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HEADQUARTERS USAF 410269

AFOAT-1

Preliminary Summary

TRITIUM CONCENTRATIONS IN THE ATMOSPHERE

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1. INTRODUCTION

1.1 Tritium activities measured in samples of atmospheric water are related to the equivalent weight of atmospheric air from which the water was extracted. The concentration of tritium per gram of air is calculated from a number of collections of whole air made from various levels within the atmosphere to altitudes of 90,000 feet mean sea level. Air collections were made by aircraft and by balloon-borne apparatus under the sponsorship and direction of AFOAT-1. Collection of these samples, analytical procedures employed, methods of calculation and the errors associated with the final values, are discussed briefly in the ensuing paragraphs.

2. COLLECTION, SAMPLING AND ANALYSIS

2.1 Collection

For sampling at altitudes of 50,000 feet and higher, the collection equipment is carried aloft on a constant altitude lift balloon. The collection equipment consists of a centrifugal collection blower, a deuterium spike dispenser capable of adding an accurately known weight of D_2O to the contents of the collection bag at the time of sampling, and a 30,000 cubic foot polyethylene collection bag.

The D_2O is kept at a constant temperature of 95°F and about 2 to 5 grams of vapor dispensed uniformly with time at a point just behind the centrifugal blower during collection of the air sample. The dispenser is weighed before and after each collection to determine the number of atoms of deuterium added as the tracer. Both bag and dispenser are effectively

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sealed at all other times. Sampling at the lower levels of ground to 40,000 feet was by means of air samples compressed into steel bottles and carried in aircraft.

2.2 Sampling

Sampling by balloon takes place in two stages. In order to effect a high percentage of recovered collections on impact with the ground, it is necessary to transfer the sample to a much stronger and smaller bag. This is done on the descent between altitudes of 30,000 and 3000 feet. It will be appreciated that losses of both water vapor condensed on the walls of the polyethylene bag and air not completely transferred can occur at this stage.

The second sampling occurs when the air is pumped out of the impact bag for use on other programs. A sample of the water vapor is frozen out of the air prior to entering the gas compressors.

2.3 Analysis

The recovered sample of water is split into two fractions. The concentration of deuterium is measured in one fraction; the concentration of deuterium in the original tracer water had been previously measured. Hydrogen gas is obtained from the other fraction by reduction over magnesium and the tritium counted in this hydrogen sample, using a geiger counter in a "cave" with anticoincidence circuitry. In general, the counting is no problem because the majority of the samples have a counting rate an order of magnitude, at least, greater than background.

3. COMPUTATION OF TRITIUM CONCENTRATION

3.1 Tritium

Provided no unknown amount of tritium, deuterium or hydrogen is

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added to one or the other of the two fractions after separation of the sample in the laboratory, the total tritium collected will be given by:

$$T \text{ (atoms)} = A_s \left(\frac{C_{d_s}}{C_{d_r}} \times C_{t_r} - C_{t_s} \right)$$

where A_s = total atoms of hydrogen (all isotopes) added as tracer (spike).

C_{d_s} = concentration of deuterium in the tracer (atoms per atom of hydrogen, all isotopes).

C_{d_r} = atom concentration of deuterium in the recovered sample.

C_{t_r} = atom concentration of tritium in the recovered sample.

C_{t_s} = concentration of tritium in the tracer.

3.2 Amount of Air Sampled

The volume of air collected is estimated by measuring the total number of revolutions of the blower calibrated under simulated conditions and by photographing the inflated collection bag to estimate its dimensions.

The mass of air sampled requires a knowledge of the precise altitude of the collection in order to assign the density.

3.3 T/H Values

It is obvious that the concentration of tritium per gram of air may be ascertained even though water is added or subtracted indiscriminately prior to the laboratory separation into the two fractions; this of course, provided any added water contains no appreciable tritium or deuterium (any tropospheric water would produce no appreciable error).

On the other hand, only subtraction of water from the original collection is permissible if T/H ratios are desired. With the enormous areas of polyethylene involved and the possibilities of exchange of the

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collected water vapor with water on this large surface either adsorbed, or as liquid not removed in the original packaging, and with a relatively high permeability of the polyethylene bag to water vapor, it cannot be maintained that any T/H value measured in the final samples is representative of the air at the collection altitude. It appears that such ratios will best be determined by an independent measure of the water vapor content of the air at high altitudes coupled with the tritium content as measured here.

4. ERRORS

4.1 Tracer

The amount of tracer added to the collection of air is estimated to be accurate to about 1 per cent.

4.2 Deuterium and Tritium

Deuterium concentrations are measurable to ± 4 per cent. Errors in the measurement of tritium amount to about 10 per cent, 5 per cent attributable to statistical counting errors and 5 per cent to calibration errors.

Careful control in the source of the tracer deuterium oxide results in a tracer containing less than 250 dpm of tritium per gram. This is normally only about 5 per cent of the total tritium in a sample and therefore uncertainty arising from tritium in the tracer is negligibly small.

4.3 Mass of Air Collected

The method of determining the volume of air collected is estimated to have a standard error of about ± 5 per cent. However, the density

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must be known to convert the volume to mass of air and due to uncertainties in the absolute altitude of the collection, the mass of air collected is not known to better than ± 15 per cent below 70,000 feet and ± 25 per cent above 70,000 feet.

Thus the error in determining the mass of air containing the tritium is the predominant error.

Combining all the sources of error, it is estimated that a single determination of the tritium concentration should be determinable to ± 30 per cent with a somewhat greater accuracy at the lower levels of the stratosphere.

5. RESULTS

5.1 Early Results

A few early results were obtained in the period between June 1955 and August 1956; all samples being collected in the neighborhood of Minneapolis, Minnesota.

These early results when added to excess carbon-14 and related to fissions through krypton-85 measurements in the same samples, give values for excess neutrons per fission in good agreement with the weighted average number of excess neutrons calculated from the known performance of the Castle devices. We may, therefore, have some confidence in these early data even though the tracing was performed by adding deuterium oxide to the collection bag before launching the balloon.

These early results are recorded here in Table 5.1.1 to show that the more recent results of 1957-1958 are not essentially lower or higher than the concentrations in the stratosphere a year to two years after

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Operation Castle. They should not, however, be used to draw any detailed conclusions as to changes in the tritium concentration in the intervening period.

5.2 Recent Results for the Stratosphere

Tritium concentrations from 50,000 to 90,000 feet in 1957-1958 as measured by the method herein outlined are given in Table 5.2.1.

5.3 Recent Results at Lower Levels

Best values for the tritium concentration of the troposphere and lower stratosphere are from samples collected in January 1958 between 70°N and 74°N. These results are summarized in Table 5.3.1.

Rather large fluctuations in the tritium content up to 20,000 feet are to be expected because of the effect of precipitation. At 40,000 feet, which was 10,000 to 13,000 feet above the tropospause, the tritium content was very similar to that found over Minneapolis, Minnesota, at 50,000 feet, also at about the same height above the tropopause.

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TABLE 5.1.1

Early Values of Tritium Concentrations in the Stratosphere Following Operation Castle

Sample	Date	Altitude (kft)	T-Atoms per Gram Air x 10^{-8}
BA 91	30 Nov 55	60	7.8
BA 103	30 Jan 56		6.2
BA 89	26 Oct 55	70	4.3
BA 102	19 Jan 56		7.2
BA 71	15 Jun 55	80	8.2
BA 78	11 Aug 55		14.0
BA 82	23 Sep 55		11.5
BA 85	10 Oct 55		8.9
BA 87	18 Oct 55		3.8
BA 111	26 Mar 56*	95	8.6
BA 135	30 Aug 56*		10.7

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* After commencement of Operation Redwing but results are probably uninfluenced by Redwing debris at Minneapolis, Minnesota, as inferred from krypton-85 and carbon-14 data.

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TABLE 5.2.1

Tritium Concentrations in 1957-1958 in the Stratosphere Above Minneapolis, Minnesota

Sample	Date	Altitude (kilofeet)	Sample* Activity (dis./min./ml.)	Total Grams of Air x 10^{-4}	T-Atoms per Gram Air x 10^{-8}
BA-163	28 May 57	50	168 ± 6	8.97	1.1
BA-167	27 Jun 57	49.6	392 ± 4	8.28	2.5
BA-168	11 Jul 57	50	54.0 ± 1.3	7.94	0.9
BA-205	6 Mar 58	50	817 ± 29	6.21	2.7
BA-211	9 Apr 58	47.4	319 ± 5	7.45	4.2
BA-175	16 Aug 57	71.0	1049 ± 19	9.31	11.8
BA-177	24 Aug 57	65.0	1118 ± 68	5.35	10.7
BA-186	26 Oct 57	64.8	2826 ± 24	5.04	11.1
BA-192	30 Nov 57	64.0	5213 ± 218	5.36	8.9
BA-198	10 Jan 58	64.2	5327 ± 154	5.24	8.9
BA-204	20 Feb 58	64.2	4615 ± 170	4.92	6.9
BA-216	7 Jun 58	61.7	654 ± 10	4.45	5.8
BA-220	20 Jun 58	60.5	285 ± 5	4.16	8.5
BA-166	20 Jun 57	76	313 ± 5	6.73	7.1
BA-170	15 Jul 57	76.5	953 ± 14	6.21	10.7
BA-176	22 Aug 57	82	486 ± 7	6.56	5.9
BA-199	22 Jan 58	77.9	3144 ± 67	5.73	6.3
BA-202	7 Feb 58	77.4	2986 ± 30	5.00	7.1
BA-207	25 Mar 58	80.0	2119 ± 35	3.54	7.3
BA-215	15 May 58	77.5	684 ± 11	4.86	6.7
BA-219	17 Jun 58	76.7	240 ± 4	2.64	15.2
BA-181	25 Sep 57	88.5	1066 ± 7	4.66	12.4
BA-188	5 Nov 57	91	1283 ± 23	3.73	7.3
BA-195	19 Dec 57	88.3	2271 ± 62	4.50	9.9

* Activity as determined from the reduced hydrogen fraction of 0.5 to 0.75 milliliters of the sample water.

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TABLE 5.3.1

Tritium Concentrations in the Troposphere and Lower Stratosphere in January 1958 (73°N Latitude)

Sampling Altitude (kilofeet)	Tropopause Height (kilofeet)	T-Atoms per Gram Air x 10^{-8}
10	27-31	0.09
		0.19
		0.61
		0.37
		Average = 0.32
20	27-31	0.12
		0.06
		0.51
		0.21
		Average = 0.23
30	31.0	0.46
	31.0	0.19
	28.1	0.46
	29.5	0.31
	29.5	0.42
	27.2	0.52
		Average = 0.39
40	26.6	1.24
	26.6	1.89
	31.1	1.82
	31.1	2.16
	31.1	1.88
	28.1	2.43
	28.1	3.55
	29.5	3.28
	29.3	2.67
	27.2	5.15
		Average = 2.61

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APPENDIX III

DATA SUPPLEMENT

TO

"DISTRIBUTION OF CARBON-14 ACTIVITY WITHIN THE STRATOSPHERE"

(FOR PERIOD FROM NOVEMBER 1957 THROUGH JUNE 1958)

16 December 1958

(Preliminary Draft)

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EXPLANATORY NOTE

The data inclosed in this report supplements data submitted previously in AFOAT-1 reports entitled "Distribution of Carbon-14 Activity within the Stratosphere," Appendix I, dated 01 April 1957, (Preliminary Draft), and Appendix II, "Data Supplement to Distribution of Carbon-14 Activity within the Stratosphere," dated 27 January 1958, (Preliminary Draft). The data in this report results from the continued collections of atmospheric whole air samples over the period from November 1957 through June 1958. This report, with the two previous reports listed above, provide the reader with continuous data over the interval from September 1954 through June 1958.

For purpose of clarity, the data in this report are divided into two tables: the data of Table I resulting from the analysis of balloon collections of stratospheric whole air from four separate sites, a collection procedure described briefly in the initial data report; the data of Table II resulting from aircraft collections of whole air made at a latitude of 74°N near Point Barrow, Alaska. For purposes of comparison, data from sampling at each location are reported in mean altitude groupings dependent upon the altitudes from which the collections were made.

The data includes the excess carbon-14 activity as observed in the CO_2 fraction of the whole air sample. The normal abundance of CO_2 in whole air is taken as 0.031 mole percent. Subtracted cosmic-ray background is 73×10^5 atoms carbon-14 per gram of air. In addition to the carbon-14 data, this report includes the associated gross (10.7 year) Kr-85 activity per gram of air as observed in the krypton fraction of the whole air collection. The abundance of krypton in air has been determined as 1.12×10^{-4} mole percent. Determination of gross Kr-85 activity in the krypton fraction of the sample is made by Argonne National Laboratory utilizing methods of gas counting quite similar to those utilized in the carbon-14 determinations. One standard deviation of the absolute error in determining the Kr-85 concentration is generally less than 5% of the measured activity.

Aircraft collections of whole air were made by utilizing a B-36 aircraft equipped with a 4 cubic feet per minute Kidde type compressor in conjunction with a high pressure bottling system. Four main altitudes of collections are reported to a maximum altitude of 40,000 feet mean sea level.

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Table I

Balloon Collections

a. From Minneapolis, Minnesota (45°N)

(1) Collections from 45,500-49,500 feet

Normal = 73×10^{5} C^{14}/g air

Sample No.	Altitude	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
BA-191	49.5	26 Nov 57	20.8 ± 6.2	23.5
BA-193	47.1	04 Dec	28.7 ± 5.9	24.9
BA-193T	47.1	04 Dec	44.3 ± 6.2	
BA-197	47.5	08 Jan 58	11.8 ± 5.2	23.6
BA-203	45.5	13 Feb	69.6 ± 5.9	25.0
BA-205	47.0	06 Mar	33.6 ± 4.5	25.0
BA-205T	47.0	06 Mar	42.2 ± 77.8	
BA-211	47.4	09 Apr	94.1 ± 6.2	26.9
BA-214	47.8	12 May	22.8 ± 6.6	30.4
BA-214T	47.8	12 May	56.4 ± 23.9	
BA-218	46.5	11 June	75.4 ± 8.0	27.9
BA-218T	46.5	11 June	73.0 ± 7.3	
(2) Collections from 60,500-64,600 feet				
BA-192	64.6	30 Nov 57	211.4 ± 5.9	23.8
BA-192T	64.6	30 Nov	213.5 ± 5.5	
BA-196	63.5	21 Dec	181.7 ± 6.9	25.0
BA-196T	63.5	21 Dec	176.1 ± 5.5	
BA-198	64.2	10 Jan 58	197.2 ± 5.2	23.6

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Sample No.	Altitude	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
BA-198T	64.2	10 Jan 58	178.9 ± 12.1	
BA-204	64.2	20 Feb	140.5 ± 14.2	20.8
BA-204T	64.2	20 Feb	177.9 ± 38.1	
BA-206	63.6	12 Mar	144.6 ± 5.5	24.0
BA-212	64.5	16 Apr	116.6 ± 5.9	24.9
BA-216	61.7	07 June	52.9 ± 6.6	26.8
BA-220	60.5	20 June	99.3 ± 6.9	28.5
BA-220T	60.5	20 June	155.7 ± 10.7	
(3) Collections from 76,700-81,100 feet				
BA-190	78.8	25 Nov 57	210.4 ± 6.9	21.0
BA-199	77.7	22 Jan 58	197.2 ± 6.6	21.5
BA-199T	77.7	22 Jan	207.6 ± 16.6	
BA-202	77.4	07 Feb	196.2 ± 9.7	21.2
BA-202T	77.4	07 Feb	213.5 ± 11.8	
BA-207	81.1	25 Mar	164.4 ± 6.9	20.3
BA-207T	81.1	25 Mar	162.6 ± 26.6	
BA-210	77.9	08 Apr	172.7 ± 5.9	21.2
BA-210T	77.9	08 Apr	190.0 ± 6.9	
BA-215	77.6	15 May	133.9 ± 6.9	21.2
BA-215T	77.6	15 May	143.9 ± 9.7	
BA-219	76.7	17 June	139.1 ± 5.5	21.0
BA-219T	76.7	17 June	194.1 ± 11.4	

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(4) Collections from 88,400-107,000 feet

Sample No.	Altitude	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-8 (atoms per gram (of air x 10^{-3}
BA-187	90.0	29 Oct 57	166.8 ± 5.5	18.1
BA-187T	90.0	29 Oct	206.9 ± 9.7	
BA-188	91.1	05 Nov	194.8 ± 5.2	19.1
BA-188T	91.1	05 Nov	232.9 ± 33.6	
BA-189	107.0	07 Nov	159.2 ± 6.9	19.3
BA-195	88.4	19 Dec	216.6 ± 6.9	20.4
BA-195T	88.4	19 Dec	212.1 ± 6.9	
BA-200	92.4	31 Jan 58	151.6 ± 6.9	19.4
BA-201	89.1	05 Feb	205.5 ± 5.2	19.9
BA-208	93.0	26 Mar	156.7 ± 6.6	18.5
BA-209	89.7	07 Apr	176.1 ± 7.3	19.9
BA-213	90.5	09 May	161.2 ± 8.7	19.1
BA-213T	90.5	09 May	178.5 ± 7.3	
BA-217	91.0	06 June	88.6 ± 5.5	24.9

b. From San Angelo, Texas (31°N)

(1) Collections from 47,500-52,400 feet

SA-122	47.5	14 Nov 57	36.0 ± 5.5	23.9
SA-126	49.0	04 Dec	0.7 ± 4.8	23.7
SA-131	50.3	09 Jan 58	13.5 ± 5.9	24.5
SA-133	50.3	01 Feb	2.4 ± 6.6	23.4
SA-140	50.6	10 Mar	34.6 ± 8.7	23.9
SA-144	49.0	08 Apr	10.0 ± 5.2	25.2
SA-145	48.0	03 May	40.1 ± 9.7	26.5

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Sample No.	Altitude	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
SA-148	49.3	10 May 58	72.3 ± 6.2	26.4
SA-153T	52.4	08 June	2.4 ± 8.7	26.4
(2) Collections from 62,000-67,000 feet				
SA-120	65.3	05 Oct 57	63.3 ± 4.8	20.8
SA-124	65.7	26 Nov	189.3 ± 5.5	24.1
SA-128	67.0	10 Dec	150.5 ± 5.5	25.9
SA-132	62.0	15 Jan 58	165.4 ± 6.6	25.5
SA-135	65.1	15 Feb	153.3 ± 5.2	23.3
SA-139	67.0	09 Mar	160.2 ± 5.5	24.1
SA-142	65.0	03 Apr	88.6 ± 5.5	23.9
SA-146	66.0	04 May	131.1 ± 4.2	23.9
SA-152	65.4	07 June	191.0 ± 6.2	26.9
SA-152T	65.4	07 June	152.9 ± 11.8	
(3) Collections from 79,800-83,700 feet				
SA-119	82.7	04 Oct 57	229.4 ± 5.5	23.0
SA-123	83.7	20 Nov	148.1 ± 6.2	20.1
SA-125	80.3	02 Dec	230.8 ± 6.2	Lost
SA-129	79.9	07 Jan 58	237.7 ± 6.2	22.5
SA-136	79.8	17 Feb	204.5 ± 5.9	20.6
SA-138	80.2	07 Mar	203.5 ± 6.9	20.7
SA-143	81.7	07 Apr	192.7 ± 8.0	20.6
SA-149	80.6	19 May	186.5 ± 8.7	20.7

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Sample No.	Altitude	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
SA-150	81.0	03 June 58	190.3 ± 6.2	19.6
SA-150T	81.0	03 June	166.4 ± 13.1	

(4) Collections from 89,200-92,300 feet

Sample No.	Altitude	Collection Date	Excess Carbon-14	Gross Krypton-85
SA-118	91.0	03 Oct 57	82.4 ± 8.0	21.6
SA-121	90.0	02 Nov	249.1 ± 6.6	Lost
SA-127	90.4	07 Dec	237.0 ± 4.8	23.3
SA-130	90.0	08 Jan 58	187.2 ± 6.6	22.7
SA-134	92.3	02 Feb	201.4 ± 5.9	21.6
SA-137	92.0	03 Mar	167.5 ± 6.6	20.5
SA-141	91.5	01 Apr	163.7 ± 9.0	19.2
SA-147	92.1	07 May	183.0 ± 5.5	19.5
SA-151	89.2	05 June	165.4 ± 5.2	19.6
SA-151T	89.2	05 June	175.1 ± 9.7	

c. From Canal Zone, Panama (9°N)

(1) Collections from 46,500-51,500 feet

Sample No.	Altitude	Collection Date	Excess Carbon-14	Gross Krypton-85
CZ-92	51.5	05 Nov 57	0.3 ± 5.5	23.0
CZ-94	46.5	01 Dec	0 ± 4.8	24.5
CZ-98	50.9	02 Jan 58	0 ± 5.5	25.6
CZ-102	51.1	02 Feb	0 ± 8.7	24.5
CZ-108	50.2	08 Mar	0 ± 5.2	25.7
CZ-111	51.6	03 Apr	0 ± 12.1	27.1
CZ-115	50.8	02 May	8.3 ± 5.5	27.0
CZ-118	51.3	01 June	0 ± 6.2	28.3

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(2) Collections from 64,900-67,400 feet

Sample No.	Altitude	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
CZ-93	66.8	06 Nov 57	61.2 ± 5.9	22.2
CZ-96	65.5	04 Dec	43.9 ± 6.9	Lost
CZ-101	65.3	15 Jan 58	151.6 ± 5.5	33.0
CZ-103	67.4	05 Feb	73.0 ± 5.9	26.5
CZ-106	66.5	03 Mar	0 ± 7.6	24.1
CZ-113	65.3	14 Apr	0 ± 6.2	23.8
CZ-114	64.9	01 May	55.4 ± 5.5	23.7
CZ-119	65.2	02 June	63.7 ± 5.5	26.2
(3) Collections from 77,000-84,600 feet				
CZ-89	80.2	30 Oct 57	91.3 ± 5.5	20.4
CZ-90	82.1	01 Nov	93.1 ± 6.6	20.9
CZ-97	77.0	15 Dec	83.4 ± 7.6	19.1
CZ-99	80.7	05 Jan 58	83.7 ± 6.6	19.3
CZ-104	81.6	10 Feb	98.6 ± 6.2	20.0
CZ-112	80.8	04 Apr	137.0 ± 8.0	21.8
CZ-116	84.6	04 May	112.8 ± 7.3	25.6
CZ-117	79.5	09 May	163.3 ± 6.2	34.4
(4) Collections from 88,700-94,000 feet				
CZ-86	88.7	08 Oct 57	197.6 ± 6.9	25.3
CZ-95	89.3	02 Dec 57	100.3 ± 5.9	23.1
CZ-100	91.5	13 Jan 58	133.6 ± 7.3	20.0
CZ-105	93.4	16 Feb 58	32.9 ± 6.6	19.0

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Sample No.	Altitude	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5}	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
CZ-107	92.2	04 Mar 58	96.5 ± 6.9	19.4
CZ-110	94.0	01 Apr	150.2 ± 15.9	19.5
d. From Sao Paulo, Brazil (23°S)				
(1) Collections from 49.600-52,300 feet				
BR-131	49.6	23 Nov 57	0 ± 7.6	22.9
BR-136	51.3	14 Dec	0 ± 5.5	23.4
BR-141	51.0	11 Jan 58	0 ± 9.0	23.4
BR-143	50.5	03 Feb	0 ± 8.7	22.8
BR-150	49.6	14 Mar	0 ± 5.5	23.3
BR-151	52.3	02 Apr	11.4 ± 6.2	23.7
BR-157	50.5	12 May	4.8 ± 5.9	24.9
(2) Collections from 64,100-68,500 feet				
BR-133	65.0	25 Nov 57	85.8 ± 5.9	16.7
BR-135	66.0	12 Dec	106.9 ± 8.7	16.6
BR-140	65.0	10 Jan 58	81.3 ± 6.9	17.8
BR-142	65.8	03 Feb	72.0 ± 6.9	18.2
BR-149	65.5	13 Mar	55.4 ± 7.6	19.0
BR-152	68.5	03 Apr	119.7 ± 7.6	16.8
BR-155	65.0	09 May	55.4 ± 11.4	21.3
BR-160	64.1	04 June	52.6 ± 6.2	20.4

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(3) Collections from 78,700-80,800 feet

Sample No.	Altitude	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
BR-130	78.7	22 Nov 57	137.7 ± 5.5	16.0
BR-132	79.9	24 Nov	111.1 ± 6.9	15.5
BR-133	80.8	10 Dec	99.3 ± 4.8	15.0
BR-138	79.0	07 Jan 58	86.5 ± 18.7	Lost
BR-144	79.8	09 Feb	148.4 ± 6.9	16.5
BR-146	80.0	05 Mar	142.2 ± 7.6	16.3
BR-148	80.0	07 Mar	159.5 ± 8.7	15.8
BR-153	80.5	04 Apr	111.8 ± 6.6	18.3
BR-159	80.1	17 May	143.6 ± 5.5	16.3
BR-162	80.4	08 June	135.6 ± 5.5	16.2
(4) Collections from 87,500-94,600 feet				
BR-129	91.6	21 Nov 57	124.2 ± 6.9	14.6
BR-137	94.6	19 Dec	165.1 ± 9.0	15.0
BR-139	89.1	08 Jan 58	133.6 ± 6.6	15.3
BR-145	89.5	10 Feb	133.9 ± 7.6	16.6
BR-147	90.0	06 Mar	143.9 ± 5.5	16.2
BR-154	90.0	05 Apr	121.1 ± 7.6	16.4
BR-156	89.0	11 May	116.3 ± 6.9	15.5
BR-158	88.0	16 May	142.9 ± 6.9	16.2
BR-161	87.5	05 June	158.1 ± 8.3	15.5

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Table II

Aircraft Samples

Collected in the Vicinity of 74°N, 156°W

a. Samples collected at 10,000 feet

Sample No.	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
PB-101	15 Jan 58	0.7 ± 6.2	32.6
PB-102	17 Jan	4.5 ± 5.5	35.3
PB-103	17 Jan	0 ± 9.3	34.9
PB-104	19 Jan	6.9 ± 8.0	31.8
PB-105	22 Jan	3.8 ± 11.4	31.6
PB-106	22 Jan	0 ± 8.3	31.9
PB-107	24 Jan	0 ± 9.7	32.0
PB-108	29 Jan	0 ± 14.2	32.7
PB-109	29 Jan	6.9 ± 8.3	34.1
PB-110	31 Jan	0 ± 4.8	33.7

b. Samples collected at 20,000 feet

Sample No.	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
PB-201	15 Jan 58	4.2 ± 10.4	31.4
PB-202	15 Jan	0 ± 5.5	30.5
PB-203	17 Jan	0 ± 6.6	32.2
PB-204	19 Jan	0 ± 6.9	32.8
PB-205	19 Jan	3.1 ± 6.9	32.7
PB-206	22 Jan	9.3 ± 7.6	31.2
PB-207	24 Jan	0 ± 6.9	31.6
PB-208	24 Jan	6.9 ± 7.3	31.4

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Sample No.	Collection Date	Excess Carbon-14 (atoms per gram) (of air x 10^{-5})	Gross Krypton-85 (atoms per gram) (of air x 10^{-3})
PB-209	29 Jan 58	0 ± 8.7	31.3
PB-210	31 Jan	4.2 ± 4.8	32.0
c. Samples collected at 30,000 feet			
PB-301	16 Jan 58	19.0 ± 5.5	24.2
PB-302	16 Jan	22.8 ± 8.3	24.4
PB-303	18 Jan	5.9 ± 5.9	26.3
PB-304	20 Jan	0 ± 5.5	28.2
PB-305	20 Jan	6.9 ± 5.5	28.8
PB-306	23 Jan	8.0 ± 6.9	26.2
PB-307	25 Jan	0 ± 6.9	28.5
PB-308	25 Jan	0 ± 6.9	28.5
PB-309	29 Jan	34.6 ± 8.7	23.0
PB-310	31 Jan	56.1 ± 6.2	26.5
d. Samples collected at 40,000 feet			
PB-401	16 Jan 58	57.8 ± 6.6	23.8
PB-402	18 Jan	61.6 ± 6.2	24.3
PB-403	18 Jan	41.9 ± 8.0	24.3
PB-404	20 Jan	130.1 ± 7.3	28.5
PB-405	23 Jan	96.5 ± 7.6	26.0
PB-406	23 Jan	92.7 ± 6.9	25.4
PB-407	25 Jan	73.7 ± 5.9	25.5
PB-408	29 Jan	150.2 ± 5.5	30.3
PB-409	29 Jan	147.8 ± 8.0	30.1
PB-410	31 Jan	114.5 ± 5.5	28.7

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DEPARTMENT OF THE AIR FORCE
HEADQUARTERS UNITED STATES AIR FORCE
WASHINGTON 25, D. C.

AFOAT-1/SWTD

13 SEP 1956

Downgrade to SECRET, CONFIDENTIAL
(on) ~~[illegible]~~ Nov 7 1956
Director, Division of Classification

CLASSIFICATION CANCELLED
BY AUTHORITY OF DOE/OC, DNA CEr. 89927 PRES/HAWTHORNE
W Strauser J Diaz 5/28/80
REVIEWED BY DATE
5/29/85

Dr. W. F. Libby, Commissioner
U. S. Atomic Energy Commission
1901 Constitution Avenue, N. W.
Washington 25, D. C.

Dear Bill:

I had hoped to get this data and report in smoother shape before you looked at it but events have prevented this. The opinion which I expressed to Dr. Scoville, copy of which letter I sent to you is based on considerations outlined very roughly in the following paragraphs.

Fig. 1 shows a map of the United States with the general areas from which we expect tritium releases. The numbers in parentheses on this map indicate the number of curies per month which we estimate are current releases from these locations based on a rough integration of past performance. Fig. 1 also shows the location of measurements which will be discussed in the following paragraphs.

Fig. 2 shows a chart of the Savannah River plant with a code of various locations at which tritium measurements have been made.

Fig. 3 is a table showing the actual measurements made in rain water, water vapor, streams, etc., at the various locations indicated in Fig. 2. These measurements were made at the Savannah River works last year (1955) and you will note that the maximum values are up around 9000 tritium atoms per 10^{18} protium atom.

Fig. 4 shows tritium measurements at Savannah River in 1956 during about the same part of the year May and June as those made in 1955. The 200H, 200F and the 100P areas are indicated in pencil on Fig. 2. There are two things to note in this Table. First, the tritium separations plant started up in February/March 1956. Second, the readings exceed those obtained in 1955 by a very large factor. The highest reading being 334,000 $T/10^{18}H$ in water vapor as opposed to the highest reading of about $T/10^{18}H$ 2000 in the previous year.

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Fig. 5 is a curve of the compilation of data from your project in Chicago for 1953 through 1956. I believe that it clearly shows a correlation between the CASTLE series and the tritium measurements in rain water. I cannot convince myself that there is any other marked correlation between the data and any of the known nuclear tests including the Soviet tests. The thing I would like to direct your attention to is the dashed line which I believe may show a growth of tritium background from 1953 through 1956 which is more likely to have been produced by the increased releases of tritium into the troposphere from production operations. I believe if we were to continue to follow the curve beyond June 1956 that the apparent increase in March through June will reveal itself as just another perturbation in the background level of tritium more likely caused by releases from production operations at some of the sources indicated in Fig. 1 with Savannah River being a real contributing source from February to June 1956.

Fig. 6 shows the tritium content in water vapor measured at Butler, Pennsylvania in the spring of 1956. The red line indicating the beginning of the REDWING series is preceded by a number of high points of tritium activity including the point of the very day of the shot which could not conceivably have been due to the first REDWING shot. I believe these points were definitely associated with production operations rather than with the tritium from nuclear tests.

Fig. 7 shows tritium in rain water at Boston. The first shot of the REDWING series is represented by a vertical red line and again you see substantial high values prior to the REDWING first shot. The high value after the REDWING series got underway is not very much higher than the peak shown in Fig. 6 which is clearly not due to REDWING. For this reason in Fig. 7 it is not possible for us to say that the highest value for middle of June is due uniquely either to production operations or to REDWING debris. In Fig. 7 you will note that the high activity produced by CASTLE in 1954 rain water at Boston diminishes very rapidly to background levels. I consider the center of gravity of the background data possibly indicates an increase of tritium background in the atmosphere near Boston due to the production operations.

Fig's. 8 and 9 represent rain water and humidry measurements in Chitosa, Japan and Tokyo, Japan. The thing that is noteworthy here is that the background level does not appear to be rising as significantly

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at these Japanese stations as at Chicago, Boston, Butler and Ottawa. Again only the CASTLE series shows up as distinct from production operations. If the tritium we are measuring is slowly filtering down from a stratosphereic tank then the buildup curve observed in your data at Chicago and our data at Boston should also be observed in Japan, and I believe that Fig's 8 and 9 show that this upward trend does not occur at Japan and, therefore, lends some weight to the argument that the increases observed in Chicago, Boston and Ottawa are due to production operation increases rather than to nuclear tests filtering from the stratosphere.

Fig. 10 is introduced to show individual peaks of activity measured at Hawaii in water vapor and rain water during the period March through June of the CASTLE series. Here it is noteworthy that the peaks produced by the shots rapidly decay to relatively low levels going up high on the next shot or two and then decaying rapidly again indicating that this tritium is in direct streaming of tropospheric air from Eniwetok and not tritium that filters down slowly from the stratosphere. We were interested in this data in the hope that the tritium activity might indicate something about the thermonuclear yield of the CASTLE shots. Unfortunately, there is no correlation in our data at Hawaii between thermonuclear yield and tritium measurements.

Fig. 11 is the only data we have on atmospheric hydrogen. I should remind you that the ordinates in this case are in tritium atoms/10^{15} hydrogen atom since we are dealing with the tritium atmospheric hydrogen rather than with hydrogen in the water vapor phase. First, I would like to point out the trend line which, starting from 1951, could be interpreted as indicating an increasing level of tritium activity in atmospheric hydrogen. Second, I would like to point out to you the three trends that are indicated by slopes a, b and c. Tritium releases from production sources are known to increase intensively just prior to test series such as GREENHOUSE CASTLE, TEAPOT and REDWING. These known releases of tritium in preparation for weapons tests then diffuse and decay as indicated by the slopes (a) and (b). However, in the case of slope (c), there appears to have been something different happening in 1956 than in any preceding year and we believe that this increasing tritium concentration in the spring of 1956 is correlated with the considerable increases in Savannah River production releases indicated in Fig. 11(top). These increased releases also produced corresponding increases in the tritium concentration in water vapor tabulated in Fig. 4. All the data that we have been able to

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get on tritium releases is plotted along the top of Fig. 11 including releases from Savannah River, Hanford, Los Alamos and Livermore. We believe the data to be complete although one must realize that complete accountability would be very difficult to achieve. For example, in the case of Los Alamos releases in 1955/56 the sum of the monthly releases does not agree with the figure of 114,000 curies listed as total releases. We believe, therefore, that the release information tabulated along top of Fig. 7 is probably in error on the low side and represents only those releases which are actually known.

I hope this may furnish background data for our discussion of this problem.

This letter is classified TOP SECRET RESTRICTED DATA because it reveals the fact that AEC production activities are releasing large amounts of tritium.

Sincerely,

Doyle

D. L. NORTHRUP
Technical Director, AFOAT-1
Office for Atomic Energy, DCS/O

1 INCL:
Figures 1 thru 11
1 cy each.

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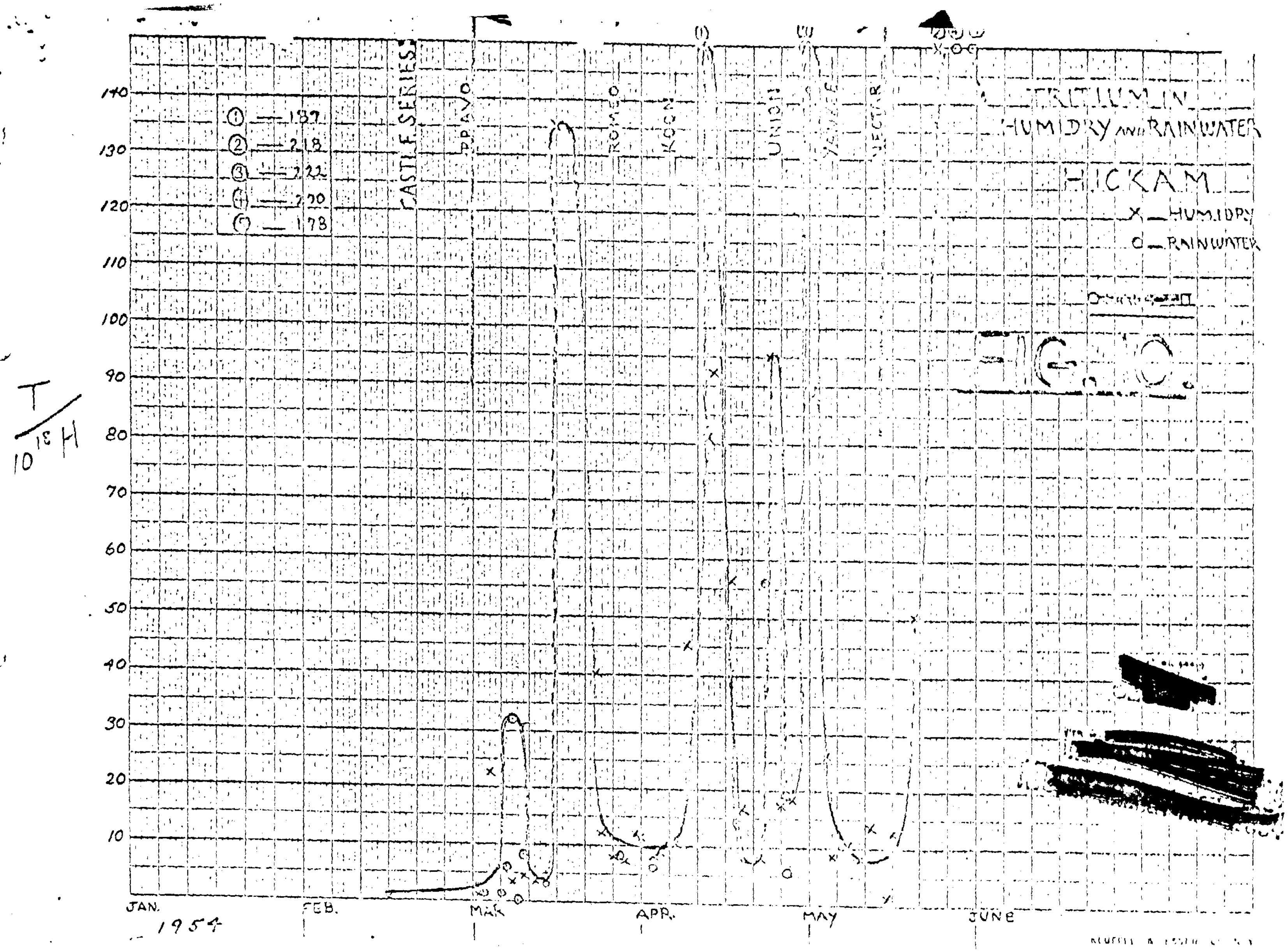

FIG. 10.

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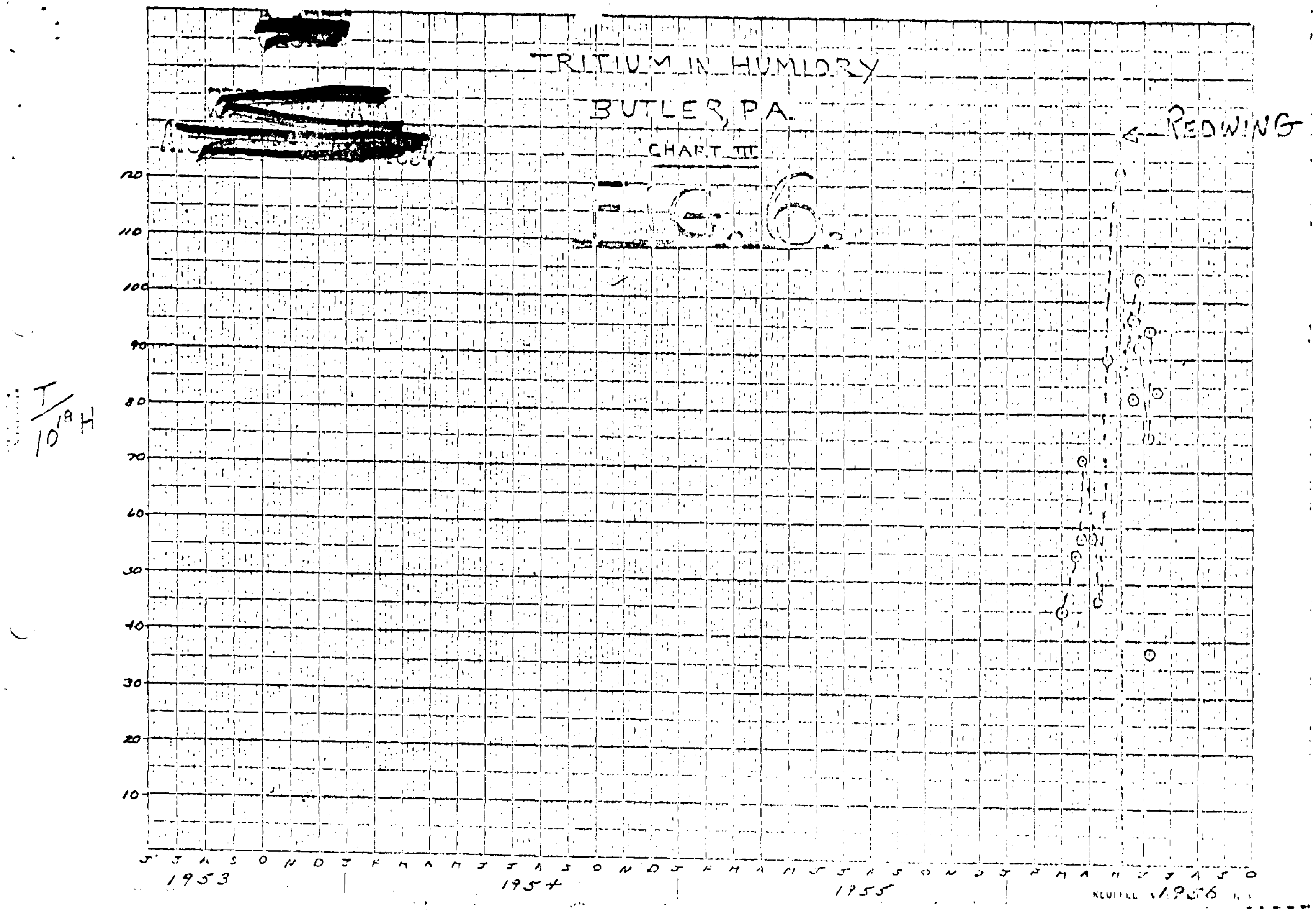

TRITIUM IN HUMIDITY

BUTLER, PA.

CHART III

FIG. 6.

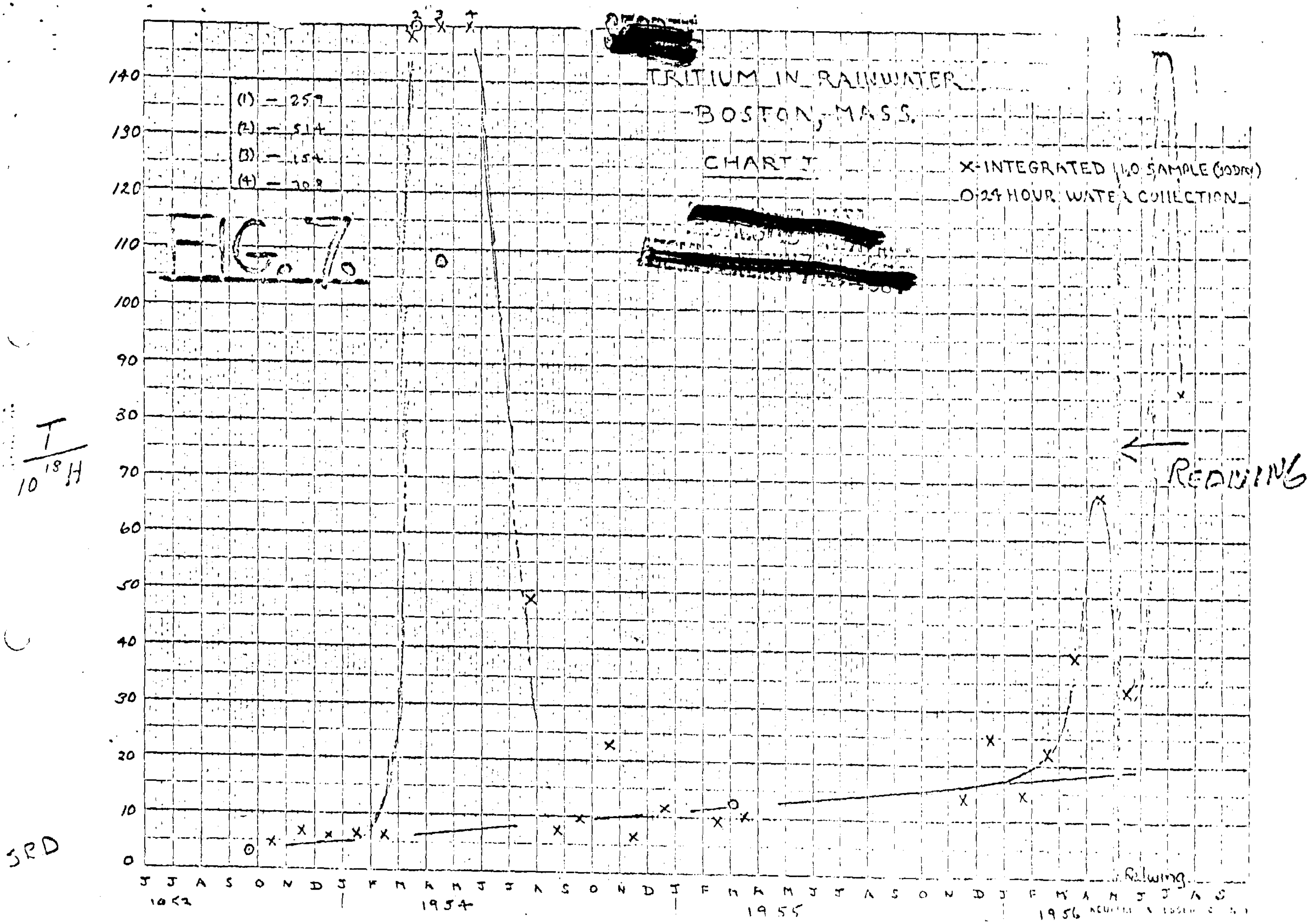

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FIG. 7

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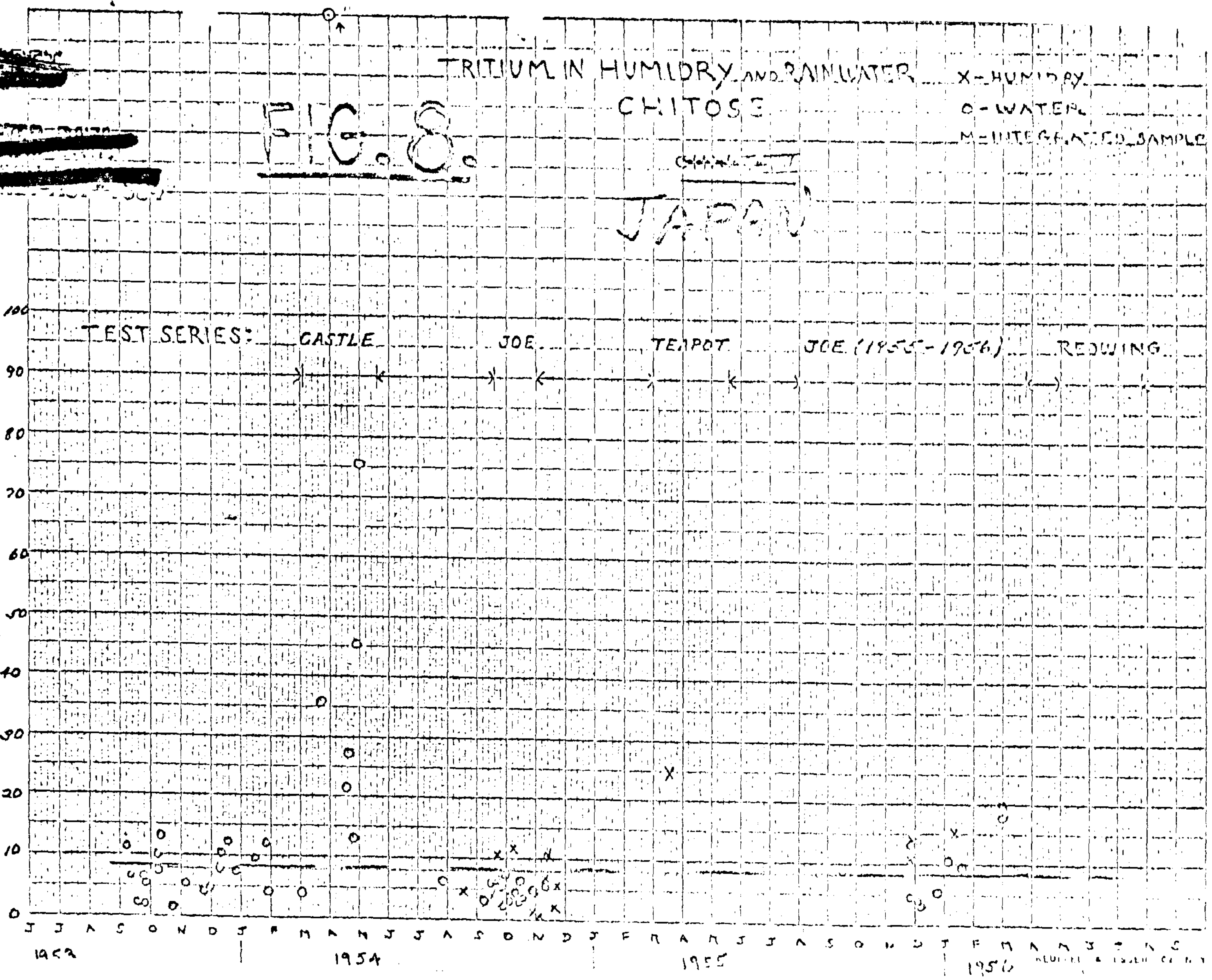
FIG. 8.



TRITIUM IN HUMIDRY AND RAINWATER

CHITOSE

JAPAN

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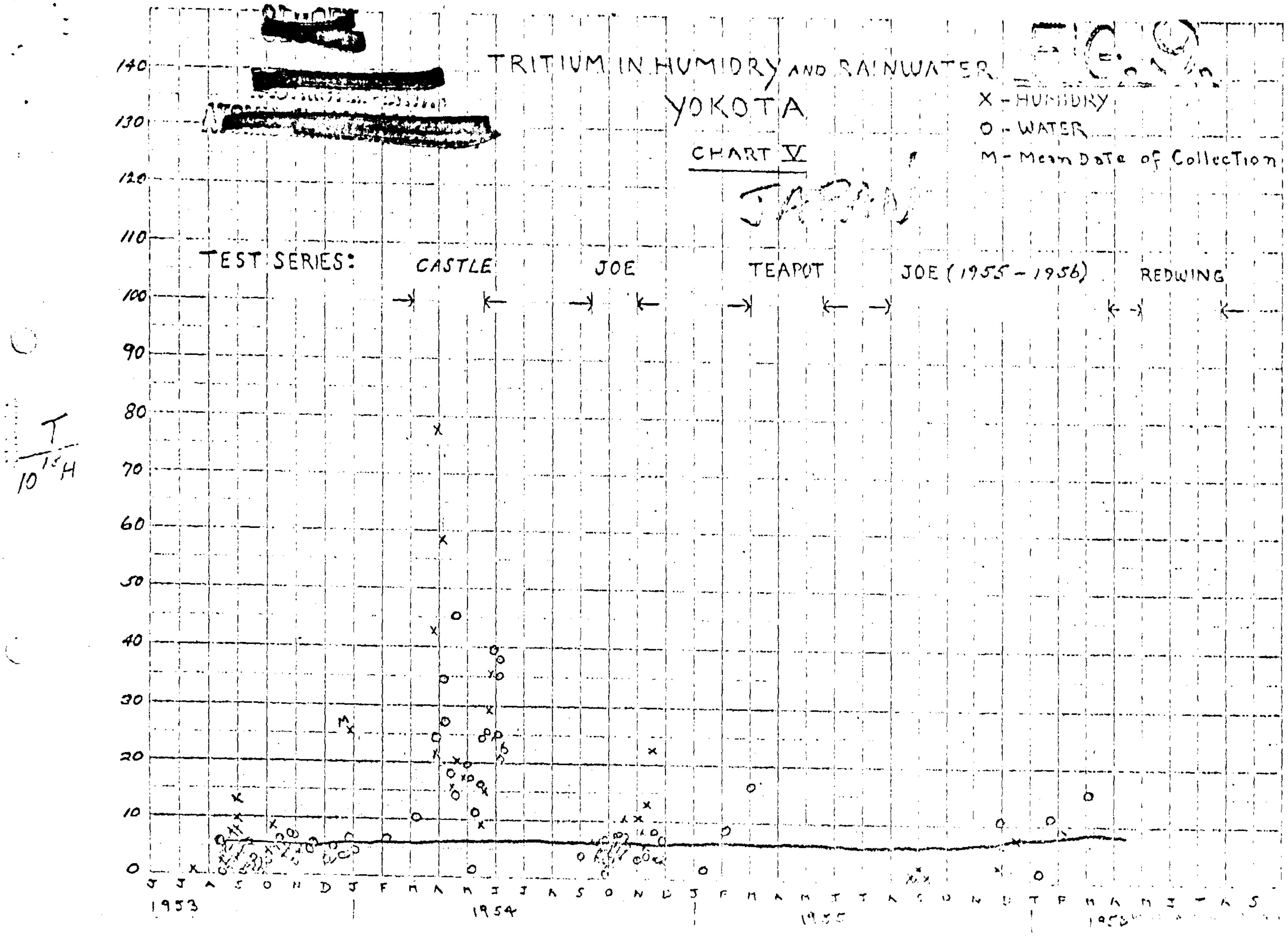

TRITIUM IN HUMIDRY AND RAINWATER

YOKOTA

CHART V

JAPAN