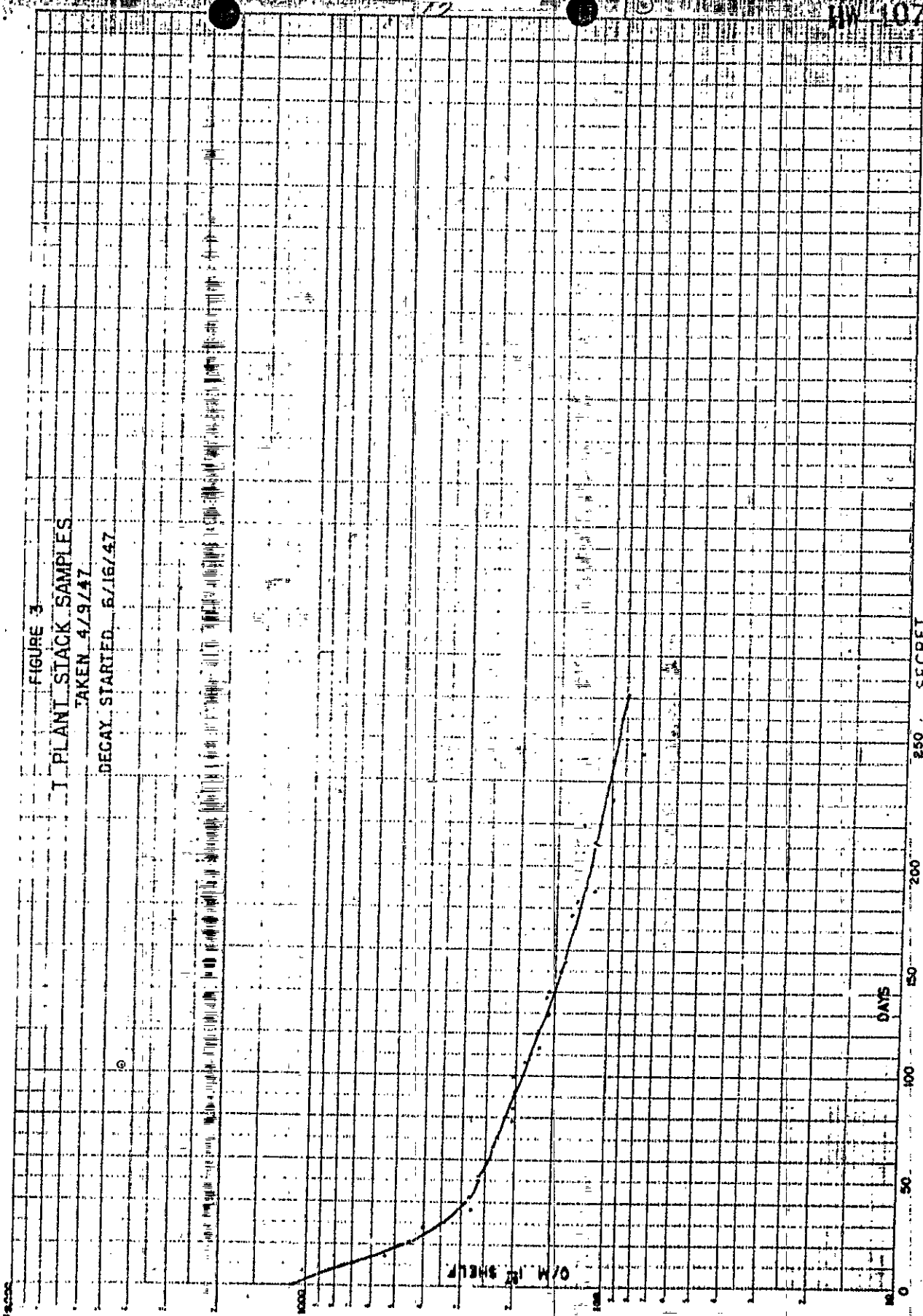
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FIGURE 3
PLANT STACK SAMPLES
TAKEN 4/9/47
DECAY STARTED 6/16/47



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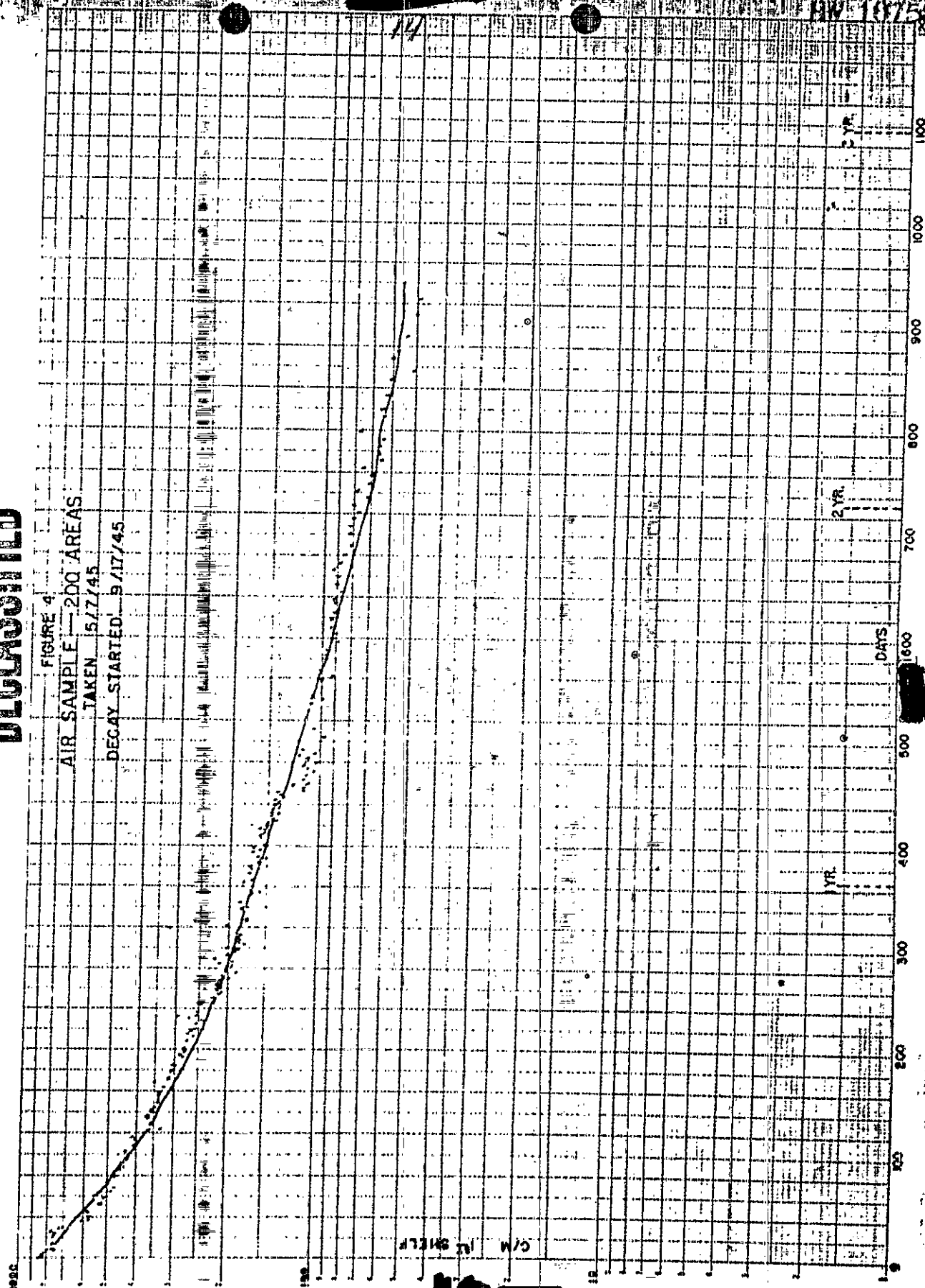
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AIR SAMPLE 1-200 AREAS

TAKEN 517145

DECAY STARTED! 9/17/45



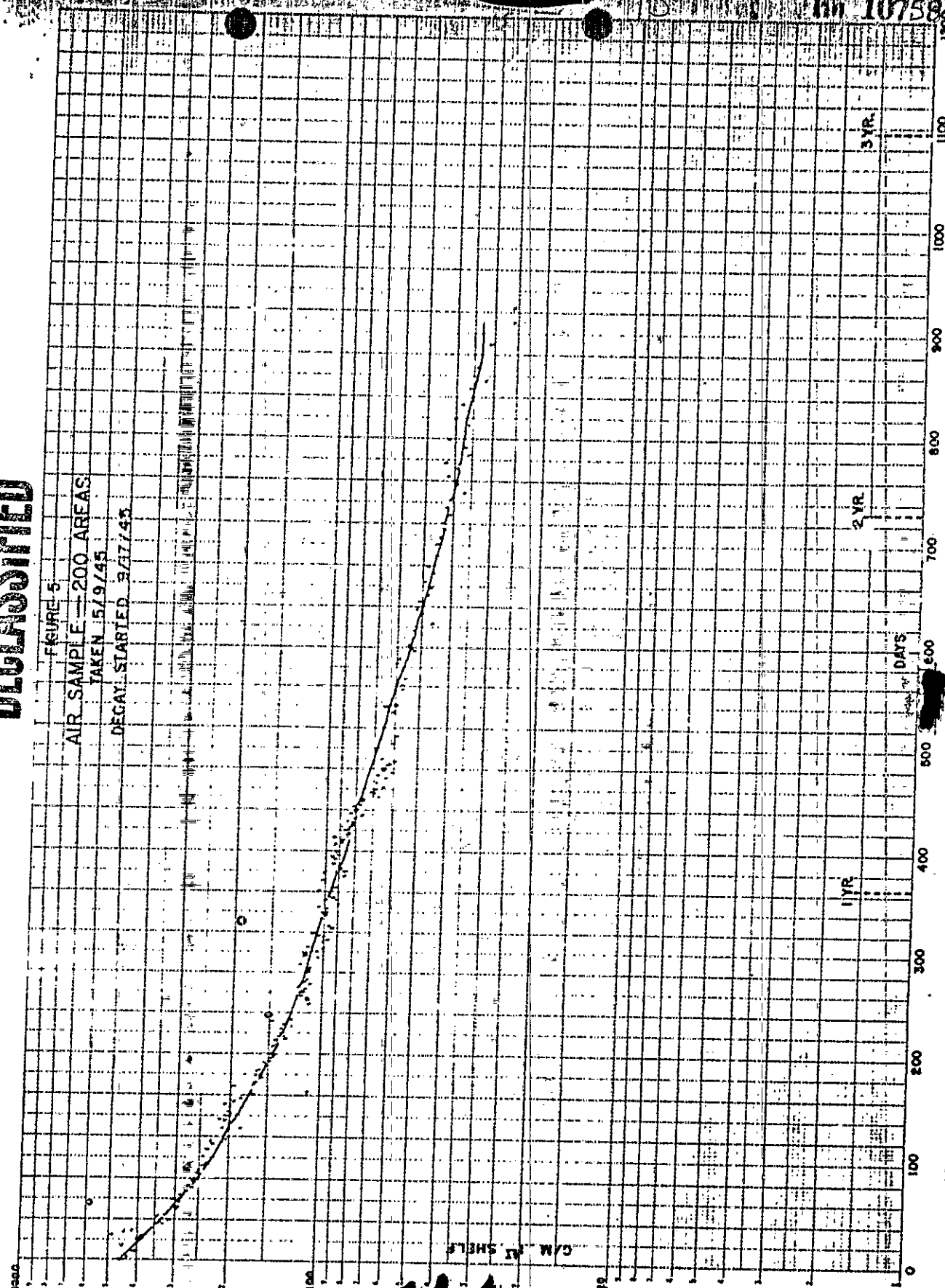
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AIR SAMPLE - 200 AREAS

TAKEN: 5/9/45

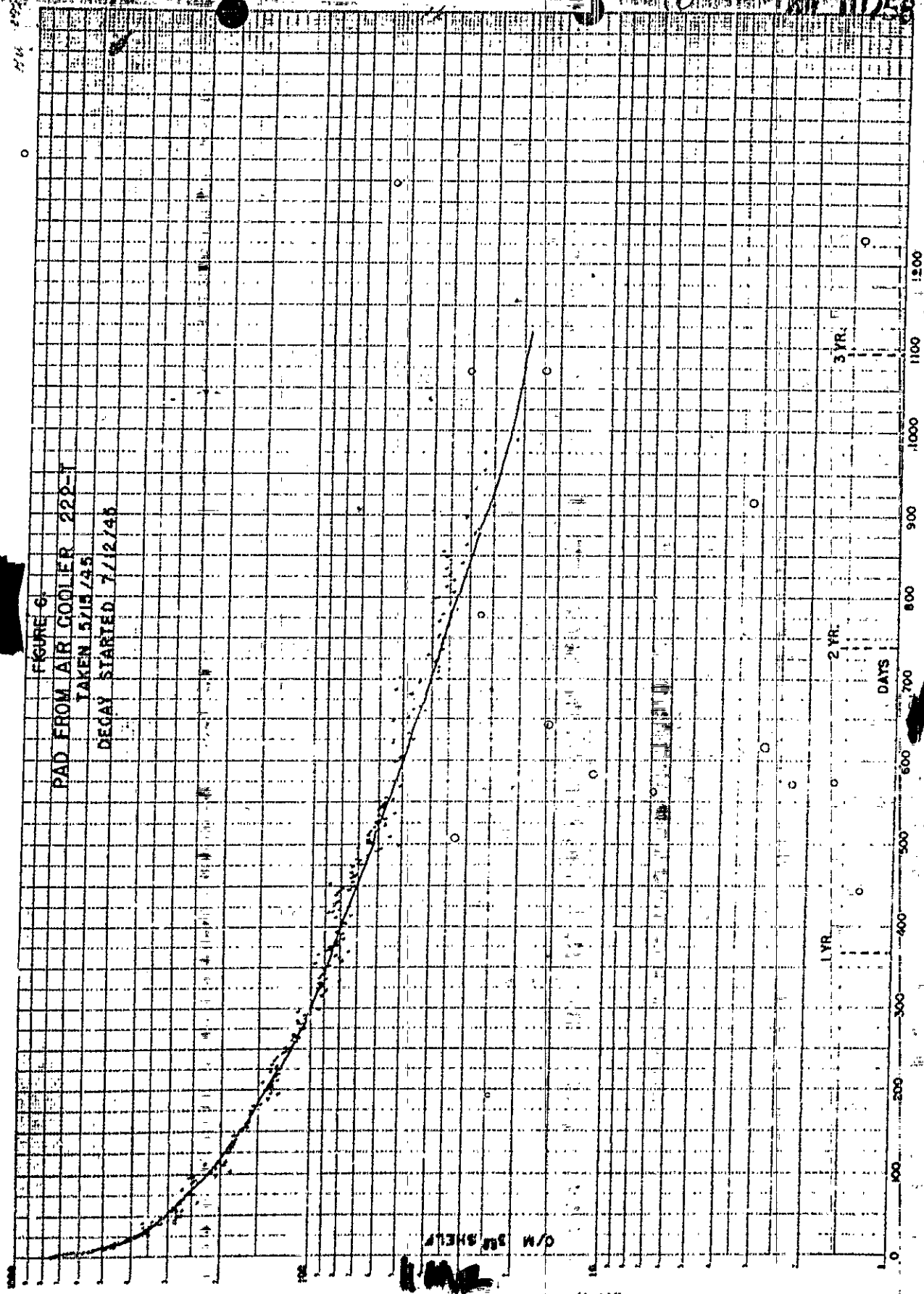
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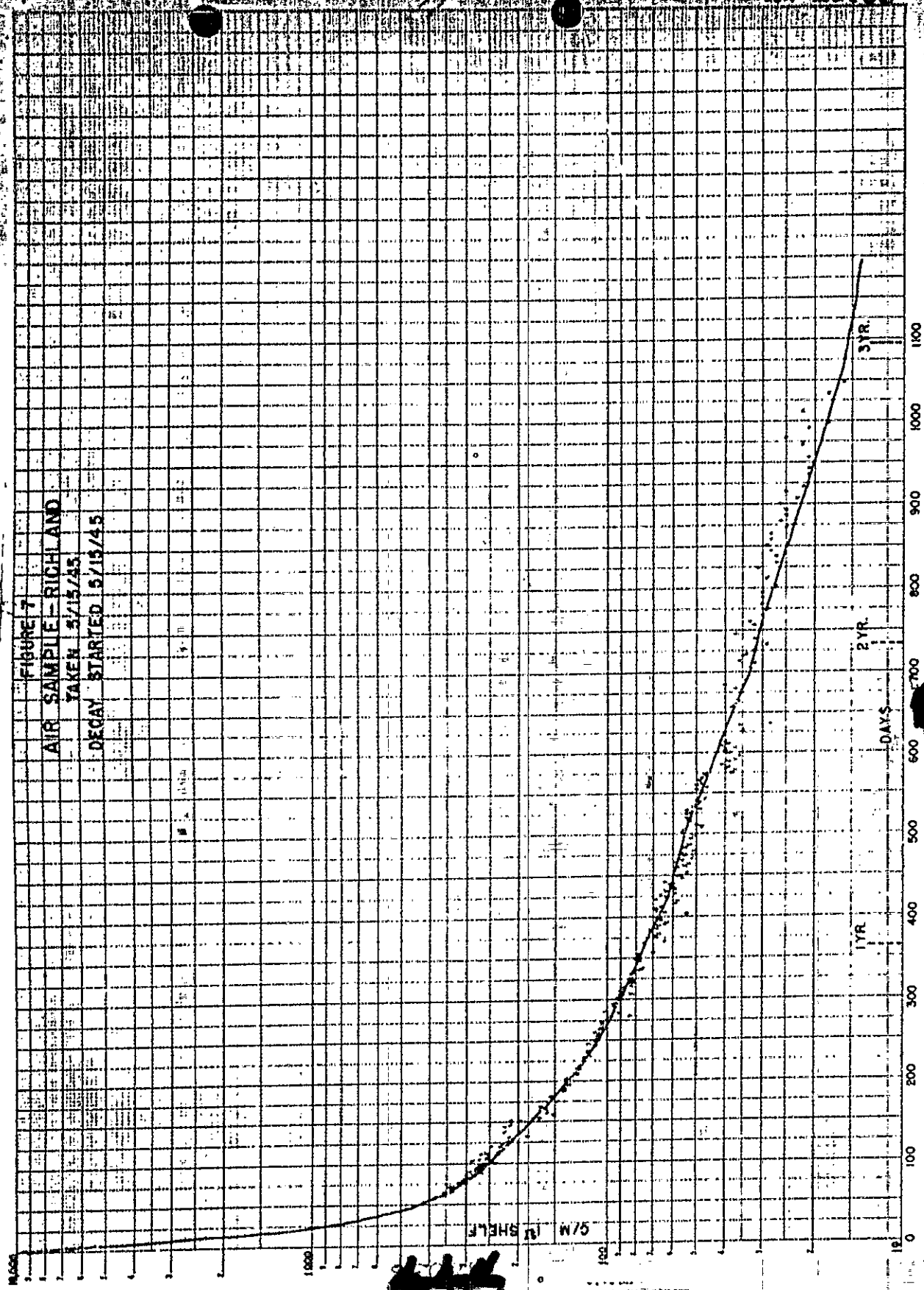
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FIGURE 6
PAD FROM AIR COOLER 222-T
TAKEN 5/15/45
DECAY STARTED 7/12/45



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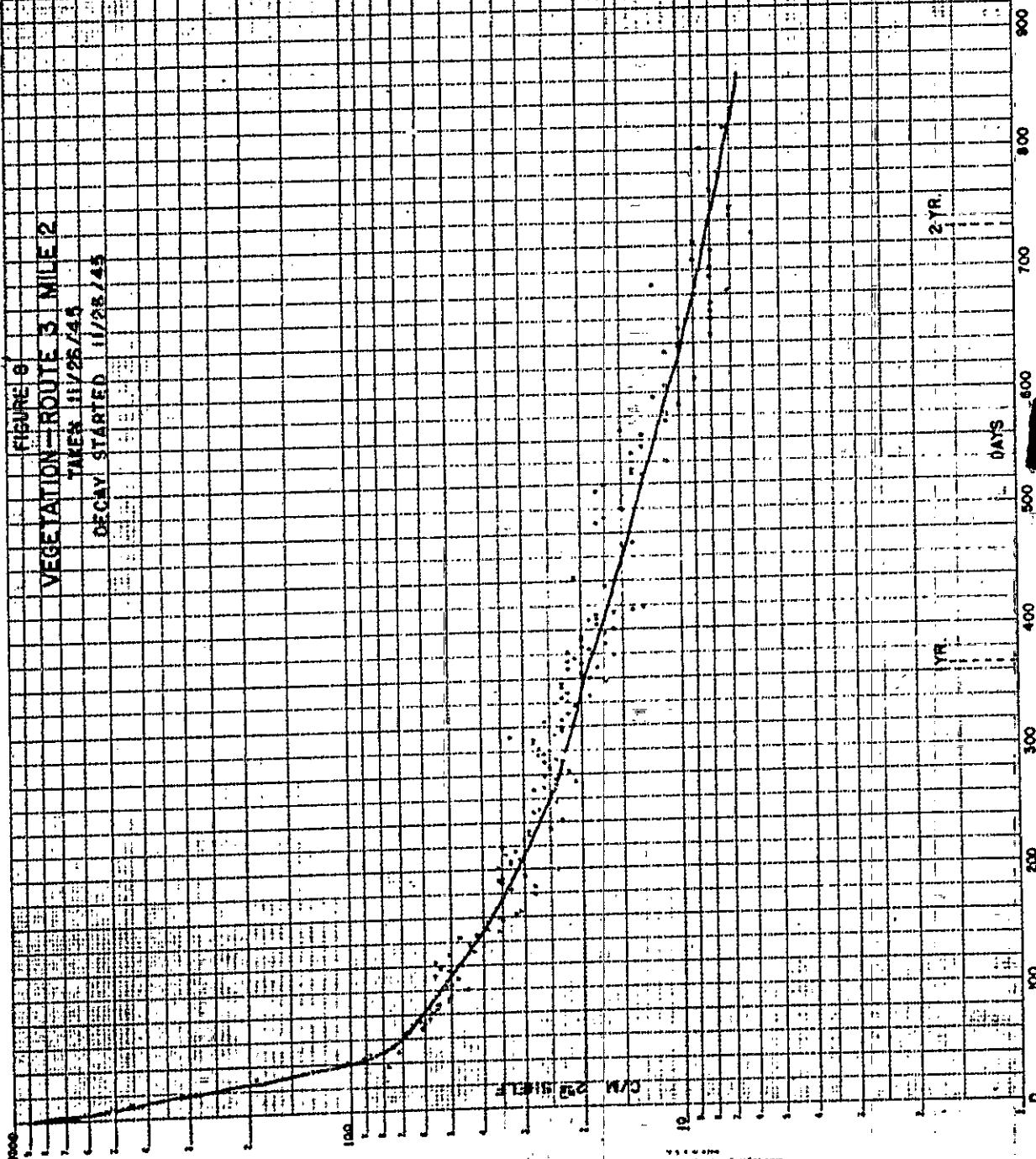
FIGURE 7
AIR SAMPLE - RICHLAND
TAKEN 5/15/45
DECAY STARTED 5/15/45



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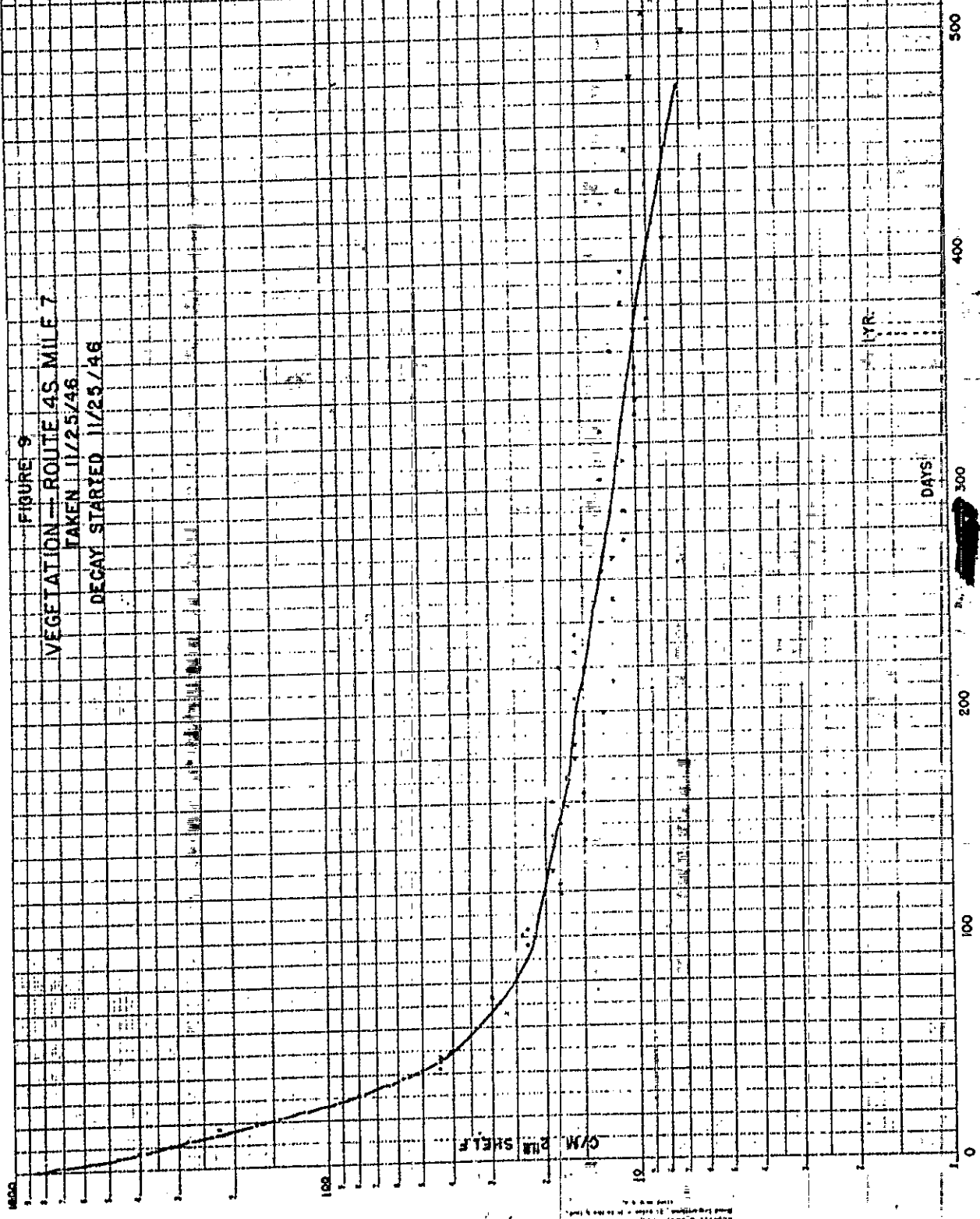
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FIGURE 9
VEGETATION—ROUTE 3, MILE 12
TAKER 11/26/45
DECAY STARTED 11/28/45



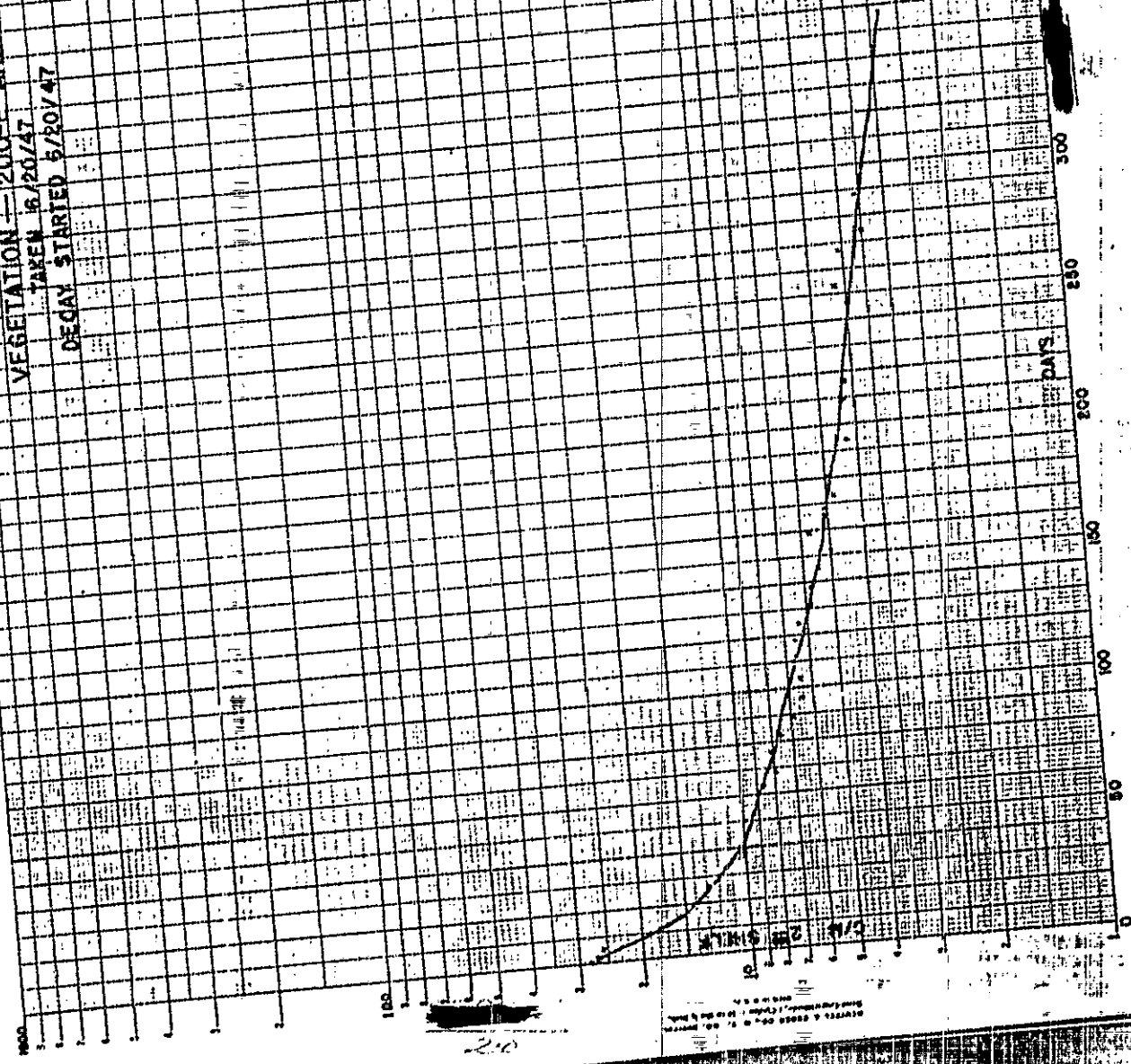
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FIGURE 9
VEGETATION - ROUTE 4S, MILE 7
TAKEN 11/25/46
DECAY STARTED 11/25/46



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FIGURE 10
VEGETATION - 200-F AREA
TAKEN 16/20/47
DECAY STARTED 5/20/47



Source: A 6000-05-01, 02, 03, 04, 05, 06, 07, 08, 09, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99, 100, 101, 102, 103, 104, 105, 106, 107, 108, 109, 110, 111, 112, 113, 114, 115, 116, 117, 118, 119, 120, 121, 122, 123, 124, 125, 126, 127, 128, 129, 130, 131, 132, 133, 134, 135, 136, 137, 138, 139, 140, 141, 142, 143, 144, 145, 146, 147, 148, 149, 150, 151, 152, 153, 154, 155, 156, 157, 158, 159, 160, 161, 162, 163, 164, 165, 166, 167, 168, 169, 170, 171, 172, 173, 174, 175, 176, 177, 178, 179, 180, 181, 182, 183, 184, 185, 186, 187, 188, 189, 190, 191, 192, 193, 194, 195, 196, 197, 198, 199, 200, 201, 202, 203, 204, 205, 206, 207, 208, 209, 210, 211, 212, 213, 214, 215, 216, 217, 218, 219, 220, 221, 222, 223, 224, 225, 226, 227, 228, 229, 230, 231, 232, 233, 234, 235, 236, 237, 238, 239, 240, 241, 242, 243, 244, 245, 246, 247, 248, 249, 250, 251, 252, 253, 254, 255, 256, 257, 258, 259, 260, 261, 262, 263, 264, 265, 266, 267, 268, 269, 270, 271, 272, 273, 274, 275, 276, 277, 278, 279, 280, 281, 282, 283, 284, 285, 286, 287, 288, 289, 290, 291, 292, 293, 294, 295, 296, 297, 298, 299, 300, 301, 302, 303, 304, 305, 306, 307, 308, 309, 310, 311, 312, 313, 314, 315, 316, 317, 318, 319, 320, 321, 322, 323, 324, 325, 326, 327, 328, 329, 330, 331, 332, 333, 334, 335, 336, 337, 338, 339, 340, 341, 342, 343, 344, 345, 346, 347, 348, 349, 350, 351, 352, 353, 354, 355, 356, 357, 358, 359, 360, 361, 362, 363, 364, 365, 366, 367, 368, 369, 370, 371, 372, 373, 374, 375, 376, 377, 378, 379, 380, 381, 382, 383, 384, 385, 386, 387, 388, 389, 390, 391, 392, 393, 394, 395, 396, 397, 398, 399, 400, 401, 402, 403, 404, 405, 406, 407, 408, 409, 410, 411, 412, 413, 414, 415, 416, 417, 418, 419, 420, 421, 422, 423, 424, 425, 426, 427, 428, 429, 430, 431, 432, 433, 434, 435, 436, 437, 438, 439, 440, 441, 442, 443, 444, 445, 446, 447, 448, 449, 450, 451, 452, 453, 454, 455, 456, 457, 458, 459, 460, 461, 462, 463, 464, 465, 466, 467, 468, 469, 470, 471, 472, 473, 474, 475, 476, 477, 478, 479, 480, 481, 482, 483, 484, 485, 486, 487, 488, 489, 490, 491, 492, 493, 494, 495, 496, 497, 498, 499, 500, 501, 502, 503, 504, 505, 506, 507, 508, 509, 510, 511, 512, 513, 514, 515, 516, 517, 518, 519, 520, 521, 522, 523, 524, 525, 526, 527, 528, 529, 530, 531, 532, 533, 534, 535, 536, 537, 538, 539, 540, 541, 542, 543, 544, 545, 546, 547, 548, 549, 550, 551, 552, 553, 554, 555, 556, 557, 558, 559, 560, 561, 562, 563, 564, 565, 566, 567, 568, 569, 570, 571, 572, 573, 574, 575, 576, 577, 578, 579, 580, 581, 582, 583, 584, 585, 586, 587, 588, 589, 590, 591, 592, 593, 594, 595, 596, 597, 598, 599, 600, 601, 602, 603, 604, 605, 606, 607, 608, 609, 610, 611, 612, 613, 614, 615, 616, 617, 618, 619, 620, 621, 622, 623, 624, 625, 626, 627, 628, 629, 630, 631, 632, 633, 634, 635, 636, 637, 638, 639, 640, 641, 642, 643, 644, 645, 646, 647, 648, 649, 650, 651, 652, 653, 654, 655, 656, 657, 658, 659, 660, 661, 662, 663, 664, 665, 666, 667, 668, 669, 670, 671, 672, 673, 674, 675, 676, 677, 678, 679, 680, 681, 682, 683, 684, 685, 686, 687, 688, 689, 690, 691, 692, 693, 694, 695, 696, 697, 698, 699, 700, 701, 702, 703, 704, 705, 706, 707, 708, 709, 710, 711, 712, 713, 714, 715, 716, 717, 718, 719, 720, 721, 722, 723, 724, 725, 726, 727, 728, 729, 730, 731, 732, 733, 734, 735, 736, 737, 738, 739, 740, 741, 742, 743, 744, 745, 746, 747, 748, 749, 750, 751, 752, 753, 754, 755, 756, 757, 758, 759, 760, 761, 762, 763, 764, 765, 766, 767, 768, 769, 770, 771, 772, 773, 774, 775, 776, 777, 778, 779, 780, 781, 782, 783, 784, 785, 786, 787, 788, 789, 790, 791, 792, 793, 794, 795, 796, 797, 798, 799, 800, 801, 802, 803, 804, 805, 806, 807, 808, 809, 810, 811, 812, 813, 814, 815, 816, 817, 818, 819, 820, 821, 822, 823, 824, 825, 826, 827, 828, 829, 830, 831, 832, 833, 834, 835, 836, 837, 838, 839, 840, 841, 842, 843, 844, 845, 846, 847, 848, 849, 850, 851, 852, 853, 854, 855, 856, 857, 858, 859, 860, 861, 862, 863, 864, 865, 866, 867, 868, 869, 870, 871, 872, 873, 874, 875, 876, 877, 878, 879, 880, 881, 882, 883, 884, 885, 886, 887, 888, 889, 890, 891, 892, 893, 894, 895, 896, 897, 898, 899, 900, 901, 902, 903, 904, 905, 906, 907, 908, 909, 910, 911, 912, 913, 914, 915, 916, 917, 918, 919, 920, 921, 922, 923, 924, 925, 926, 927, 928, 929, 930, 931, 932, 933, 934, 935, 936, 937, 938, 939, 940, 941, 942, 943, 944, 945, 946, 947, 948, 949, 950, 951, 952, 953, 954, 955, 956, 957, 958, 959, 960, 961, 962, 963, 964, 965, 966, 967, 968, 969, 970, 971, 972, 973, 974, 975, 976, 977, 978, 979, 980, 981, 982, 983, 984, 985, 986, 987, 988, 989, 990, 991, 992, 993, 994, 995, 996, 997, 998, 999, 1000

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