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SANITARY ENGINEERING ASPECTS OF LONG-RANGE FALLOUT FROM NUCLEAR DETONATIONS (Thesis)

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ABSTRACT

Results of a study of the sanitary engineering aspects of fallout from nuclear weapons are reported.

Data were obtained from:

a. Weekly surface water samples from 22 stations on streams and reservoirs in Eastern Massachusetts, collected from March, 1952 to July, 1953, and assayed for radioactivity.

b. Weekly rain samples from March to July, 1953.

c. Stream mud and soil samples collected before and after a large fallout.

d. Results of fallout activity measurements by others based on samples from Massachusetts and the vicinity of Rochester, New York.

Plots are presented showing the relations of the radiological data to the timing of detonations and the daily precipitation and showing daily deposition of activity and concentration in runoff.

Runoff coefficients for fallout radioactivity are calculated.

Low decay rates of fallout deposited during the Worcester tornado in the summer of 1953 suggest that the tornado scavenged high altitude debris from the November, 1952 "Ivy" detonation.

Variability of decay rates with the nature of the sample and the time of collection indicated some fractionation of the fission products in nature.

The efficiency of removal of fallout radioactivity by rapid sand filter water treatment plants in the study area was found to be between approximately 10% and 55%, the higher efficiency occurring soon after a detonation and diminishing with time. The efficiencies were lower than expected on the basis of theory and pilot plant experience.

PREFACE IV

This is a report of work done at Harvard University under Atomic Energy Commission Contract AT(30-1)841. It was originally written as a thesis in partial fulfillment of the requirements for the degree of Doctor of Science in Sanitary Engineering.

Many of the unpleasant aspects of the impact on civilization by the utilization of nuclear energy must be ameliorated by the sanitary engineer. Some of the most important of the problems to be solved are the ultimate disposal of radioactive wastes, especially those from power reactors; the restoration of water supplies and drainage facilities after bombardment; and the monitoring of the environment for unexpected radioactive contamination. Although only one phase of the last-mentioned problem is the subject of this thesis, the author believes that many of the techniques and viewpoints expressed, particularly those concerned with sample preparation and measurement, are closely related to general radioactive waste problems.

As this thesis is the first on a new topic, it was necessary to attempt a synthesis more extensive than usual of results of research in related fields. The author is fortunate in having been given for analysis fallout measurements taken by Mr. B. L. Rosenthal of the Massachusetts Department of Public Health and also a large number of measurements on fallout samples taken by others in the vicinity of Rochester, New York.

The author did the research and wrote the first draft of this thesis in close association with his faculty advisor, Professor H. A. Thomas, and is greatly indebted to him.

The author also wishes to express sincere gratitude to Mrs. Jane Maclachlan for her careful work in preparing most of the samples measured at Harvard and computing their count rates. Mr. F. L. Parker deserves much credit for his help, especially on occasions when the author was most in need. Under the direction of Dr. Thomas, Mr. Parker and Mr. R. S. Kleinschmidt had developed a large portion of the sample preparation and counting procedures before the author's phase of the work began.

Other persons connected with this research who were most helpful were Mrs. R. S. Kleinschmidt, Mr. Gerald Parker, Mr. Joe Murphy, Mr. K. C. Hannah, and Mrs. Joan K. Rothman.

On the Harvard faculty, Professor D. W. Batteau was most helpful in the electronic design of the automatic evaporator; Professors H. W. Emmons and C. H. Berry gave useful advice on the evaporation process, as did Professor J. C. Morris on several chemical problems. Dr. G. Gordon, who did his doctoral research in the Department of Physics at Harvard, was most kind in giving the author measurements of long-range fallout incidentally recorded by his cosmic ray instruments. The staffs of the United States Weather Bureau and the United States Geological Survey in Boston were also of considerable help.

Appreciation is expressed to Mr. A. E. Gorman and Mr. J. Lieberman, both of the Atomic Energy Commission, the sponsor of the project in which research for this thesis was conducted.

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SYNOPSIS

"Fallout", a new term in our language, can be defined as airborne radioactive debris created by nuclear detonations and eventually deposited on the earth. Since it is formed in vast quantities and spread by the winds over the face of the earth before deposition, the study of fallout cannot but be of interest to persons considering public health problems, or for that matter, the welfare of mankind. In this thesis, the necessary first phases of study of the sanitary engineering aspects of fallout are presented as a basis for future research on such vital questions as the histories in nature of individual radioactive nuclear species from long-range fallout.

From March, 1952, to July, 1953, the count rates of radioactivity of samples collected weekly from all or most of twenty-two stations located on streams and reservoirs in eastern Massachusetts were measured automatically with Geiger-Müller tubes. Between March and July, 1953, simple rain-collection devices were placed at most of the stations and rain samples were also collected weekly and measured automatically. Soil samples were collected both from stream banks and stream bottoms before and after a large fallout.

In addition to making these measurements, the author was permitted to analyze data collected on fallout radioactivity at the Lawrence Experiment Station of the Massachusetts Department of Public

Health and also data on a large number of fallout samples collected in the vicinity of Rochester, New York. At Harvard some 70,000 measurements were made on approximately 4,400 samples; at Lawrence some 6,000 measurements on approximately 600 samples; and near Rochester some 12,000 measurements on approximately 6,000 samples.

Fallout sample collection and preparation procedures are described. The problems of water sample evaporation are discussed with a review of the techniques used for this thesis. Circuit diagrams and photographs are included of a sample evaporation device built for this research that is controlled electronically by the change in bias on the grid of a triode tube caused by the leakage of current through the sample being evaporated. This device was found to be reliable and an improvement over the commonly used steam bath for non-boiling evaporation of samples.

Master plots are presented for the eastern Massachusetts and the Rochester, New York, areas relating the announced detonation dates, daily precipitation, concentration of radioactivity in the rain, concentration of radioactivity in the surface waters, calculated daily areal deposition of radioactivity, and calculated daily areal concentration of radioactivity in the runoff from the watersheds. The plotted measurements indicate that during a Nevada test series a single rain in the northeastern part of the United States can be more than two orders of magnitude more radioactive than any other precipitation during the same series and that the deposited radioactivity from a large fallout can be detected in the surface waters by gross beta ray

measurement on evaporated samples for more than two months following the fallout.

The areal concentrations of fallout (in terms of beta ray count rate per square mile) were calculated and are graphed, as is the areal concentration of radioactivity in the runoff during the periods in which samples were collected in eastern Massachusetts and near Rochester, New York. For this thesis, a coefficient of runoff for radioactivity is defined as the ratio of the sum of disintegrations of beta radioactivity in the runoff (from a unit area of watershed over a period of approximately three months following a large fallout) to the radioactivity deposited by the fallout (on the same area computed as at the time of deposition). This ratio reflects retention of radioactive material by the watershed and decay of the radioactive material between the times of deposition and collection from the surface waters, as well as such effects as sedimentation of the radioactivity in the receiving waters. In eastern Massachusetts the runoff coefficients for seven streams after the April 7-8 fallout were found to have a geometric mean of 1.2% with 95% confidence limits of 0.27% and 5.1%. The runoff coefficient for the Genessee River at Rochester, New York, is estimated also to have a geometric mean of 1.2% but with 95% confidence limits of 0.14% and 9.2%

In the spring and summer of 1953, eighteen integrated rain samples were collected weekly at stations located in eastern Massachusetts. The concentrations of radioactivity in those samples collected following the large fallout on April 7-8 were found to vary widely

between samples. At the time of deposition, the geometric mean count rate was estimated to be 1.4×10^5 counts per minute per liter and the geometric standard deviation of the distribution, 6.4.

A single set of data indicated that the concentration of fallout radioactivity deposited on the ground during a rainstorm is quite variable with time of deposition. There was no indication of an initial large deposition with subsequent depositions decreasing in concentration exponentially with time after the beginning of the storm.

Analyses are presented of the die-away of measured long-range fallout external gamma radiation with time after the large April 7-8, 1953, deposition in eastern Massachusetts. Decay measurements with cosmic ray Geiger-Müller tubes and counts of photographed ionization tracks in a cosmic ray cloud chamber of electrons ejected by fallout gamma rays followed hyperbolic decay formulations with rate constants quite close to 1.2, which contrasts with the rate constants averaging approximately 1.4 for the beta decay of evaporated rain samples from the same fallout. The gamma ray measurements indicated that the fallout radioactivity was not moved by rains following initial deposition.

Fallout samples collected during the Worcester tornado in the summer of 1953 were found to have a relatively low concentration of radioactivity, but some of the samples decayed at a surprisingly low rate. It is suggested that the tornado scavenged the debris from the November, 1952, Eniwetok "Ivy" detonation from above the tropopause.

Surface water decay rates following the April 7-8 fallout in 1953 were examined and found to be quite different depending on time

of collection after the fallout. The differences in the decay rates of the groups of samples were found to be statistically significant, indicating that there was a relatively strong preferential uptake of fallout radioactivity in nature.

Approximately 2,200 samples for radioactivity measurement were taken in the various stages in three water treatment plants located at Cambridge and at Lawrence, Massachusetts, and near Rochester, New York. All plants employed alum coagulation and rapid sand filtration. A system of small reservoirs, a river, and Lake Ontario were the respective water sources.

It was found that within about two weeks after a Nevada detonation approximately 45% of the gross long-range fallout radioactivity entering the rapid sand filter plants passed completely through them. From two to ten weeks after detonation about 53% passed through the plants. For periods beginning at least ten weeks after detonation practically all of the radioactivity (partially of natural origin) passed through the plants. After the first three days following fallout, the rapid sand filters were able to remove only a small portion (approximately 10%) of the radioactivity passing the sedimentation process. In all cases, the average removal efficiencies of the various stages of the plants were lower than those of pilot scale plants using pile-produced fission products and much lower than reported estimated removals based on theoretical considerations.

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CHAPTER I

GENERAL DISCUSSION OF LONG-RANGE FALLOUT

A. NUCLEAR DETONATIONS

As nuclear detonations take place, the hot gases produced may rise rapidly at least to the top of the troposphere (1) and perhaps on into the stratosphere (2,3). The fireball is made up of fission products; unfissioned plutonium (Pu^{239}) or uranium (U^{235}); products of the material used to set off the fission detonation, which may be assumed to be the same as those from conventional explosives (4); usual constituents of the atmosphere; steel from the support tower if one is used; soil if the detonation takes place low enough for the fireball to come close to the ground; and any other material touched by the fireball.

Upon cooling, many vaporized materials making up the detonation's fireball go to their oxide state, that is, much of the material condenses into solids to form particles. As the debris rises, almost all of the residual radioactivity from fission products is located in the head of the mushroom cloud (5). Unless the detonation takes place at a high altitude, the detonation shock wave and winds cause various amounts of earth to be forced up into the debris cloud. The radioactive particles in the cloud, predominantly metallic oxides, are reported to adhere to soil particles (6).

This radioactive debris is distributed vertically in the atmosphere by the rising hot gases and is subject thereafter to such forces as gravity and electrical attraction, as well as air mass movements. Depending on atmospheric conditions and the characteristics of the products, the debris is deposited at various places on the earth. Both this debris and its deposition are called "fallout", a new but well-established term.

1. Size of the detonations

"Experimental nuclear device" is a better term than "weapon" or "bomb" to describe the types of apparatus detonated in the Nevada tests, since these experiments have been conducted to gain information for weapon design, not to test a finished weapon type. As the best theoretical calculations have misjudged the size of the Nevada fission detonations by 50% (7), it may be assumed that they are quite variable in size.

It has been announced (8) that there are now fission devices that release twenty-five times the energy of the "nominal" Hiroshima-type bomb, that is, they release an amount of energy equivalent to that in the detonation of 0.5 million tons of TNT. Also, there now exist fusion devices that release "millions" of tons of equivalent TNT energy, that is, some multiple of fifty times the nominal bomb energy. W. F. Lawrence has reported (9) that the November, 1952, Eniwetok detonation is generally unofficially known to have been larger in energy released than 5,000,000 tons (5 megatons) of TNT. When the size of nuclear detonations and the great distances the debris is carried are considered,

it is apparent that more knowledge of long-range fallout, its health significance, and methods of solving the sanitary engineering problems created is essential.

2. Nuclear species created in fission

A large portion of newly-created fission fragments are not physically stable. These species, however, instead of again undergoing fission, tend toward more stable states by disintegration, a spontaneous nuclear transformation that takes place with the emission of mass (beta particles) and/or energy (gamma and/or X-rays).

At one minute after a nominal bomb detonation, the fission debris is undergoing 1.8×10^{24} gamma-producing disintegrations per minute (0.82 megamegacuries) (1). This disintegration follows a law that may be written $A_t = A_1 t^{-1.2}$, that is the radioactivity A_t of the products at any time t after fission detonation is equal to the radioactivity of the fission products at unit time after detonation (one minute, one hour, or one day, for example) divided by the time after fission taken to the 1.2 power in the same units as those chosen for A_1. For slow neutron fission, Hunter and Ballou estimate (10) that this formulation gives values that lie within about 10% of the correct values.

No time limit for this formulation is given in The Effects of Atomic Weapons (1), nor is any indication given that the average energy of the fission product beta and gamma particles changes with time; Figure 8.17 in this book indicates that the ionizing radiation from fission debris follows this formulation for at least forty days.

Instead of producing two equal halves, uranium (U^{235}) and plutonium (Pu^{239}) divide to give a distribution of more than sixty primary fission fragments which are forms of thirty-four elements that range in atomic number from 30 (zinc) to 64 (gadolinium). The distribution of the mass numbers of the fission products as a function of their percentage compositions in the total yield just after fission is distinctly bimodal. About 97% of the fission material lies within the two ranges of mass numbers 85 to 104 and 130 to 149. On an average, each primary fission fragment undergoes approximately three disintegrations before reaching a stable state. In a short time after the initial detonation, more than three hundred radioactive species are known to have been created, as well as a large number of stable nuclear species. Sixty-one atom percent of the fission products have a half-life greater than eight days and less than one year. Strong gamma ray emitters amount to some forty-five atom percent and strong beta ray emitters amount to some thirty-three atom percent of the fission products (11). These are the nuclear species that are of principal concern from a health standpoint.

B. LONG-RANGE FALLOUT

1. Physical and chemical nature of long-range fallout

Radioactive particles that are deposited at long distances from their origin range from less than a micron to more than one hundred microns in size (6). Particles up to ten microns in size are reported often to be made up of fission products only; larger particles are more likely to be a combination of fission material and dust. Particles of

twenty-two microns in diameter were detected at the Oak Ridge National Laboratory in Tennessee on March 19, 1953. It was shown by the die-away of the radioactivity in the particles that they originated in the Nevada detonation of March 18, 1953 (12).

A knowledge of the particles' size distributions, shapes, densities, surface characteristics, solubilities, porosities, and resistances (to weathering, crushing, and erosion) is necessary for a complete study of long-range fall-out by sanitary engineers. It would also be of interest to know how homogeneously the radioactivity is distributed in the particles and if solid solution effects are important. Such information is necessary for a study of retention and run-off of fallout radioactivity from water sheds, uptake by algae and suspended solids in the water, passage through water treatment plants (in coagulation, settling, filtration, and softening), and retention by the filter sand and the pipe walls of the distribution system.

A knowledge of the chemical states of the radioactive elements in fallout is necessary for these studies. A listing has been made (4), based on chemical-thermodynamic considerations, of the most likely chemical compounds formed as the material in the fireball cools. In this estimation, no consideration was given to nuclear reactions that take place in the fission products. For example, $Rb^{89}O_2$ may decay and form $Sr^{89}O_2$, rather than go to the more chemically stable $Sr^{89}O$; the former reaction product is not listed in this table.

Table 1 gives the individual nuclear fission species that contribute more than 1% to the total disintegration rate at their peaks after the first day following fission, their half-lives (102); the time of these peaks (10); the percentage contribution at their peaks (10); the

types and energies of radiation (10); their tolerance values in water (14); and the most likely chemical compounds in which they would exist (4). Table 2 gives data on radioactive species other than fission products that could be in long-range fallout, which would consist of species of neutron-induced radioactivity and the unfissioned fuel material. As fusion detonations would have an intense neutron flux (11,15), induced radioactivity could become a more important constituent of long-range fallout.

2. Carriage of detonation debris

An estimation was made (1) for the rate of total fallout following the Alamogordo "Trinity" test in which it was assumed that the fission products were taken uniformly upon the surface of a dust having a size distribution like that collected above the Sahara Desert. It was calculated that 51% of the debris from the Alamogordo detonation (which took place on a tower) remained suspended in the atmosphere for more than eight hours. Deposition of fission products on a target from a high altitude burst of a bomb or device is of negligible importance unless it is dropped in a rainstorm (11).

One radioactive cloud is often carried, at different layers, in various directions from the detonation site (6). The newly-formed cloud (or clouds) may be visible for about an hour and specially-equipped monitoring aircraft are often able to follow it for about six hundred miles. In one case, a cloud from a Nevada detonation was tracked by monitoring planes to Ohio (16).

TABLE 1

PERTINENT DATA ON FISSION SPECIES (102)

(Listed in Order of Occurrence After Fission When at Maximum Proportion of Total Fission Product Radioactivity)

Species	Half-Life	Time of Maximum (days)	%*	Type of Radiation		Tolerance ($\mu c/ml\ H_2O$)	Probable Chemical Compound
				Beta-Mev.	Gamma-Mev.		
Xe^{135}	9.13 hr.	1.0	12.5	0.905	0.250	1×10^{-3}	Xe
Zr^{97}	17.0 hr.	1.3	9.2	1.91	0.747	-	ZrO_2
Nb^{97}	72.1 min.	1.3	10.0	1.267	0.665	-	Nb_2O_5
I^{133}	20.5 hr.	1.8	8.6	1.3, 0.4	0.53, 0.85, 1.4	-	I_2, IBr
Ce^{143}	33 hr.	2.5	10.2	1.39, 1.09, 0.71	0.035 - 0.72	-	CeO_2
Rh^{105}	36.5 hr.	2.6	2.2	0.570, 0.25	0.322, 0.157	1.5×10^{-2}	Rh_2O_3, Rh
Pm^{149}	54 hr.	3.5	2.9	1.05	0.285, ~1.3	-	Pm_2O_3
Tc^{99}	6.04 hr.	4.0	1.33	-	0.1403, 0.1423	-	TcO_3
Mo^{99}	67 hr.	4.1	12.3	1.23, 0.45	0.040, 0.181, 0.367, 0.741, 0.780	14	MoO_3
Te^{132}	77.7 hr.	4.3	7.8	0.22	0.231	-	TeO_2

* "%" is percent of total activity at time of maxima.

TABLE 1—Continued

Species	Half-Life	Time of Maximum (days)	γ^*	Type of Radiation		Tolerance ($\mu c/ml$ H_2O)	Probable Chemical Compound
				Beta-Mev.	Gamma-Mev.		
I^{132}	2.4 hr.	4.3	8.0	2.2, 0.9	0.69, 1.41, 2.0	-	I_2, IBr
Xe^{133}	2.3 dy. 5.270dy.	7.2	12.0	0; 0.345	0.233; 0.081	4×10^{-9}	Xe
I^{131}	8.141dy.	11.5	6.8	0.815, 0.608, 0.335, 0.250	0.080, 0.163, 0.284, 0.364, 0.637, 0.722	3×10^{-5}	I_2, IBr
Nd^{147}	11.3 dy.	15.5	5.15	0.83, 0.60, 0.38	0.0918, 0.309	-	Nd_2O_3
La^{140}	40.0 hr.	18.0	13.9	1.32, 1.67, 2.26	0.093, 0.3286, 0.4867, 0.8151, 1.596,	1	La_2O_3
Ba^{140}	12.80 dy.	18.0	12.0	1.022, 0.480	0.0296, 0.132, 0.162, 0.304, 0.537	2×10^{-3} (with La^{140})	BaOH, $Ba(OH)_2$
Pr^{143}	13.7 dy.	21	12.0	0.932	-	0.4	Pr_6O_{11}
Ce^{141}	33.1 dy.	40	11.6	0.581, 0.442	0.145	-	CeO_2
Ru^{103}	39.8 dy.	72	7.4	0.217, 0.698	0.498	-	RuO_2
Rh^{103}	57 min.	72	7.3	-	0.0400	-	Rh_2O_3, Rh
Sr^{89}	53 dy.	99	10.6	1.463	-	7×10^{-5}	SrO, $Sr(OH)_2$ $SrCO_3$

TABLE 1--Continued

Species	Half-Life	Time of Maximum (days)	f^a	Type of Radiation		Tolerance ($\mu c/ml$ H_2O)	Probable Chemical Compound
				Beta-Mev.	Gamma-Mev.		
Y^{91}	61 dy.	110	12.7	1.537	1.2, 0.2	0.2	Y_2O_3
Zr^{95}	65 dy.	130	15.4	0.371, 0.84	0.721,	-	ZrO_2
Nb^{95}	35 dy.	199	25	0.160	0.745	4×10^{-3}	Nb_2O_5
Ce^{144};Pr^{144}	282 dy.	1.5 yr.	32.3	0.300, 0.170	0.0337, 0.054, 0.0807, 0.100, 0.134	4×10^{-2}	CeO_2
Ru^{106};Rh^{106}	1.0 yr.; 30 sec.	2 yr.	3.4	0.0392; 3.53, 3.1, 2.44, 2.0	-; 0.513, 0.624 0.87, 1.045, 2.41	0.1	RuO_2
Pm^{147}	2.6 yr.	5 yr.	22	0.223	-	1	Pm_2O_3
Kr^{85}	9.4 yr.	9.5 yr.	1.7	0.695, 0.15	0.54	-	Kr
Sm^{151}	73 yr.	17 yr.	2.6	0.076	0.019	0.2	Sm_2O_3
SrY^{90}	19.9 yr.; 61 hr.	20 yr.	24	0.61; 2.18	- ; -	8×10^{-7}	See Sr^{89}, Y^{91}
Cs^{137};Ba^{137}	33 yr.; 260 min.	100 yr.	-	0.523, 0.6616	-	1.5×10^{-3}	CsO_2, CsI, CsBr, CsOH, $CsCO_3$

TABLE 2 10

PERTINENT DATA ON IMPORTANT SPECIES FROM INDUCED RADIATION AND UNFISSIONED MATERIAL THAT CAN BE PRESENT IN LONG-RANGE FALLOUT OF DETONATION DEBRIS (102)

Species	Half-Life	Type of Radiation		Tolerance (μc/ml H_2O)	Probable Chemical Compound
		Beta-Mev.	Gamma-Mev.		
H^3	12.46 yr.	0.01795	-	0.2	H_2O
C^{14}	5.568 yr.	0.155	-	3×10^{-3}	CO_2
P^{32}	14.30 dy.	1.701	-	2×10^{-4}	P_2O_4, $PO_4^{\equiv}$
S^{35}	87.1 dy.	0.1670	-	5×10^{-3}	SO_2, SO_3, $SO_4^{\equiv}$
Cr^{51}	27.8 dy.	K	0.330	0.5	Cr_2O_3
Fe^{59}	45.1 dy.	0.460, 0.257	1.295, 1.097	1×10^{-4}	Fe_3O_4, Fe_2O_3
Fe^{55}	2.94 yr.	K	-	4×10^{-3}	Fe_3O_4, Fe_2O_3
Ni^{59}	8×10^4 yr.	K	-	0.25	NiO
Cu^{64}	12.8 hr.	0.571	γ with E.C.	1×10^{-4}	CuO
Zn^{65}	250 dy.	0.325	1.120	6×10^{-2}	ZnO
Na^{24}	15.06 hr.	1.390	1.3679, 2.7535	8×10^{-3}	-

TABLE 2—Continued

Species	Half-Life	Type of Radiation		Tolerance (μc/ml H_2O)	Probable Chemical Compound
		Beta-Mev.	Gamma-Mev.		
U^{235}	7.13×10^8 yr.	4.58	0.094, 0.143, 0.184, 0.289, 0.386	-	U_3O_8
Np	-	-	-	-	NpO_2
Pu^{239}	24, 360 yr.	5.150	0.039, 0.0531, 0.100, 0.124, 0.384	1.5×10^{-6}	PuO_2

The air masses which carry detonation debris may move at surprisingly high speeds. It is quite common for an air mass above 20,000 feet altitude to move, for example, from Nevada to New England in forty to fifty hours. Jet streams often travel along the same general path but usually at a higher altitude at more than twice this speed. With the general west-to-east air movement across the United States, the majority of the Nevada detonations' long range-fallout is deposited to the east of the test site. Often particles from detonations can be detected in virtually any part of the United States, and some particles have been measured that have traveled a large portion of the distance around the earth. For example, the debris from the "Able" detonation at Bikini on July 1, 1946, was detected twenty days later over Paris, France (17). The sharp increase in the radioactivity of the rains falling in late August and early September, 1953, in Cambridge, Massachusetts, undoubtedly was due to the Russian detonations which occurred about two weeks previously. Although these long carriages and wide distributions take place, it is important to note that, because of eddy diffusion, it is possible for debris from a detonation to be deposited in one area and be completely absent from neighboring areas, even if the atmosphere is turbulent.

It was the author's experience that an appreciable fallout was always accompanied by a rainfall. Stefanizzi listed (18) ordinary rain, thunder showers, and snow as increasing in this order in their ability to remove natural radioactivity from the air. From a study of radon (Em^{222}) in rain he concluded that at least a part of the radon (Em^{222}) was acquired by the precipitation in its fall through the air after leaving the cloud.

As an illustration of the movement of fission debris from the Nevada test site, a brief review will be made of the histories of the fission cloud and the rainstorm involved in the largest fallout observed in eastern Massachusetts during the period of continuous monitoring for this thesis from May, 1952, until December, 1953. The cloud of the detonation of April 6, 1953, rose to 40,000 - 50,000 feet in a few minutes (19) and there was no fallout in the test area.

3. Trajectories of the debris clouds

Estimated trajectories of portions of the radioactive fission debris cloud were drawn for the 150, 200, 300, and 500 millibar heights as shown in Fig. 1. Calculations for these trajectories were based on data and maps of the United States Weather Bureau. Also shown is the path of the center of a low-pressure area that accompanied the rainfall in eastern Massachusetts on April 7-8. Starting April 6 in eastern Texas, this area moved through the Tennessee Valley and up the Atlantic coast. Rain began in Massachusetts after midnight on April 7 and ended the morning of April 8, giving an average precipitation of 1.4 inches. As the storm was of a type that probably had clouds that reached 30,000 feet, it is reasonable that the rain scavenged and precipitated detonation debris from this level. Also, portions of the cloud at the 300 millibar level could have been mixed with the air of the low-pressure area, swept over Massachusetts, and deposited with the rain. The plots shown are the estimated centers of portions of the radioactive fission cloud for the stated pressure elevations. Naturally, in almost two days of travel there was appreciable diffusion from these centers. Surface water samples were collected at fourteen eastern Massachusetts stations

(the same ones at which weekly monitoring samples were taken for fourteen months) just after this large fallout. They registered an average of 212 net counts per minute per liter on instruments that measured principally beta radiation. Rain samples from thirteen eastern Massachusetts stations gave an average count rate of 71,000 counts per minute per liter. These values are believed to be more than one hundred and more than ten thousand times natural levels, respectively.

Time		Designation	Pressure Height	Approx. Elevation
0	1530 GCT April 6	—●—	150 millibars	45,000 ft.
A	0330 GCT April 7	—×—·—×—	200 millibars	39,000 ft.
B	1530 GCT April 7	—+—·—+—	300 millibars	31,000 ft.
C	0330 GCT April 8	—□···—□—	500 millibars	19,000 ft.
D	1530 GCT April 8	--△--------△--	Cyclone center track at sea level (101)	
E	0330 GCT April 9			

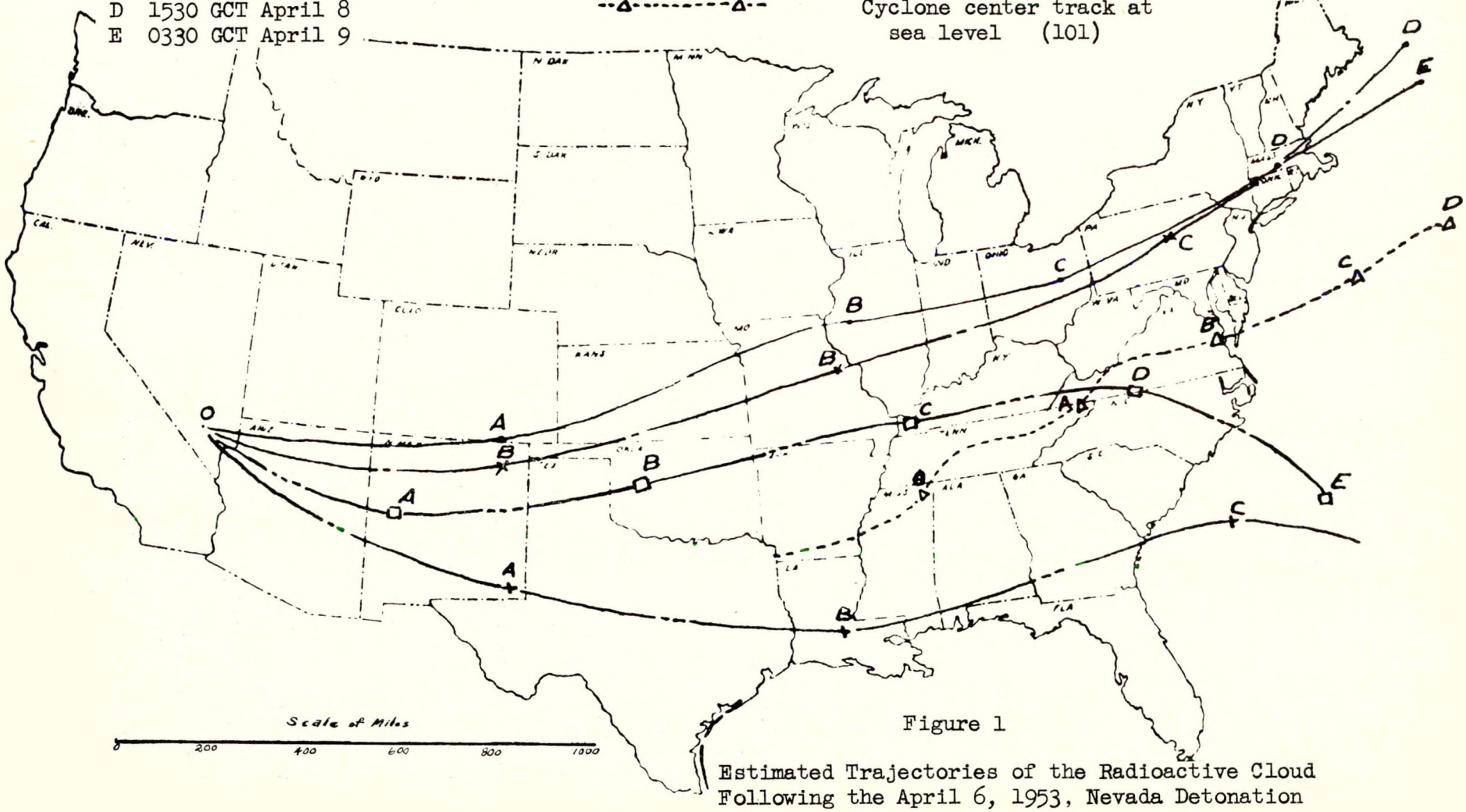

Figure 1

Estimated Trajectories of the Radioactive Cloud Following the April 6, 1953, Nevada Detonation

CHAPTER II

HAZARDS DUE TO LONG-RANGE FALLOUT

A. HEALTH HAZARDS OF LONG-RANGE FALLOUT

Three effects should be considered in evaluating the health significance of long-range fallout. They are (a) radiation sickness (which is caused principally by intense penetrating radiation, (b) long-term tissue damage (that would be due principally to internal radiation), and (c) genetic changes (from either external or internal radiation). Sanitary engineering is concerned principally with monitoring and controlling waterborne radioactive material that would cause internal radiation damage if consumed. This chapter is centered on that aspect of the topic.

1. Maximum permissible levels

a. Basis. Tolerance levels for ionizing radiation are based in part on each of three considerations: the threshold mechanism of the damage that is caused, the recovery power of the body, and an agreed

acceptable low incidence or degree of harm (20). Limits are usually given as "maximum permissible" levels rather than "tolerance" levels, in order to emphasize keeping exposure to the lowest practical value rather than just below a set maximum. They are usually set on the basis of working life (for occupational exposure) or lifetime exposure (for environmental exposure) (20, 14). Long-range fallout effects would be based on the latter.

Tables of the maximum permissible concentration values for many of the nuclear species likely to be encountered by the sanitary engineer have been published by the National Bureau of Standards (14). The values were compiled by the Subcommittee on Permissible Internal Dose of the National Committee on Radiation Protection, which advises that "because of the uncertainties involved in the present values and in determining the actual accumulation and potential hazard of radioisotopes in the human body, it is strongly recommended that exposures be kept to a minimum insofar as it is practicable" and "a factor of safety that may be as large as ten be used in the design and operation of permanent installations where large quantities of radioactive materials are involved".

b. Permissible levels. A provisional limit of 0.1 micromicrocurie per milliliter of water has been set for material of unknown nuclear species that is outside any area controlled by the United States Atomic Energy Commission. This concentration value is believed to be safe for exposure to any of the radioisotopes for a few months; here the word "safe" means not producing any readily detectible biological changes.

1) External radiation. Maximum permissible weekly doses (14) in the blood-forming organs are: 0.3 roentgen per week (r/week) for gamma rays and X-rays, 0.3 roentgen equivalent physical per week (rep/week) for beta rays, and 0.015 roentgen equivalent physical per week for alpha rays.

Radiation is measured for health purposes in units of the roentgen or its equivalents. A roentgen (r) is that amount of gamma or X-radiation which produces, in one cubic centimeter of dry air at standard conditions, electrically-charged particles (2.083×10^9 ion pairs) carrying a total of one electrostatic unit of charge of either sign. A milliroentgen is 1/1000 of this value. Two supplementary units are used for the ionization of human tissue. A roentgen-equivalent man or mammal (rem) is that quantity of radiation which when absorbed by man produces an effect equivalent to the absorption by man of one roentgen of gamma or X-radiation. The roentgen-equivalent physical (rep) is the amount of ionizing radiation that would result in the absorption in tissue of 83 ergs (equivalent to 1.615×10^{12} ion pairs) per gram of tissue. Some authorities have suggested 93 ergs per gram for this value, which would be about two ionizations per cubic micron. The two roentgen-equivalent values do not have to be for ionization caused by photon radiation but can be for beta or alpha radiation. These three types of radiation are of importance in the study of long-range fallout. Owing to the relatively short range of other radiations present in fallout, gamma rays are of primary concern in external radiation. In fallout, their mean energy is 0.7 million electron volts (Mev) (1).

Neutrons that are produced but not used in the chain reaction of fission are captured by many of the non-fissioning elements present

at the detonation, such as those in earth and the device's casing. Of these nuclear species formed by neutron capture, many are radioactive, and sodium (Na^{24}) is perhaps the most dangerous because of the amount produced and the strong gamma rays emitted when it undergoes beta decay. Sodium (Na^{24}) has a half-life of only 15.06 hours; therefore, it can be important only in the early stages of long-range fallout.

When received externally, the alpha radiation from long-range fallout is less hazardous than beta rays and still less dangerous than gamma rays. The range of alpha particles is only an inch or two in air and they are completely absorbed by ordinary clothing. Almost all are stopped by the skin's epidermal layer. As only one species produced in nuclear fission, samarium (Sa^{147}), has been found to emit alpha rays, it can be assumed that most of the alpha radiation would come from fission material not detonated.

Few of the fission products emit beta radiation having a mean energy greater than two million electron volts. Only a small portion of these beta rays can penetrate more than approximately one centimeter of tissue; therefore, most cannot penetrate to those parts of the body that are especially sensitive to radiation, such as the blood, the bone marrow, and the spleen.

2) Internal radiation. It has been stated (1) that if external radiation from fallout is at a safe level, then in general there is little danger from internal radiation by fission material fixed in the human body. Relatively small amounts of fission debris would enter the body soon after detonation of a nominal bomb; however, since much more

powerful nuclear devices have been constructed, it is important to examine the health hazards of long-range fallout on the basis of long-term exposure of large portions of the race to relatively low levels of radioactivity.

The damage caused by externally received X-rays or gamma rays has been studied for more than fifty years and many observations have accumulated. Determinations of acceptable concentration levels for internal emitters, except radium (Ra^{226}), have short histories. The mechanisms of uptake, retention, and elimination by the body are all extremely complex. So many variables are present that, even when the most careful investigations are repeated, there are often important differences in the reported results (14).

The bases for estimating the hazards caused by internal radiation have been established on knowledge of the effects of externally-originating X-rays and gamma rays, natural (including cosmic) radiation suffered by the human body in average conditions, results of experiments with animals, and observations on persons who - through ignorance, carelessness, or accidents - have taken large doses of radioisotopes into their bodies. Also, in research on the most hazardous nuclear species, it is sometimes possible to substitute for injection into volunteers less hazardous species that are isotopic with, and in the same chemical form as, those under study. In some instances this substitution allows sufficient margins of safety for these important determinations.

The total radiation man receives from cosmic rays and natural radioactivity is between 0.1 and 0.7 milliroentgens equivalent physical

(mrep) per day. Also, within the average man occur approximately 13,000 disintegrations per minute of radium and its daughter species; 150,000 disintegrations per minute of carbon (C^{14}) (21); and 470,000 disintegrations per minute of potassium (K^{40}). Thus, man receives and always has received appreciable quantities of radiation, both external and internal.

Relatively few determinations have yet been made on the radium (Ra^{226}) content of the bodies of persons believed never to have taken special doses of radium (Ra^{226}) - such as those prescribed for a time by physicians - in order to find the amount of radium (Ra^{226}) a person normally has in his body from natural sources. The mean content determinations by three independent investigators range in value from 0.12 to 7.5 millimicrograms per skeleton, or sixty-fold. The high values could be due to the persons' having drunk water from radium springs (20).

Like most other poisons, radioactive fission debris generally enters the body through the respiratory system and/or through the gastrointestinal tract. Theirring concludes (22) that about 0.1 millicurie of those fission products having half-lives averaging about one month, if retained in the body, would be likely eventually to produce death. Retention of inhaled particles within the respiratory system is highly dependent on particle size. Maximum retention appears to occur at a diameter of 2 microns (5) and "respirable size" dust is that having a diameter between 0.5 and 3 microns (23).*

Radioactivity from internal disintegrations is more hazardous than rays from the same number of disintegrations of the same nuclear

*Drinker and Hatch (103) note important retentions at less than 0.5 μ and a maximum size for retention of at least 5 μ.

species that are received from outside the body, because (a) the geometry for internal emitters is often nearly perfect for radiation absorption, and the geometry for external emitters is usually relatively poor; (b) the radioactive species are carried within the body until they decay or are eliminated, the rate of elimination for some of these nuclear species being very slow; and (c) the nuclear species are not distributed evenly throughout the body but can be concentrated, depending on their chemical states, in specific parts of the body. The precise distribution of the fallout nuclear species within the body would be difficult to measure. Iodine (I^{131}), for example, when in the iodide state goes to the thyroid gland; plutonium (Pu^{239}) and strontium (Sr^{89}, Sr-Y^{90})* are bone-seekers (as is radium [Ra^{226}], which is not produced in fission).

2. Genetic hazards of long-range fallout

When radiation that causes ionization or electron excitation passes through the reproductive cells (24), there is an acceleration in the mutation rate of the organism that is directly proportional to the dosage received no matter how low the dose. Within wide limits this change is independent of the radiation's wave length and time-intensity distribution (25). Dose for dose, heavy particles such as

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* Strontium (Sr-Y^{90}) stands for the fission-created nuclear species of mass number 90 of the element strontium and its product of beta ray decay, yttrium, also of mass number 90, which in turn decays by beta ray emission. This notation will be used in this thesis.

alpha rays usually have effects several times more damaging than those of electrons, for the more intense ionization along the paths of the heavier particles causes more frequent chromosome rearrangement. Photon-induced mutations are similar to those that occur spontaneously. Spontaneous mutations are believed to occur a thousand-fold too frequently to be caused by natural (including cosmic) radiation.

a. Genetic "tolerance". Evans (26) has estimated that a person receiving 250 roentgens (r) before reproducing with a non-irradiated mate will thereby increase by 1.5% the inherited anomalies in his children caused by recessive gene mutation. Making certain assumptions, he concludes that for dominant mutations induced into one parent by 250 r the anomalies would be increased by approximately 25% in the next generation. The difficulty with these calculations is that the data on which they are based have by no means been agreed upon by the specialists, even for Drosophila melanogaster, the species of fruit fly on which most of the experiments have been made. Relatively few measurements have been made on man. Evans takes the spontaneous mutation probability as 10^{-5} per gene per generation. Wright (27) states that no doubt probabilities of 10^{-6} or 10^{-7} are more common and that there can well be those that have values of 10^{-8} or 10^{-9}. Also, according to Muller (25), one of the primary needs in genetics is information of the relative frequency of mutations that are not lethal or characterized by readily visible effects; these are in the end as much or more damaging than the lethal mutations. All of the mutation probability calculations yet made

have been based only on anomalies that are readily distinguished. Depending upon the spontaneous mutation rate assumed, very different induced mutation probabilities can be calculated for a given amount of radiation received.

Genetic changes are cumulative, not only in individuals, but also through generations (1). Well over 99% of the mutations are harmful (20). The rate at which these mutations are eliminated from the population is in a large measure dependent upon the average grade of dominance of those mutations that are classed as recessive. More information on this phase is classed by Muller (25) as one of the primary necessities in genetics at present. Wright concludes (27) that the gaps in knowledge of genetics at present are so large that estimates of genetic radiation damage to man cannot be taken very seriously, but that it is quite possible that exposures of 300 r could well be genetically important and 30 r exposures not negligible. A reduction factor of 100 has been considered (20) as a tentative basis for the exposure of a population of 50,000,000. Calculated using the widely-accepted maximum permissible levels for beta and photon radiation, this factor would set the maximum permissible exposure for blood-forming organs at 0.3 r/100 = 3 milliroentgens per person per week, which is only about one to four times that received from natural causes.

b. Internal radiation. Little attention has been paid specifically to genetic effects from internal radiation. Perhaps the condition most likely to be harmful to reproduction is that in which the gonads are irradiated by bone-seeking nuclear species deposited in the

pelvic region. Despite its short maximum range in tissue (approximately one centimeter), the beta radiation from strontium ($Sr\text{-}Y^{90}$) has been suggested (20) as a possible chief offender.

In the study of genetics, one large field that is only in its beginning is the effect of radiation on the somatic (body) cells (20). For example, nothing is known of the effect produced on descendants conceived several years after the gonads have received a dosage of radiation. Muller notes the inference (25) that the genetic damage to the somatic cells is the source of most of the delayed and long-term injury to tissues and concludes that it is necessary to review all of the biological effects of radiation on this basis.

These gaps in present knowledge alone are justification for investigations by sanitary engineers of practical means of minimizing levels of radioactivity in water supplies.

3. Principal hazardous species in long-range fallout

Of the three radioactive materials - strontium ($Sr\text{-}Y^{90}$), plutonium (Pu^{239}), radium (Ra^{226}) - which have tolerance concentrations less than the provisional general standards for air and water outside an area controlled by the Atomic Energy Commission (14), one, strontium ($Sr\text{-}Y^{90}$), is present in a large proportion in long-range fallout and another, plutonium (Pu^{239}), can be present if it is used for fission material. A nominal (20,000-ton TNT equivalent) bomb has been considered in calculations by others (1, 28) to have a maximum of 100 kilograms of uranium (U^{235}) or plutonium (Pu^{239}) of which only one kilogram

undergoes fission. Ninety-nine kilograms of plutonium (Pu^{239}) have a disintegration rate of 5,720 curies, which is less than the disintegration rate of the debris from the one kilogram that undergoes fission even ten years after detonation. Uranium (U^{235}) has a long half-life and does not tend to concentrate in the more sensitive organs of the body. Its health hazard is comparatively negligible.

The nuclei of mass numbers 89 and 90 each form about 6% of the fragments of the uranium (U^{235}) or plutonium (Pu^{239}) atoms that undergo fission (29). The krypton (Kr^{90}) formed decays by beta ray emission through rubidium (Rb^{90}), strontium (Sr^{90}), and yttrium (Y^{90}) to stable zirconium (Zr^{90}) (30, 102):

$$Kr^{90} \xrightarrow[33\ sec.]{\beta^- 3.2\ Mev.} Rb^{90} \xrightarrow[2.74\ min.]{\beta^- 5.7\ Mev.} Sr^{90} \xrightarrow[19.9\ yr.]{\beta^- 0.54\ Mev.} Y^{90} \xrightarrow[61\ hr.]{\beta^- 2.18\ Mev.} Zr^{90} \qquad (2\text{-}1)$$

In this reaction, the values above the arrows are the maximum beta ray energies of disintegration in millions of electron volts and the values below the arrows are the half-lives of the species.

The decay scheme for bromine (Br^{89}) is:

$$Br^{89} \xrightarrow[4.51\ sec.]{\beta^-} Kr^{89} \xrightarrow[3.18\ min.]{\beta^- 4.0,\ 2\ Mev.} Rb^{89} \xrightarrow[15.4\ min.]{\beta^- 4.5\ Mev.} Sr^{89} \xrightarrow[53\ dy.]{\beta^- 1.48\ Mev.} Y^{89} \qquad (2\text{-}2)$$

Yttrium (Y^{89}) is stable.

If a nominal bomb (28) were made of plutonium (Pu^{239}), the ratio in curies of strontium (Sr^{90}) to unfissioned plutonium (Pu^{239}) soon after detonation would be approximately one; the ratio of strontium (Sr^{89})

to plutonium (Pu^{239}) would be approximately ten. Thus, on this basis, a nominal bomb's fission products are probably a more important hazard than the undetonated fission material.

For fallout material immediately following a bomb blast the Atomic Energy Commission has given the Civil Defense Administration (31, 32) an acceptable risk value of 0.09 microcuries per milliliter of drinking water for a ten-day consumption period. A more specific tolerance limit, given in a recent Atomic Energy Commission publication (6), is 5,000 micromicrocuries of three-day-old total fission product debris per milliliter of water. Water meeting this standard may be considered safe for drinking at any period of time.

The difference between the tolerance value given by the National Bureau of Standards and the ten-day emergency value given the Civil Defense Administration is probably based on considerations such as (a) the fact that newly-created fission debris contains, in terms of disintegration rates, a small proportion of those nuclear species that are most hazardous and (b) the assumption that these ten-day maximum periods of consumption will take place only rarely, in contrast to the values listed in the National Bureau of Standards publication, which are based on lifetime consumption.

4. Fission debris decay parameters and maximum permissible levels of radioactivity in drinking water

If the 5,000 micromicrocuries per milliliter limit at three days is used for calculation of the total radiation received in

drinking water, it can be shown that the decay parameter of the fission debris strongly affects the total exposure over a lifetime.

Neglecting biological elimination and assuming a lifetime of (70 years) (365 days) = 25,500 days,

$$5000 \int_{3}^{25,500} t^{-1.2} dt = c \int_{3}^{25,500} t^{-n} dt, \text{ where } c \text{ is the maximum}$$

permissible $\mu\mu c/ml$ at 3 days. (2-3)

Thus $$c = \frac{(n-1)\ 16,800}{\frac{1}{3^{n-1}} - \frac{1}{25,500^{n-1}}}$$

Example: Use $n = 1.5$. Therefore $$c = \frac{(0.5)\ (16,800)}{\frac{1}{3^{0.5}} - \frac{1}{25,500^{0.5}}} = 14,700 \ \frac{\mu\mu c}{ml}.$$

As can be seen in Equation (2-3), the value of c is strongly dependent on the value of n employed.

Much more study of the sanitary engineering aspects of the chemistry of several of those elements that have fission-produced radioactive species is necessary for accurate estimates of the hazards of fallout. Because of the wide range of maximum permissible levels for human consumption of these species, it is reasonable that first emphasis should be given to those most hazardous. Much study should be given to the history of the elements strontium, zirconium, iodine, cesium, barium, and promethium as they are turned free into nature, since these elements have especially hazardous radioisotopes that are produced in large quantities in nuclear fission (33).

5. Reason for low tolerance limits for certain nuclear species

Two radioisotopes of strontium (Sr^{89} and Sr-Y^{90}) have low tolerance values because strontium, which is in Group 1-A in the Periodic Table and acts chemically much like calcium, is deposited on and retained by the bone. Bone marrow, which produces red blood cells, is very sensitive to radiation. The radiation from each strontium (Sr^{89} and Sr-Y^{90}) isotope is powerful enough to cause a large amount of ionization. Also, some investigators claim that strontium is not uniformly deposited on the bone but is preferentially taken up by the epiphysial (end) part of long bones over the diaphysis (shaft part) (20), which means that the radiation on these more sensitive areas is concentrated and thus more likely to cause bone tumor. Both of the hazardous radioisotopes of strontium (Sr^{89} and Sr^{90}) have half-lives long enough to permit them easily to reach the bone after ingestion and still short enough to have high specific activities.* Strontium (Sr^{89}) is considered (32) the most hazardous species of newly created fission debris; whereas, strontium (Sr^{90}) is considered (35) probably to be the most hazardous when older fission products are ingested or the products from one detonation are ingested over a long period of time.

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* Isotope specific activity of a radioactive nuclear species may be defined as the total radioactivity of a gram of the species. The gram element specific activity is based on a gram of the element measured (34). Another definition of specific activity is the ratio of the number of radioactive atoms to the total number of atoms of the element in the sample (30).

6. Comparison of hazards of strontium (Sr^{89}, $Sr\text{-}Y^{90}$) with those of plutonium (Pu^{239})

It may be assumed that vegetation can be contaminated by long-range fallout, either by direct contact or by being grown in soil that contains fallout radioactivity. In experiments in which a large number of reactor-produced radioisotopes were used, only strontium was reported (35) as being carried by plants from their roots to their leaves. The ratio of leaf concentration to soil concentration was found to be more than 3,500 times as much for strontium as for plutonium.

Whereas the unfissioned 99 kilograms of plutonium (Pu^{239}) from a nominal bomb detonation have a disintegration rate of about 5,700 curies, the portion of this bomb that is fissioned will produce about 57,000 curies of strontium (Sr^{89}) and about 4,200 curies of strontium (Sr^{90}) as soon as the short half-lived precursors of these species die away. However, the estimation of relative contamination due to long-range fallout is difficult because of the uncertainty of the proportion which escapes fission. Any definite statement would indicate the efficiency of the bomb and is, therefore, not available in the unclassified literature (28, 32). A very early estimate gives a uranium (U^{235}) bomb a range of efficiencies of 1 to 5% and a range of sizes from 2 to 100 kilograms. If these efficiencies are taken for plutonium (Pu^{239}) fission and applied to the tolerance factors for drinking water, the estimated danger due to strontium ($Sr\text{-}Y^{90}$) will range from equality to six times as great as for plutonium (Pu^{239}).

7. Solubility of long-range fallout

Although fallout particles have been reported to be quite insoluble (4), the high passages of radioactive material through filtration plants reported in Chapter VII of this thesis indicate a fairly high effective solubility of the material. Most of the fission products, as well as uranium (U^{235}) and plutonium (Pu^{239}), are believed to be in the oxide form and, therefore, in most cases comparatively insoluble in the body fluids. As the acidity of the secretions into the normal human stomach is as high as pH 2.3, it is possible that much of the fallout material not soluble in surface waters would become soluble in the stomach and be able to pass through the stomach walls.

An interesting experiment was conducted in the Rochester, New York, area in 1951. Thirty-eight liters of rain collected June 3-4 were allowed to settle for eight days; the supernatant was evaporated to 730 milliliters in a glass container, then filtered through a Whatman # 40 paper; the filtrate was placed in a cellophane bag suspended in a cylinder containing 500 milliliters of distilled water so that the heights of water inside and outside the bag were equal. This was allowed to stand for five days with occasional stirring. Then the solutions inside and outside the bag were evaporated and counted on June 18, 1951, with a Geiger-Müller tube having an end-window of 2.3 milligrams per square centimeters.

The cellophane bag was tested for leaks by suspending it in distilled water as before and also adding a few drops of India ink to the water inside the bag. No migration of ink was detected. In the

same type of experiment, using a solution of potassium iodide (KI) in distilled water instead of rain, it was found that the potassium iodide reached equilibrium on each side of the membrane within three days. This result suggests that the settled, filtered rain-carried fallout in the cellophane bag was in ionic form. On June 4, the day of collection, four liters of the same rain were filtered through Whatman # 40 paper. The evaporated filtrate registered a net count rate of 49 counts per minute per liter and the ashed filter paper registered 44 counts per minute per liter. Results are summarized in Table 3.

Another experiment was conducted at Harvard on January 6 - 9, 1953. Five 100-milliliter samples of rain were run through Millipore filters (Type HA, Lot # 350604, plain).* The filtrate was evaporated directly onto planchets and counted. Unfiltered duplicates were run in each case and comparisons of count rates are listed in Table 4.

It is believed that the radioactive material of the first three samples came from the Eniwetok experiments and the last two from Nevada tests. If a large part of fallout radioactivity can pass through cellophane and Millipore filters, it is quite possible that it can go through the walls of the human stomach with ease.

8. Reported measurements of long-range fallout

By far the most extensive reported study on long-range fallout

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* Manufactured by the Lovell Chemical Company, 36 Pleasant Street, Watertown, Massachusetts.

TABLE 3

RESULTS OF DIALYSIS EXPERIMENT ON LONG-RANGE FALLOUT

	Weight	Count Rate	Count Standard Deviation	Time of Count	Background Count Rate
Inside Bag	0.120	283 cpm	5.4	14:30 hr.	25
Outside Bag	0.124	279 cpm	5.4	14:40 hr.	25

TABLE 4

PASSAGE OF RAIN-PRECIPITATED FALLOUT THROUGH MILLIPORE FILTERS

Date of Rain or Snow	Filtered		Unfiltered		Ratio of Net Count Rate, Filtered to Net Count Rate, Unfilt'd
	Net Count	Count Std.Dev.	Net Count	Count Std.Dev.	
Dec. 5, 1952	4.66	0.92	6.78	0.85	0.69
Nov. 22, 1952	25.70	1.49	39.40	1.50	0.65
Jan. 3, 1953	4.67	0.76	4.32	0.67	0.92
June 10, 1952 (settled, decanted)	-0.09	0.78	2.60	0.85	-
June 10, 1952 (unsettled)	1.93	0.78	5.25	0.85	0.37

has been made by the New York Operations Office of the Atomic Energy Commission. Their highest reading measured during the test series of the fall, 1951, and spring, 1952, was 10 - 20 milliroentgens (physical) per hour for depositions of seven to fifteen hours' age (5). These readings were made within the 200-500-mile annulus of the Nevada Proving Grounds.

Independent of this group, surprising levels of ionizing radiation due to Nevada detonation debris have been reported at Chicago, Illinois (36), and Troy, New York (37, 38). An average radiation concentration of 1.5 milliroentgens per hour was observed at the University of Chicago. When rain had collected in ground depressions, 15 milliroentgens per hour was measured. At Troy, New York, on April 26, 1953, an average radiation intensity of 0.75 milliroentgens per hour was reported with an intensity of 20 milliroentgens per hour near a roof drain.

The tolerance value for indefinite exposure to radiation is 100 milliroentgens per working day and 300 milliroentgens per week. On this basis alone, the measurements in the preceding paragraph appear dangerous. However, ionizing radiation from fission products, whether due to gamma rays or beta rays or both, follows the same die-away formulation as does the disintegration rate (1). This formulation states that $I_t = I_1 t^{-1.2}$, where I_1 is the dosage rate at unit time after detonation and I_t is the dosage rate at time t after detonation. The age of the fission debris measured at Troy was reported to be about fifty hours. Thus, if it is assumed that all the radioactive

material that was measured near the roof drain is fixed in place and that there is no separation of the radioactive constituents of the debris between detonation and deposition, the total radiation from the time of measurement is

$$20 \frac{\text{mr.}}{\text{hr.}} (50 \text{ hr.})^{1.2} \int_{50}^{\infty} \frac{dt}{t^{1.2}} = 5{,}000 \text{ milliroentgens} = 5 \text{ roentgens.}$$

For the first week after fallout, this radiation rate integrates to 1.3 roentgens. If it is assumed that the general radiation at the time of fallout is 0.75 milliroentgen per hour, the cumulated first week dose is $1300(0.75)/20 = 50$ milliroentgens. If the tolerance value of 300 milliroentgens per week (based on an indefinite exposure) is accepted, it is obvious that no danger from external radiation existed under these conditions.

One of the many factors in considering relative tolerance values of long-range fallout is that of the length of time fallout remains in an area once it is deposited on the ground or in a surface water. It is certain that the long-range fallout from a single detonation is not deposited uniformly over even a portion of the earth. The length of time that deposited long-range debris from any detonation remains in place could well be a major factor in the magnitude of its hazard.

B. EFFECTS OTHER THAN THOSE DIRECTLY ON HEALTH

1. Effect on the manufacture of film

It is interesting to note that the first discovery of radioactivity (by Becquerel in 1896) and the first reported discovery of long-range fallout (by Webb [2] in 1945) were both from the unintentional exposure of photographic film. On August 6, 1945, in the preparation of strawboard that was later used by the Eastman Kodak Company to pack film, a manufacturer in Vincennes, Indiana, used water from the Wabash River in the process. In this water was fallout from the "Trinity" detonation, which took place on July 16, 1945. It was found that this strawboard contained radioactivity that was intense enough to ruin X-ray film stored in it and sufficiently active to be used in experiments that were conducted two to four months after the detonation.

Owing to this contamination problem, it is necessary for film manufacturers to monitor the air, raw materials, containers, and water used in production. It is also necessary for the manufacturers of paper used for containing and packing film to monitor their process water and supplies. Often the paper stock is made at still another plant, where the materials must also be monitored for radioactivity.

2. Effect on the monitoring procedure at plants where large amounts of radioactive material are processed

In the careful measurements for radioactivity in the air, vegetation, soil, and surface waters near the large Atomic Energy Commission installations, such as at Hanford, Washington, (21) and

Brookhaven, New York, (39) valuable information has been accumulated on natural levels of radioactivity. This study of natural radioactivity is necessary so that the small amounts of artificial radioactivity turned loose in nature can be precisely determined. As long-range fallout can increase these readings far above natural levels, it can so obscure the routine measurements that, at least for a few days, detection of increases of some radioactive materials accidentally released on the site could be difficult.

3. Effects on the measurement of naturally-occurring radioactive nuclear species

The free neutrons that exist in the atmosphere are caused principally by cosmic rays. If these neutrons are captured by nitrogen (N^{14}), radioactive carbon (C^{14}) is produced (40). Also, tritium (H^3) is produced by cosmic radiation in the earth's atmosphere (41, 42, 43). This knowledge has led to the establishment of useful techniques for age determinations of certain samples, as well as tracer studies with these naturally-occurring nuclear species. As neutrons are released in nuclear fission (28) and in large numbers in fusion detonations, nuclear explosions interfere with carbon (C^{14}) determinations (44) and can be expected to interfere with tritium (H^3) measurements in nature.

4. Effects on cosmic ray measurements, uranium prospecting, and weather

Interference by long-range fallout on cosmic ray studies is discussed in Chapter VI.

There have been complaints from prospectors that fallout has interfered with their measurements in hunting for uranium. Presumably these complaints are due to increases in the non-sample count rate ("background") caused by fallout and are probably important for only a few days after each large fallout.

Complaints and discussions have taken place in the press, in the Congress of the United States, and even in popular publications (45) on the ability of long-range fission debris to cause tornadoes. A short discussion of tornadoes and long-range fallout will be given in Chapter VI.

CHAPTER III

PROCEDURES FOR COLLECTION, PREPARATION, AND MEASUREMENT OF SAMPLES

A. SAMPLE COLLECTION

1. Location of the collection stations

Figure 2 is a map of eastern Massachusetts showing the locations from which rain and surface water samples were taken on a routine basis for this thesis. Table 5 gives (46) the geographical location of the sampling points, which are on streams and reservoirs chosen to yield representative samples of eastern Massachusetts surface waters. Wherever practical, the sampling points were located near United States Geological Survey stream-gaging stations or sampling stations of the Massachusetts Department of Public Health. During the first year, samples were collected at the stations designated by Arabic numerals 1 through 14 and at the rain collection station on the Harvard campus in Cambridge and at Lawrence, Massachusetts. At the end of March, 1953, it was decided to add eight more collection stations to the group. These are designated by Roman numerals from I through III and from VI through X. Stations Number 13 and Number 14 have the alternate designations of Number V and Number

TABLE 5

LOCATION OF SAMPLING STATIONS OF THE HARVARD UNIVERSITY-U. S. ATOMIC ENERGY COMMISSION PROJECT 1952-1953 (45)

No.	Station	Latitude	Longitude	Area of Watershed Sq. Mi.
1	Charles River at Newton Falls	42° 19' 06"	71° 13' 43"	145.4
2	Framingham Reservoir #2, outlet	42° 16' 59"	71° 26' 44"	28.5
3	Framingham Reservoir #2, inlet	42° 15' 34"	71° 27' 09"	27.2
4	Blackstone River, Rts. 122a and 20	42° 12' 43"	71° 47' 14"	46.8
5	Salisbury Pond, Worcester	42° 16' 35"	71° 48' 07"	9.0
6	Quoboag River, Rt. 19	42° 14' 03"	72° 09' 47"	107.1
7	Ware River at Ware	42° 17' 05"	72° 12' 59"	150.5
8	Quabbin Reservoir, outfall	42° 17' 46"	72° 18' 26"	185.9
9	Quabbin Reservoir at Windsor Dam	42° 16' 50"	72° 20' 56"	185.9
10	Ware River at S. Barre	42° 22' 48"	72° 06' 53"	110.5
11	Stillwater River at W. Stirling	42° 26' 0"	71° 48' 40"	4.9
12	Wachusett Reservoir, outlet	42° 24' 33"	71° 41' 0"	107.7
I	Concord River at Riverside	42° 32' 05"	71° 17' 59"	
II	Assabet River above Concord	42° 28' 15"	71° 21' 47"	
III	Concord River at Concord	42° 28' 0"	71° 21' 23"	
V	Assabet River at Concord	42° 26' 27"	71° 25' 48"	118.4
IV	Sudbury River at Concord	42° 27' 34"	71° 22' 0"	134.4
VI	Assabet Brook at Maynard	42° 25' 22"	71° 28' 41"	
VII	Hog Brook and Trip Pond at Hudson	42° 23' 23"	71° 34' 50"	
VIII	Sudbury at Cordaville	42° 15' 58"	71° 31' 13"	
IX	Sudbury at Framingham Center	42° 18' 0"	71° 25' 39"	
X	Sudbury River at Route 20	42° 21' 47"	71° 23' 0"	
	Harvard Yard	42° 22' 41"	71° 07' 02"	
	Lawrence Experiment Station	42° 42' 25"	71° 08' 34"	

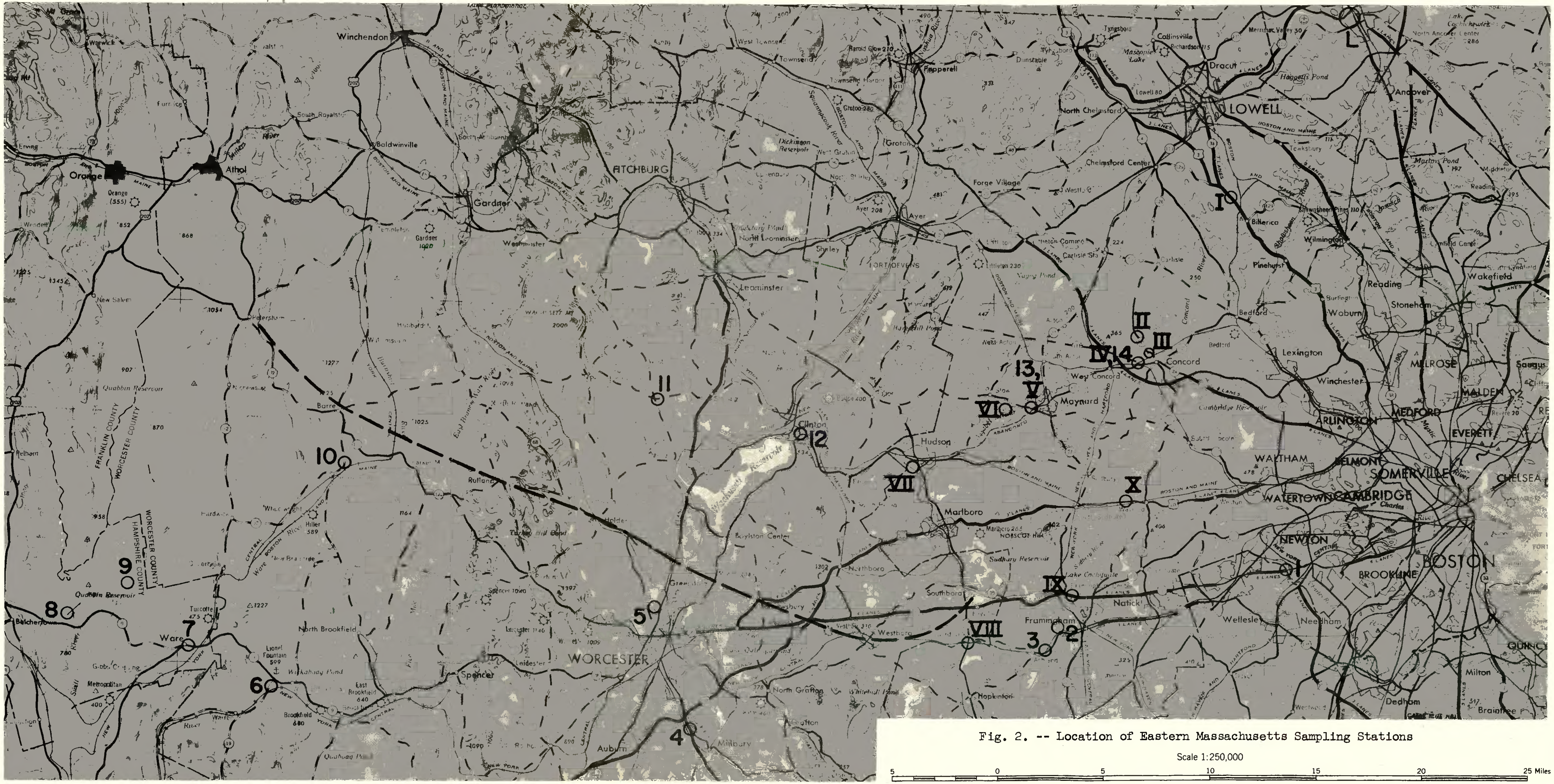

Fig. 2. -- Location of Eastern Massachusetts Sampling Stations

Scale 1:250,000

5 0 5 10 15 20 25 Miles

5 0 5 10 15 20 25 30 35 Kilometers

IV , respectively. It was possible to visit all of these stations by automobile and collect samples in about ten hours.

2. Rain collection apparatus

Beginning March 1, 1953, rain collection funnels (as shown in Figure 3) were placed at nineteen of the sample stations. The collectors were made of sheets of 30-inch by 32-inch by 26-gage galvanized iron. In the fabrication, a hole was bored in the center of the sheeting. It was then cut from the center to one side, lapped to form a cone, put together with sheet metal screws, attached to a small aluminum funnel by screws, and spray-painted on the inside with clear acrylic plastic and on the outside with black shellac. Because the collectors were unprotected, a stenciled sign was sprayed on them designating ownership. The collectors were seldom disturbed, with the exception of those located at stations Number 1 and Number 5, where populations are concentrated.

All precipitation collected for this project was in the form of integrated samples. That is, the rain collectors at the stations were in place continuously, and sample jugs that contained rain at the time the stations were visited for collection were replaced immediately by empty clean jugs to await the subsequent fallout. Usually there was rain between collections. Integrated precipitation samples at Harvard were taken during almost every precipitation. In a small number of cases the amount of rain that fell at a collector between the times that samples were gathered was enough to overflow the jug. No attempt was made to evaluate the error introduced thereby.

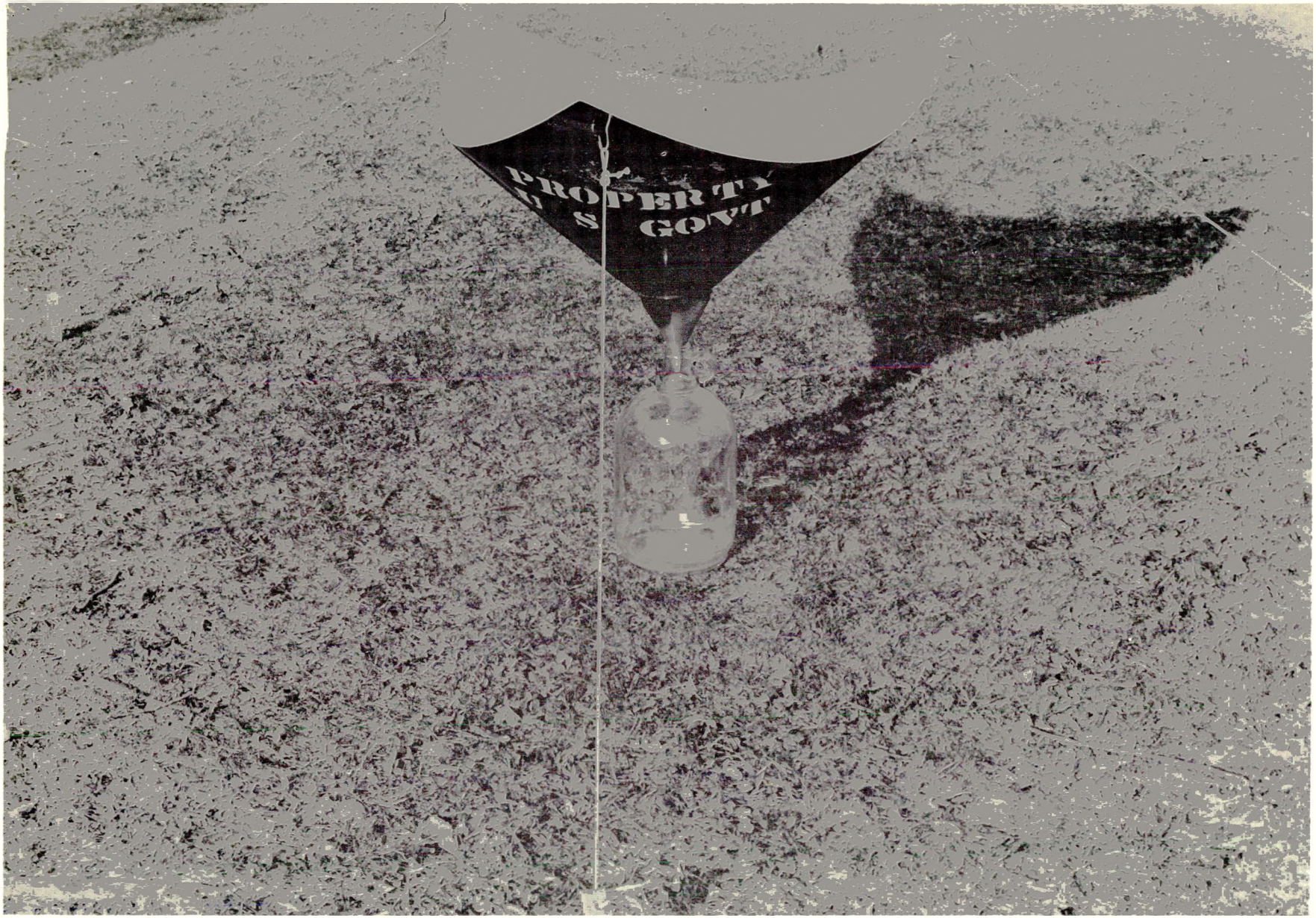

FIGURE 3 RAIN COLLECTION APPARATUS USED TO COLLECT SAMPLES IN EASTERN MASSACHUSSETTS FROM MARCH 1, 1953 TO DECEMBER 31, 1953

The main disadvantage in using these devices is that they cannot easily be adapted to collection of snow samples for fallout measurement. Mr. R. S. Kleinschmidt, who has worked on the Atomic Energy Commission project at Harvard, constructed three precipitation collectors in which heat is distributed to the collection surface from electric heaters by means of Freon in copper tubing. The electric element is regulated by a thermostat, so that the temperature of the collector surface and the storage chamber never falls below a degree or so above freezing. Mr. Kleinschmidt plans to publish a description of this device.

Other methods for the collection of fallout include the use of air filters and gummed paper. The latter has been used extensively by the New York Operations Office group of the Atomic Energy Commission (5). Although it is known that a large proportion of the long-range fallout particles are less than ten microns in size (6), it is claimed that even when these particles are brought to earth by rain they are effectively retained on the surfaces of exposed gummed sheets, which give results that can be reproduced. However, no data have been published to show the accuracy attained when this method is used to estimate the areal concentration of rain-precipitated fallout.

3. Sample containers

Soft lime glass jugs, usually one gallon in size, were used to collect samples of both rain and surface waters. An investigation was conducted to compare the absorption of long-range fallout by the walls

of glass jugs with its absorption by the walls of polyethylene jugs. Duplicate surface water samples in polyethylene and soft lime glass jugs were collected at Massachusetts stations Number 1 through Number 12 on February 14, 1953. The samples from each process were prepared and counted by the standard techniques. The mean count rate of those samples collected in glass jugs (counted twice on February 18 and twice on February 22) was 1.86 counts per minute per liter. The mean count rate of the duplicate samples collected in polyethylene bottles (counted twice on February 21 and twice February 24) was 2.08 counts per minute per liter. The mean of the differences in count rates between the two methods indicated that the samples collected in polyethylene bottles had a higher count rate by 0.23 counts per minute per liter with a standard deviation of 0.32 counts per minute per liter. The median of these differences indicated that the samples collected in glass jugs had a higher count rate by 0.15 counts per minute per liter. This disparity is due to a single sample of unusually high radioactivity. The net count rates of this experiment are too low to form a basis for firm conclusions as to the merits of polyethylene compared to glass collection jugs. However, the test does not indicate that there is a large difference between the materials in this respect. No comparisons of this type were made during periods of high fallout radioactivity.

4. Soil-sampling

The Department of Soil Mechanics of Harvard allowed the author

to use some of its soil-sampling devices, which were eight-inch sections of commercial two-inch steel pipe with one end beveled for driving into the ground. These were painted with lacquer to aid in the extraction of the soil from the tubes. This removal was accomplished with a special device that extracted the soil in as nearly an undisturbed state as practical. Soil samples at depths up to seven inches were collected from the eastern Massachusetts sampling stations by this technique. Planchets full of these samples were measured and the means of the net count rates obtained are given in Table 6.

B. SAMPLE PREPARATION

1. Evaporation procedure and apparatus

On a routine basis, one-liter of each of the samples of eastern Massachusetts surface waters was evaporated, transferred to a planchet, and automatically counted for gross beta radiation with a Geiger-Müller tube. One-liter volumes of rain were also evaporated and counted unless the count rate was high enough to justify testing smaller volumes. Samples measured at Lawrence were usually 100 milliliters in volume, those measured in the Rochester, New York, area usually four liters in volume. At Harvard, the one-liter samples were evaporated in porcelain dishes over steam baths in about ten hours. The samples were not allowed to boil while evaporating. A new rubber policeman and a small amount of distilled water were used carefully to transfer the solids from evaporation into a standard E-20 stainless steel planchet (1" diameter by 5/16" high) manufactured by Tracerlab, Incorporated. The solids

TABLE 6 49

GROSS BETA COUNT RATES OF SOIL SAMPLES FROM THE EASTERN MASSACHUSETTS COLLECTION STATIONS

Counts per minute per planchet-full, corrected by standard

Date Collected	Feb. 21, 1953 (AR)				Feb. 28, 1953 (AS)				Mar. 28, 1953 (AW)			
Dates Counted	Feb. 26-Mar. 14, 1953				Mar. 14-Mar. 22, 1953				Apr. 6 - Apr. 28, 1953			
	Mean Net CPM	No. of Samples	Mean Net CPM	No. of Samples	Mean Net CPM	No. of Samples	Mean Net CPM	No. of Samples	Mean Net CPM	No. of Samples	Mean Net CPM	No. of Samples
Depth (inches)	Bottom		Bank		Bottom		Bank		Bottom		Bank	
0-1/4					5.79	13	5.71	14	6.87	13	6.61	13
1/4-1/2	6.24	13	5.09	14	6.00	13	6.16	14	6.43	13	6.32	13
1/2-3/4					5.55	13	5.76	14	6.43	13	6.08	14
3/4-1					6.50	13	6.78	14	7.22	13	6.44	14
1-2	6.94	13	5.44	14	5.66	13	6.26	14	6.50	13	6.16	14
2-3	5.32	13	6.08	14	6.30	12	6.09	14	7.56	13	5.44	14
3-4	6.52	9	5.96	14	6.83	10	5.76	14	6.93	13	10.74	14
4-5	5.38	6	6.53	12	6.29	5	5.88	6	6.24	9	5.87	14
5-6	7.20	1	6.60	6	6.63	2	6.40	6	6.59	5	6.87	11
6-7			5.94	3					7.02	3	6.89	6
7-8			7.47	1								

TABLE 6--Continued

Date Collected	Apr. 13, 1953 (AY)				May 27, 1953 (BE)			
Dates Counted	Apr. 30-May 4, 1953				Jne. 20-Jly. 13, 1953			
	Bottom		Bank		Bottom		Bank	
Depth (inches)	Mean Net CPM	No. of Samples	Mean Net CPM	No. of Samples	Mean Net CPM	No. of Samples	Mean Net CPM	No. of Samples
0-1/4	6.34	13	10.8	14	7.09	12	9.45	14
1/4-1/2	7.16	13	6.74	14	5.81	12	8.55	14
1/2-3/4	7.27	13	7.92	14	5.99	12	6.84	14
3/4-1	6.17	13	6.26	14	7.63	12	6.34	14
1-2	7.05	13	7.59	14	7.98	12	5.77	14
2-3	6.39	12	6.62	14	6.50	12	7.15	14
3-4	5.70	10	7.29	14	8.05	8	6.84	14
4-5	5.43	6	6.70	12	8.94	5	6.03	13
5-6	6.74	3	6.19	7	7.55	1	7.55	7
6-7			6.28	3			7.31	1
7-8								

were then dried without boiling under a heat lamp. Some of the mineral acids and bases and a few organic solvents were tried as aids in removing the solids from the evaporating dishes, but none was found that did not cause the solids to become bulky when dried in the planchet. It usually was necessary for the dish to be scrubbed with the policeman and rinsed thoroughly three times for effectively complete transfer. Undoubtedly a portion of the mixed fission product ions present was preferentially absorbed by the glass jug, the porcelain evaporating dish, and the rubber policeman, depending on the chemical species present and their oxidation states. Important first steps have been made in the quantitative determination of the errors that are introduced by absorption such as in the sampling and measuring of radioactivity in water (47, 48, 49). Watson (47) found the following relative decontamination factors* for reactor-produced fission product mixtures in water: Polythene, 10,000; Teflon, 5,963; plate glass, 783; lead sheet, 501; and Number 347 mirror-finish stainless steel, 6. Unfortunately, as Polythene melts at 106° centigrade, its use is limited in the evaporation of samples. Tests should be made to investigate the practicality of using Teflon-coated metal evaporating dishes.

One series of tests was conducted after the large fallout of April 8, 1953, to determine the amount of recently created nuclear

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* "Decontamination factor" is the ratio of the amount of undesired radioactive material initially present to the amount remaining after a suitable processing step has been completed. Decontamination factors may refer to the reduction of some particular type of radiation or of the gross measurable radioactivity.

debris that was retained by the evaporating dish that could be removed by subsequent scrubbings and the amount retained by the rubber policeman. Table 7 summarizes the measurements obtained. It may be concluded that additional scrubbing would have removed little more of the recently-deposited fallout radioactivity from the evaporating dishes.

2. Automatic evaporation

As can be surmised from the previous discussion, there is a need for a method or device for the automatic evaporation of water samples containing low levels of radioactivity - ideally one that will evaporate samples directly onto the counting planchet. Also, more practical devices for the continuous monitoring for radioactivity in water supplies are needed. This need will be especially pressing in the event that reactors become a practical energy source with the resulting large increase in radioactive waste production.

Several hydraulically-fed automatic devices (50, 51) for the direct evaporation of water samples onto planchets were constructed and found both unreliable and too slow. Figure 4 shows one device that was built during research and found impractical. It worked on a principle similar to that commonly used for feeding water to chickens. A device that appears practical for the evaporation of small samples of liquid (52, 53) did not seem to be applicable to the rapid evaporation of the relatively large volumes necessary for the measurement of very low levels of radioactivity in water.

An investigation was therefore begun of a device that controlled the level of water in the container in which evaporation took place

TABLE 7

RETENTION OF LONG-RANGE FALLOUT FISSION DEBRIS BY A PORCELAIN EVAPORATING DISH AND A RUBBER POLICEMAN
(RAIN COLLECTED APRIL 8, 1953)

Items Measured for Count Rate	Count Rate (cpm/l) on April 10,1953	Relative Count Rate (Per Cent)
1 Liter Evaporated Rain Sample (First Treatment)[a]	1.86×10^5 (from an extrapolation of the sample decay curve)	100
Fallout Transferred From Same Dish (Second Treatment)	822.0	0.44
Fallout Transferred From Same Dish (Third Treatment)	37.6	0.02
Rubber Policeman Used in First Treatment[b]	20.1	0.011

a "Treatment" means three vigorous scrubbings and rinsings of the dish into the counting planchet. The transferred solids were then dried in the planchet (without boiling) under a heat lamp. This was the standard sample transfer technique for this thesis.

b The rubber policeman was sliced and placed in a standard planchet for counting.

FIGURE 4 AUTOMATIC SAMPLE EVAPORATION DEVICE CONTROLLED HYDRAULICALLY

(the evaporation dish or planchet), using a simple vacuum tube circuit. The evaporation was accomplished by a vertical stream of hot air impinging on the surface of the water and by heat supplied by a stove below the container. Although the device was never advanced to the point that, on a routine basis, it could replace the technique using an evaporating dish over a steam bath, the author believes it can be developed to do so. Figure 5 and Figure 6 show two versions of this device.

The electronic control circuit of this evaporator is shown in Figure 7. The principle of the device is the interruption of a slow leakage of current through the water in the evaporation dish. This interruption allows water to refill the dish until contact is again made permitting current leakage. As can be seen in the figure, through a 2.7 megohm resistor a 3 to 10 volt potential is placed by a grounded battery both on an electrode inserted in an evaporation dish and on the grids of a 6SL7 triode tube. The cathodes of the vacuum tube are also grounded. If the container in which evaporation takes place is not a good conductor, it can be replaced by an additional grounding electrode inserted into the water. The tube's cathode and anode have on them a potential of 150 to 200 volts and are in a circuit with a rather sensitive relay that is energized by a current flow of about three milliamperes. In turn, the contacts of the relay are in a circuit conducting alternating current at a potential of 115 volts through a solenoid which operates a simple clamp valve against a spring. Current flows through this circuit when the relay

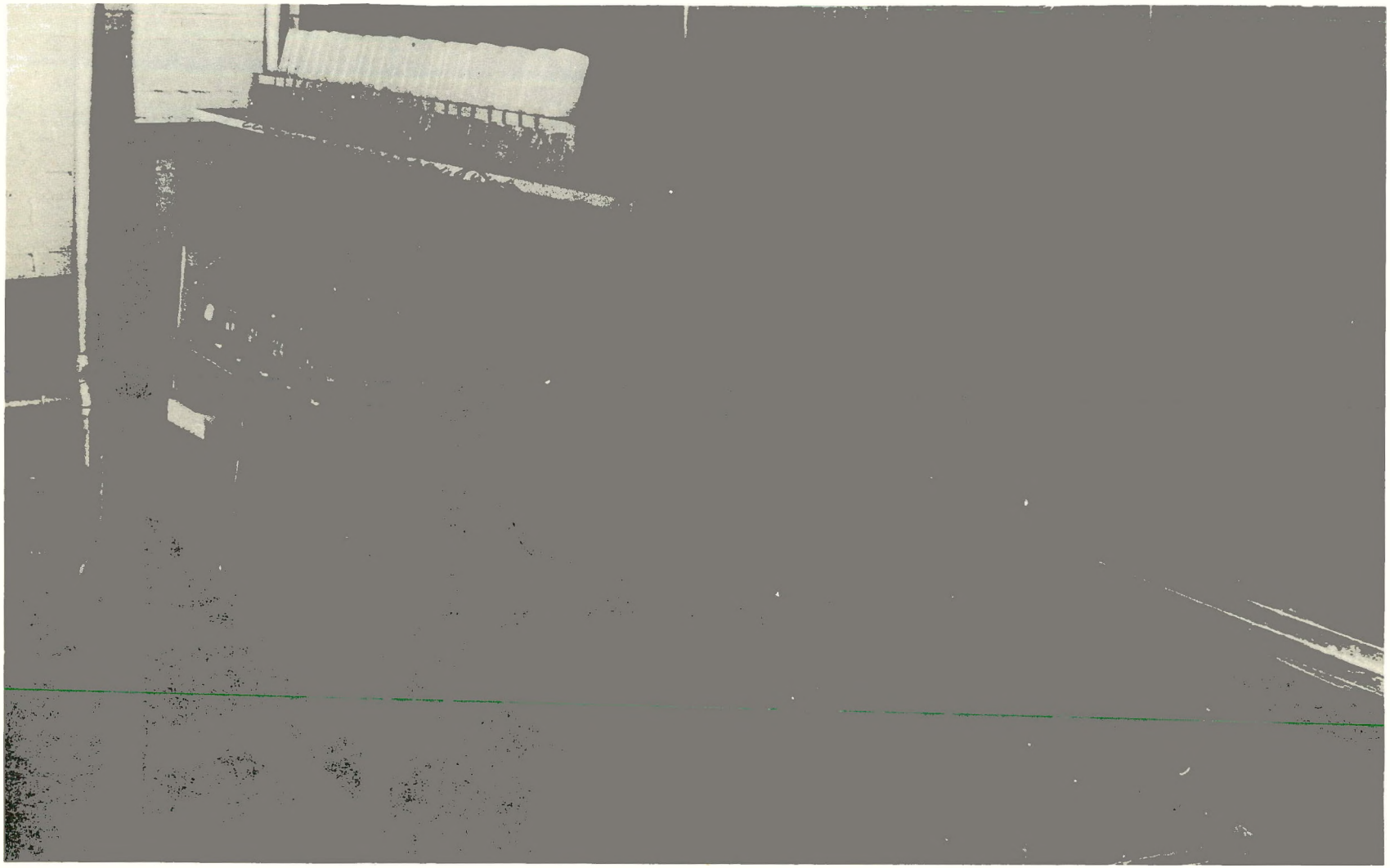

FIG. 5 DEVICE FOR THE EVAPORATION OF TWENTY-TWO WATER SAMPLES DIRECTLY ON TO PLANCHETS. CONTROLLED ELECTRONICALLY.

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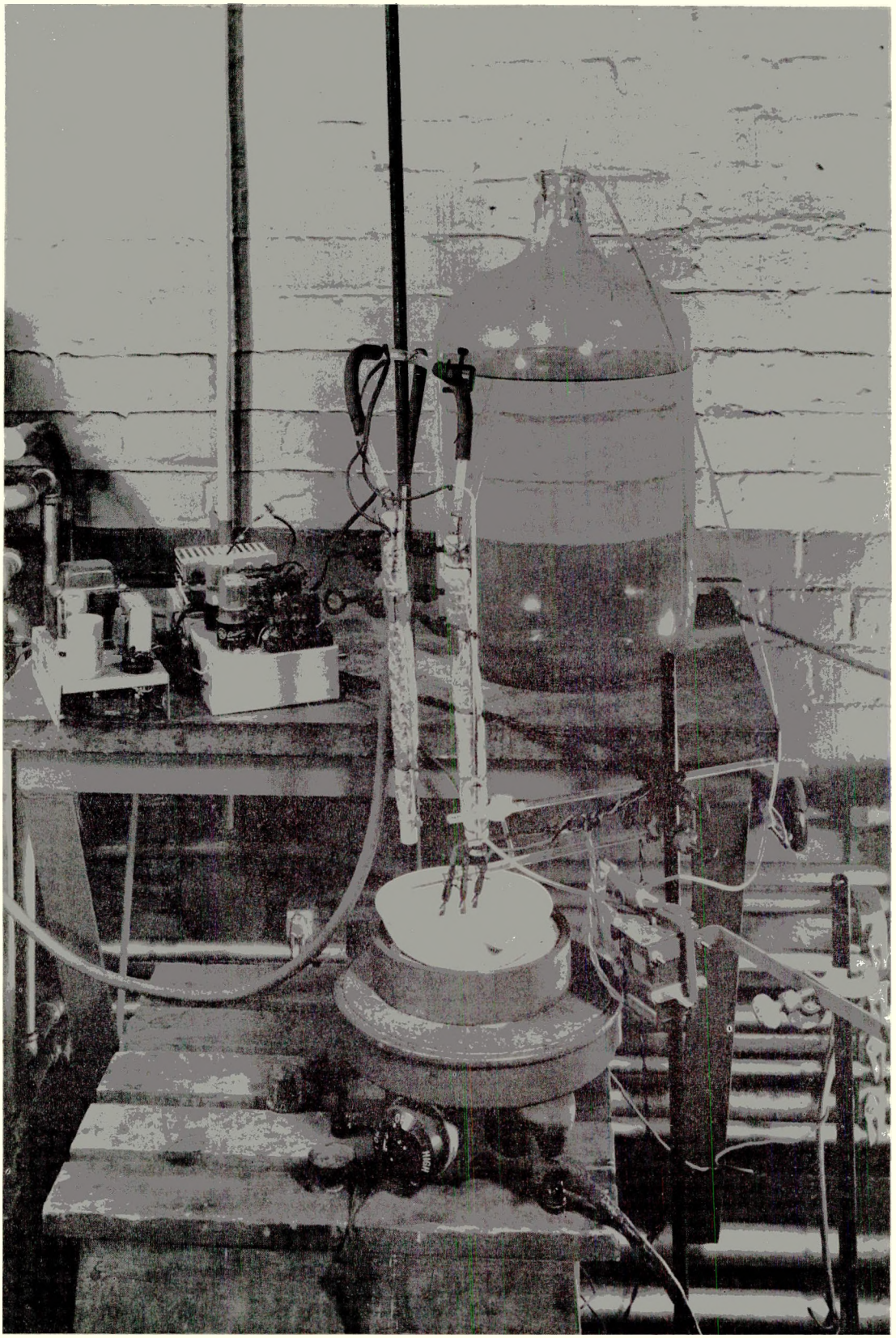

FIGURE 6 DEVICE FOR THE EVAPORATION OF WATER SAMPLES. CONTROLLED ELECTRONICALLY

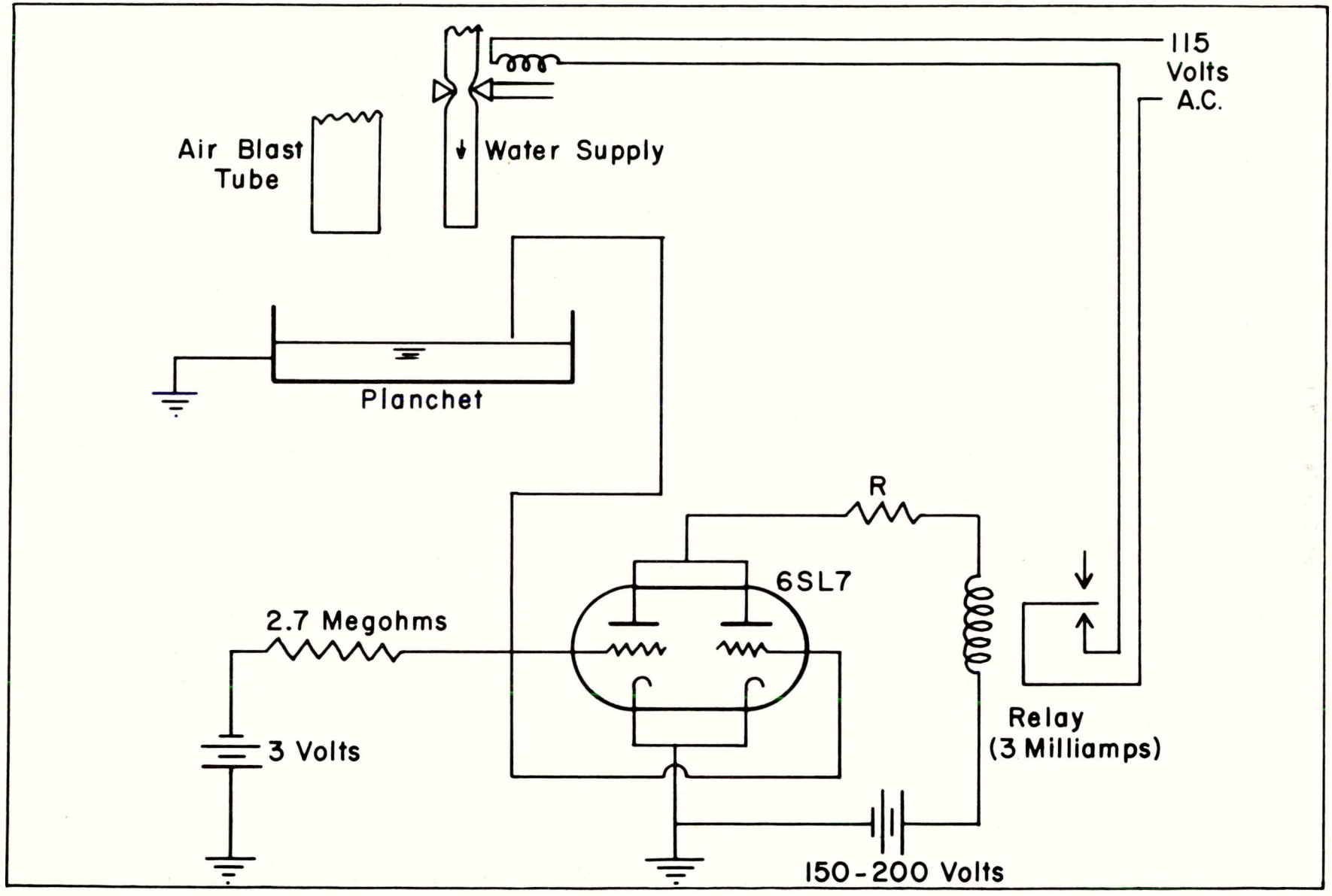

Fig. 7. — Schematic Diagram of the Liquid Level Control for the Automatic Evaporation of Samples.

is not activated. A polyethylene tube that feeds water to the evaporation dish passes from the reservoir through the solenoid-activated clamp valve.

When evaporation proceeds to the point at which the level in the planchet or dish drops below the end of the platinum electrode, the leakage of the current (about one microampere) from the battery through the water to ground is prohibited. This permits a potential to build up on the grids of the vacuum tube, reducing the flow of electrons from the cathodes to the anodes and consequently through the relay circuit. In turn, this allows the circuit containing the solenoid to close, thus opening the solenoid clamp valve against the spring and permitting the evaporation dish to be refilled with water to the level of the platinum electrode, when the circuit again automatically shuts off the flow.

The device is satisfactory for evaporation without boiling of water samples between approximately 10 and 100 milliliters in volume directly onto standard planchets (one inch in diameter by 5/16 inch in height). For smaller volumes, evaporation under a heat lamp would probably prove more convenient. For larger volumes, the time required for evaporation by this device directly onto planchets is too long. In the experiments for this thesis, the highest rate at which samples were evaporated directly onto planchets was one liter in eleven hours. The air was preheated to 260° centigrade and it was obvious in this test that the system was strained under these conditions. The measured count rate per liter by this technique was slightly lower than that obtained when the water bath and porcelain evaporating dish were used.

Of course, in this automatic evaporation technique, the sample would be ruined if the planchet overflowed or was allowed to run dry.

Another automatic machine was built (Figure 5) incorporating the same electronic circuit. It evaporated twenty-two samples directly onto planchets simultaneously. The air preheating arrangement located inside a piece of thick-walled Transite pipe with Transite ends consisted of a Nichrome wire radiator, which could produce up to six kilowatts of heat. The preheated air was distributed through stainless steel tubes to blow directly onto the surface of the water being evaporated in each of the planchets. A single electronic circuit of the type described above was used in conjunction with a stepping switch and a timing device, so that about every five seconds the circuit was switched from one planchet to the next. When the level of the water in the planchet dropped below the end of the platinum electrode, the circuit allowed it to be refilled before passing to the next planchet. In this apparatus, no heat was supplied beneath the planchet, and the evaporation rate for each sample was only about thirty milliliters per hour, so that it was useful only for sample volumes between ten and one hundred milliliters.

The same type of circuit was used in constructing the device shown in Figure 6 in which the evaporation took place in a porcelain dish six inches in diameter which was heated from beneath by an electric stove. A vertical blast of air heated to 260° centigrade impinged on the surface of the evaporating water. The evaporation rate by this method was about five hundred milliliters per hour.

The photograph of the automatic evaporator (Figure 6) shows a liquid-level control that actually consists of two of the above described circuits in parallel, so that if one circuit failed, the other would take control before the dish overflowed. This precaution did not prove necessary. The tube that delivered the water to the evaporating dish was made of polyethylene, which, with proper placement, delivered the water without being melted by the hot air blast. It has recently been found that this plastic can be treated by intense nuclear radiation to be made more heat-resistant and, therefore, will possibly have wide usage in the evaporation of water samples for the measurement of their radioactive components.

Although experimentation was not carried further, the next reasonable step would have been to increase the supply of heat beneath the dish and to the airstream. At the evaporation rate of 500 milliliters per hour, the temperature of the water was only 84° centigrade. A small thermocouple control could be submerged in the water in the porcelain evaporating dish, so that the temperature of the water could be maintained automatically just below the boiling point. It is quite possible that the evaporation rate could be greatly increased if the temperature of the water were kept near the boiling point (54).

C. SAMPLE MEASUREMENT

1. Radioactivity measuring instruments

The instruments used for the measurement of radioactivity in this thesis are listed in Table 8. Practically all measurements were made with automatic sample-changing equipment manufactured by Tracerlab, Incorporated.

TABLE 8

RADIOACTIVITY MEASUREMENT INSTRUMENTS USED FOR THIS THESIS

Instrument	Number Used	Serial Numbers
SC-6C Automatic Sample Changer (62)	2	315 and 135
SC-1B and SC-1C Autoscaler (62)	2	460 and 753
SC-5A and SC-5C Tracergraph (62)	2	220 and 346
SC-2A "64" Scaler (62)[a]	1	1-103
503 D Anticoincidence Analyzer[b]	1	101

[a] This instrument is manufactured by Tracerlab, Incorporated, of Boston, Massachusetts, and was used for only a few measurements.

[b] This instrument is manufactured by the Atomic Instruments Company of Cambridge, Massachusetts, and was used to a small extent.

Mr. R. S. Kleinschmidt designed a special six-inch steel shield and built special anti-coincidence Geiger-Müller tubes for use with the anti-coincidence analyzer. This arrangement is described briefly in a paper by Thomas, et al. (55). Tracerlab TGC-2 Geiger-Müller tubes

(which have a mica end-window of about two milligrams per square centimeter) were used for practically all of the measurements taken at Harvard and Lawrence, Massachusetts, for this thesis. When tubes of this type were placed in a lead shield two inches thick (as for the automatic measurements), a background count rate of about twenty counts per minute was recorded. When one of the tubes was placed in the steel shield and surrounded by the anti-coincidence tubes, it registered a background count rate of about five counts per minute on the Atomic Instruments Anticoincidence scaler. Mr. Kleinschmidt found that about one count per minute was due to contamination of the TGC-2 tube, presumably from radioactive potassium(K^{40}) in the glass and mica window. A special tube was made by Tracerlab, Incorporated, quite similar to the TGC-2 except that the walls were made of brass instead of glass. Unfortunately, this did not reduce the background count rate appreciably. Mr. Kleinschmidt also constructed a one-inch mercury shield to go between the counter and anti-coincidence tubes, but this modification also did not reduce the background count rate.

2. Future development of other devices for sample preparation, sample measurement, and monitoring

In the routine counting of large numbers of samples, it would be of considerable advantage to have an instrument that combines anti-coincidence tubes and circuits with automatic sample changers. As an additional improvement, the elimination of the loss caused by the absorption of radiation from the sample passing through the air gap and

mica window of the end-window Geiger-Müller tube could be effected by using a gas flow, internal Geiger-Müller counter in combination with the anti-coincidence and automatic sample-changing instruments. These complications in instrumentation could prove to be well worthwhile, especially in routine monitoring near large installations of the Atomic Energy Commission, such as nuclear power reactors. If nuclear reactors become an economical source of power, it is quite possible that thousands of samples annually of water, soil, and air will have to be measured around reactor sites all over the world. In this event, it seems reasonable that there will also have to be frequent or continuous measurement of radioactivity of the surface waters treated by municipal plants.

An evaporator that could be useful for the concentration of large volumes of water containing low levels of radioactivity is the Kleinschmidt distillation device (56), which is used on submarines and many other ships of the United States Navy. The shipboard model, powered by an electric motor, is designed to distill sea water at the rate of 1,000 gallons per day; the land model, powered by a gasoline engine, is designed to distill fresh water at the rate of 10,000 gallons per day. Thermodynamically, the units are far more efficient than other evaporators. The removal of the residue of evaporation (the material that would be used for radiochemical analysis) from the apparatus is a somewhat time-consuming routine procedure. The tubing for these stills is made of cupronickel, and the adsorptive properties of this tubing would have to be considered in any particular application for

the concentration of samples containing low levels of radioactivity. Only an insignificant amount of fission-produced radioactivity would be lost in the water vapor in proper evaporation.

One of the problems that the sanitary engineer probably will face is that of continuous monitoring of radioactivity in the water entering a treatment plant. Some commercially available ion exchange resins are able to undergo a large number of sorptions and desorptions and still retain their exchange properties. It may be worthwhile to investigate the practicality of using a continuous belt (or belts), holding cationic and (or) anionic resins, that would pass through the water being monitored, then pass a radioactivity measuring device, and finally go through a desorption liquid before again re-entering the water being monitored. It is possible that low levels of ionized radioactive material in water could be detected in this manner. If this device proved to be practical, the belts could be mass-produced and discarded when necessary.

3. Counting procedure

No attempt will be made to review the statistical theory of counting. Several references (30, 57, 58, 59, 60, 61, 62) give most of the fundamental mathematics required for the counting techniques used in this thesis.

Figure 8 is a copy of a typical calculation page. The procedure was to place the samples on the automatic scaling apparatus (listed in Table 8), so that each of the planchets containing the residue from the

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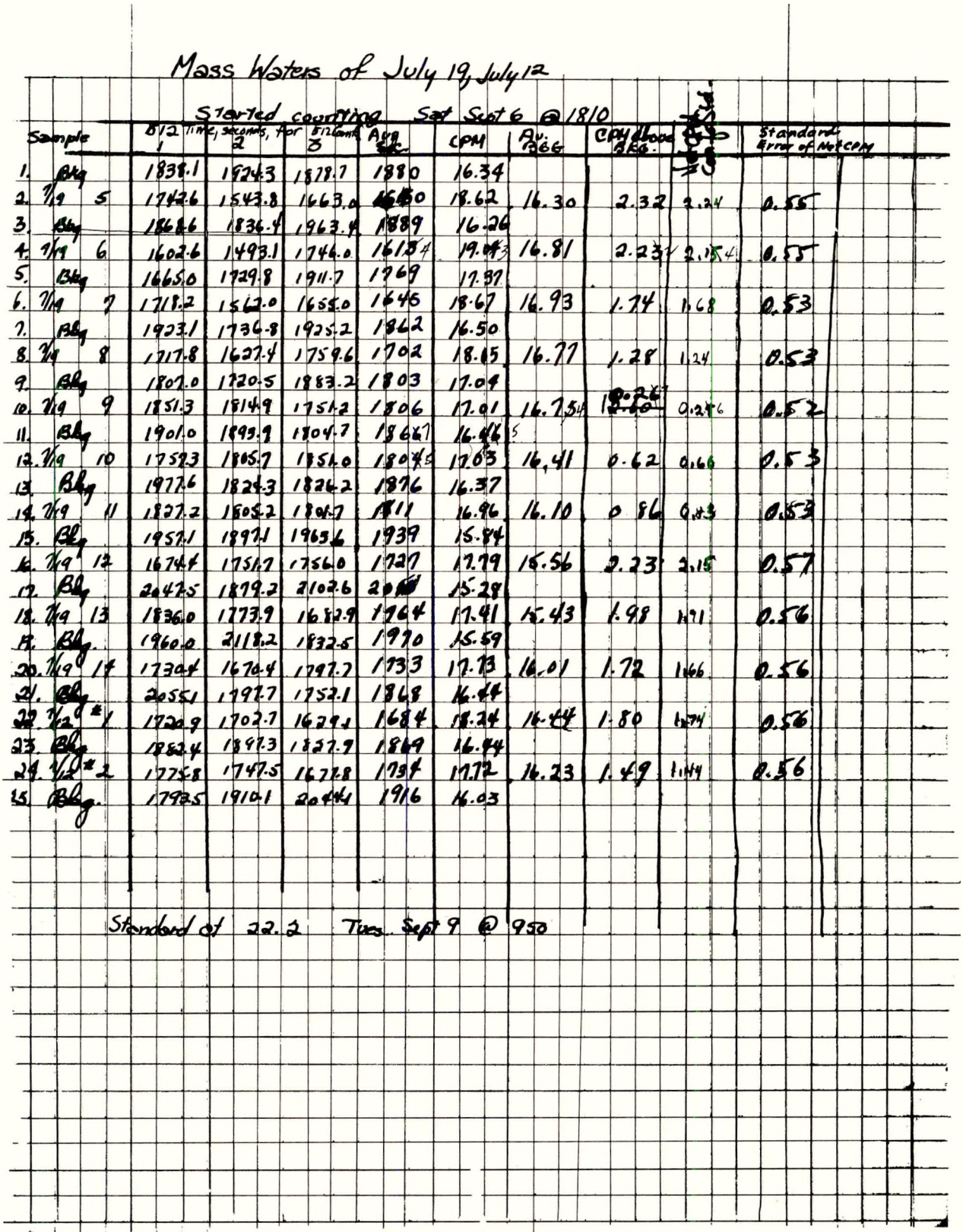

Mass Waters of July 19, July 12

Started counting Sat Sept 6 @ 1810

Sample	B12 Time, seconds, for Blank 1	2	3	Avg	CPM	Av. Beg	CPM above Beg.		Standard Error of Net CPM
1. Blg	1838.1	1924.3	1878.7	1880	16.34				
2. 7/19 5	1742.6	1543.8	1663.0	1650	18.62	16.30	2.32	2.24	0.55
3. Blg	1866.6	1836.4	1963.4	1889	16.26				
4. 7/19 6	1602.6	1493.1	1746.0	1613.4	19.08	16.81	2.27	2.18	0.55
5. Blg	1665.0	1729.8	1911.7	1769	17.37				
6. 7/19 7	1718.2	1562.0	1655.0	1646	18.67	16.93	1.74	1.68	0.53
7. Blg	1923.1	1736.8	1925.2	1862	16.50				
8. 7/19 8	1717.8	1627.4	1759.6	1702	18.65	16.77	1.28	1.24	0.53
9. Blg	1807.0	1720.5	1883.2	1803	17.09				
10. 7/19 9	1851.3	1814.9	1751.2	1806	17.01	16.75	0.26	0.26	0.52
11. Blg	1901.0	1899.9	1804.7	1868	16.46				
12. 7/19 10	1752.3	1805.7	1854.0	1804	17.03	16.41	0.62	0.66	0.53
13. Blg	1977.6	1824.3	1826.2	1876	16.37				
14. 7/19 11	1827.2	1805.2	1804.7	1811	16.96	16.10	0.86	0.83	0.53
15. Blg	1957.1	1897.1	1963.6	1939	15.84				
16. 7/19 12	1674.4	1751.7	1756.0	1727	17.79	15.56	2.23	2.15	0.57
17. Blg	2042.5	1879.2	2102.6	2008	15.28				
18. 7/19 13	1836.0	1773.9	1682.9	1764	17.41	15.43	1.98	1.91	0.56
19. Blg	1960.0	2118.2	1832.5	1970	15.59				
20. 7/19 14	1730.4	1670.4	1797.7	1733	17.73	16.01	1.72	1.66	0.56
21. Blg	2055.1	1797.7	1752.1	1868	16.44				
22. 7/12 #1	1720.9	1702.7	1629.1	1684	18.24	16.44	1.80	1.74	0.56
23. Blg	1862.4	1897.3	1827.9	1869	16.44				
24. 7/12 #2	1775.8	1747.5	1677.8	1734	17.72	16.23	1.49	1.44	0.56
25. Blg.	1793.5	1910.1	2044	1916	16.03				

Standard of 22.2 Time Sept 9 @ 950

FIGURE 8

TYPICAL CALCULATION PAGE SHOWING THE PROCEDURE OF COMPUTING COUNT RATES FOR THIS THESIS

evaporation of a sample of water was separated in its counting order from the next by an empty planchet. It is not possible to measure the radioactivity of samples without measuring "background" radioactivity at the same time. This background measurement is principally due to cosmic radiation and the ubiquitous naturally-occurring radioactivity.

The counting equipment used is designed so that it automatically changes samples and records the time required for a cumulation of counts reaching any preset binary number between 2 and 2^{12}. With low level samples of variable radioactivity, such as most of those measured for this thesis, a machine that could count to a preset time would give more economical use of counting time than one, such as used for this thesis, that measures the time required for the counts to cumulate to a preset number. However, the complexity of instrumentation for the former technique makes its use impractical. A reasonable number - 512 counts - was selected as that cumulated by the machine before it automatically recorded the elapsed time and inserted the next planchet for counting. A cumulation to only 256 counts would make necessary a computation procedure almost twice as long as with 512 counts. A cumulation to 1,024 counts would allow only one counting of each of the planchets every twenty-four hours, because of the low count rates usually encountered in most measurements for this thesis, thereby decreasing the chance of detecting errors due to spurious counts. When the radioactivity of the samples is sufficiently high, two or more sample planchets separate the background planchets in counting order on the automatic machines, as must be done for optimum use of the total counting time.

4. Measurement of background radioactivity

Many investigators who use instruments for which the samples must be changed by hand find it convenient to measure count rates only with samples in the machine during the day and to leave the instrument recording background overnight. A statistical comparison was made between this method and that of alternating samples with empty background planchets in the machine, as was done for this thesis. Data recorded by one of the automatic instruments on two occasions were recalculated as if they had been taken from an instrument that recorded the count rates on successive samples during the usual work day and the cumulated count rate due to background during the night. Two statistical comparisons were made, and the dates of measurement for these were chosen to coincide with reported severe geomagnetic storms (63) on the assumption that these storms would be likely to make the background count rate more variable. A period of low fallout radioactivity, that shortly before the November, 1952, Eniwetok experiments, was chosen for these calculations. The dates in this period during which severe geomagnetic storms occurred are September 7 and October 3. Both sets of data were recorded by the older automatic counting device.

Those background count rate values recorded after 17:00 hours and before 8:00 hours during each of the counting periods were averaged and called "night background". On each occasion this average value was subtracted from each of the measured count rates of the samples with their accompanying backgrounds, whether counted during the day or night. The net count rates of the repeated measurements of the same sample

were then averaged. Twelve samples alternating in counting order between thirteen empty "background" planchets were measured, each twice to 512 counts on October 2 and each three times to 512 counts on September 7. The individual net count rates calculated by this method were subtracted from those computed from the same data by the procedure usually followed for this thesis. (The computation procedure usually followed for this thesis is shown in Figure 8.) Each of these individual differences was then divided by the net count rates as usually calculated. Table 9 summarizes the results.

TABLE 9

EFFECTS OF BACKGROUND MEASUREMENT TECHNIQUES ON NET COUNT RATES
(A Comparison of the Alternate Measurement of Background and Sample with the Measurement of Samples during the Day and Background at Night)

Date of Samples	Median Difference in Net Count Rates between Alternate and Overnight Background Measurement Technique	Range of Values in the Groups of Differences	Median of Ratios (Difference in Net Count Rates) ÷ (Net Count Rate Based on Alternate Background Measurement Technique)
Sept. 7, 1953	-0.20 cpm	1.51 cpm	-11%
Oct. 2, 1953	-0.39 cpm	3.42 cpm	-26%

It would be expected that the differences between the two methods of measurement would be greater than shown in Table 9, since in this computation sample measurements recorded at night were included in the calculation simulating work-day sample measurement and overnight back-

ground measurement. If this table indicates the true situation, then one can conclude that using this type of instrument during magnetic disturbances (in a measuring procedure in which background is counted only at night) introduces a significant bias when the net count rate of the sample is less than about five counts per minute. In locations where various interferences cause the background count rate to fluctuate more radically, especially where interfering machines and apparatuses are run during the work-day only, this bias easily could be increased. For sample count rates larger than about 25% of the background count rate the difference between the two methods of counting probably is relatively unimportant. However, the main reason for measurement at this low level in work on the sanitary engineering aspects of radioactive debris and waste is to detect increases in the concentration of radioactivity in water, either by measuring gross count rates or isolated radioactive species, as early as possible so that corrective or preventive steps can be taken.

5. Fluctuation in counting sensitivity of the instruments

Corrections for daily fluctuations in the sensitivity of the measuring instruments were made by taking frequent (usually daily) count rates on a standard Tracerlab R-6 uranium X (U^{238}+Th^{234}+Pa^{234}) reference source. Figure 9 shows the daily fluctuations in these count rates of the number of seconds required for each of the automatic measuring devices to register 4,096 counts. All calculated net count rates, including those of calibration (Chapter V), were corrected by

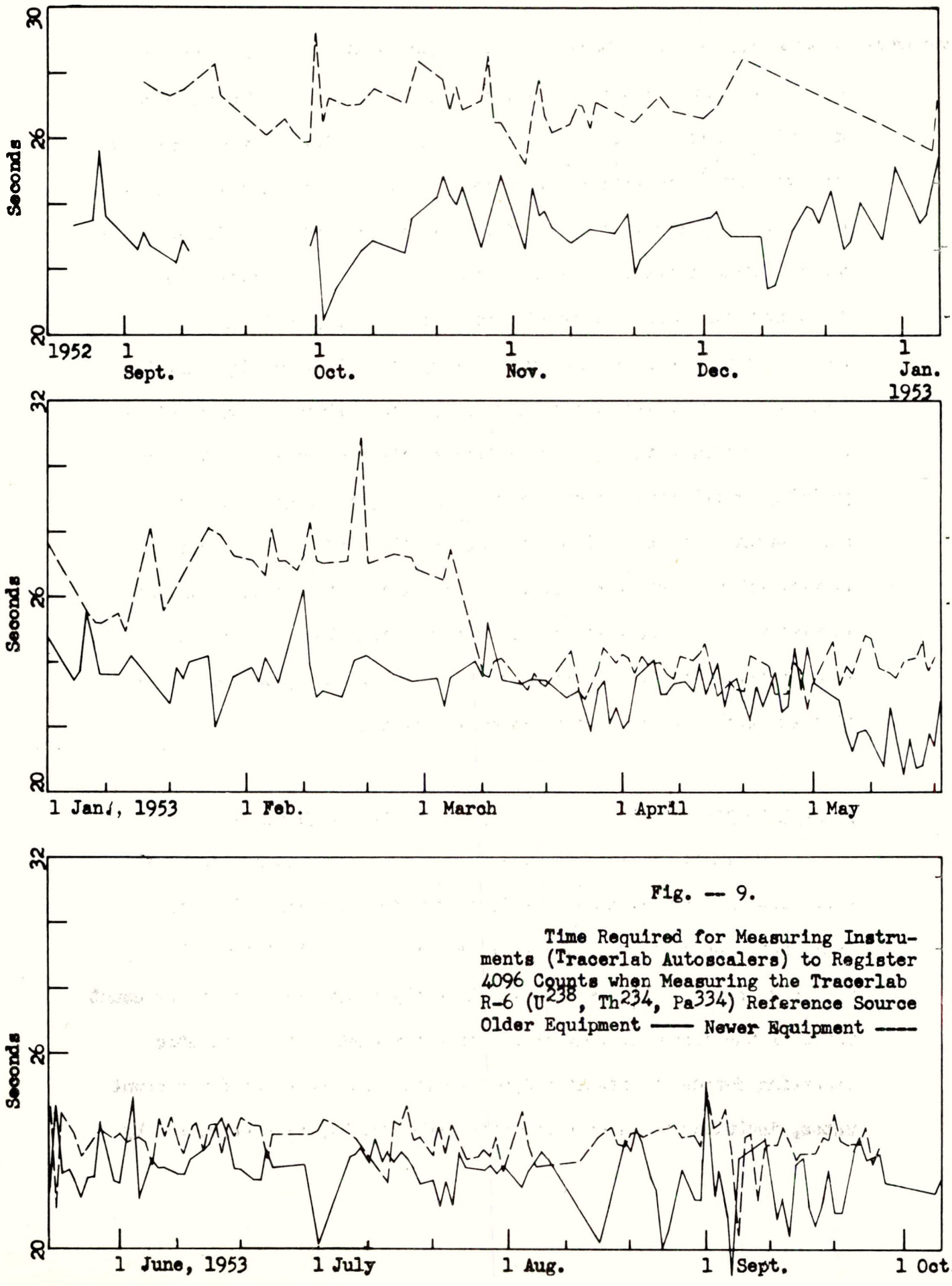

Fig. — 9.

Time Required for Measuring Instruments (Tracerlab Autoscalers) to Register 4096 Counts when Measuring the Tracerlab R-6 (U^{238}, Th^{234}, Pa^{334}) Reference Source
Older Equipment —— Newer Equipment ----

a factor that brought the count time for the R-6 standard arbitrarily to twenty-three seconds for 4,096 counts, which is approximately the average of the times required for this standard and these instruments. The R-6 standard (64) produces beta rays more energetic - 95% with a maximum of 2.3 million electron volts (Mev.), 5% with a maximum of 1.5 Mev.- than those from fission debris (1), which have an average maximum energy of 1.5 Mev., overall mean energy of 0.4 Mev; therefore, the R-6 standard is not ideal for use with fission debris. Of the samples of calibrated radioactivity now issued by the National Bureau of Standards, the thallium (Tl^{204}) standard, when placed as the evaporated water samples in a planchet, would apparently make the most satisfactory basis for comparisons.

6. Effect of power line surges on low level count rates

The effect of power line surges on very low net count rates was checked by a series of measurements that lasted a total of ninety hours. Measurements were made on the Atomic Instruments anti-coincidence counter, which was used to take some of the sample count rates for this thesis and is listed in Table 8. With this instrument, background count rates were measured consecutively for thirty-minute periods with (a) a "Sola" sine wave corrected constant-voltage transformer, (b) a "Raytheon" constant-voltage transformer that was not designed for sine wave correction, and (c) no power line stabilization other than that which was built into the anti-coincidence counter. If normality of the distributions from which the measurements were taken is assumed and if the differences

between successive readings are compared by the "Student" t-test (65) over the period, it can be stated that at a probability level of approximately 85% each of these transformers decreases the spurious count rates caused by line surges. It was not possible to demonstrate the advantage of one transformer over the other by this test.

7. Aids to calculation of net count rates

As an aid in the calculations of the total count rates from the values of the number of seconds required to cumulate to the preset 512 counts, conversion tables were made to change the seconds cumulated to counts per minute for each second of time over the range from 10 to 2,000 seconds. Later, when the timing device for the automatic recorder was changed for mechanical reasons to record in minutes instead of seconds, a similar conversion table was computed on this basis for each 0.01 minute between 0.01 and 34 minutes.

Figure 10 gives curves that can be used in a graphical solution of the standard error calculation for a sample with its adjacent background measurements when each is measured to a cumulation of 512 counts. The chart is used by locating the net sample count rate on the abcissa, going vertically to the curve representing the average background count rate, then going horizontally to the proper scale on the left chosen for the number of times the sample planchet with its adjacent empty background planchets was measured to 512 counts. The standard error is read directly.

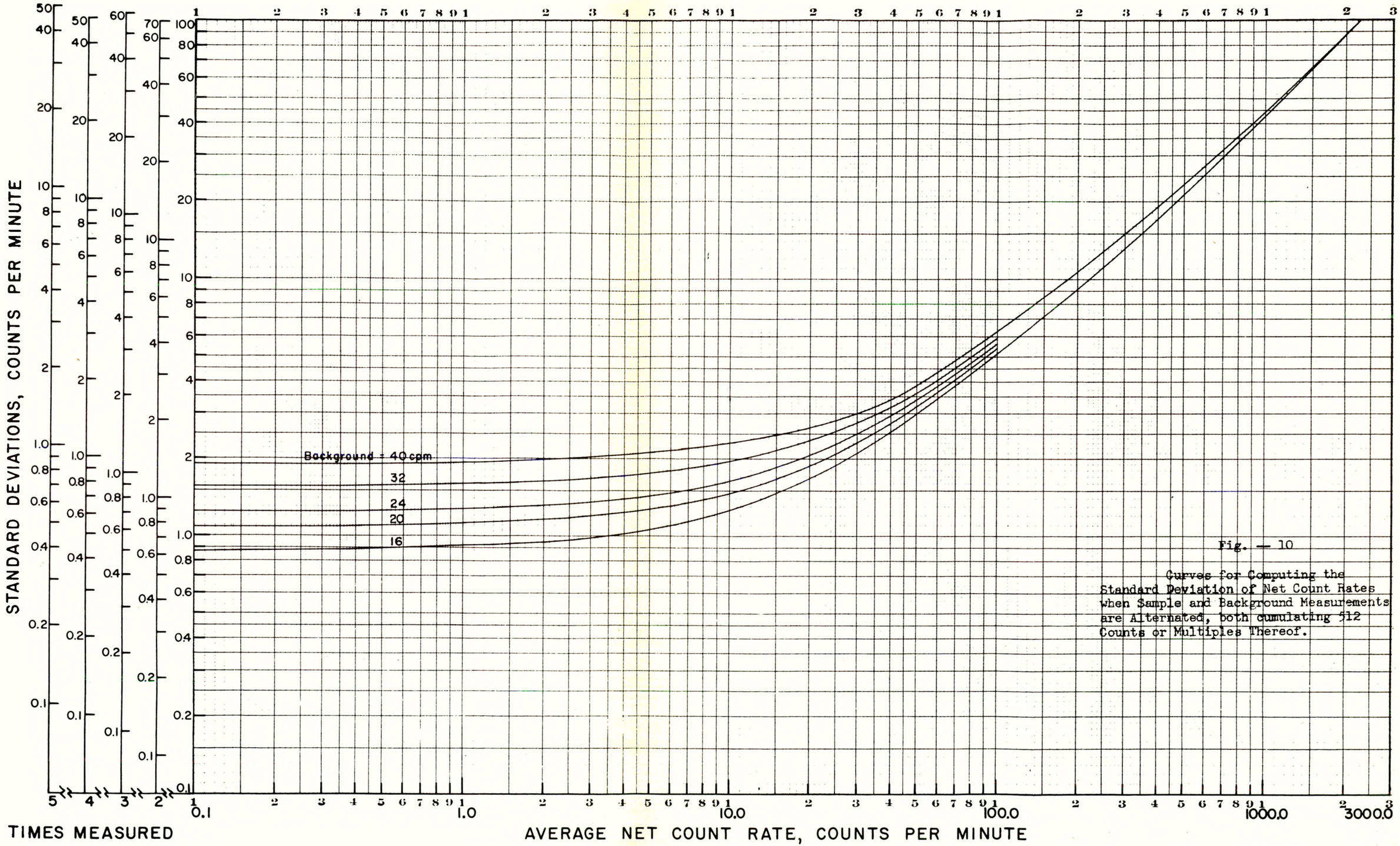

Fig. — 10

Curves for Computing the Standard Deviation of Net Count Rates when Sample and Background Measurements are Alternated, both cumulating 512 Counts or Multiples Thereof.

CHAPTER IV

NATURAL RADIOACTIVITY

A. NECESSITY OF MEASURING NATURAL RADIOACTIVITY

The problem of the measurement of natural radioactivity is complex, the components and energies of this radiation varying widely. In the measurement for this thesis of gross beta radiation at very low levels from fission debris in air, water, and soil, it was usually necessary to consider and make allowances for the count rates caused by extra-sample radiation (background), as well as those caused by radiation from the naturally radioactive elements in the samples. There have been many articles published on natural radioactivity and radiation that are of interest to the sanitary engineer (18, 21, 39, 66, 67, 68, 69, 70). The principal naturally-occurring elements that cause count rates in water, soil, and air samples are uranium and its daughter products, thorium and its daughter products, and potassium.

Until the establishment of centers by the Atomic Energy Commission in various parts of the United States for the production and handling of huge quantities of radioactive materials, there was little incentive for measuring the radioactivity of streams and reservoirs. When research on the disposal of nuclear wastes in streams was begun, it was found that a knowledge of the natural radioactivity present was

essential for the measurement of very low levels of artificial radioactivity. Before this time, a large portion of the measurements of natural radioactivity in water had been made on springs for the purpose of advertising and selling the waters produced for their supposed therapeutic value. For example, the list of remedies of the American Medical Association as reported in 1922 (69, 71) stated that no radium solution for internal use was acceptable if the dosage was less than two microcuries per day. It is fortunate that no spring in the United States (72) has been found to contain a sufficient concentration of radium to allow this dosage in drinking water. Other surface waters have been found to contain far less radium than the most radioactive springs.

B. PREVIOUS MEASUREMENTS IN EASTERN MASSACHUSETTS

The information on natural radioactivity of surface and ground waters in Massachusetts is not very extensive. Almost forty years ago, measurements of the radioactivity of Massachusetts waters were made for radium content by using an electroscope, but these values appear to be unreliable. In 1916, Dr. S. C. Brooks of Harvard (73) measured the radioactivity in the water from fourteen wells in the Boston, Massachusetts, area and reported a range of 0.057 to 0.223 microcuries per liter of initial radioactivity. These values appear to be much too high. He took similar measurements on water from the surface of Fresh Pond, from near the bottom of Fresh Pond, and from a tap on the Cambridge distribution system. None showed significant radium by the method used.

These measurements were made ten years before the National Bureau of Standards started calibrating radioactive samples.

Of forty-six samples of water from various parts of the world (principally from the United States), the radioactivities of which were reported by Skinner (69) in 1922, a sample from a warm spring in Massachusetts gave the second highest electroscope reading. This sample gave a reading equivalent to 0.188×10^{-9} grams of radium (Ra^{226}) per liter at collection and was described by Skinner as "temporary", which presumably means that it was principally due to radon (Em^{222}) rather than the parent radium (Ra^{226}). The sample of eastern Massachusetts water having the highest content of "permanent" radioactivity was that from a spring which registered 2×10^{-12} grams of radium (Ra^{226}) per liter reported in the form of radium salt. Calculations based on the preparation and counting techniques used in measuring the surface water samples for the present Atomic Energy Commission-Harvard project show that 2×10^{-12} grams of radium would give approximately sixteen beta-ray-emitting disintegrations per minute, or about three counts per minute per liter, which is probably more than ten times higher than the average estimated uranium daughter radioactivity in eastern Massachusetts streams and reservoirs. This estimate is based on measurements remote in time from previous nuclear detonations.

C. RECENT MEASUREMENTS IN EASTERN MASSACHUSETTS

The National Bureau of Standards, in cooperation with the Massachusetts Institute of Technology and the Carnegie Institution of

Washington, has made a precise measurement of the radium (Ra^{226}) content of granite taken from a quarry near North Chelmsford, Massachusetts, and the bureau sells the pulverized granite as a standard (No. 789). The radium (Ra^{226}) content is given as $2.96 \times 10^{-6} \pm 0.08$ grams of radium (Ra^{226}) per gram of rock. The indicated variation is the probable error and includes the variation caused by the heterogeneity of the sample. The sample contains natural radioactivity other than that of the radium, its precursors and daughters. The geochemical analysis of the samples shows the granite has a potassium content of 5.55% (reported as K_2O). The naturally-occurring radioactive potassium (K^{40}), therefore, produces about ninety-four beta rays per minute per gram of granite and must be the dominant radioactivity measured with a Geiger-Müller tube having a thin end-window.

For this thesis, an estimate of the self-absorption of the radiation of this granite was obtained by analyzing the measured net count rates of samples filling the standard planchets to various depths. As the self-absorption should follow a formulation like that of the first-stage biochemical oxygen demand, a rapid method of determining the self-absorption equation's parameters was used (74) and the plot for Chelmsford granite is shown in Figure 11. It should be noted that this formulation also takes into account changes in geometry caused by differences in thicknesses of samples. Series of self-absorption measurements on material of constant specific activity were also run on a sample of mixed earth and evaporated ocean water and a sample of surface soil. Neither of these samples should have had a large proportion of long-range fallout radioactivity. Figure 12 and Figure 13 and Table 10 summarize these measurements.

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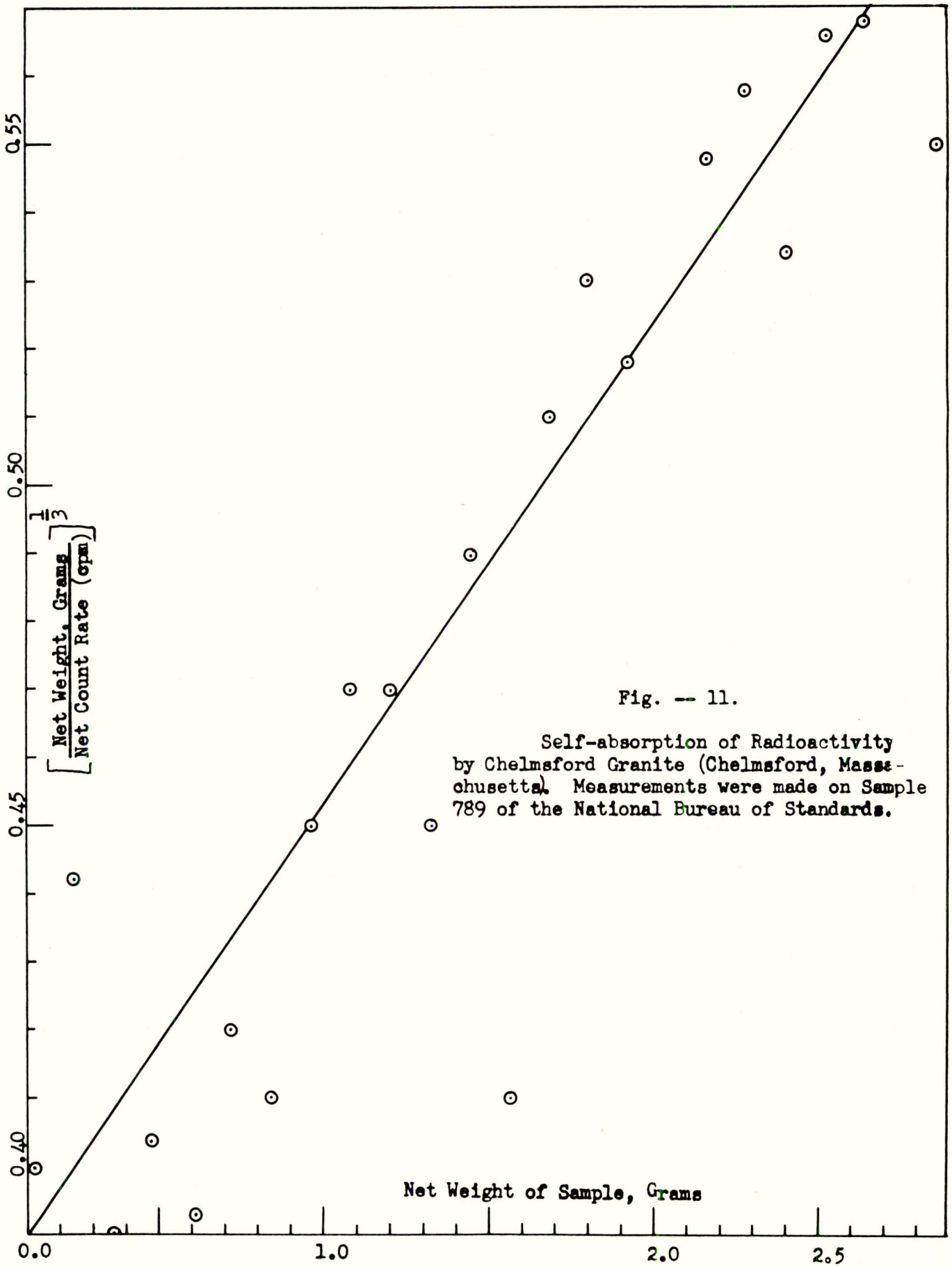

Fig. -- 11.

Self-absorption of Radioactivity by Chelmsford Granite (Chelmsford, Massachusetts). Measurements were made on Sample 789 of the National Bureau of Standards.

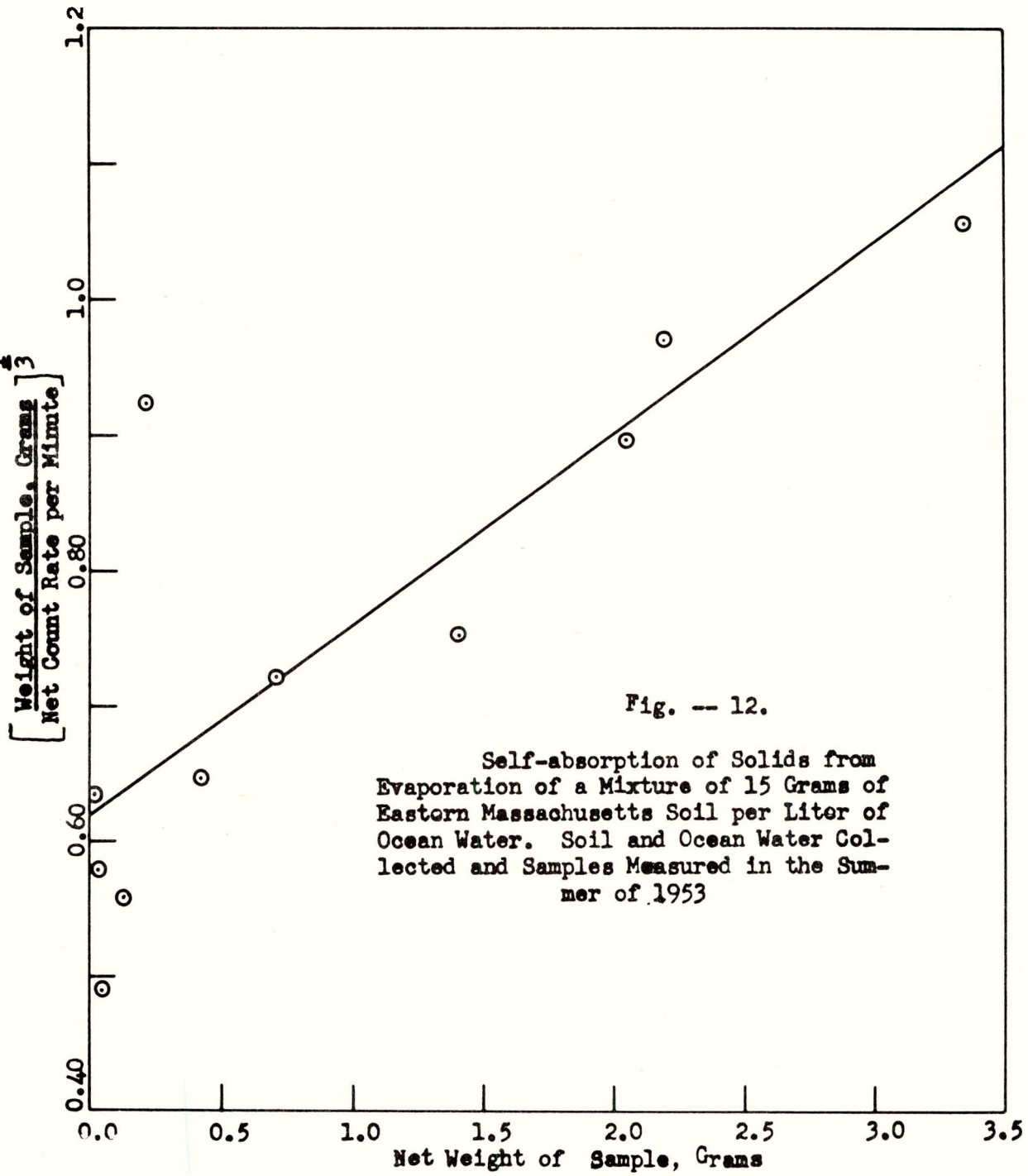

Fig. -- 12.

Self-absorption of Solids from Evaporation of a Mixture of 15 Grams of Eastern Massachusetts Soil per Liter of Ocean Water. Soil and Ocean Water Collected and Samples Measured in the Summer of 1953

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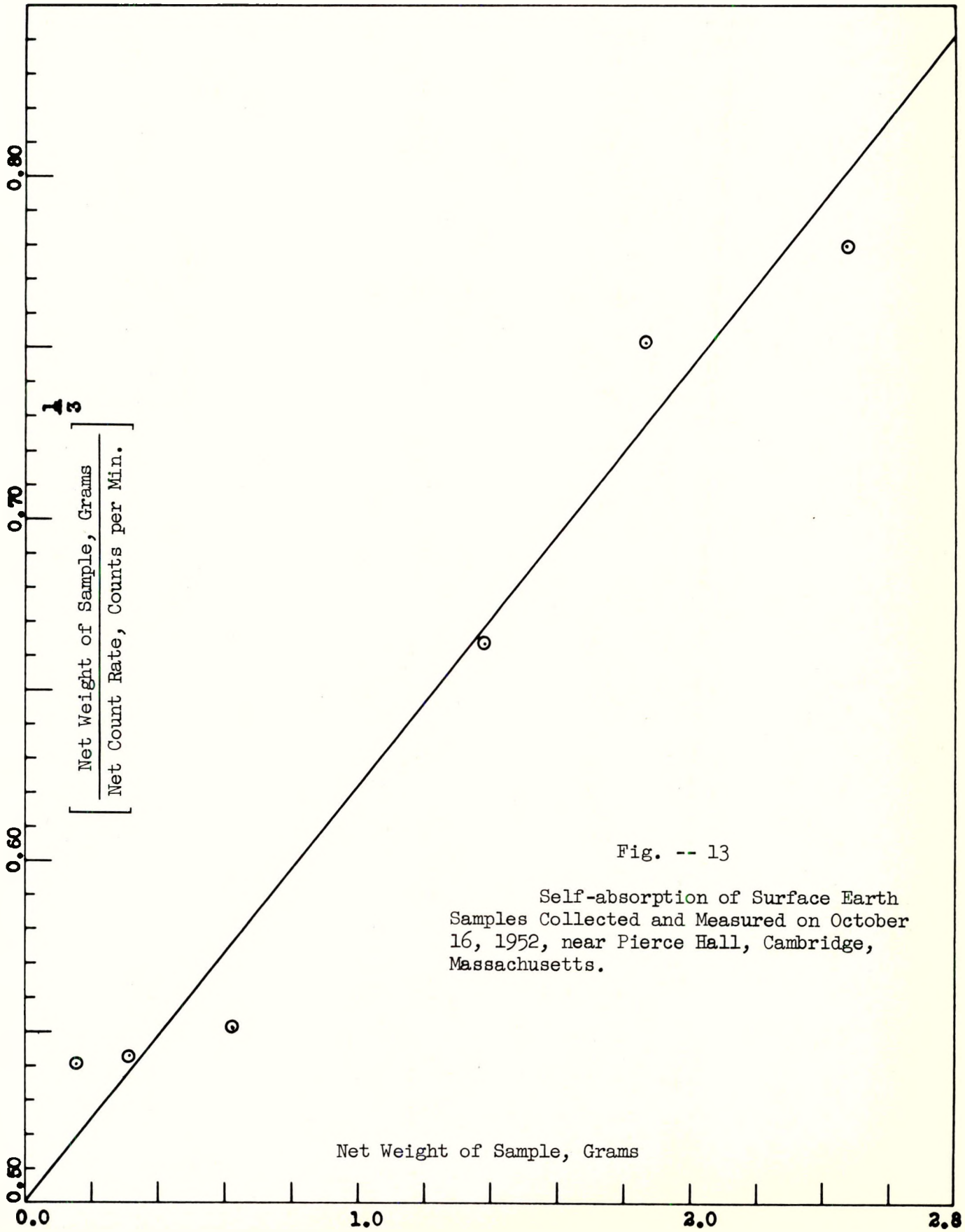

Fig. -- 13

Self-absorption of Surface Earth Samples Collected and Measured on October 16, 1952, near Pierce Hall, Cambridge, Massachusetts.

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TABLE 10

SELF-ABSORPTION FORMULATIONS FOR VARIOUS NATURALLY RADIOACTIVE MATERIALS FOUND IN EASTERN MASSACHUSETTS *

Material	Location	Self-absorption Formulation
Chelmsford Granite	North Chelmsford, Mass. (U.S. Bureau of Standards)	$cpm = 16.7\ (1 - e^{-1.13w})$
Solids from Evaporation of Clean Ocean Water	Crane's Beach, Mass. (Summer, 1953)	$cpm = 3.06\ (1 - e^{-1.36w})$
Surface Soil	Harvard Yard, Cambridge, Mass. (Collected Oct. 16, 1952)	$cpm = 5.46\ (1 - e^{-1.13w})$

* The formulation gives the net count rate registered on the automatic scaling equipment for various net weights, "w", in grams of the samples. The number to the left of the parenthesis is the calculated beta ray count rate that would be measured if the sample were infinitely thick. Standard steel planchets one inch in diameter and 5/16 inches high were used.

D. NATURAL RADIOACTIVITY OF EASTERN MASSACHUSETTS SURFACE WATERS

No chemical separations were attempted to determine the relative amounts of natural radioactivity in the samples collected for this thesis. As no detonation was announced between June and November, 1952, and since the samples collected from August 1 to November 1, 1952, were the lowest in measured radioactivity during the entire collection period, the mean level of radioactivity of the residue (from evaporation of 156 one-liter surface water samples taken from the fourteen principal eastern Massachusetts sampling stations) was found to be only 1.16 counts per minute per

liter at first count on the automatic equipment. This count rate is presumed to be due principally to natural radioactivity. Also, the average count rate of 315 soil samples collected from sampling Stations Number 1 through Number 14 just before the large fallout of the spring, 1953, series was only 5.93 counts per minute per planchetful (Table 6). Healey's values (21) for natural radioactivity, the mid-ranges of which are about three times as high as found in this research for eastern Massachusetts, are probably based on water and soil samples collected near Hanford, Washington. The igneous rock deposits of the Rocky Mountains could easily cause the radioactivity of the Hanford samples to be three times as high as those collected in eastern Massachusetts.

Between July 6 and September 21, 1951, Mr. B. L. Rosenthal of the Lawrence Experiment Station, using equipment like that used for this thesis (Tracerlab Autoscalers, TGC-2 Geiger-Müller tubes, and E-20 planchets), made measurements of the net count rates on the evaporated residues of 100-milliliter samples of tap water collected in eighty-eight eastern Massachusetts communities. These samples gave a mean net count rate of 1.78 counts per minute per liter. This mean value is higher than the average count rate of the eastern Massachusetts surface waters between August 1 and November 1, 1952. This difference is probably due to a part of the tap samples being taken from well supplies, which have more opportunity to pick up radioactivity from the ground.

The period between August 16 and October 4, 1952, was chosen for a test by analysis of variance to compare the relative count rates

of the samples from the various streams on successive collections (Table 11). The previous announced detonation had occurred early in June, 1952, and this period had the lowest fallout radioactivity between April, 1952, and January, 1954. From this test, it can be concluded that during this period the radioactivities observed at the various stations differ from each other at the 99% level of significance. This fact indicates that the observed count rates, although averaging only 5% above the background count rate, were not statistically homogeneous between stations and between collection dates.

The method of Tukey (75) was used to show that the two extreme mean net count rates (0.27 counts per minute per liter at Station Number 9 and 2.00 counts per minute per liter at Station Number 7) alone were significantly different from the means of the other twelve stations and thus can be considered derived from different statistical populations. This difference is reasonable in as much as Station Number 9 is on Quabbin Reservoir. The higher ranking values are on streams below factories. Certain industrial wastes could contain small quantities of radioactivity, presumably of natural origin, able to cause this difference. However, it is more likely that the watersheds drained by streams having higher count rates are of geological formations containing more naturally radioactive material, which was picked up by the streams.

TABLE 11

DATA FOR F-TEST ON NET COUNT RATES OF EASTERN MASSACHUSETTS SURFACE WATERS, 1952

Counts Per Minute Per Liter

Date / Station	16 Aug.	23 Aug.	30 Aug.	6 Sept.	13 Sept.	20 Sept.	27 Sept.	4 Oct.	Σ	m	Σ sq.
1	1.50	1.16	1.42	0.87	0.60	3.10	2.15	1.34	12.14	1.52	22.77
2	2.64	1.41	1.40	1.97	1.36	0.83	2.46	1.31	13.38	1.67	25.11
3	1.91	1.19	0.86	1.58	1.11	2.73	2.08	0.94	12.40	1.55	22.20
4	2.14	2.28	1.70	1.86	0.41	1.31	1.45	2.06	13.21	1.65	24.36
5	4.50	1.11	0.90	1.88	0.81	1.82	2.05	1.57	14.64	1.83	36.45
6	0.90	1.75	1.28	0.72	1.78	1.92	0.81	1.70	10.86	1.36	16.44
7	2.28	2.45	1.98	1.46	2.48	2.63	0.77	1.94	15.99	2.00	34.67
8	1.84	0.99	1.44	-0.83	-0.28	0.57	1.56	-0.12	5.17	0.65	9.97
9	0.49	0.43	0.29	-0.10	-0.39	1.39	0.36	-0.32	2.15	0.27	2.82
10	2.78	0.85	1.00	-0.38	-0.10	-0.45	1.04	0.21	4.95	0.62	10.92
11	1.02	0.80	0.19	1.51	0.17	0.64	0.51	0.65	5.49	0.69	5.12
12	0.76	-1.07	1.89	1.40	-0.14	0.22	1.03	0.35	4.44	0.55	8.50
13	2.15	1.35	0.80	2.02	2.24	0.65	-0.35	0.96	9.82	1.23	17.64
14	1.24	1.09	0.82	0.45	0.68	0.27	0.44	0.42	5.41	0.68	4.50
	26.15	15.79	15.97	14.41	10.73	17.63	16.36	13.01	130.05		
m	1.87	1.13	1.14	1.03	0.77	1.26	1.17	0.93		1.16	
sq.	62.85	26.92	22.05	26.14	19.36	36.79	27.82	19.54			241.47
Sq.	683.82	249.32	255.04	207.65	115.13	310.82	267.65	169.26			2258.69

E. CORRELATION OF COUNT RATES WITH CHEMICAL CONSTITUENTS IN SURFACE WATERS

In addition to the routine measurement of the gross beta radiation from the evaporated residue of one liter of each of the surface water samples, several water chemistry analyses were also run on many of the samples collected in 1952. The results of the correlation studies of these tests are summarized in Table 12. In all of the groups of tests, the probability that no correlation existed between the net count rate and the mineral content of the water (iron, total hardness, total solids, and chloride content) was small. In the measurements of the samples collected before the largest fallout in the spring of 1952, a considerably better product moment correlation between the net count rate and the total iron content was obtained than between the net count rate and the total solids content. The high correlations obtained in these two tests indicate that the net count rates measured before the deposition of the majority of the fallout from the spring series of the Nevada tests in 1952 was due principally to natural radioactivity. The lower product moment correlation coefficient between the iron content and the net count rate for the total period is due to the interfering increase in net count rates of the samples caused by fallout from the Nevada detonations. The product moment correlation between the samples' net count rates and their total hardness was found to be just less than significantly different from zero at the usual two-standard deviation level.

TABLE 12

CORRELATION[a] OF THE NET COUNT RATES[b] OF EASTERN MASSACHUSETTS SURFACE WATER SAMPLES WITH THEIR TOTAL IRON, TOTAL HARDNESS, TOTAL SOLIDS, AND CHLORIDE CONTENTS

Correlation of Net Count Rates With -	Date of Samples From To (1952)	No. of Samples	Product Moment Correlation Coefficient	Minimum Value of r for $P\{r \geq 0\} = 0.975$
Total Iron	April 26 - May 17	52	0.80	0.39
Total Iron	June 28 - August 23	126	0.45	0.25
Total Iron	April 26 - September 6	238	0.47[c]	0.18
Total Hardness	May 3 - July 5	112	0.27	0.28
Total Solids	August 30 - November 15	91	0.61	0.29
Chloride Content	April 26 - August 23	293	0.29	0.16

[a] The product moment correlation coefficient was calculated as suggested by Dixon and Massey (76, p. 233).

[b] Net counts per minute per liter at the first counting of samples, usually three days after collection.

[c] This value is based on data used in the first two calculations. An independent calculation using most of those data gave r = 0.40 for 178 samples.

F. ADDITIONAL MEASUREMENTS FOR NATURAL RADIOACTIVITY

Several measurements were made of other samples whose radioactivity was thought to be almost entirely of natural cause. Twelve planchets of the same earth samples that were reported by Thomas, et. al., (55) were measured. Their median weight was 1.55 grams with a range of 0.94 grams, and their median count rate was 5.57 counts per minute with a range of 2.98 counts per minute.

Between 1942 and 1944 a large number of samples were collected in various places in the United States by the Department of Soil Mechanics of the Harvard Graduate School of Engineering. One-half gram samples of twelve of these soils, when measured for this thesis in the fall of 1952, gave a mean net count rate of 3.26 counts per minute with a standard deviation of 1.40 counts per minute. The soil samples were placed in standard planchets and counted by the usual procedure.

Twelve samples of soil were collected from beneath basements of buildings on the Harvard campus at Cambridge in order to get measurements of Massachusetts soils that had had little possibility of contamination by long-range fallout. The mean sample weight was 2.04 grams with a standard deviation of 0.32 grams and the mean net count rate was 4.15 counts per minute with a standard deviation of 0.86 counts per minute.

Count rates were measured of six samples of activated carbon (made from wood pulp) of the type used in water treatment. The mean net count rate (0.03 counts per minute with a standard deviation of 0.20 counts per minute for samples that averaged 0.20 grams in weight)

indicates by the test used that the activated carbon did not contain a significant amount of radioactivity. It was estimated from the calibration data and the reported abundance of radiocarbon (C^{14}) in nature (40) that a planchet full of activated carbon would give a net count rate of only 0.04 counts per minute; therefore, naturally-occurring radiocarbon (C^{14}) was not detected to any appreciable extent by the techniques used for this thesis.

CHAPTER V

DETERMINATION OF DISINTEGRATION RATES FROM COUNT RATES OF LONG-RANGE FALLOUT

A. THE PROBLEM OF ESTIMATING DISINTEGRATION RATES

One of the important phases of the study of detonation debris by sanitary engineers is the accurate estimation of the disintegration rate of a sample based on its measured count rate. A nuclear disintegration may be defined as a spontaneous transformation characterized by the emission of mass (particulate matter) and/or energy (photons) by the nucleus. The allowable radioactivity limits with which the sanitary engineer is principally concerned, that is, the tolerance limits for drinking water, are given in disintegrations per unit of time. The conventional way of reporting this rate is in terms of the curie, which is defined as 3.7×10^{10} disintegrations per second. The microcurie (2.2×10^{6} disintegrations per minute) and the micromicrocurie are the conventional subdivisions used.

Owing to differences in decay rates and radioactive daughter production, the predominant species of radioisotopes in long-range fallout change rapidly with time during the first three months after fission. This period covers the interval in which long-range fallout samples can have relatively high disintegration rates per unit volume or weight.

When long-range fallout is measured with an end-window Geiger-Müller tube, the predominant radiation detected is beta particles. Disintegrating fission debris gives off approximately one gamma ray for each two beta rays emitted (1). However, only approximately 1% of the gamma rays entering the sensitive volume of a Geiger-Müller tube of the type used for this thesis causes it to register a count, although practically all beta rays do so. No attempt was made to measure alpha rays. Previous reports of measurements of long-range fallout (38, 77) have indicated that its alpha radiation was not significantly above natural levels.

The beta ray energies of fission products vary over a wide range. Those emitted by one nuclear species in one type of disintegration are not monoenergetic but come from the nucleus in a distribution which ranges from a definite maximum value, by which the radiation energy spectrum is usually characterized, to zero. Radioactive fission debris of less than a few months' age consists of a large number of nuclear species, most of which are beta emitters; many of these decay by production of beta rays belonging to more than one energy distribution.

The most accurate method of measuring beta radiation energy is with a beta ray spectrometer. However, as a specific activity sufficient to give 10^6 counts per minute per gram of sample is considered a weak source for this technique (78), it is not likely to be useful in most problems met in sanitary engineering. In practice, measurements are made by the insertion of absorbers, usually made of aluminum, between the radioactive sample and the Geiger-Müller tube. Bethe, Rose,

and Smith state (79) that fast electrons in a beam scatter but little when the beam enters an absorber. With decreasing energy, however, scattering increases until a diffusion stage is reached where the direction of motion is almost random. It has been found that a beam of electrons, all of equal velocity, is definitely not absorbed exponentially. However, the nuclear disintegration beta rays are distributed in velocity spectra and "fortuitously" have been found to be absorbed in an exponential fashion over a fairly wide range (30, 80, 81). This exponential absorption is due to superpositioning of the velocity values in each energy spectrum as the beta rays pass through the absorber (82).

B. INTERVENING ABSORBERS

The linear portion of a logarithmic-arithmetic plot of net count rate versus the mass per unit area of an intervening absorber may be used in determining an absorption coefficient μ_{al}, which usually is determined with aluminum and expressed in square centimeters per milligram, in the following equation:

$$\frac{C_x}{C_o} = e^{-\mu_{al} x} \qquad (5\text{-}1)$$

where

C_o is the count rate with no absorber
C_x is the count rate for radiation passing through the absorber
x is the mass of the absorber in milligrams per square centimeter
e is the base of Napierian logarithms

(As an illustration, a plot of laboratory measurements of net count rates versus various quantities of added aluminum absorber is shown later in this chapter in Figure 19.)

C. SELF-ABSORPTION

When the radioactive material is homogeneously distributed within a medium having appreciable mass, this medium itself acts as an absorber. If the exponential absorption law applies, the following formulation can be made (refer to Figure 14):

Let u_o be the sample specific activity* in disintegrations per minute per gram of the sample

ρ be the density of sample in milligram per cubic centimeter

A be the surface area of the sample in square centimeters

dy be the thickness of an incremental layer of sample in centimeters

y be the distance of the layer below the surface in centimeters

Thus, ρyA is the absorbing mass above the layer dy in milligrams

ρy is the absorbing mass per unit area above the layer in milligrams per square centimeter

$A u_o \rho y$ is the radioactivity of the layer in disintegrations per minute.

k is a geometry and backscatter factor accounting for the proportion of the radiation directed to the sensitive volume of the geiger tube.

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* For this thesis, sample specific activity is defined as the disintegration rate of the sample in disintegrations per minute per gram of sample.

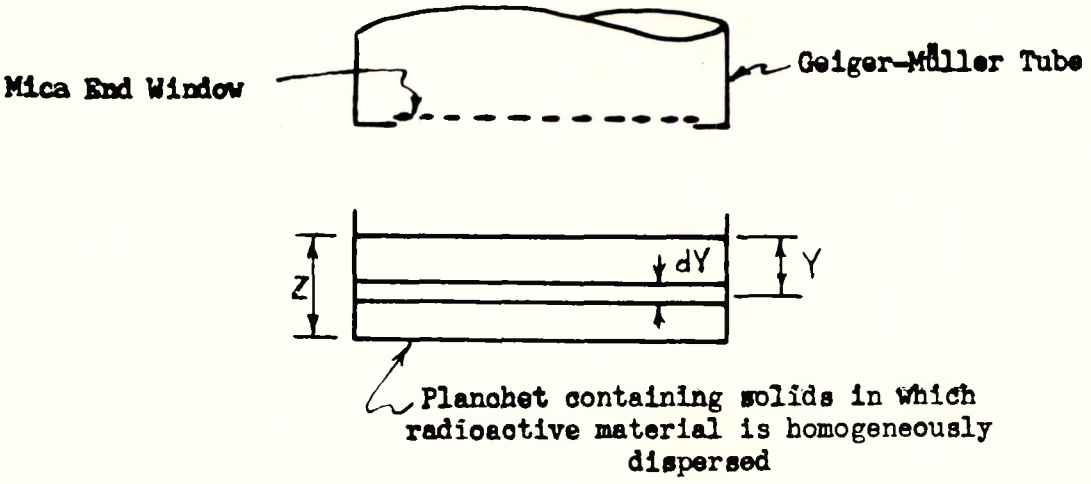

Fig. 14. — Sketch for the Self-absorption Formulation

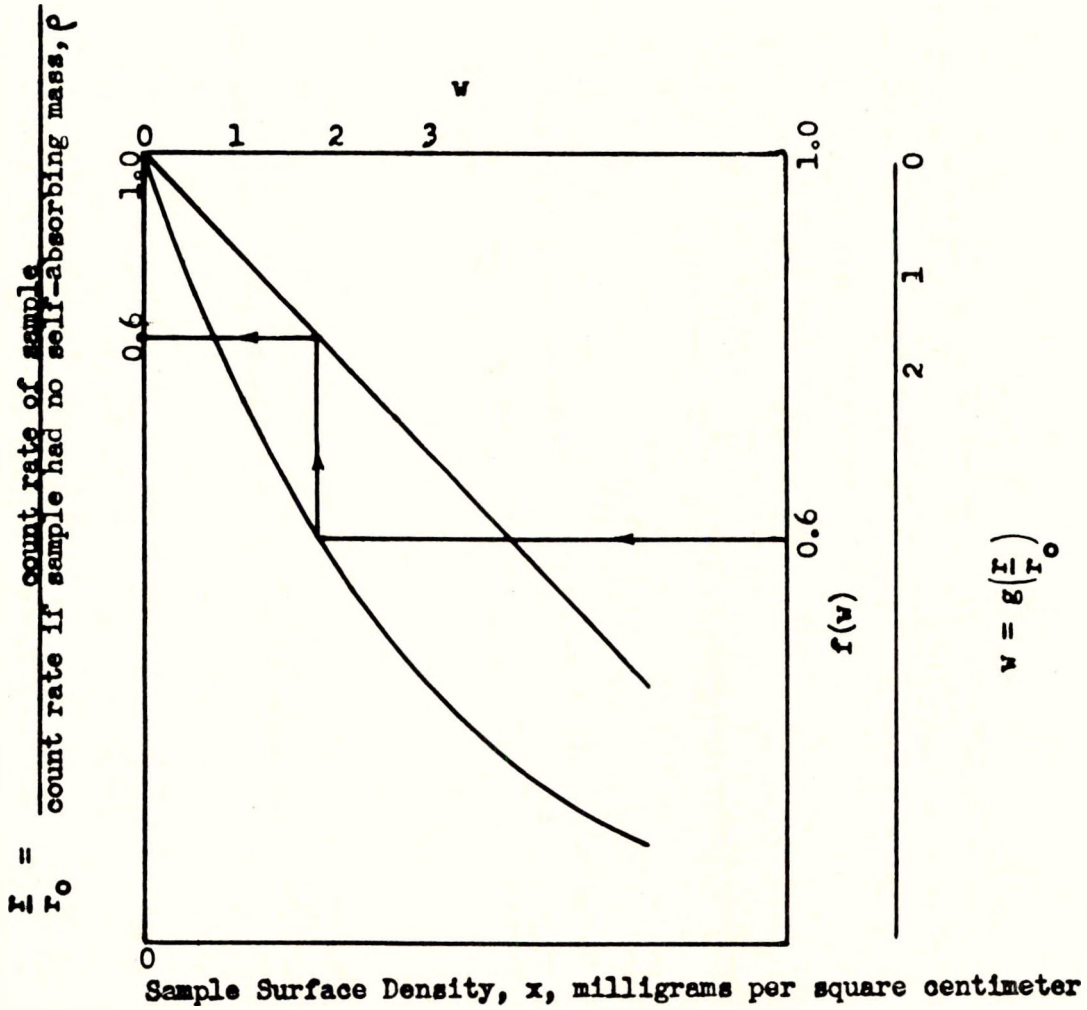

Fig. 15. — Illustration of the Construction of "Self-absorption" Paper

Since the scattering characteristics for beta radiation when the absorber is near the sample are different than when the absorber is near the Geiger-Müller tube, a different absorption coefficient, μ_{sa}, is used in the self-absorption determination. Thus, the amount of the radioactivity in the layer that reaches the surface of the sample is

$$k\, u_o\, \rho\, A \quad e^{-\mu_{sa}\rho y}\, dy \tag{5-2}$$

and the sum of all of the beta rays reaching the surface is

$$k\, u_o\, \rho\, A \int_0^Z e^{-\mu_{sa}\rho y}\, d\, y = \frac{k\, u_o\, A}{\mu_{sa}} \left(1 - e^{-\mu_{sa}\rho Z}\right) \tag{5-3}$$

If no self-absorption takes place, that is, if $\mu_{sa} = 0$, the radioactivity reaching the surface would be $k\, \rho\, Z\, A\, u_o$. The self-absorption factor, which is defined as the ratio of the count rate with the μ_{sa} of the self-absorbing medium to the count rate for $\mu_{sa} = 0$, is

$$\frac{r}{r_o} = \frac{u_o\, A}{\mu_{sa}\, u_o\, A\, \rho\, Z} \left(1 - e^{-\mu_{sa}\rho Z}\right) = \frac{1 - e^{-\mu_{sa}\rho Z}}{\mu_{sa}\, \rho\, Z} \tag{5-4}$$

It is convenient to define the sample thickness in units of areal density, $x = \rho Z$, the "surface density" in milligrams per square centimeter. Thus,

$$\frac{r}{r_o} = \frac{1 - e^{-\mu x}}{\mu x} \tag{5-5}$$

The formula may be linearized for plotting purposes by a transformation. A dimensionless variable, $w = \mu_{sa}x$, will be used. Thus,

$$f(w) = \frac{1 - e^{-w}}{w} = \frac{r}{r_o} \quad . \tag{5-6}$$

The inverse of this function is

$$w = g\left(\frac{r}{r_o}\right) \quad . \tag{5-7}$$

Therefore,

$$\mu_{sa} = \frac{1}{x}\, g\left(\frac{r}{r_o}\right) \quad . \tag{5-8}$$

A projection of points from this curve upon a straight line of convenient slope passing through the coordinates $\left[w = 0 , f(w) = 1 \right]$, yields the ordinate value r/r_o. On Figure 15 are plotted both the ordinate values of w and the values of $f(w)$.

The self-absorption coefficients for the calibrated isotopes and rain samples were obtained in the following manner. Equal portions of each sample were mixed with varying amounts of self-absorption medium, evaporated, and transferred to planchets. Their net count rates, r, were then measured. A plot of the logarithms of the net count rate versus the self-absorbing masses in milligrams per square centimeter was made in order to obtain the value of r_o, the net count rate that would exist if there were no self-absorption. An extension of the line fitted through these points to a self-absorbing mass of zero gave the value of r_o.

The self-absorption coefficient was obtained by making a plot on "self-absorption" paper of the mass in milligrams per square centimeter versus its corresponding r/r_o value. A line of best fit through the origin (self-absorbing mass = 0 , $r/r_o = 1$) was used to obtain the coefficient from the relationship

$$\mu_{sa} = \frac{g^{\left(\frac{r}{r_o}\right)}}{x} \quad . \qquad (5\text{-}9)$$

D. CALIBRATION

In radiochemistry and in tracer studies it is often possible to obtain the desired results by measuring relative radioactivities rather than absolute disintegration rates. The latter are much more difficult to evaluate. Indeed, so many are the variables and complex interrelationships that absolute beta counting is an ideal that can only be approached. For some of the objectives of this thesis it has been desirable to estimate actual disintegration rates. This has been done by taking into consideration most of the important factors involved. However, it has not been possible to allow for all factors and it should be emphasized that disintegration rates reported are only approximate estimates.

Disintegration rates were determined by taking into consideration the following factors that affect Geiger-Müller tube measurement of radioactive samples:

1. Geometry, the relative position of sample and Geiger-Müller tube, or more exactly, that portion of the total solid angle about the sample that is subtended by the sensitive volume of the measuring device. In almost all of the readings taken for this thesis the same geometry was maintained. In all of the aluminum absorption and self-absorption calibration tests the geometry was identical with that of the sample measurements. Nearly all of the measurements made for this thesis at Harvard were with instruments claimed by their manufacturer to have no significant source of error owing to the lack of reproducibility of geometry.

2. Self-absorption, the absorption of radiation of the sample by the mass of the sample, a complicated function of the energy and type of radiation and of the mass and atomic weight of the sample material.

3. Self-scattering, or self-focusing, the ability of the mass of the sample to deflect the radiation from its original direction of motion away from or into the sensitive volume of the Geiger-Müller tube. The details of the mechanism of this effect are not yet well understood.

4. Backscattering, the reflection by the sample support of the sample rays into the sensitive volume of the Geiger-Müller tube. In the work at Harvard, steel Tracerlab cup-planchets were used in all cases, so that the variability in backscattering was not important in the measurements for this thesis. However, a series of runs was made

using calibrated samples of pure nuclear species with various sample support materials in order to generalize the calibration procedure. Results are shown in Figure 16. As has been previously found (83), the backscattering increases with the atomic number of the support material. No attempt was made to separate the radiation coming directly from the support material from that reflected by the backscattering material.

5. Reflection of radiation, from the walls of the sample container and the shielding and support material. Nearly all samples measured at Harvard were with the same types of containers and shields, so that this factor was usually not an important source of variance in the measurements.

6. Absorption of radiation by the air gap and the thin end-window of the Geiger-Müller tube as the rays go from the sample surface to the sensitive volume of the tube. Corrections for these factors were made by assuming an absorption of radiation similar to that obtained by added aluminum absorbers.

E. ABSORPTION OF RADIATION FROM SAMPLES OF CALIBRATED NUCLEAR SPECIES BY ALUMINUM

The National Bureau of Standards sells samples containing nuclear species whose disintegration rates per unit volume of sample have been accurately determined. The equipment used at Harvard was calibrated by using radioactive carbon (C^{14}), Phosphorous (P^{32}), strontium ($Sr\text{-}Y^{90}$), thallium (Tl^{204}) and cobalt (Co^{60}) - all originally in liquid

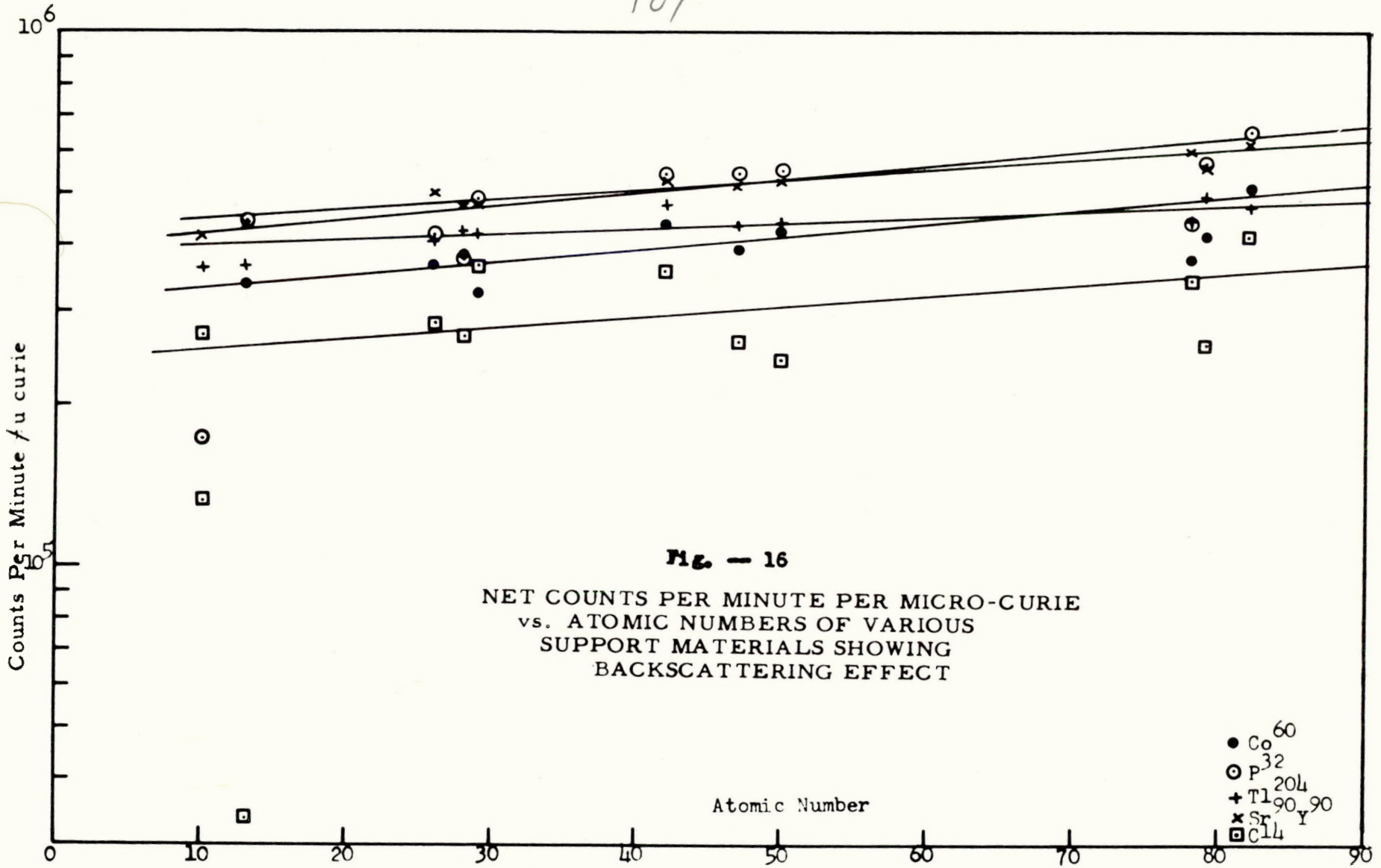

Fig. — 16

NET COUNTS PER MINUTE PER MICRO-CURIE
vs. ATOMIC NUMBERS OF VARIOUS
SUPPORT MATERIALS SHOWING
BACKSCATTERING EFFECT

solution and calibrated in disintegrations per minute per milliliter of solution. Also, an accurately calibrated radium D + E (Pb^{210} + Bi^{210}) standard was used for an aluminum absorption coefficient determination and for estimating the ratio of counts per minute obtained to disintegrations per minute for the beta ray energy distribution it produces. Of the calibrated samples of nuclear species currently issued by the Bureau, all were used except iodine (I^{131}).

Micro-pipettes were used to transfer carefully measured quantities of each of the samples of calibrated nuclear species to planchets. These solutions were dried and used to make aluminum absorption measurements. Sheets of aluminum foil were cut into convenient-sized pieces and cleaned, measured for area, and weighed individually. Various combinations of these aluminum foil pieces were then attached to the planchet holder in the automatic sample changer, so that increasing masses of absorber were inserted between the sample and counter. Except in a few instances in which the large mass of absorbing material had caused the net count rate to drop to a prohibitively low value, all measurements of the calibrated samples of nuclear species were made long enough to cumulate 40,000 counts and insure statistical accuracy. This number of counts reduces the coefficient of variation of the measurement to about 0.5%. Figure 17 shows the curves of the logarithms of the counts per minute versus the surface density, in milligrams per square centimeter, of aluminum absorber obtained for the various calibrated radioactive samples. The non-linearity of the strontium (Sr-Y^{90}) curve is expected because the beta ray spectrum of the yttrium (Y^{90}) has a much higher energy than does the strontium (Sr^{90}). The sharp

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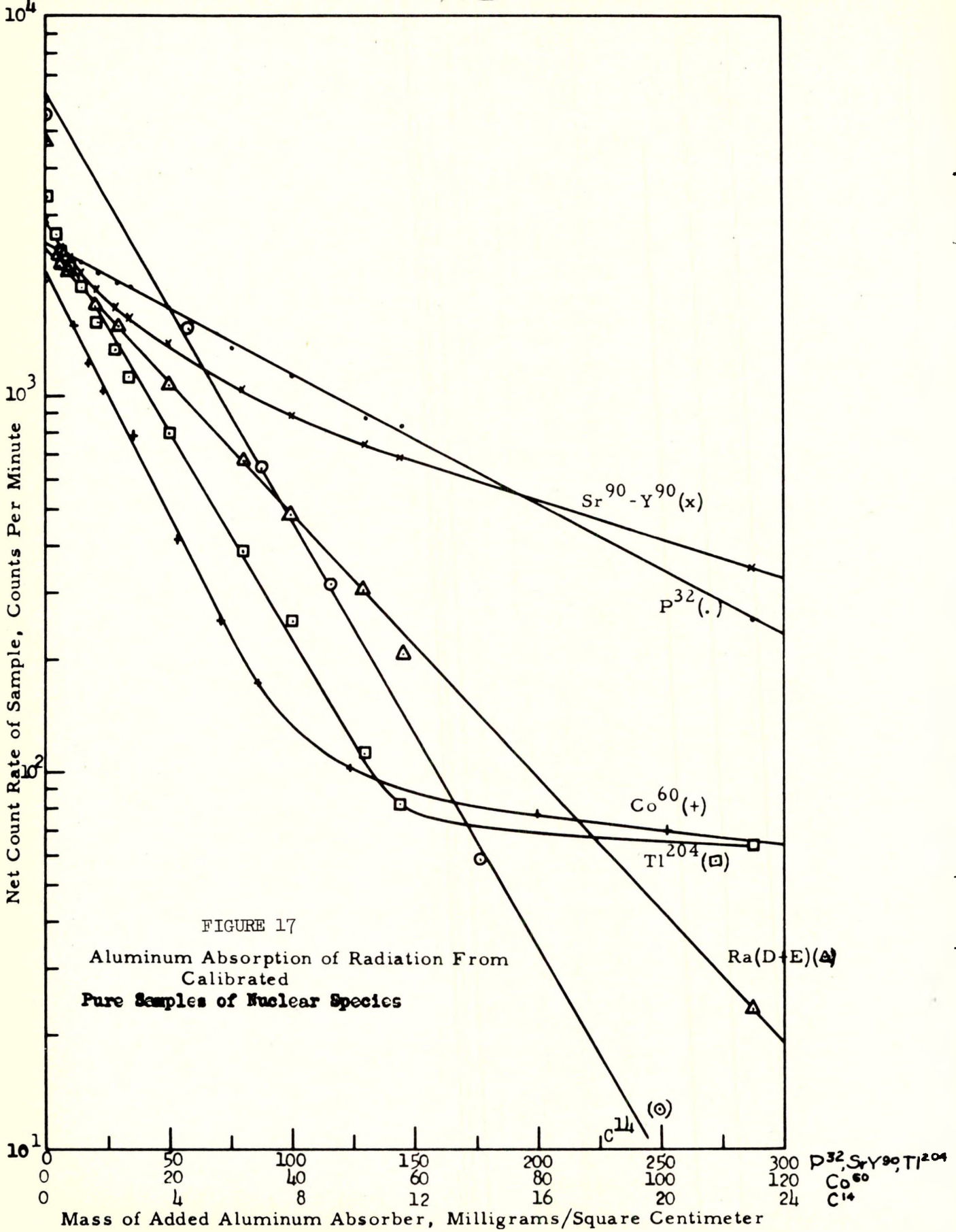

FIGURE 17

Aluminum Absorption of Radiation From Calibrated
Pure Samples of Nuclear Species

curvature of the lower section of the cobalt (Co^{60}) curve probably is due to the gamma radiation emitted in disintegration of the cobalt (Co^{60}). The curvature at the bottom of the thallium (Tl^{204}) plot could be due to Bremsstrahlung (secondary photon emission caused by the deceleration of the beta particles in the nuclear field of the absorber). Separate calibrated samples of thallium (Tl^{204}) and strontium ($Sr\text{-}Y^{90}$) were obtained from the National Bureau of Standards and duplicate runs were made as a check.

That portion of the strontium ($Sr\text{-}Y^{90}$) curve beyond the maximum range of strontium (Sr^{90}) beta particles in aluminum (about two hundred and twenty milligrams per square centimeter) was found to be fairly linear and was assumed to be due to yttrium (Y^{90}) beta rays alone. A line drawn through the plotted points caused by the yttrium (Y^{90}) beta rays alone was extrapolated to the vertical axis and was used to separate the net count rates caused by the beta rays of the strontium (Sr^{90}) alone in order to get a strontium (Sr^{90}) aluminum absorption coefficient. At a given absorber mass, the difference in the net count rates between the point on the extrapolated yttrium (Y^{90}) line and the actual measured count rate value was assumed to be caused by strontium (Sr^{90}) alone. A plot (Figure 18) of the logarithms of these differences versus their respective absorber densities is approximately linear.

Table 13 summarizes all of the aluminum absorption and self-absorption coefficients obtained from measurements for this thesis of the calibrated samples of nuclear species, as well as values for comparisons that were obtained by other investigators or were calculated from appropriate empirical formulas (80).

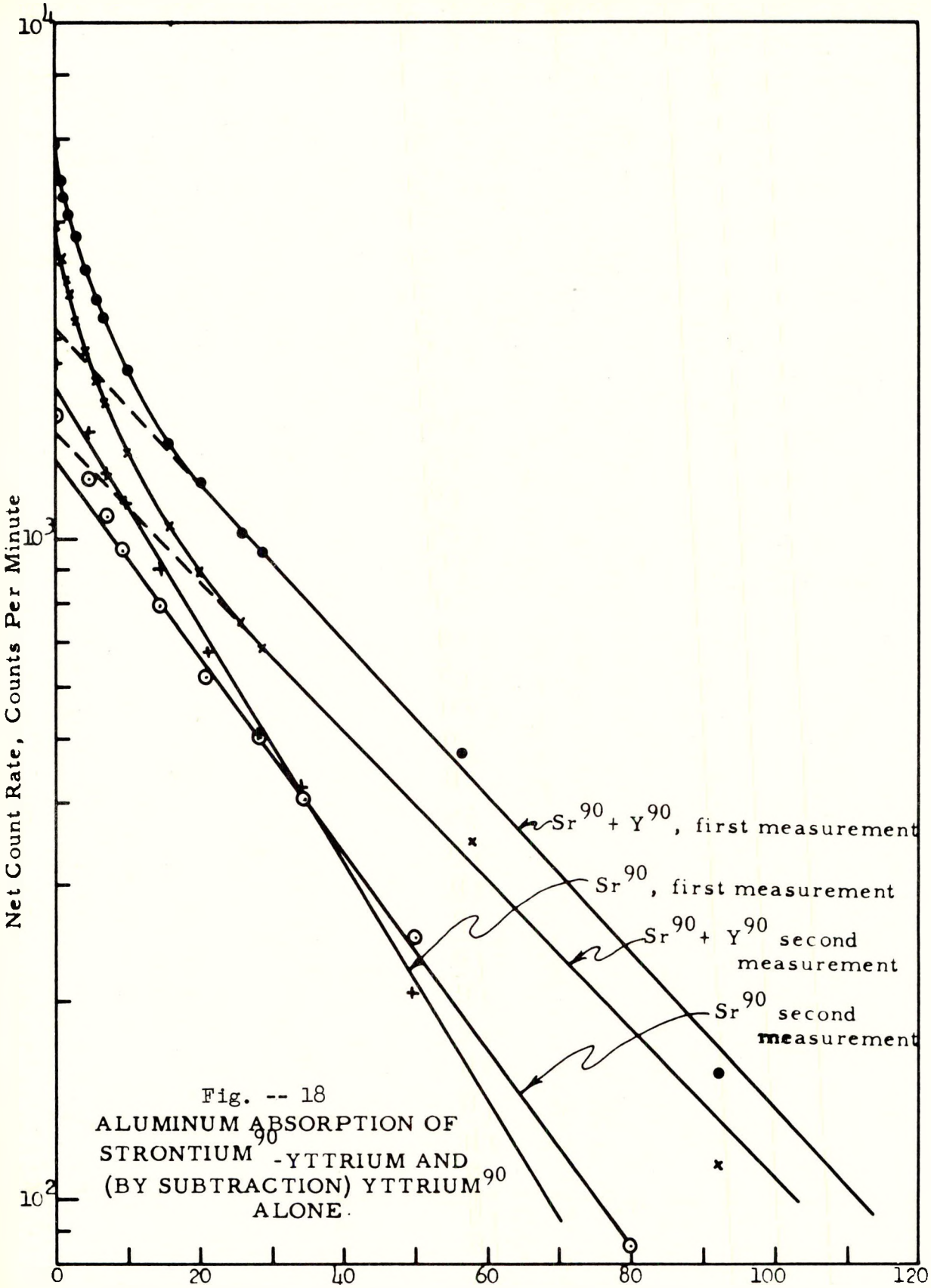

Fig. -- 18
ALUMINUM ABSORPTION OF STRONTIUM^{90}-YTTRIUM AND (BY SUBTRACTION) YTTRIUM^{90} ALONE.

TABLE 13

ABSORPTION COEFFICIENTS FOR SAMPLES OF CALIBRATED PURE NUCLEAR SPECIES

Nuclear Species	Maximum Energy, Mev.	Measured Aluminum Abs. Coef. ($\mu_{al.}$)	Found by Other Investigators (μ_{al})	Measured Self-Abs. Coef. (μ_{sa})
C^{14}	0.014	0.328	0.39[a]	0.136[g]
Co^{60}	0.30	0.0647	0.079[b]	0.0575
Tl^{204}	0.87	0.0242 0.0245	0.0256[b] 0.0292[f]	0.0363 -
Y^{90}	2.6	0.00520 0.00544	0.0047[b]	- -
Sr^{90}	0.65	0.0344 0.0407	0.039[c]	-
P^{32}	1.69	0.00792	0.0053[d]	0.00433
Ra(D+E)	1.17	0.0162	0.0179[e]	-

[a] Calculated using Libby's formula and the median "constant" of Table 5 in (85).

[b] From Table 7 in (80, p. 72) but assuming that the data for Tl^{204} have been reversed with those of Tl^{206}.

[c] Calculated using formula in (80, p. 58).

[d] From Table 7 in (80), a water absorption coefficient.

[e] Calculated using formula in (80, p. 58), but with the range in aluminum taken from Friedlander and Kennedy (13).

[f] Calculated from plot of data in which the same type of equipment was used as that for this thesis (59).

[g] Other investigators have gotten 0.285 and 0.29 for this value (86).

A project was conducted at the Radiation Laboratory of the University of California (84) in which count rates of carefully deposited salts containing pure beta-emitting nuclear species were measured. No theoretical equation was found that would fit the experimental data beyond a sample mass of twenty milligrams per square centimeter.

F. ABSORPTION OF RADIATION FROM LONG-RANGE FALLOUT BY ALUMINUM

The solids from evaporated rain that contained easily measurable amounts of fission radioactivity were used to make several aluminum absorption curves. Figure 19 and Figure 20 are examples of the plots obtained and Table 14 summarizes the resulting coefficients. The mean aluminum absorption coefficient is 0.0140 with a standard deviation of the measurements of 0.0044 square centimeters per milligram. No correlation was found to exist in these data between the fission material age (assuming appropriate detonation dates) and its aluminum absorption coefficient. Therefore, the average value has been used in the calibration.

Under measuring conditions quite similar to those used at Harvard, Clark and his associates (88) found an average aluminum absorption coefficient of 0.012 ± 0.002 square centimeters per milligram for Nevada fission products collected in Troy, New York, during the 1953 weapons test period.

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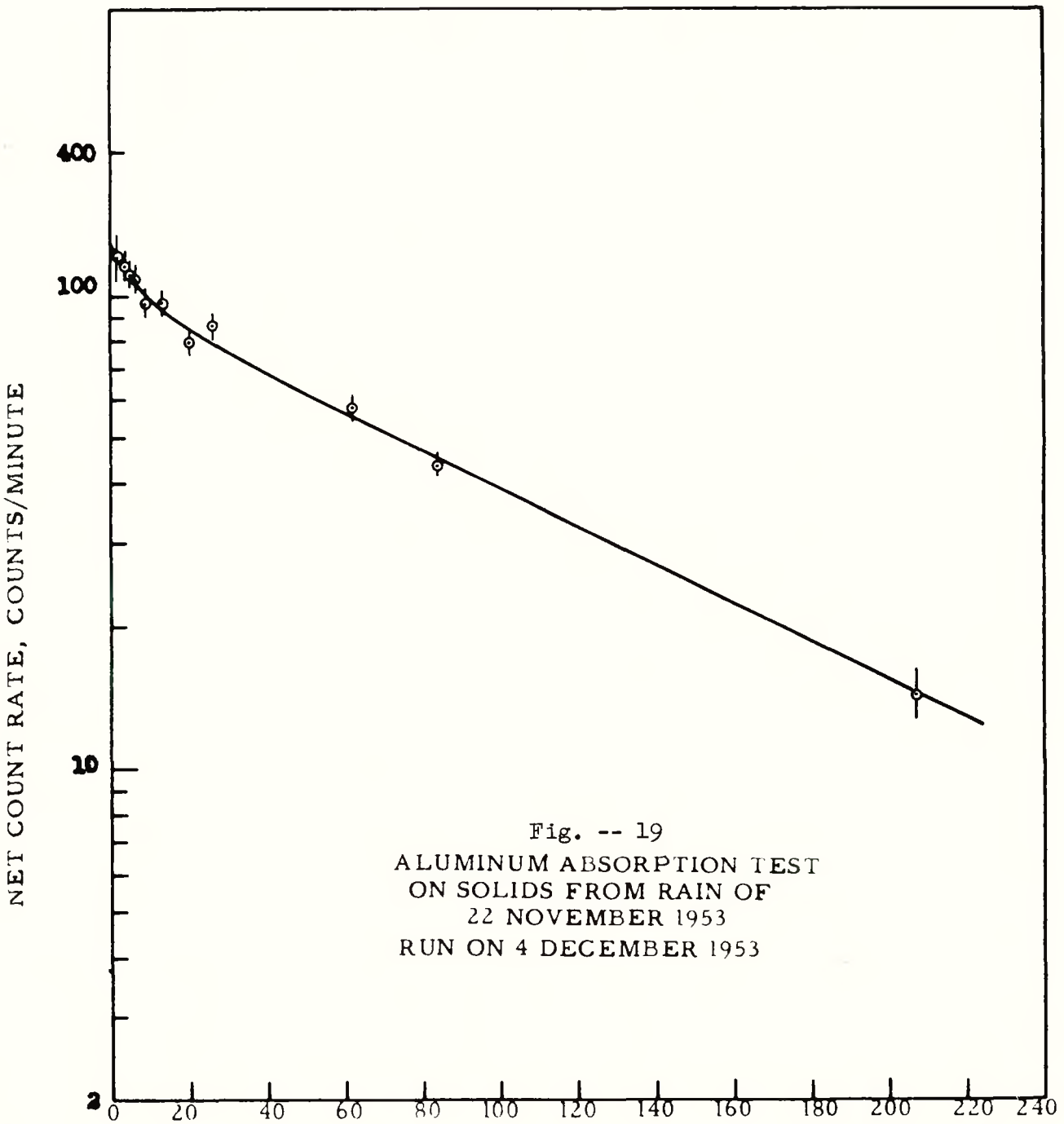

Fig. -- 19
ALUMINUM ABSORPTION TEST
ON SOLIDS FROM RAIN OF
22 NOVEMBER 1953
RUN ON 4 DECEMBER 1953

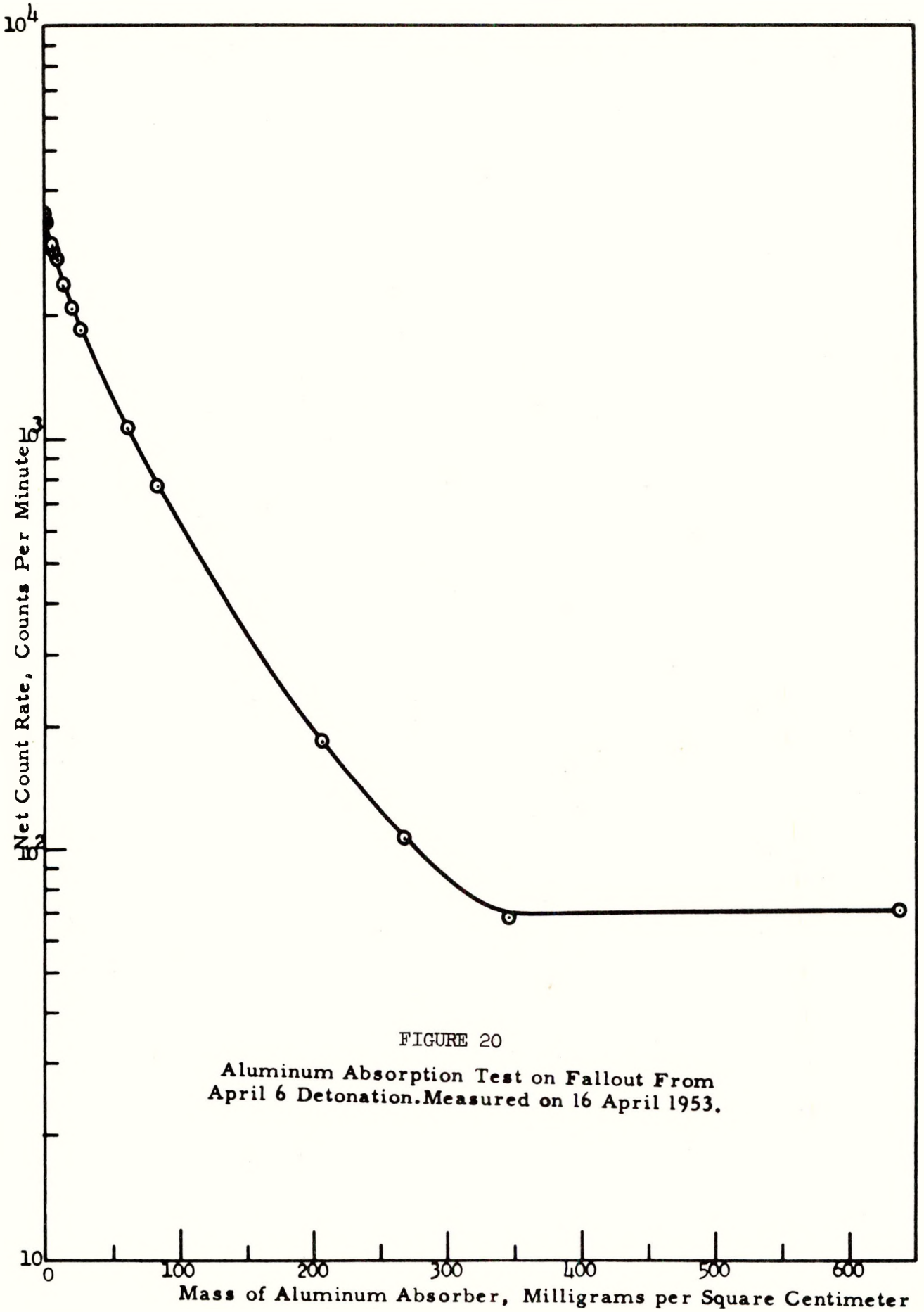

FIGURE 20

Aluminum Absorption Test on Fallout From
April 6 Detonation. Measured on 16 April 1953.

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TABLE 14

MEASURED ALUMINUM ABSORPTION COEFFICIENTS OF FALLOUT SAMPLES

Probable Blast Date	Source of Fallout Sample	Date Sample Collected	Date Sample Measured	Age of Sample (Approx.)	Aluminum Absorption Coefficient (μ_{al} cm^2/mg)
May 6, 1952	Rain, Cambridge, Mass.	May 12, 1952	May 26, 1952	20 days	0.0161
Nov.?, 1952	Filtrate, 5 gal. rain Rochester, N.Y., area	Nov. 17, 1952	Nov. 20, 1952	1 month	0.0105
Nov.?, 1952	Solids of 5 gal. rain Rochester, N.Y., area	Nov. 17, 1952	Nov. 20, 1952	1 month	0.0098
Nov.?, 1952	Filtrate, 5 gal. rain Rochester, N.Y., area	Nov. 17, 1952	Nov. 20, 1952	1 month	0.0102
Nov.?, 1952	Rain, Cambridge, Mass.	Nov. 22, 1952	Dec. 4, 1952	1 month	0.0098
Apr.6, 1953	Rain, Cambridge, Mass.	Apr. 8, 1953	Apr. 17, 1953	11 days	0.0141
Apr.6, 1953	River Water, Eastern Mass.	Apr. 9, 1953	Apr. 29, 1953	23 days	0.0188
Apr.6, 1953	River Water, Eastern Mass.	Apr. 9, 1953	Apr. 21, 1953	15 days	0.0223
			Mean		μ_{al} = 0.0140* cm^2/mg
			Standard Deviation of		μ_{al} = 0.0044 cm^2/mg

* From (87) the empirical formula $\mu = 22/E_m^{1.33}$, relating the aluminum absorption coefficient to the maximum energy, yields an average maximum energy $E_m = (22/14)^{1/1.33} = 1.43$ Mev., which agrees well with the value of 1.4 in Reference (1).

G. MEASUREMENTS OF SELF-ABSORPTION OF RADIATION FROM CALIBRATED SAMPLES

Self-absorption coefficients for fallout radioactivity were obtained by mixing equal quantities of each of the calibrated samples obtained from the National Bureau of Standards with various quantities of solids and measuring the net count rates obtained. The solids used were chosen to simulate those remaining after evaporation of eastern Massachusetts surface waters. Two types of solids were used. The first consisted of the residue from evaporation of a mixture of fifteen grams of finely-ground eastern Massachusetts surface soil in a liter of clean ocean water. The second was a synthetic mixture of solids having a chemical composition close to that of the eastern Massachusetts surface waters. When the soil-ocean water mixture was used, it was necessary to evaporate suitable quantities of the mixture to obtain sufficient solids for the self-absorption calibration. The residue from this evaporation was transferred from porcelain evaporating dishes to planchets; the calibrated radioactive nuclear species solutions were then added; the mixture was stirred with a small rod of polyethylene and then placed under a heat lamp for evaporation to dryness. It was difficult to keep the concentrated soil-ocean water material from "climbing" over the planchet wall while drying. Also, the solids from the evaporated ocean water had sufficient natural radioactivity to be a factor of error, especially when self-absorption calibrations of fallout of low radioactivity were made. In the self-absorption measurements made with the calibrated samples, parallel controls were run on samples having

the equivalent amount of solids but no added radioactivity. The count rates used in plotting the self-absorption curve were obtained by subtracting the count rate of the control from that of the respective sample containing added radioactive material.

It was found that a better approach to the problem would be to use the mixture of synthetic solids as a self-absorption medium. In October, 1952, which was during the fourteen-month period in which weekly samples of eastern Massachusetts surface waters were taken for this thesis, a series of chemical analyses of many of these same surface waters was made by the United States Geological Survey. The analyses for seven streams (the Nashua, Merrimack, Concord, Charles, Blackstone, Quoboag, and Westfield) were used to obtain an average chemical composition for making the synthetic solids. Five of the stations from which water samples were collected weekly were located on these streams. The ratios of the amounts of mineral ions of different types for each stream relative to its total ion concentration was found to be remarkably consistent for all of the seven streams. The average ionic composition for those ions present to the extent of more than one part per million concentration in these streams was then reproduced by mixing the necessary laboratory chemicals in their proper proportion in distilled water. A solution containing a small, flaky, dense, brown precipitate was obtained. This synthetic solid did not try to "climb out" of the planchet when being dried under a heat lamp. Table 15 gives the average of the predominant constituents of the surface waters and the proportion of chemicals used in the synthetic solids. Figure 21 gives the curves that

TABLE 15

AVERAGE MINERAL COMPOSITION OF SEVEN MASSACHUSETTS STREAMS
AND COMPOSITION OF SYNTHETIC SOLIDS USED

Ion	Avg. Amt. E. Mass. Streams (ppm)	Milli-equivalents	Lab Chemicals Used	Milli-equivalents	Grams of Each Chemical Used in Mixture
Fe(total)	0.45	0.0162	$NaHCO_3$	0.443	72.10
Ca	8.633	0.433	K_2SO_4	0.064	10.82
Mg	2.00	0.165	Na_2SO_4	0.203	27.94
Na	14.74	0.646	$Fe_2SO_4 \cdot 7H_2O$	0.016	4.30
K	2.49	0.0614	$Mg(NO_3)_2 \cdot 6H_2O$	0.038	5.46
$CO_3^=$	1.86	0.0614	$MgSO_4 \cdot 7H_2O$	0.127	30.32
HCO_3^-	23.51	0.382	$CaCl_2 \cdot 2H_2O$	0.391	42.04
Cl	14.00	0.391	SiO_2		8.30
SiO_2	3.83				

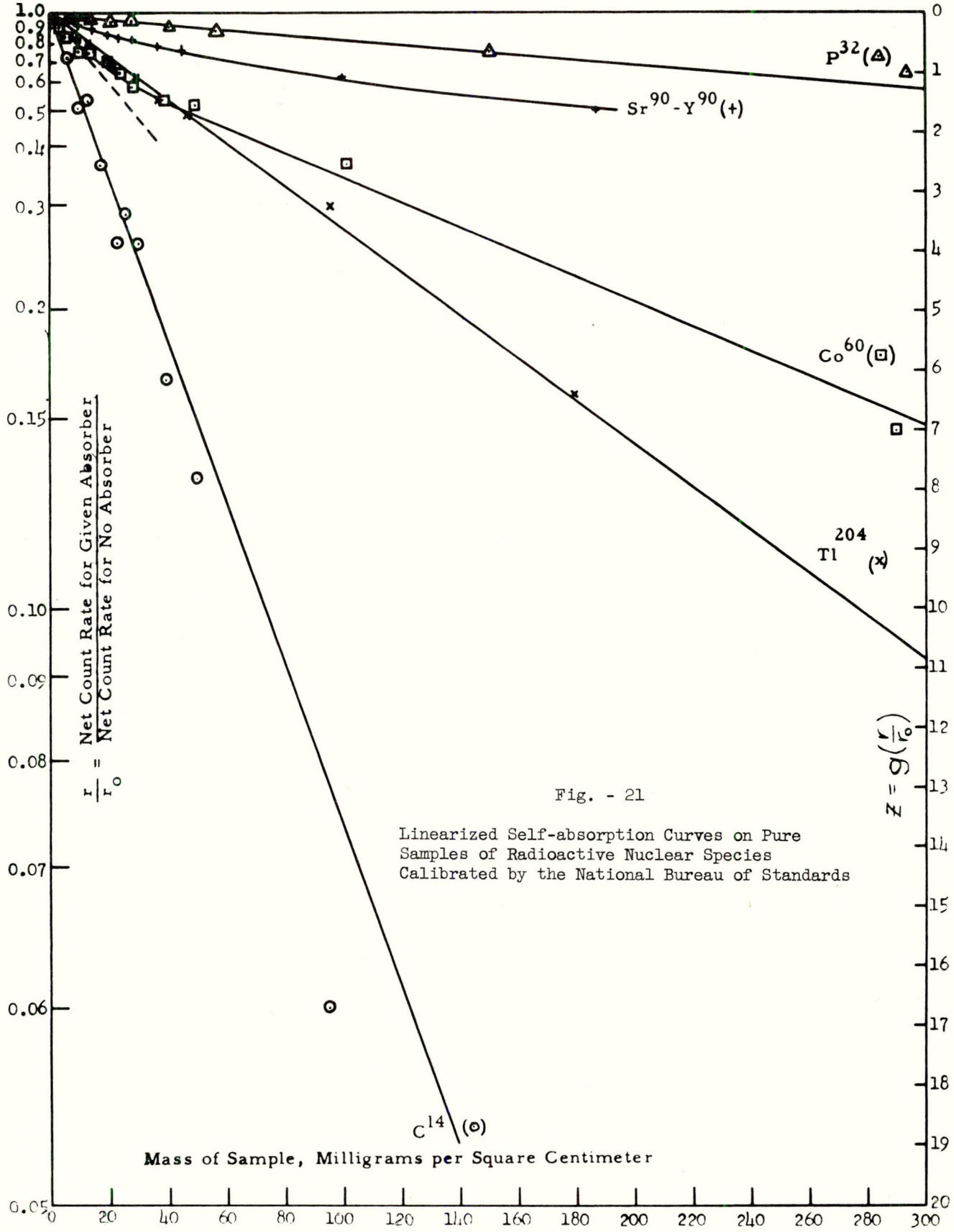

Fig. - 21

Linearized Self-absorption Curves on Pure Samples of Radioactive Nuclear Species Calibrated by the National Bureau of Standards

were obtained with these solids in self-absorption measurements using the calibrated samples. The plots are on "self-absorption" paper. Figure 22 gives an illustration of the plots that were made in order to obtain the r_o values for the self-absorption determination. Only four of the calibrated samples could be used to obtain a self-absorption coefficient, because no method was devised for the separation of the strontium (Sr^{90}) from the yttrium (Y^{90}) counts, as was done in the aluminum absorption determination. The cobalt (Co^{60}) self-absorption coefficient was taken from a straight line of best fit drawn through measurements taken on the five samples of the lowest mass, in order to minimize the influence of the cobalt (Co^{60}) gamma rays.

Two self-absorption experiments were run with long-range fallout material in which the ocean water-soil mixture was used for the absorbing solids. One was made with "old" debris collected in rainfall following a Nevada detonation. The other was made using the most recent fallout available at the time of these measurements. This was fission material contained in a rain collected a few weeks after the announced approximate dates of Russian detonations.

A third experiment, in which the rain of September 20, 1953, was used, was made with the mixed synthetic solids as the absorbing medium. The number of self-absorption experiments using fallout material (summarized in Table 16) was small, and the low count rate of the samples made from the "old" fallout fission debris did not allow a precise determination of its self-absorption coefficient. The variability of the measured count rate of these samples probably is due in part to the

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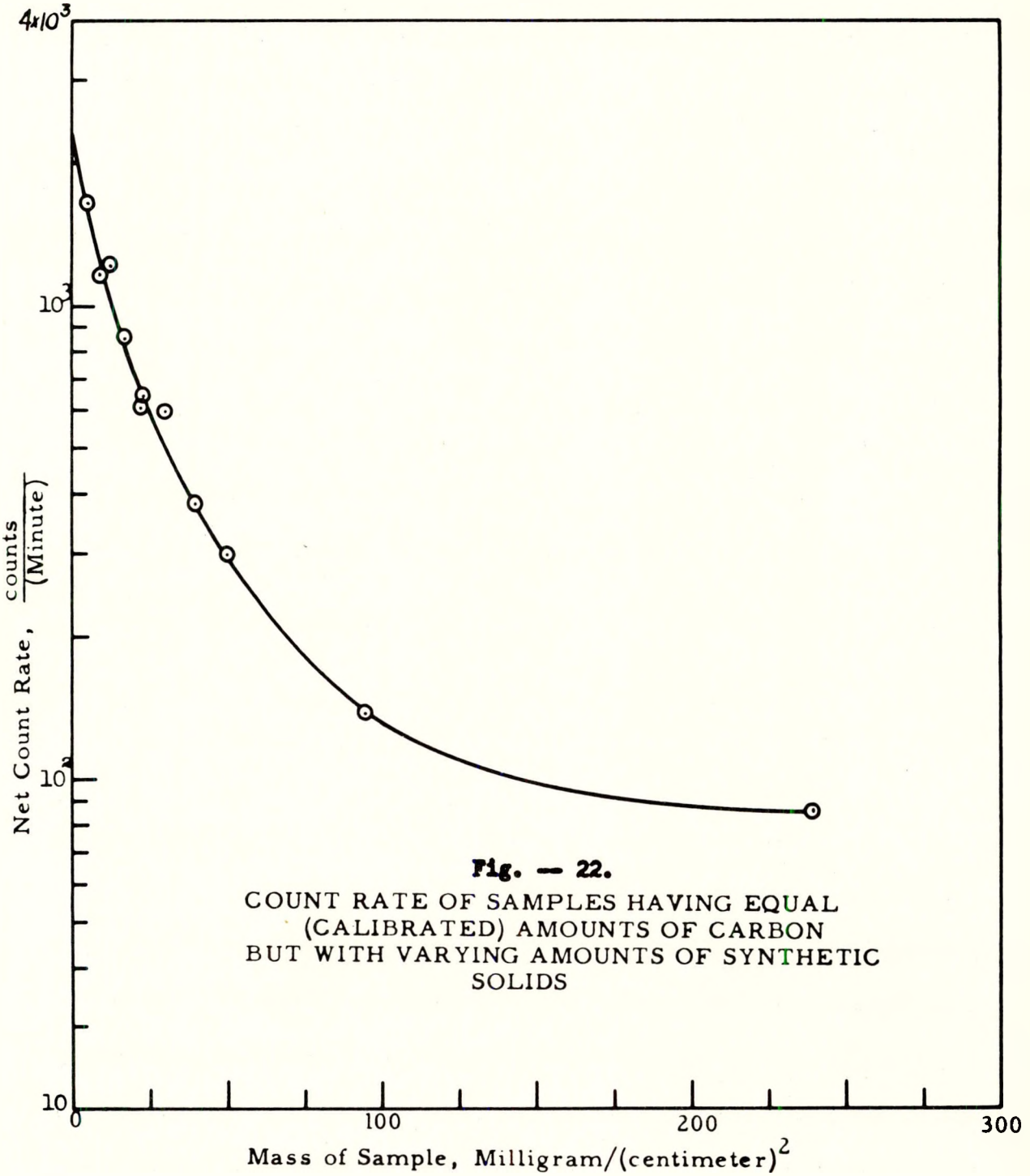

Fig. — 22.

COUNT RATE OF SAMPLES HAVING EQUAL (CALIBRATED) AMOUNTS OF CARBON BUT WITH VARYING AMOUNTS OF SYNTHETIC SOLIDS

particulate nature of the fallout material. These factors suggest that the fallout measurements that were made are not adequate for use in calibration. (The data are presented as a guide to the methods used.)

TABLE 16

MEASURED SELF-ABSORPTION COEFFICIENTS OF LONG-RANGE FALLOUT

Assumed Place of Detonation	Probable Date of Detonation (1953)	Date of Measurements (1953)	Age of Sample, Days (approx.)	Type of Solids Used	μ_{sa} cm^2/mg
Nevada	April 6	August 16	132	Soil-ocean salt	0.0175
Russia	Mid-August	September 2	20	Soil-ocean salt	0.025
Russia	Mid-August	September 30	47	Synthetic	0.037

Owing to the factors just mentioned, it was decided that a line fitting the points of the plot on double logarithmic paper of the aluminum absorption versus self-absorption coefficients would be used as a basis for the relation of these parameters (Figure 23). It was felt that a curved line of best fit through these points would unevenly weight the estimation in favor of the nuclear species having higher and lower absorption coefficients (C^{14} and P^{32}). Thus, a line having a 45° slope was fitted to the four points by eye. This presumes that a simple direct proportionality exists between the two types of absorption

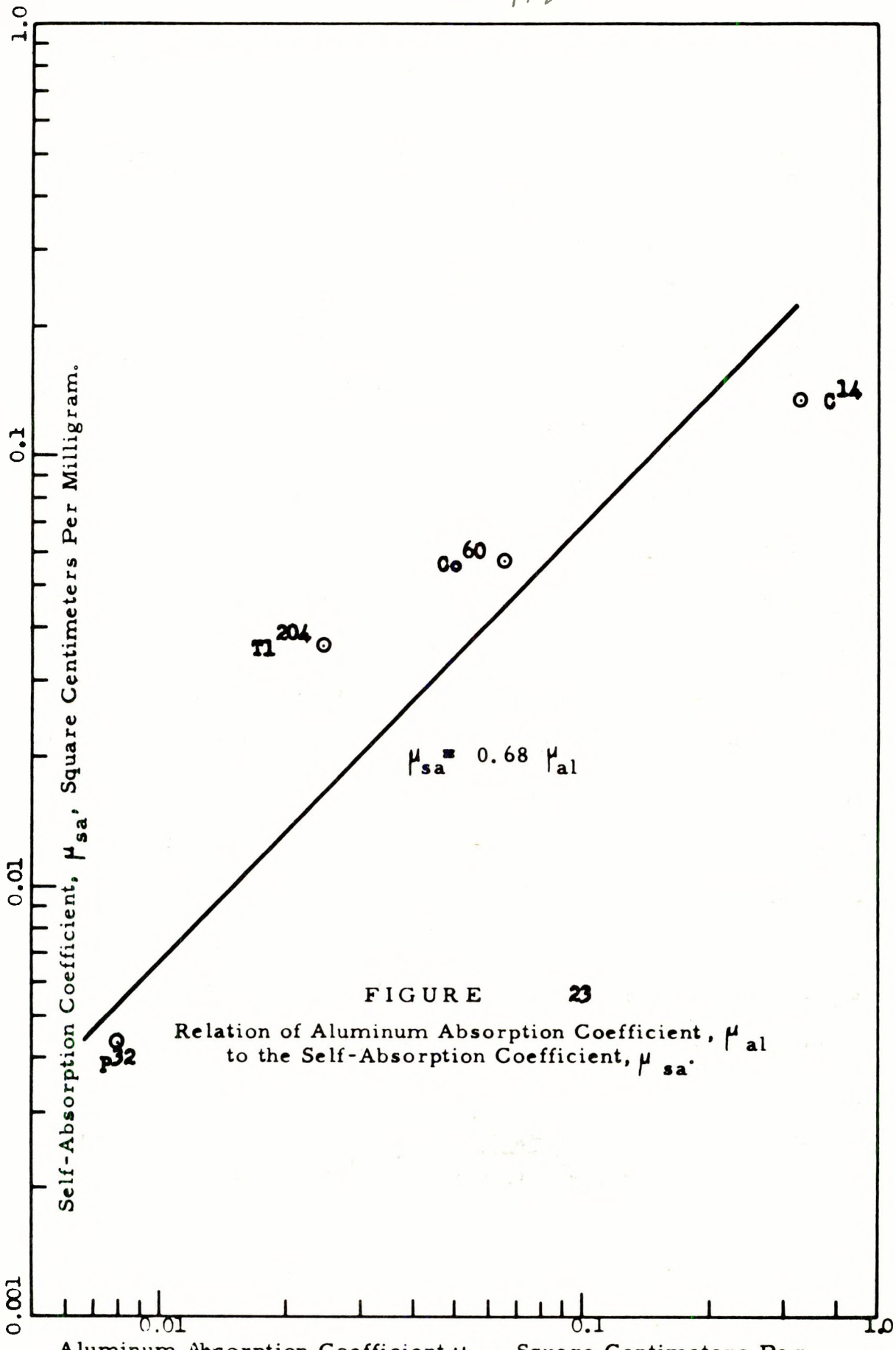

FIGURE 23

Relation of Aluminum Absorption Coefficient, μ_{al} to the Self-Absorption Coefficient, μ_{sa}.

coefficients. As the mean aluminum absorption coefficient for fallout material was found to be 0.014 square centimeters per milligram, a self-absorption coefficient of 0.0094 square centimeters per milligram, obtained from Figure 23, was used for purposes of calibration.

A plot was made of the aluminum absorption coefficients versus the logarithms of the ratio of the counts per minute at zero equivalent aluminum absorber to the known disintegration rate of these samples of calibrated nuclear species (Figure 24). The vertical axis intercept of a straight line of best fit drawn through these points gives the ratio of the counts per minute to the microcuries per sample if the equivalent aluminum absorption coefficient of the air gap and mica window were zero, that is if there were no air in the gap and no mica window in the counting instrument. The slope of this line (Figure 24) was used to calculate the mass of the air gap and mica window in terms of the added aluminum absorbers. This calculated value, 7.67 milligrams per square centimeter, is quite different from the actual mass of the air gap and end window (4.6 milligrams per square centimeter) and probably is due to the geometric and atomic weight differences.

A formula can, therefore, be obtained for converting fallout samples of a known net weight and for a given geometry from counts per minute to a disintegration rate for beta rays.

$$a = \frac{r_o \mu_{sa} x}{a\, e^{-m\mu_{al}} (1 - e^{-\mu_{sa} x})} = r_o F \qquad (5\text{-}10)$$

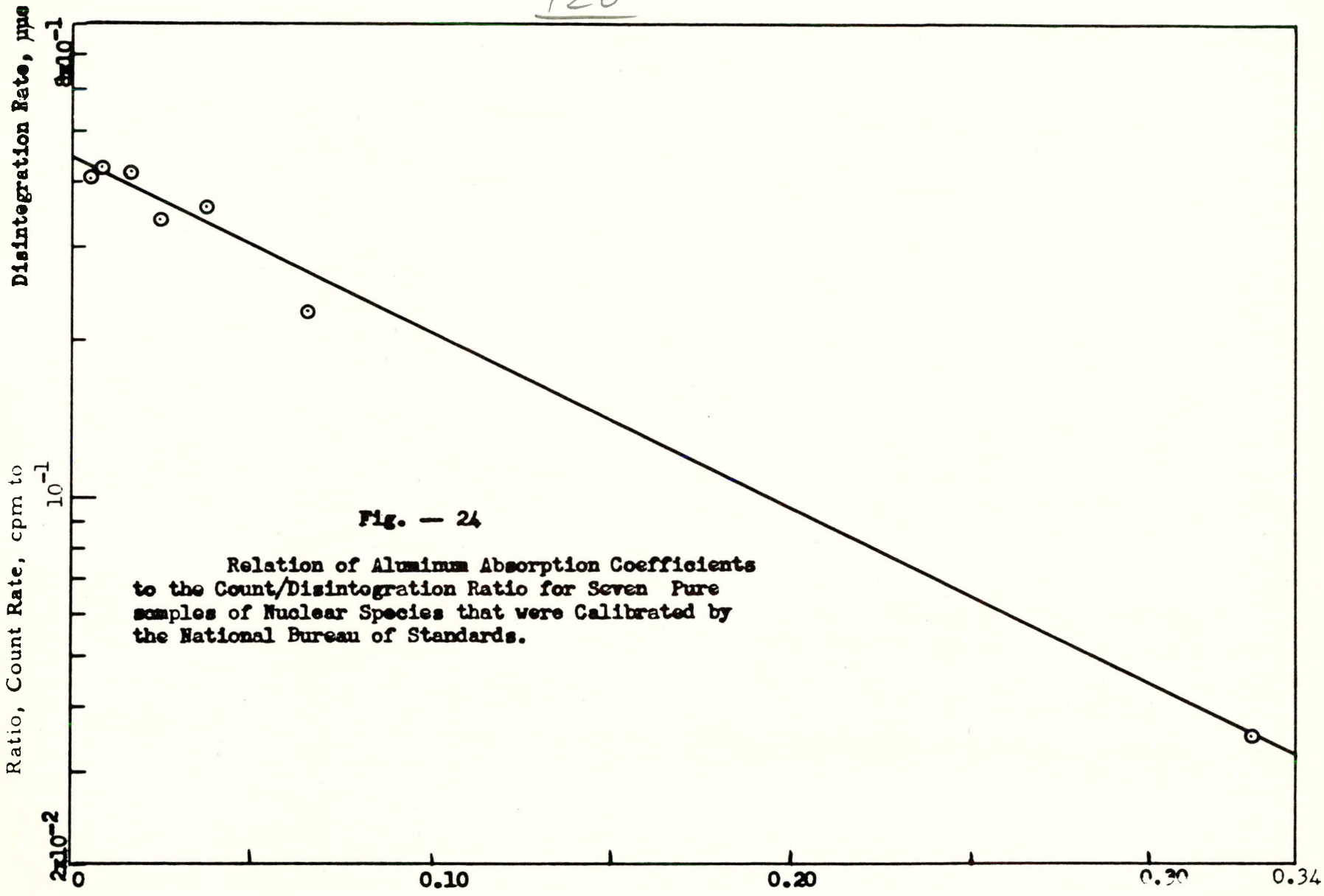

Fig. — 24

Relation of Aluminum Absorption Coefficients to the Count/Disintegration Ratio for Seven Pure samples of Nuclear Species that were Calibrated by the National Bureau of Standards.

where r_o is the net count rate of the sample in counts per minute

a is the vertical axis intercept of the line of best fit in Figure 24

α is the disintegration rate of the sample in micromicrocuries

m is the equivalent aluminum absorber mass of the mica and air gap in milligrams per square centimeter

μ_{al} is the mean aluminum absorber coefficient of long-range fallout in square centimeters per milligram

μ_{sa} is the estimated self-absorption coefficient of long-range fallout in square centimeters per milligram

x is the mass of the sample in milligrams per square centimeter

Calculations were made and a curve plotted which related the sample weight in milligrams per square centimeter to the factor F to facilitate the conversion to the sample disintegration rate in micromicrocuries. Similar curves were also drawn using an aluminum absorber coefficient of the mean (0.0140 square centimeters per milligram) plus or minus one standard deviation (0.0044 square centimeters per milligram). In order to avoid false accuracy, the weights and F factors were grouped in intervals set by one-third of this standard deviation (Figure 25 and Table 17). No important loss in precision came from this grouping (89).

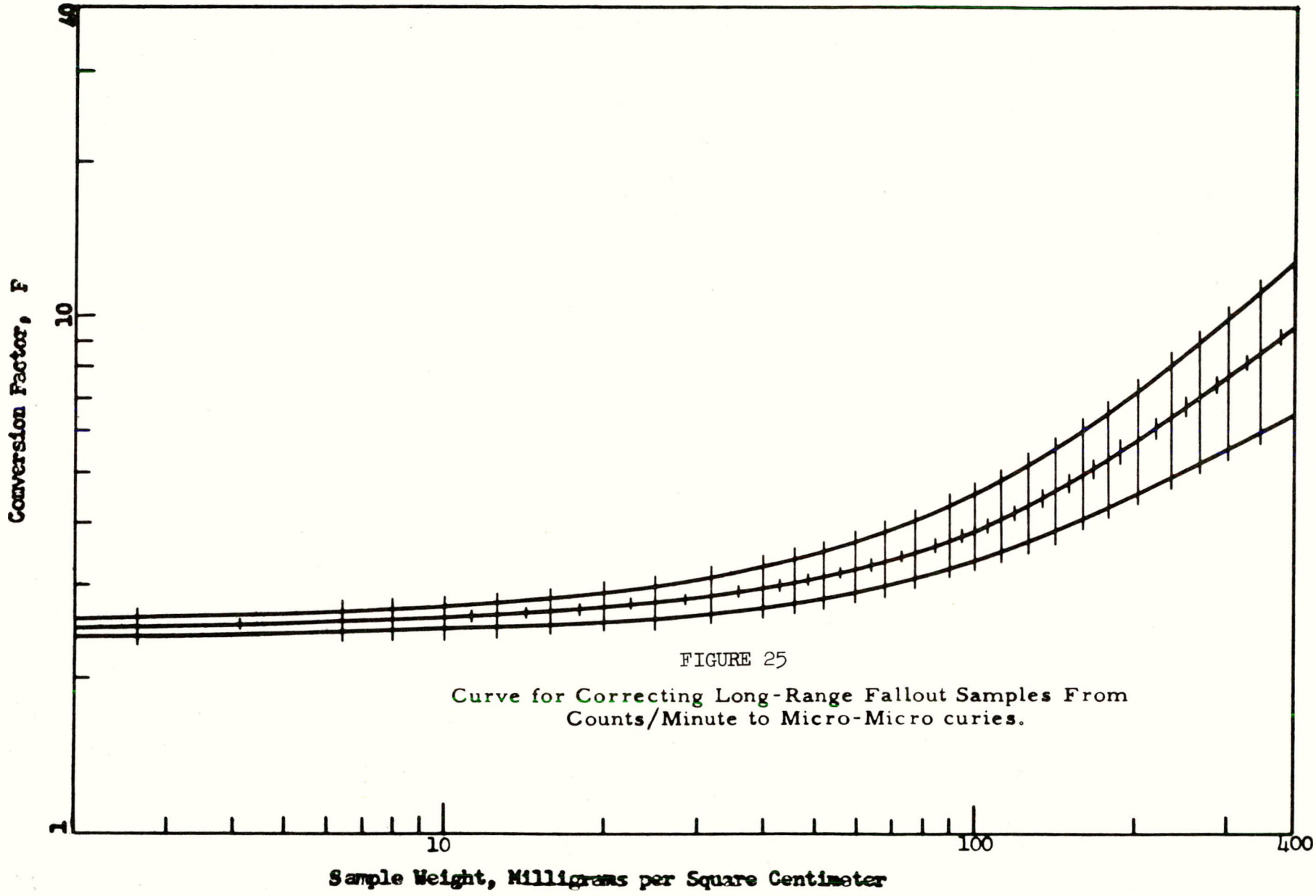

FIGURE 25

Curve for Correcting Long-Range Fallout Samples From Counts/Minute to Micro-Micro curies.

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TABLE 17

FACTORS FOR CONVERSION FROM NET COUNTS PER MINUTE TO DISINTEGRATION RATES

Net Wt. Sample (grams)	F	Net Wt. Sample (grams)	F	Net Wt. Sample (grams)	F
0.000-0.010	2.50	0.200-0.225	3.02	0.620-0.640	4.44
0.010-0.020	2.51	0.225-0.250	3.10	0.640-0.780	4.78
0.020-0.050	2.58	0.250-0.290	3.20	0.780-0.880	5.08
0.050-0.065	2.63	0.290-0.330	3.31	0.880-1.000	5.48
0.065-0.080	2.68	0.330-0.380	3.42	1.000-1.150	6.07
0.080-0.100	2.72	0.380-0.440	3.60	1.150-1.300	6.75
0.100-0.120	2.78	0.440-0.490	3.75	1.300-1.450	7.39
0.120-0.160	2.85	0.490-0.550	3.95	1.450-1.700	8.10
0.160-0.200	2.95	0.550-0.620	4.18	1.700-1.950	9.10

CHAPTER VI

MEASUREMENTS OF LONG-RANGE FALLOUT

A. SURVEY PLOTS

1. Eastern Massachusetts

In order to show the effect of nuclear detonations on the surface waters of eastern Massachusetts, the author made a survey plot (Figure 26) that relates the dates of announced detonations with (a) the estimated daily precipitation at Cambridge, Massachusetts; (b) the concentrations of beta radioactivity that fell on the Cambridge campus of Harvard University; (c) the average concentrations of radioactivity in the surface waters of the collection area; (d) the daily areal deposition of radioactivity in Cambridge; and (e) the areal "runoff" of radioactivity from watersheds in the collection area.

At the top of Figure 26 are noted the dates of announced detonations from April, 1952, to the end of 1953. "N" stands for detonations in Nevada, "E" for Eniwetok, "S" for Siberia, and "A" for Australia. It is probably incomplete, for the United States Atomic Energy Commission discontinued, at least temporarily, the publication of the detection of foreign tests after announcing the first two detonations of the 1953 series of the Soviet Union.

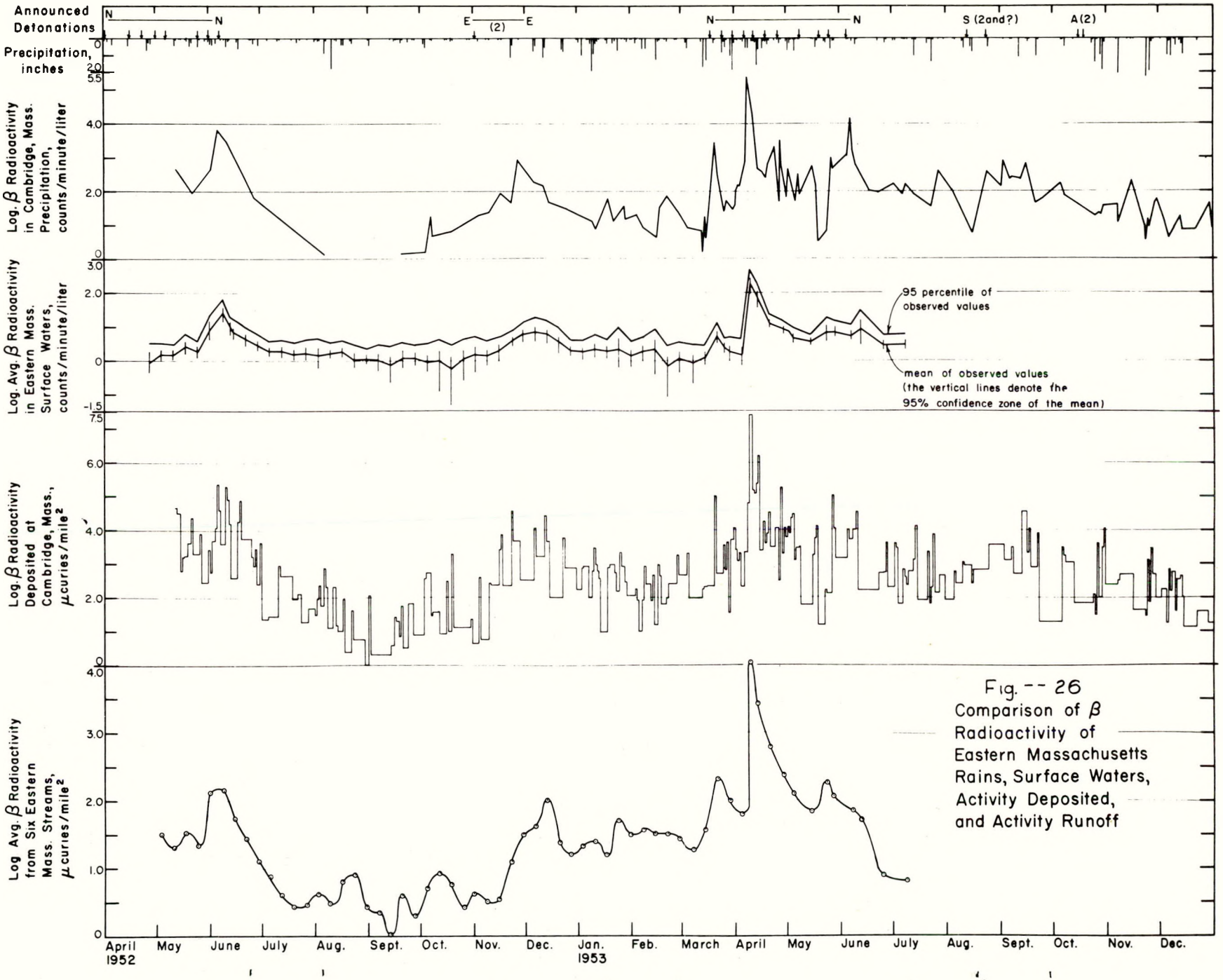

Fig. -- 26
Comparison of β Radioactivity of Eastern Massachusetts Rains, Surface Waters, Activity Deposited, and Activity Runoff

a. Precipitation. Just below the bomb detonation plot is shown the estimated daily precipitation in inches at Cambridge, Massachusetts. These values were obtained by averaging the United States Weather Bureau's measurements of daily rainfall at Logan International Airport, Blue Hill Observatory, and Waltham, Massachusetts.

b. Radioactivity of precipitation. The plot below that of the rainfall shows a line connecting the plotted values of the concentrations of radioactivity in terms of counts per minute per liter of precipitation in samples collected at Harvard at the time of first counting as registered by the automatic scaling equipment described in Chapter III. With the exception of a few rains in the spring and summer of 1952, samples were collected and measured from all precipitations from May 12, 1952, through December 31, 1953. The time of first counting of these samples was almost always from one to three days after the precipitation, which is sufficient for the short-lived radium A, B, C, and C' (Po^{218}, Pb^{214}, Bi^{214}, Po^{214}, respectively) to decay to low concentrations, as well as thoron (Em^{220}) and its short-lived daughters. Although repeated measurements were made on each sample of precipitation (as on almost all samples collected), no correction for the sample radioactivity decay between collection and first counting was made for this plot. The mass of the evaporated precipitation sample residue was so low that self-absorption caused an average decrease in count rate of less than 10%. No rinses of the collecting apparatus for dry fallout were made. At least a portion of the dry fallout that occurred must have been deposited in the collection apparatus (described in Chapter

III) and washed into the sample jug by the following rain. The experience of the author indicates that most long-range fallout radioactivity is brought to the ground by precipitation. However, good measurements have been made at Rochester, New York, that show areal concentrations of dry fallout almost as large as the largest recorded there accompanying rain.

In a comparison of the dates of detonation with the dates of high concentrations of radioactivity in the eastern Massachusetts rains (in counts per minute per liter), several effects are apparent. As mentioned in Chapter I, it is here shown that not only do individual detonations fail to cause equal concentrations of radioactivity in long-range fallout but also that some detonations caused no deposition in eastern Massachusetts, at least at the levels of sensitivity of these measurements. This result could be expected, since it is quite possible that air masses over Nevada at the time of a detonation could take paths such that virtually none of the debris could be deposited in eastern Massachusetts, at least not until after circling the earth. Another effect is that the build-up to the peak concentration of radioactivity in the fallout is faster for relatively nearby detonations (Nevada) than for distant ones (Eniwetok and the Soviet Union). Also, six of the nineteen Nevada detonations gave higher counts per minute per liter of precipitation than either of the 1952 Eniwetok or any of the 1953 Soviet detonations. The peaks of November 15 and 29, 1953, could be from the Australian or the Siberian experiments. As with all plots of measured radioactivity, the ordinate values are base-10 loga-

rithms of the measured values. It is important that the concentrations of radioactivity in the precipitation samples were sufficiently high and the sample preparation and counting techniques used good enough that - with a single exception - no negative average net count rate of precipitation sample was obtained.

c. Radioactivity of surface waters. Below the rain concentration plot is shown the average beta radioactivity concentration in samples of surface water collected from streams and reservoirs once each week at Stations Number 1 through Number 14 (Chapter III). The lower of these two transverse lines connects plotted points representing the average beta radioactivity concentrations in terms of the net count rates at first counting. The first count rate per liter was made on the autoscaler, almost always three days after collection. Again, for this plot no correction was made for radioactivity decay between the time of collection and first counting, although decay measurements of all of these samples were made for other studies. Also, no correction was made for this plot for self-absorption of these samples (Chapter V). An error of less than 10% was introduced, because this correction was ignored. The ends of the vertical lines on this plot represent the 68% confidence zone of the mean of the statistical population from which these samples were drawn. The upper of these two transverse lines represents the upper 95% confidence limit of the mean net count rate. The concentrations of radioactivity in the surface waters were usually one to two orders of magnitude lower than those in the rain, and during one period in which there were no announced detonations (August-October,

1952), the levels fell to an arithmetic mean of about 1.16 counts per minute per liter. In both the plot of the rain count rates per liter and that of the average surface water count rate per liter, the lines connecting the plotted points are added so as better to show these changes in concentration. From the sampling and counting techniques used, it can be seen that the debris from the Nevada detonations was still detectable in the surface waters about three weeks after the end of each of the series. The increased net count rate caused by the November, 1952, Eniwetok detonations appears to have maintained the level of radioactivity in eastern Massachusetts surface waters above pre-detonation values for more than two months.

d. Areal concentration of precipitation radioactivity. The second graph from the bottom of Figure 26 shows a curve connecting plotted points representing the logarithms of the calculated daily deposition of radioactivity in units of microcuries per square mile at Cambridge. These values were obtained by multiplying the net count rates per liter of rain at first counting by the amount of rain deposited per square mile each day. The quantity of rainfall was calculated by averaging the daily measurements on the precipitation gages located at Boston's Logan International Airport, Waltham, and Blue Hill Observatory, as reported by the United States Weather Bureau. For periods in which there was no rain, the radioactivity of the previous rainfall (in microcuries per square mile) was averaged over the entire period, including the day the rain occurred. No correction was made for natural radioactivity, self-absorption, or decay of radioactivity of the samples between collection and counting.

e. Areal concentration of runoff radioactivity. The lowest plot on Figure 26 shows the average areal runoff of beta radioactivity in microcuries per square mile as observed for streams in the collection area. The calculations for this plot were made using the surface water runoff data for the six streams -the Assabet, Charles, Concord, Quaboag, and Ware Rivers and Kettle Brook (formerly called the Blackstone River)- in the sample collection areas that are gaged by the United States Geological Survey. This average areal runoff value, in liters per square mile per day, was multiplied by the average count rate of samples from these same streams in counts per minute per liter and converted to microcuries (Chapter V). The values of the radioactivities of these streams used for this plot were the count rates per liter at the time of first counting of the samples. No correction was made for measured natural radioactivity of the streams, which probably averaged less than one count per minute per liter.

The coefficient of runoff of radioactivity is defined for this thesis as the ratio of the cumulated areal runoff of radioactivity caused by fallout as measured in the receiving streams to the areal concentration of radioactivity at the time of deposition. A rough computation of this coefficient was made using the data for the lowest curve (Figure 26) and the histogram above it. These data give an estimated overall value of the "runoff coefficient" of 0.0099 or about 1% for fallout beta radioactivity. The results of more accurate calculations are given in Table 29 for the individual and combined streams during and following periods of high fallout radioactivity.

2. Rochester, New York, area

Figure 27 gives a comparison of various radioactivity measurements conducted by other investigators in the Rochester, New York, area between 1951 and 1953.

a. Detonations. As in Figure 26, the detonation dates are shown at the top of the plate.

b. Precipitation. The daily precipitations in inches at the Rochester, New York, airport are shown in the graph just below that of the announced detonations.

c. Radioactivity of precipitation. Below the plot of detonation dates is a broken line connecting points representing the logarithms of sample fallout radioactivities, both dry and precipitation-borne, in counts per minute per 100 square feet, as measured at their first counting and plotted for the day of collection. In almost all cases the sample was measured on the day of collection or the day following. During and immediately following test periods and often in periods remote from detonations, the collection basis was rinsed with distilled water and this rinse water was processed and counted in the same fashion as the rain and snow samples. Details of the techniques used in the measurements for this figure are given in Chapter III.

An inspection of this plot shows that the detonations at Eniwetok in 1951 raised the levels of radioactivity in the precipitation and apparently in Lake Ontario to a small extent. The first announced Soviet

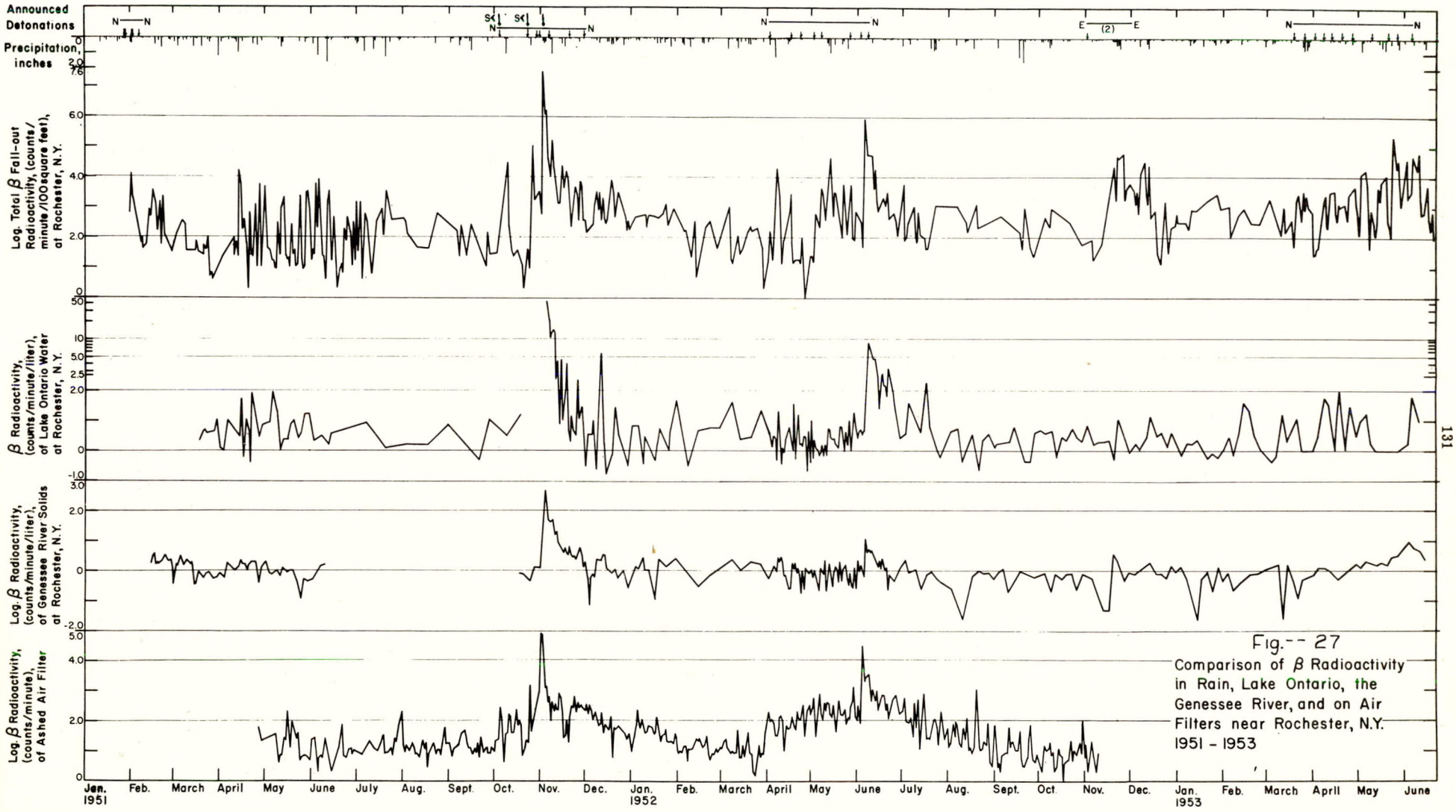

Fig.-- 27
Comparison of β Radioactivity in Rain, Lake Ontario, the Genessee River, and on Air Filters near Rochester, N.Y. 1951 - 1953

detonation in their test series in the fall of 1951 was registered by the increased net count rates of rain and air filter samples.

It is interesting to note that in the fall of 1951 there was one fallout that caused a sample count rate (per volume of rain) more than two orders of magnitude higher than any other. To a lesser extent one fallout was dominant over all the others that occurred in eastern Massachusetts during the Nevada spring series of 1953. In the Nevada spring series of 1952 the highest concentration of fallout in rain was more than one order of magnitude above the next highest. Debris from the November, 1952, experiments at Eniwetok was detected in the precipitation. From the start of the monitoring of rain early in 1951 until April 16, 1953, the values shown are of the total beta activity. After this date the values represent the amount of beta radioactivity (in counts per minute per liter) that was retained by a Whatman # 40 paper after filtration of the rain.

d. Radioactivity of Lake Ontario water. Below the plot of the areal concentration of fallout beta radioactivity is a line connecting points representing the beta radioactivity of Lake Ontario water in counts per minute per liter. During periods remote in time from detonations, the levels of radioactivity in the lake often fell to low values; with the counting techniques used, the statistical fluctuations allowed the recorded net count rate per liter occasionally to fall even to negative values. Because this curve and the one preceding it are principally for display purposes, it was decided to choose a scale arithmetic below 2.5 counts per minute per liter and logarithmic above this value.

The debris from the 1951 Eniwetok experiments apparently raised the count levels in Lake Ontario slightly. In the Nevada test the precipitation count rate peak of November 1, 1952, was followed within two days by an increase in count rate in the lake water 117-fold higher than the last reading taken before the fallout. Unfortunately, no measurement was made of the radioactivity in Lake Ontario water between mid-October and November 3, 1951. The graph shows that fission-produced gross beta radioactivity in the lake had decreased below the levels of sensitivity of measurement about a month after the end of this series.

In the spring series of 1952, the large fallout of June 4 was followed two days later by a peak count rate in the samples of Lake Ontario water. The lake water samples for these measurements were taken from the treatment plant inlet (Chapter VII) that receives water directly from a pipe, the intake of which is located 1.5 miles from shore and fifty-five feet below the water surface. In this case, the mechanisms that transported the radioactivity appear to have taken about two days to cover this vertical distance of fifty-five feet.

One consideration at this point is of interest: although the ratio of the beta radioactivity of the Genessee River to that of Lake Ontario was about 5.6 on November 3, 1951 (following the large November 1, 1951, fallout), the ratio of these measurements on June 6, 1952, was only about 0.63. Therefore, the two-day delay between the peak fallout of June 4, 1952, and the peak radioactivity of the lake on June 6 could be explained by the supposition that a still larger fallout occurred in the lake beyond the water treatment plant intake than at Rochester or in the Genessee River near Rochester.

e. Radioactivity of Genessee River water. Just below the middle of Figure 27 is shown a plot connecting points representing the logarithms of the beta radioactivities in the Genessee River, which flows through Rochester, New York. The 1951 Eniwetok experiments caused little increase in these measurements, but the large depositions of November 1, 1951, and June 4, 1952, gave significant peaks. Surprisingly, the relatively large deposition in November, 1952, hardly increased the gross beta count rates above those measured before this test series. This failure in increase could be explained as the occurrence of a relatively intense local fallout at Rochester with little deposition on the watershed of the Genessee River upstream from Rochester. After March 22, 1953, the river, lake, and treatment plant samples were processed by the same techniques but in a different laboratory in the Rochester, New York, area.

f. Radioactivity of ashed air filters. The lowest plot on the figure connects points representing the logarithms of the net beta count rates of ashed air filters. Outside air was pulled through the thirteen-by-fifteen-inch Airway filters for from one to three days; then the filter was removed, ashed, counted, and reported as the net count rate at first count. For cases in which air was drawn through a filter for more than one day, the measured net count rates are plotted on the median of the dates that the filters were used. It is seen that this plot indicates the same major peaks of fallout as do the plots of the rain, lake, and river samples. The fluctuations, other than those inherent in the statistics of counting, for periods neither during nor

immediately following anncunced detonations probably are ceused to a large extent by temporary increases in concentrations of naturally-occurring radon (Em^{222}) and thoron (Em^{220}) and their radioactive decay products. This increased concentration of natural radioactivity can be caused by temperature inversions in the atmosphere.

It can be seen from these two survey plots that rain, surface water, and air filter measurements can all be used in monitoring for fallout.

B. DECREASE IN RADIOACTIVITY OF SAMPLES WITH AGE

1. Precipitation containing long-range fallout

As discussed in Chapter I, for a given amount of fission debris, the emission rate of gamma rays or beta particles varies as a constant power of the number of time units after detonation. That is, $A = A_o t^{-1.2}$, where A is the emission rate at any time after detonation, A_o is the emission rate at unit time after detonation, and t is the number of time units after detonation. A, A_o, and t must all be based on the same time units, such as seconds, hours, days. In all cases for this paper, the unit of time is a day. If a detonation date is assumed, the elapsed time after detonation of repeated measurements of the same sample can be plotted versus their respective net count rates on double logarithmic paper. The date one day after fission is given a value of 1, that ten days after fission, a value of 10, etc. If the date of fission is properly chosen and if the decay formulation stated above holds, a

line of best fit through the plotted points should give an approximately straight line; the radioactivity of the sample obtained one day after fission may be estimated by extrapolation of this line of best fit to the time t_1. If the detonation date actually occurred later than assumed for the plot, the line should be concave upward.

a. Intercontinental carriage of radioactive debris. Although many decay measurements were made in research for this thesis, principal discussion will be limited to the fallout from two periods. The decay of rain-precipitated samples from detonations that took place at great distances from the collection site is illustrated by measurement of the rains occurring at Cambridge, Massachusetts, from late August to early October, 1953; these will be discussed. This period follows the two announced Siberian detonations in August. The first of these detonations was stated to be of the fusion type. Presumably these were only the first in a test series.

Figure 28 shows the decay of two samples taken from the thunder shower that occurred at Cambridge August 24, 1953. If a Cambridge date of August 12 is assumed for the detonation, it is seen that the decay follows well the above-discussed formulation. Decay curves for the rains of September 2, 6, 7, 12-13, 15, 16-17, 20-21, October 5, and October 7 are shown in Figure 29, Figure 30, and Figure 31. All of these curves are based on an assumed August 12 detonation date. The possibility that the radioactivity in the September 7 rain could have come from a detonation occurring on August 23 was tested by plotting the measured radioactivities of the sample on this basis (Figure 30). It can be seen by

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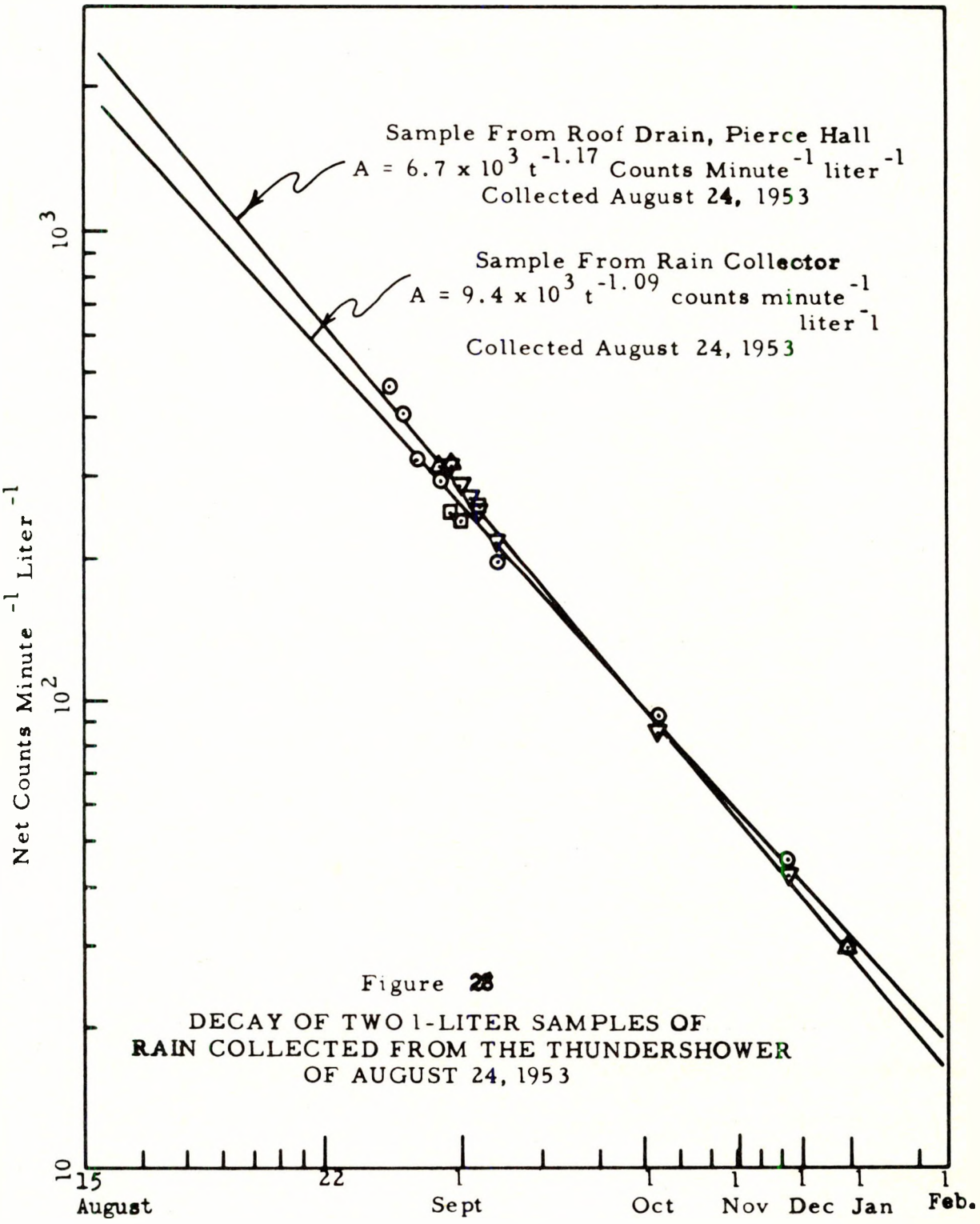

Figure 28

DECAY OF TWO 1-LITER SAMPLES OF RAIN COLLECTED FROM THE THUNDERSHOWER OF AUGUST 24, 1953

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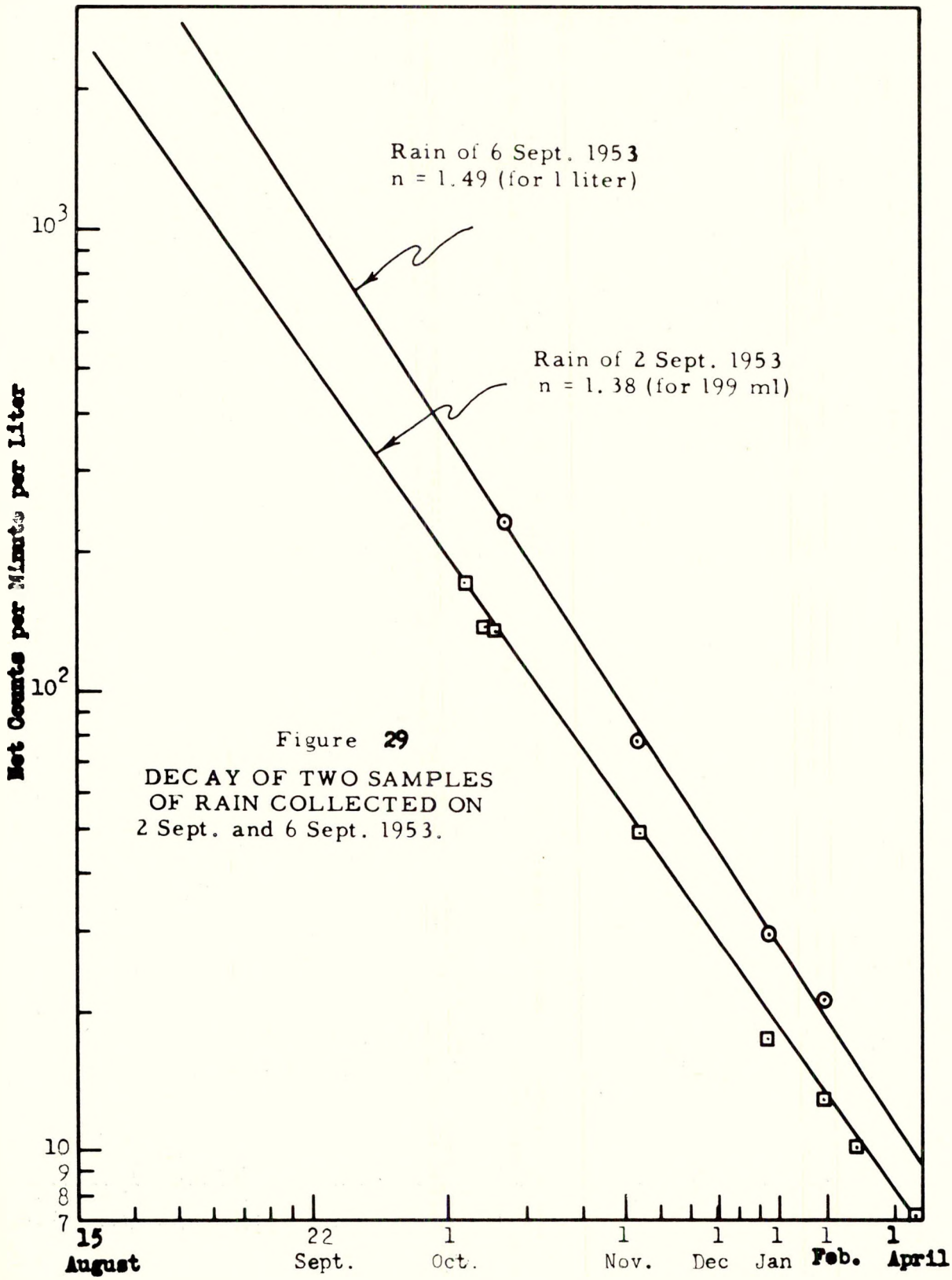

Figure 29

DECAY OF TWO SAMPLES OF RAIN COLLECTED ON 2 Sept. and 6 Sept. 1953.

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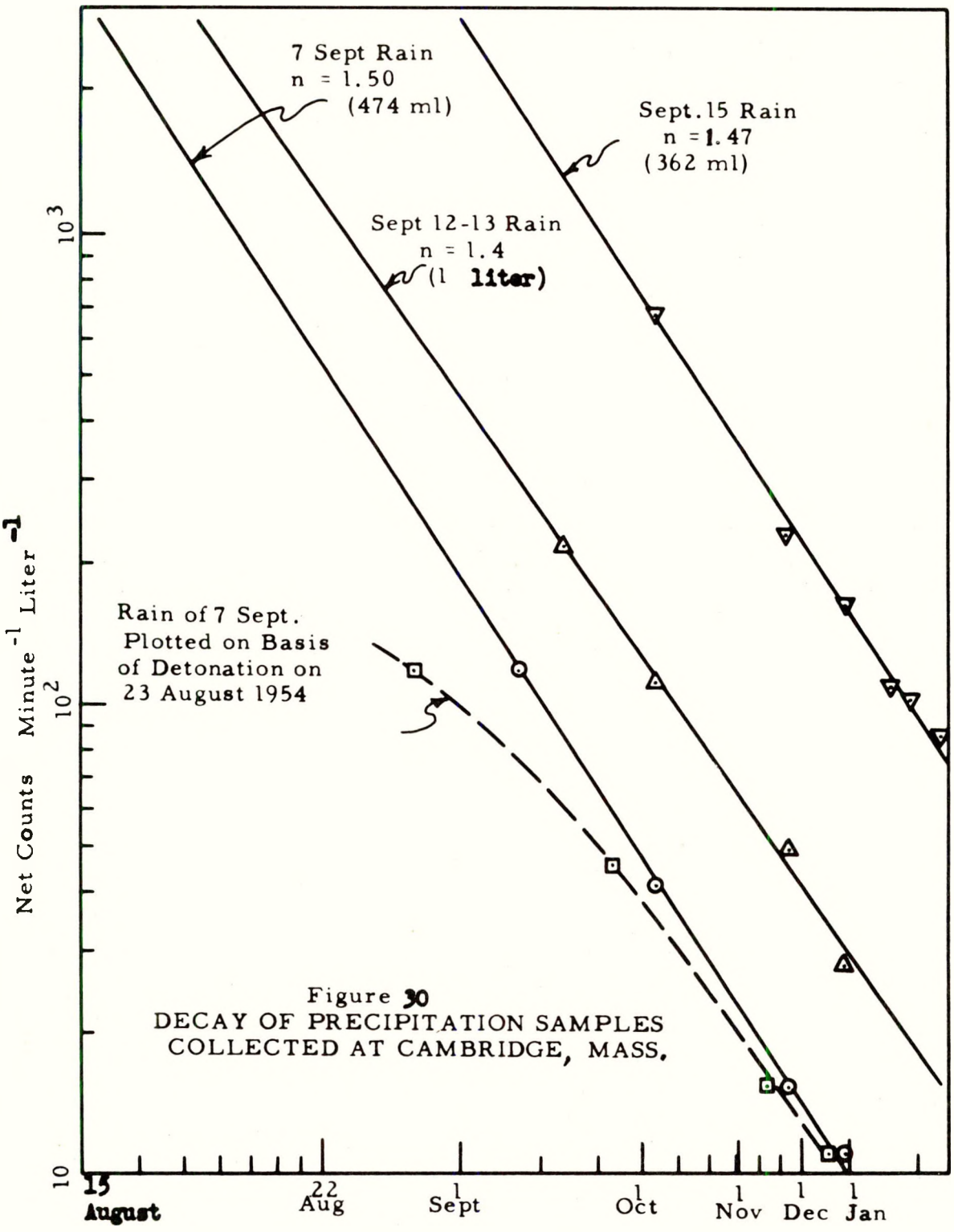

Figure 30
DECAY OF PRECIPITATION SAMPLES
COLLECTED AT CAMBRIDGE, MASS.

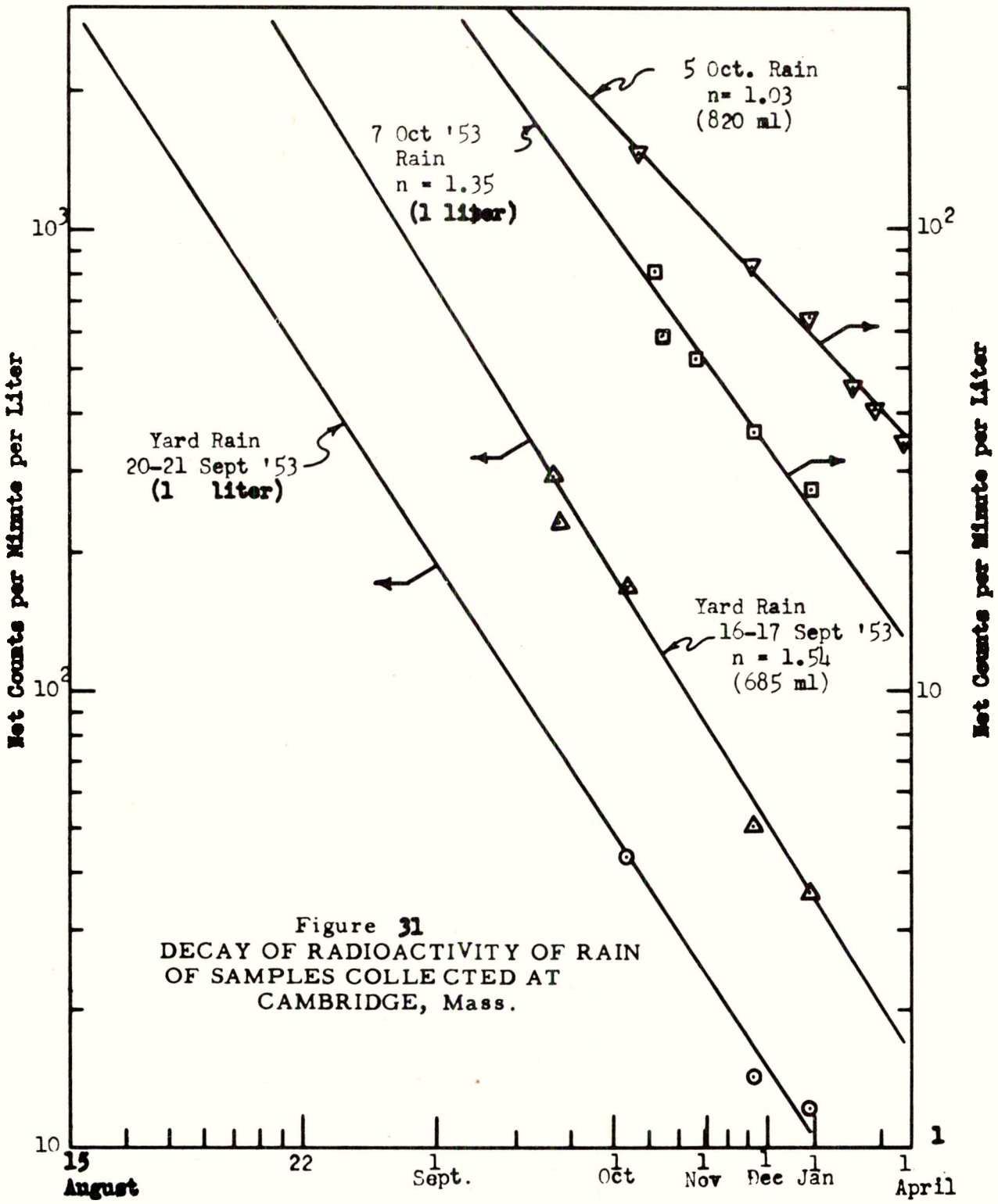

Figure 31
DECAY OF RADIOACTIVITY OF RAIN
OF SAMPLES COLLECTED AT
CAMBRIDGE, Mass.

the non-linearity of this plot that August 12 is a more reasonable detonation date. No correction was made for natural and antecedent detonation radioactivity, which averaged 130 counts per minute per liter on first count for the eight rains previous to August 24, which occurred in July and August, 1953.

From these observations it can be concluded that fallout from detonations on the other side of the earth, could, if in sufficient number and/or size, raise the levels of radioactivity in rain and surface waters to such a concentration as to merit the attention of sanitary engineers.

b. Transcontinental carriage of fallout. The largest single deposition of radioactivity detected in eastern Massachusetts during the monitoring period from April, 1952, to January, 1954, followed the April 6, 1953, Nevada detonation. Many of the conclusions drawn in this chapter are based on that fallout and it will be discussed as an example of fission debris transported over a distance of about two thousand miles before deposition. As will be shown, there is reason to believe that the fallout characteristics are similar to those described in Reference 1.

Figure 32 shows the decay measurements on two samples collected at Harvard University at approximately 16:00 hours April 8, 1953. Coincidence corrections were made on the first count rate taken on the 100-milliliter sample. The decay follows the formulation $A = A_1 t^{-n}$; however, the difference in the n value decay parameter from 1.2 as given in (1) cannot be due to statistical errors of counting alone.

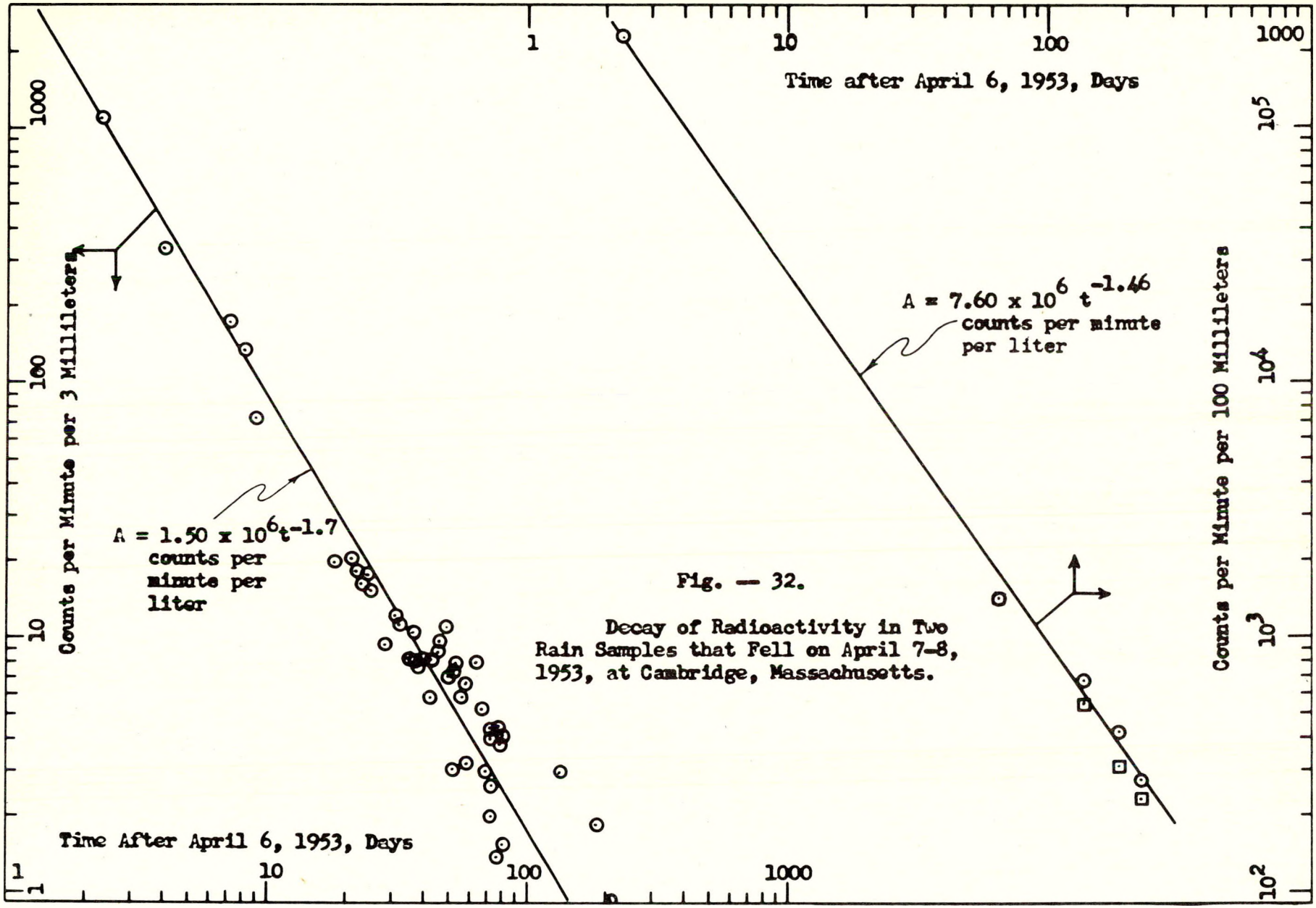

Fig. — 32.

Decay of Radioactivity in Two Rain Samples that Fell on April 7-8, 1953, at Cambridge, Massachusetts.

Heslep (90) has observed a deviation from the hyperbolic formulation for a fallout that occurred in California shortly after a Nevada detonation in which it decayed more slowly than by the parameter $n = 1.2$ between the third and fourth day and thence more rapidly for ninety-six days. The samples were collected on air filters during Nevada tests. This early effect was not observed by the author.

Table 18 summarizes the n values obtained in the study of decay of rains following the April 6 detonation. These rains were collected routinely on the watersheds of the same eastern Massachusetts streams and reservoirs sampled for this thesis. Figure 33 and Figure 34 show the decay plots of the rains collected on April 8 and 9 and on April 13. For the rains of April 7 and 8, which were collected on April 8 and 9, the "average" formulation (calculated mean count rate at one day after detonation and mean n-value) for the decay of samples from eighteen stations was found to be $A = 3.22 \times 10^{5} t^{-1.40}$ counts per minute per liter. Mr. B. L. Rosenthal of the Lawrence Experiment Station of the Massachusetts Department of Public Health made a series of measurements to determine the decay of radioactivity in 100 milliliters of rain collected in the local gage during the night of April 7-8. When his eight die-away measurements (between April 8 and July 21, 1953) were plotted, the line of best fit gave the formula $A = 3.5 \times 10^{5} t^{-1.47}$ counts per minute per liter. These measurements were made using the same brand and type of equipment and sample container as used in the author's study.

Figure 35 shows the percentile distribution of logarithms of the decay parameters for rains collected April 8 and 9, 1953, in the eastern Massachusetts sampling area.

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TABLE 18

PARAMETERS FOR THE DECAY CURVES (n-VALUES) OF RAIN SAMPLES COLLECTED ON EASTERN MASSACHUSETTS WATERSHEDS

Station*	April 8		April 13	
	A_1 Counts per min. per liter	n	A_1 Counts per min. per liter	n
2	5.6×10^4	1.24	3.1×10^5	2.20
3	4.5×10^4	1.43	1.4×10^6	2.77
4	1.43×10^5	1.33	9.3×10^5	2.47
6	3.0×10^5	1.38	2.4×10^6	2.98
7	8.0×10^4	1.41	2.1×10^7	2.02
8	1.0×10^6	1.46	9.9×10^5	2.33
9	2.4×10^6	1.30		
10	1.17×10^5	1.76	6.6×10^5	1.19
11	8.8×10^4	1.27	3.1×10^7	2.72
12	3.4×10^4	1.12	8.5×10^3	1.37
	April 9			
I	1.18×10^4	1.39	1.8×10^5	2.19
II	5.95×10^3	1.19	1.4×10^4	1.37
IV(14)	8.0×10^3	1.29		
V(13)	2.93×10^5	1.92	1.5×10^5	1.75
VII	1.6×10^4	1.28	1.1×10^6	2.63
VIII	7.4×10^4	1.61		
IX	1.58×10^4	1.42	1.2×10^5	2.07
Mean		1.40		2.21
Standard Deviation		0.19		0.58
Standard Deviation		0.046		0.15
Mean (log n)		0.143		0.328
Standard Deviation (log n)		0.066		0.15

* See Chapter III for detailed descriptions of the station locations.

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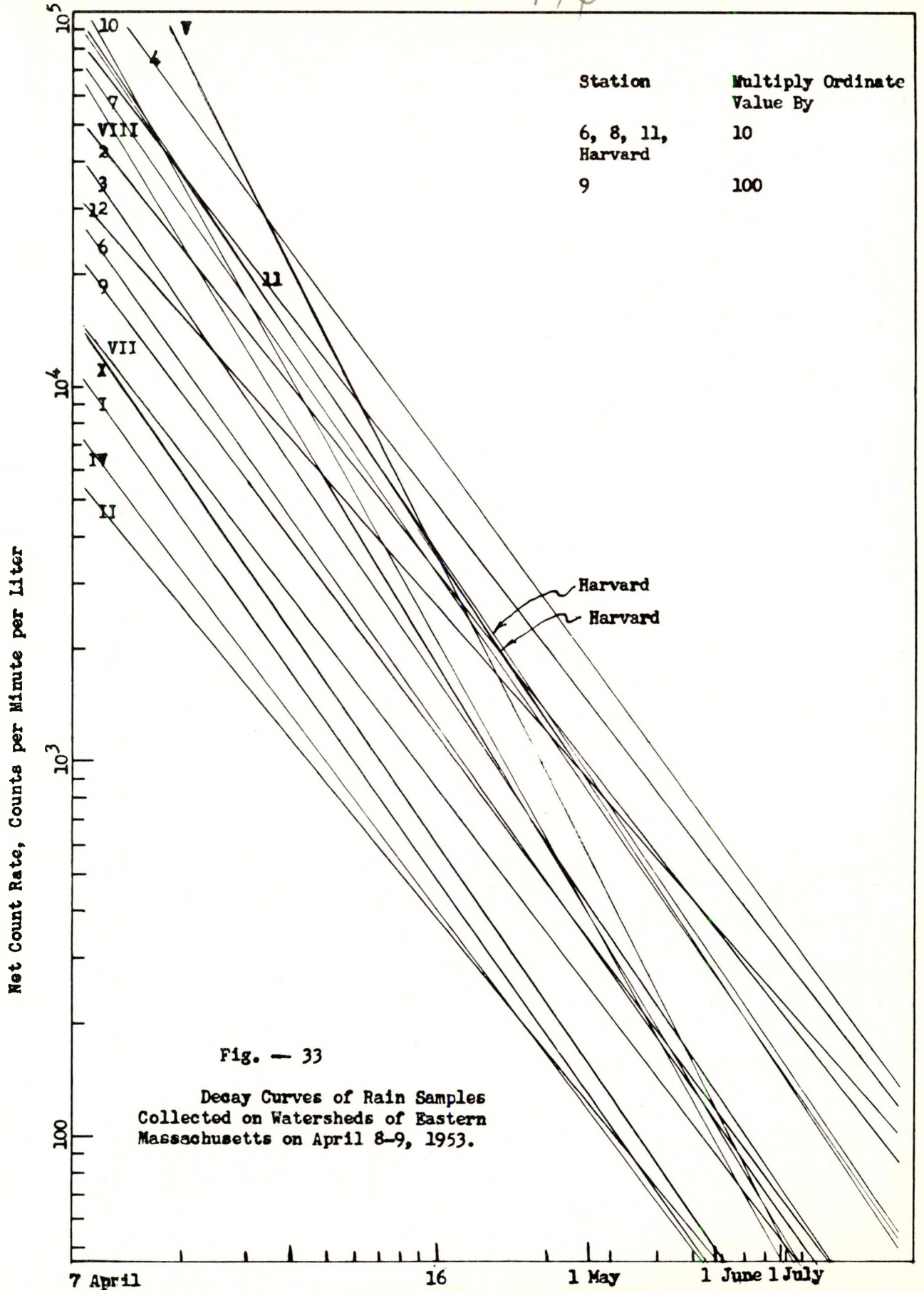

Fig. — 33

Decay Curves of Rain Samples Collected on Watersheds of Eastern Massachusetts on April 8-9, 1953.

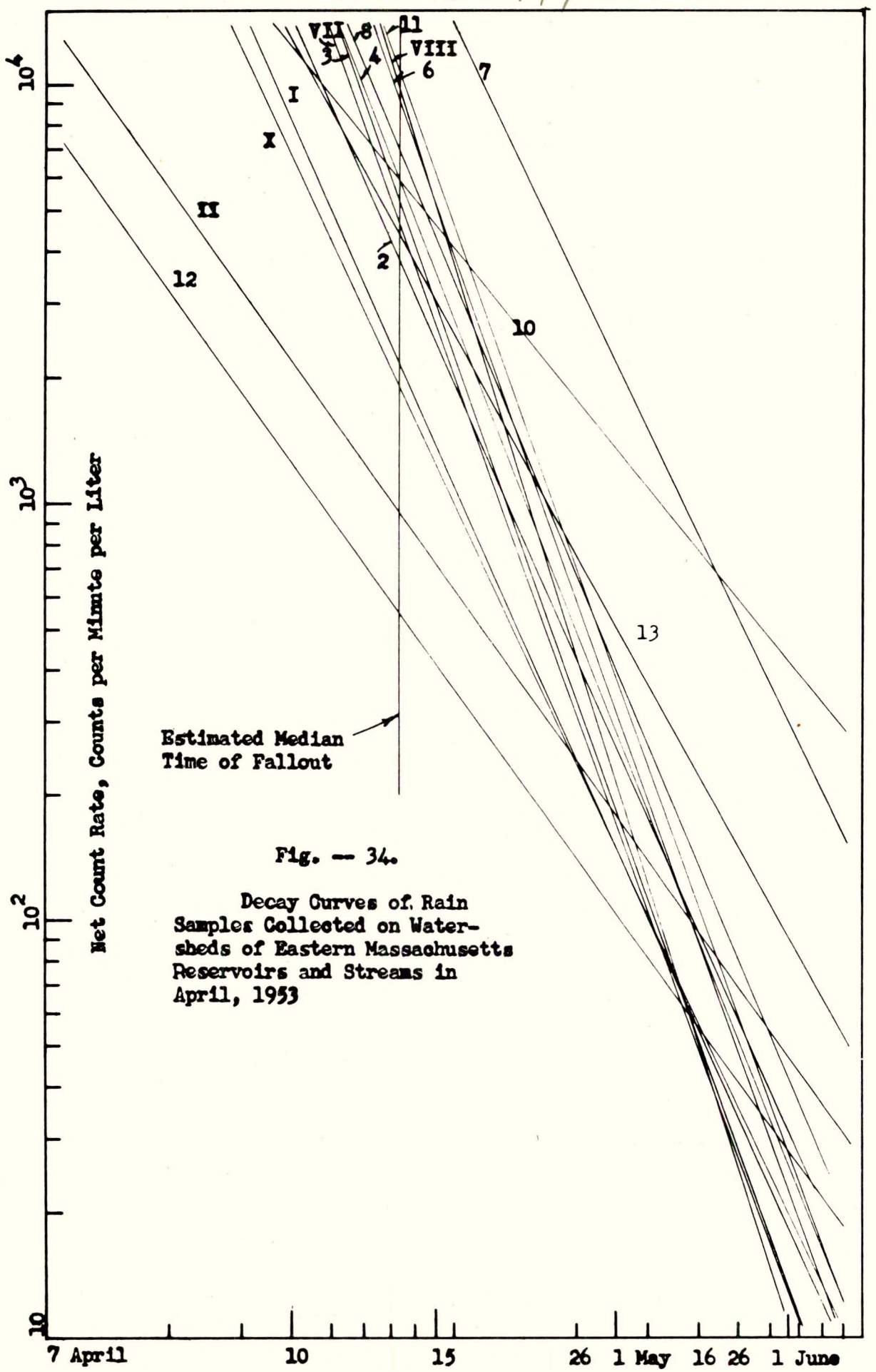

Fig. — 34.

Decay Curves of Rain Samples Collected on Watersheds of Eastern Massachusetts Reservoirs and Streams in April, 1953

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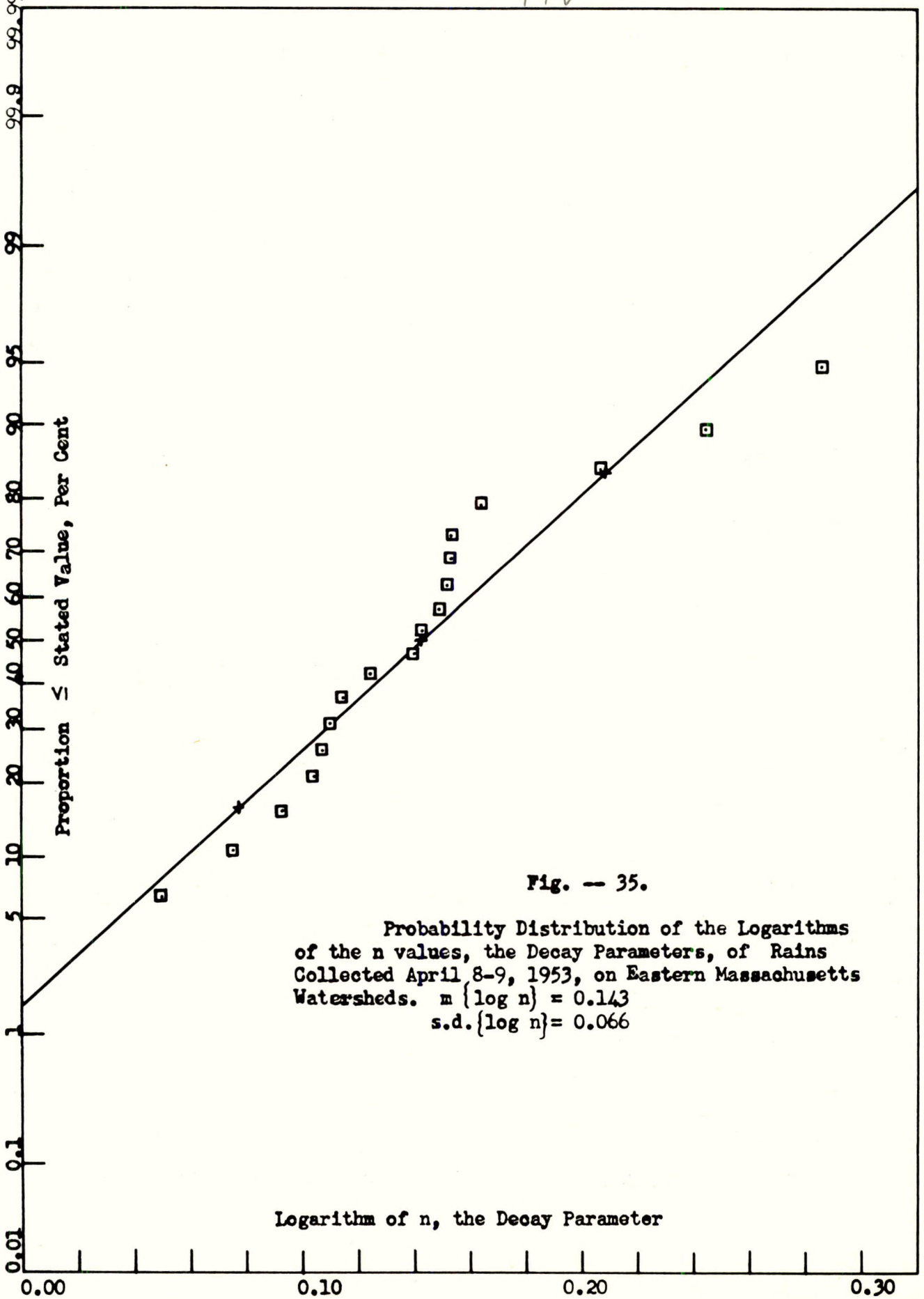

Fig. — 35.

Probability Distribution of the Logarithms of the n values, the Decay Parameters, of Rains Collected April 8-9, 1953, on Eastern Massachusetts Watersheds. m {log n} = 0.143
s.d.{log n}= 0.066

C. DISTRIBUTION OF CONCENTRATIONS OF RADIOACTIVITY IN A SINGLE FALLOUT

The distribution of concentrations of radioactivity in samples collected at the various stations following the large fallout in eastern Massachusetts on April 7 and 8 was studied. A calculation can be made of the average concentration of radioactivity in the rain at the time of deposition by extrapolating the measured decay curve. As will be shown later in this chapter, preliminary measurements indicate that the concentration of fallout radioactivity is not constant with time during a storm. Also, the measurements indicate that the radioactivity is not deposited on the ground in an exponential fashion.

The median time of the rain over the collection area on April 7 and 8 is estimated, principally from the rainfall data of the United States Weather Bureau, to be about thirty-eight hours after the April 6 Nevada detonation. Table 19 gives these estimated beta disintegration rates at time of fall in both count rates and micromicrocuries per liter. These values were ranked and the probabilities of the disintegration rates were computed according to Thomas (91) for plotting on probability paper. Figure 36 shows that these rates fit a logarithmic normal distribution rather closely. A calculation was made, according to Hald (92), that showed that there was no reason to doubt that this was a typical logarithmic normal distribution.

1. Hazards in drinking rain water

a. Eastern Massachusetts. If a small extrapolation of the

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TABLE 19

ESTIMATED RADIOACTIVITY OF RAIN SAMPLES IN EASTERN MASSACHUSETTS AT TIME OF FALL, APRIL 7-8, 1953

Station	cpm/l at fall	μμc/l at fall	log μμc/l	Rank	P%
2	3.7×10^4	9.2×10^4	4.96	7	35
3	2.8×10^4	7.0×10^4	4.84	6	30
4	9.2×10^4	2.3×10^5	5.36	12	60
6	1.9×10^5	4.8×10^5	5.68	16	80
7	5.0×10^4	1.2×10^5	5.08	9	45
8	6.2×10^4	1.5×10^5	5.18	10	50
9	1.6×10^6	4.0×10^6	6.60	19	95
10	6.4×10^4	1.6×10^5	5.20	11	55
11	5.8×10^5	1.4×10^6	6.15	17	85
12	2.4×10^4	6.0×10^4	4.78	5	25
I	7.3×10^3	1.8×10^4	4.25	3	15
II	4.0×10^3	1.0×10^4	4.00	1	5
IV(14)	5.2×10^3	1.3×10^4	4.11	2	10
V(13)	1.57×10^5	3.9×10^5	5.59	15	75
VII	1.0×10^4	2.5×10^4	4.40	4	20
VIII	4.3×10^4	1.1×10^5	5.04	8	40
X	9.7×10^4	2.4×10^5	5.38	13	65
Harvard Yard	6.2×10^5	1.5×10^6	6.18	18	90
Lawrence Expt. Station	1.33×10^5	3.3×10^5	5.52	14	70

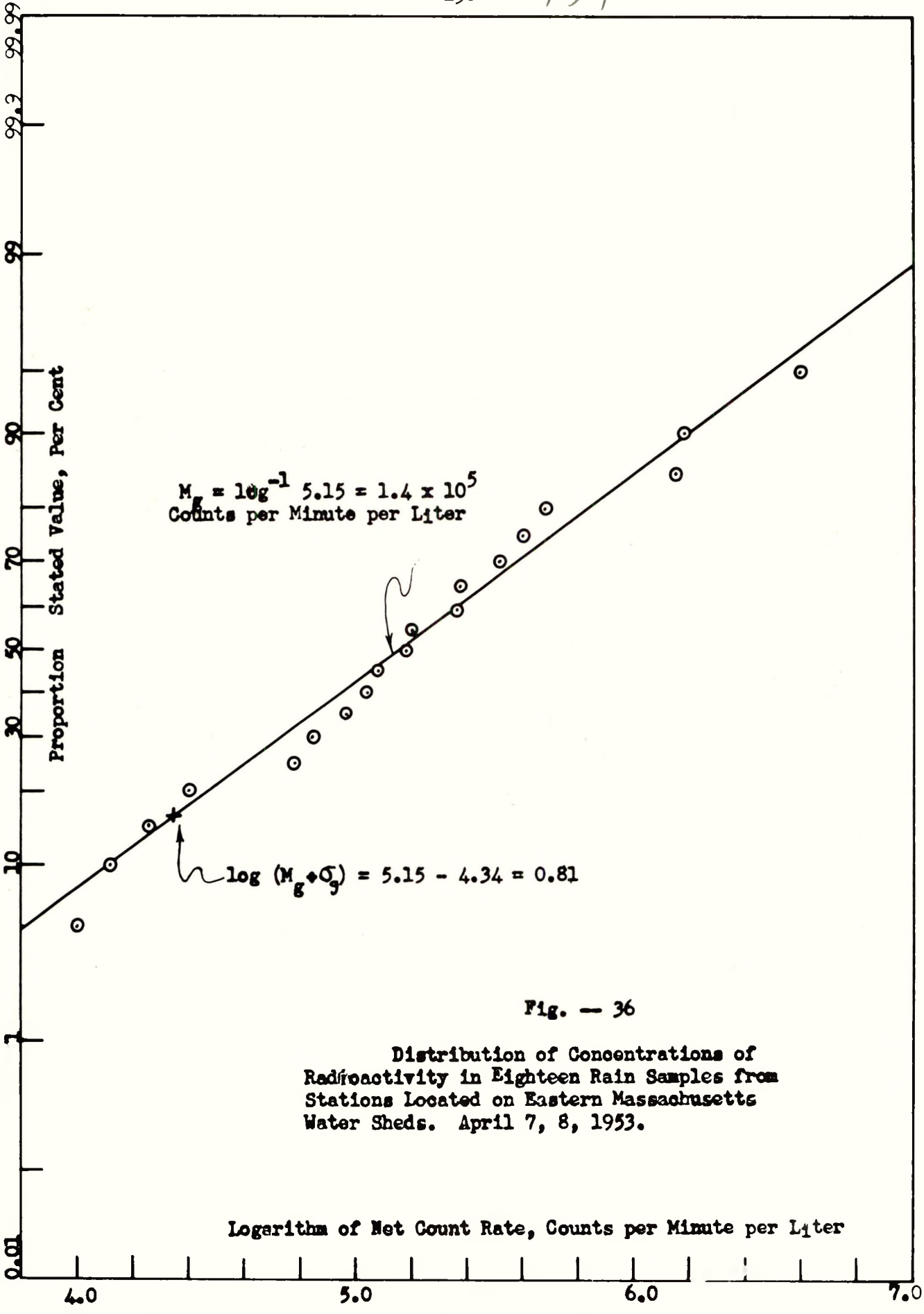

Fig. — 36

Distribution of Concentrations of Radioactivity in Eighteen Rain Samples from Stations Located on Eastern Massachusetts Water Sheds. April 7, 8, 1953.

above-discussed logarithmic distribution curve of the average beta radioactivity in the rain is allowed, a parametric calculation can be made to estimate the portion of the rain samples that had a disintegration rate above some of the published maximum permissible levels. Of course, this rain could have been collected and held in cisterns for drinking. The most definite standard for fallout concentrations states (6) that water containing 5,000 micromicrocuries of three-day-old total fission product radioactivity can be safely consumed for any period of time. In this standard it was recommended that each of the sample radioactivity decay curves be extrapolated to three days after fission. In this instance, it is more conservative and seems more reasonable to extrapolate the maximum acceptable value to the median time at which the rain fell by the widely used $A = A_1 t^{-1.2}$ formulation. If this is done, a level of 4.92×10^6 counts per minute per liter or 1.23×10^7 micromicrocuries per liter at thirty-eight hours after detonation is obtained. If water containing fission debris of this age and quantity is drunk by an individual at the rate of one liter per day for ten days, he will consume material disintegrating at about 5.7 microcuries. If, instead of the nineteen integrated samples actually collected, a large number of samples from this rain had been measured, about 3% of them would have exceeded this maximum permissible level. As data presented later indicate, the fallout radioactivity of samples is quite variable during a storm, and rain samples collected for a short time can exceed the average value considerably. This value of 3% is thought to be low rather than high, for absorption

could have taken place on the rain collection funnel, the collection jug, the evaporating dish, and the rubber policeman with which most of the samples came into contact in being processed for counting. No consideration of the exact beta ray energy spectrum was made for these measurements, which probably introduced a fairly large error, since end-window counters were used exclusively and, therefore, the weakest beta ray components were not recorded. Also, during the fission age in which the fallout was deposited, processed, and first counted, radioisotopes of iodine (principally I^{135}, I^{133}, I^{132}, I^{131}) make up an appreciable portion of the radioactivity, and quite likely some of this iodine sublimates in the preparation of samples for counting. No attempt was made to determine if the fallout contained "total" fission product as specified in the permissible level.

Seven of the rain samples collected April 8, whose measured radioactivity makes up part of the values shown in Table 19, were evaporated by the automatic device described in Chapter III. These samples were from Stations No. I, II, IV, V, VII, VIII, and X. All others with the exception of the one measured at Lawrence, Massachusetts, were of small volume (three milliliters) and evaporated directly in the counting planchets under a heat lamp. The count rate data from those eastern Massachusetts stations whose samples were not evaporated on the experimental automatic device were used in a calculation to check the results given in Subsection B-1-b. The decay curves were extrapolated to the estimated mean time of the radioactive fallout; then the count rates were evaluated for this mean time, corrected for self-

absorption, and converted to micromicrocuries per liter according to Table 17, Chapter V. The logarithms of these values were plotted according to Thomas (91). A calculation was made to show that there was no reason to doubt the normality of this distribution. If the same Atomic Energy Commission acceptable risk level (5,000 micromicrocuries per milliliter at three days after detonation) is used as before, it can be estimated that about 2.2% of a large number of integrated samples of this size drawn from this rain would have exceeded the permissible level. It is possible, however, that relatively large errors were introduced in this estimate in extrapolating the decay curves.

Another limit was given to the Civil Defense Administration by the Atomic Energy Commission and is called the Proposed Acceptable Risk Maximum Beta Activity Level (32). It applies to the radioactive contamination of drinking water in a period immediately following a bomb blast. For a ten-day consumption period the value is 3.5 microcuries per liter. Since the fallout immediately following a nuclear detonation decays rapidly at first, this limit is actually higher than the above-discussed three-day value. If (a) one liter per person per day of the April 6-7, 1953, eastern Massachusetts rain, having a radioactivity equal to the average measured for this rain, had been consumed, beginning at fallout, for ten days and if (b) the average measured decay parameter of 1.4 is used, it can be shown that the total per capita intake would have been approximately one microcurie at the time of consumption. The maximum ten-day consumption as given by the Atomic Energy Commission - Civil Defense Administration limit would be thirty-five microcuries.

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The distribution of the concentration of the April 7-8, 1953, fallout may be further examined by employing statistical tolerance limits (65), which are non-parametric. Based only on a knowledge of the number of samples (from nineteen different locations), the statement can be made that these samples range over an expected 90% of the population that would have been found if samples from a very large number of stations had been measured. If the range of the extreme count rate values in the nineteen samples is assumed to be nearly normally distributed, then it can be said that, at a probability level of about 68%, the proportion of the population's count rates that fall outside the observed range lies between 6% and 14%.

b. Rochester, New York, area. The highest recorded reading taken at Rochester, New York, was 9.34×10^5 counts per minute per liter at first count and was of the precipitation collected November 1, 1951. This is calculated to be 2.4 microcuries per liter at the time of first count. If it is assumed that the detonation on October 28 created the radioactivity, this rate would be (using a decay parameter of 1.2) 3.3 microcuries per liter at the time of deposition. If it is also assumed that all of the above-mentioned losses in processing are negligible, and if there were no coincidence losses in counting the sample, then it can be said that this sample was below the Atomic Energy Commission - Civil Defense Administration limit and the more recent Atomic Energy Commission standard at three days. However, if the same variability in concentration of radioactivity held for this fallout as was found for the April 7-8 fallout in eastern Massachusetts,

it is obvious that an appreciable portion of the samples taken from this rain would have contained radioactivity that was above the Atomic Energy Commission standard (6).

2. Time variation of the concentration of radioactivity in precipitation

Without access to measurements, one would assume that radioactive material is scavenged from the sky in an exponential fashion and likewise deposited on the ground. A set of samples was taken from a single rain-precipitated fallout that contradicts this supposition. During all of the Nevada test series that occurred between the fall of 1951 and the summer of 1953, Mr. B. L. Rosenthal of the Lawrence Experiment Station of the Massachusetts Department of Public Health worked on the fallout problem in close cooperation with the Atomic Energy Commission project at Harvard. His equipment and sample preparation techniques were the same as those used at Harvard in almost all cases. On November 7, 1951, he conducted an experiment in which he measured the radioactivity (counts per minute per 100 milliliters) of the residue after evaporation of rain samples that were collected at short intervals following the beginning of the rainstorm. The average of the hourly precipitation of three United States Weather Bureau recording rain gage stations near Lawrence, Massachusetts, are given in Table 20. The results of this experiment are given in Table 21.

Unfortunately, hourly precipitation rates are not measured at Lawrence. To show that the maximum precipitation rates probably occurred at the time when the rainfall contained the maximum concentration

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TABLE 20

HOURLY PRECIPITATIONS AT RECORDING GAGES NEAREST LAWRENCE, MASS., ON NOV. 7, 1951

Time	Birch Hill Dam	Rockport	Sterling	Average
7:00-8:00*	0.00	0.00	0.08	0.03
8:00-9:00	0.00	0.05	0.06	0.04
9:00-10:00	0.00	0.05	0.20	0.08
10:00-11:00	0.01	0.06	0.31	0.13
11:00-12:00	0.34	0.04	0.37	0.25
12:00-13:00	0.38	0.02	0.28	0.68
13:00-14:00	0.02	0.14	0.00	0.05
14:00-15:00	0.01	0.48	0.02	0.17
15:00-16:00	0.01	0.04	0.00	0.02

* There was no rain before 7:00 hours and little rain after 16:00 hours.

TABLE 21

TIME DISTRIBUTION OF THE CONCENTRATION OF RADIOACTIVITY WITHIN THE RAINSTORM AT LAWRENCE, MASS., NOV. 7, 1951

Time	Net cpm/l at first count	First Count std. error cpm/l	Net cpm/l at median time of storm	Decay Parameter n
8:00-8:30	2,488	7.4	3,600	1.40
8:30-10:00	1,233	4.4	1,800	1.40
10:00-11:00	408	2.0	590	1.40
11:00-12:00	2,438	7.2	3,500	1.40
12:00-13:00	6,320	16.8	10,000	1.76*
13:00-14:00	1,790	5.7	2,600	1.40
14:00-15:00	755	2.5	1,350	1.40
			m = 3,320	
Integrated 100 ml sample from rain gage	1,482		2,600	1.40

* Value from decay measurements of sample. Other values from decay measurements of integrated samples.

(count rate per unit volume) of radioactivity, the author made an investigation of the time of occurrence of the maximum hourly precipitation rates for the six, twelve, and eighteen stations nearest Lawrence. In each group, the median time of the maximum hourly precipitation rate was found to be either at 13:00 hours or 13:30 hours on November 7, which indicates that the maximum rainfall rate in the storm probably occurred at Lawrence at the same time and that the rates of rainfall near Lawrence were correlated in this storm.

In addition to the initial measurements of the concentration of radioactivity in the hourly and integrated samples, four measurements for decay were made between November 28, 1951, and January 7, 1952, on the integrated sample and that collected between 12:00 and 13:00 hours on November 7, 1952. The resulting decay parameters (n values) are shown in Table 21. For extrapolation of count rates of samples in this experiment other than the 12:00-13:00 hours sample, the n value of the integrated sample (1.40) was assumed to apply. This assumption made it possible to place all of the count rates of the samples at a common age for comparison.

These results indicate that there is no reason to think there is a scouring and precipitating mechanism that puts the largest portion of the radioactivity on the ground during the first part of a rainstorm and decreasing amounts later in the storm. If rain takes up long-range fallout from the sky in an exponential fashion, then there must have been sufficient mixing between scavenging and deposition to conceal the effect in these rain samples.

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The apparent interrelation of the concentration of radioactivity in the rain and the average hourly rate of rainfall at nearby stations that have recording gages prompted a statistical analysis that yielded a correlation coefficient (ρ) of 0.854 (Table 22). It was found by using Fisher's z transformation that $P\{\rho \leq 0\} = 0.002$. Therefore, the data indicate that an increase in the rate of rainfall from this storm, which contained newly-created fission debris, was accompanied by an increase in the concentration of radioactivity. A possible cause of this result could have been a more radioactive, shorter, harder rain that fell at approximately 12:00 hours from a higher cloud through the lower, less radioactive rain cloud that was the source of the rain for most of the storm (93).

An array of the data was made for the count rates of the samples taken at various times during the storm of November 7, as counted on November 8, and it was found that they followed the geometrically normal distribution (M_g = 1750, SD_g = 2.83) quite well. This single set of data indicates a surprising non-uniformity of radioactivity concentration during a rain-accompanied fallout.

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TABLE 22

PRODUCT MOMENT CORRELATION BETWEEN THE ESTIMATED RATE OF RAINFALL AND THE CONCENTRATION OF RADIOACTIVITY IN THE RAIN FOR VARIOUS PERIODS IN THE NOV. 7, 1951 RAINSTORM AT LAWRENCE, MASS.

Time	X Rain (0.01)(inches/hour)	Y Counts/minute/liter on Nov. 8
8:00-8:30	4	2,490
8:30-10:00	7	1,230
10:00-11:00	13	410
11:00-12:00	25	2,440
12:00-13:00	68	6,320
13:00-14:00	5	1,790
14:00-15:00	7	930

D. OBSERVED EXTERNAL RADIATION IN LONG-RANGE FALLOUT

1. Measurements with Geiger-Müller tubes

Mr. G. Gordon, who did research at Harvard on cosmic rays in 1953 and 1954, followed a procedure at the time of the Nevada spring test series of 1953 that included taking daily count rate readings on a bank of ten large, thick-walled cosmic ray Geiger-Müller tubes located in a thin-roofed shed on the Cambridge campus. The daily readings of April 1, 2, 3, and 7 were found to average 2,899 ± 89 counts per minute. On taking the reading on April 8, 1953, he observed a 123% increase above the average of the previous four observations. After this date, the readings gradually decreased and at the end of approximately one month returned close to the pre-April 8 level. If it is assumed that this change was caused by the long-range fallout from the Nevada detonation of April 6, 1953, there is basis for plotting this increase over the average count rate during the week before April 8 versus the time after the blast on double logarithmic paper. This plot is shown in Figure 37. Only one of the differences during this period of one month (that of April 27) was negative. The curve of best fit (by eye) through the plotted points gives a formula whose exponent agrees well with the "1.2" law given in Reference 1.

The walls of these Geiger-Müller tubes are of brass, 1/32 inch in thickness. As the energies of fission debris beta rays are such that only a few per cent of them could have penetrated this mass into the sensitive volume of the tubes, it is quite likely that the predominant radiation measured was gamma rays from the long-range

fallout. It is interesting that no increases in the decay rate (Figure 37) were observed that were caused by rains subsequent to the April 7-8 fallout. This indicates that the deposited radioactive material was held in place, presumably on the roof of the building.

2. Spots observed in a cloud chamber

Also on April 8, there was observed in the same laboratory an increase in the number of spots in the photographs taken automatically of the Wilson cloud chamber used for the cosmic ray studies. A reasonable explanation is that this increase was caused by condensations on the paths of electrons that were detached from molecules in the chamber when struck by external gamma rays from fallout. For the cosmic ray investigation, a strong magnetic field (4,800 oersteds) was maintained on the chamber, so that each detached electron was held in a small region until its energy was dissipated in ionizing collisions with gas molecules in the chamber. Thus, a white spot was formed when condensation was brought about in the chamber. The same electron ejection and ionization takes place when the human body receives external gamma ray radiation, this being one of the mechanisms of radiation damage to tissue. Figure 38 shows representative photographs of the cloud chamber on days before and after the April 7-8 fallout. The photograph taken on April 8 shows the newly-formed condensation path of one of these electrons.

Each of the 1616 photographs of the cloud chamber taken from April 1 to April 24, 1953, was scanned and the number of electron tracks counted in order to study the change in the production of these electron

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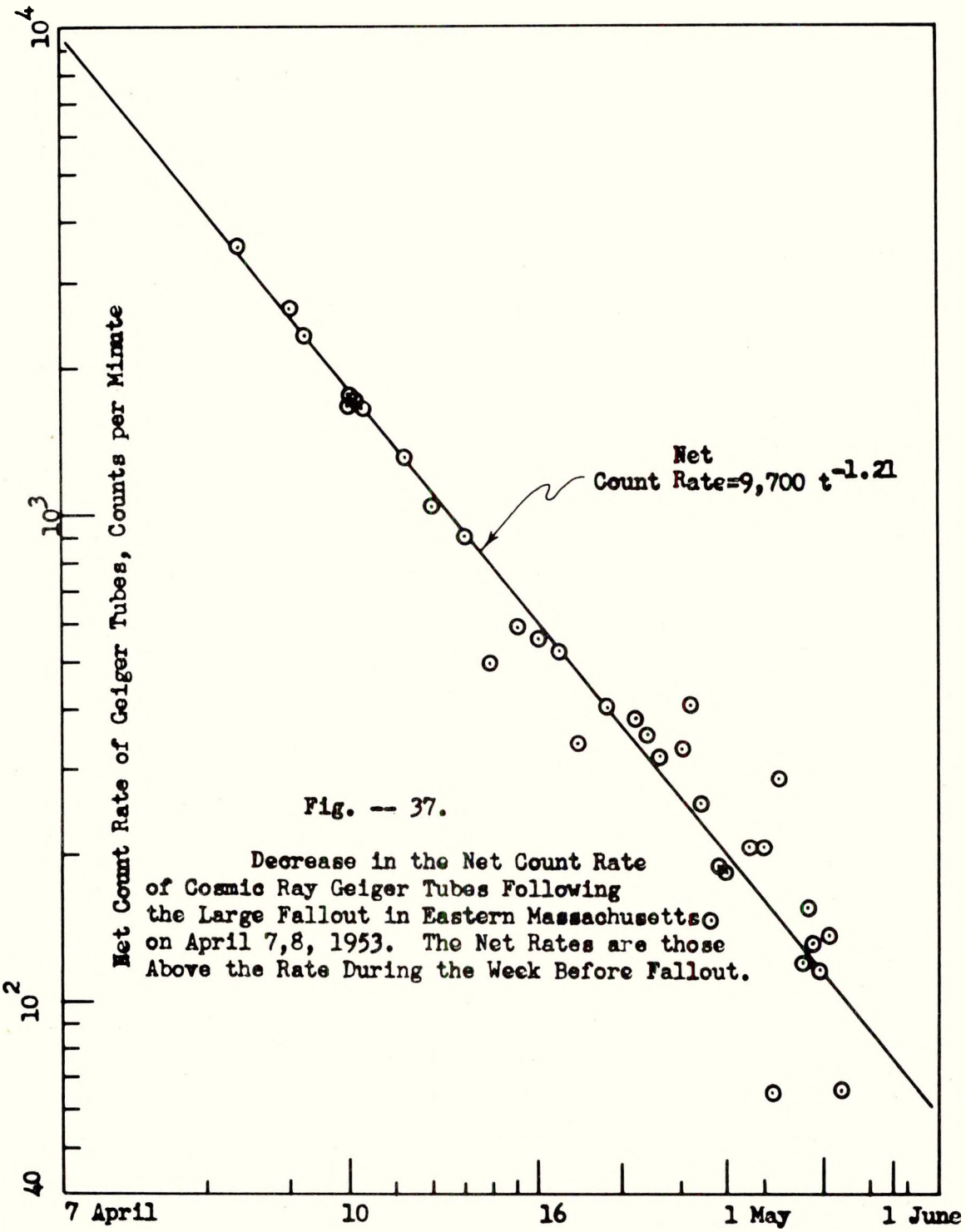

Fig. -- 37.

Decrease in the Net Count Rate of Cosmic Ray Geiger Tubes Following the Large Fallout in Eastern Massachusetts on April 7,8, 1953. The Net Rates are those Above the Rate During the Week Before Fallout.

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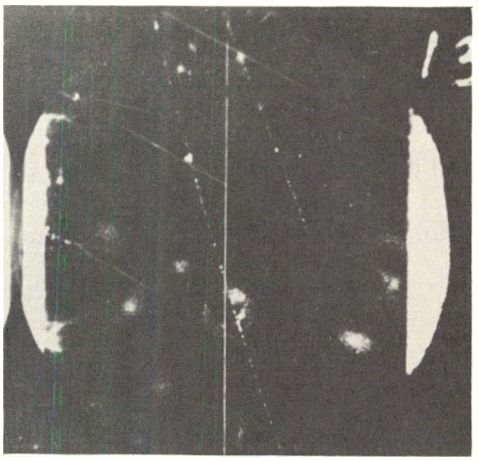
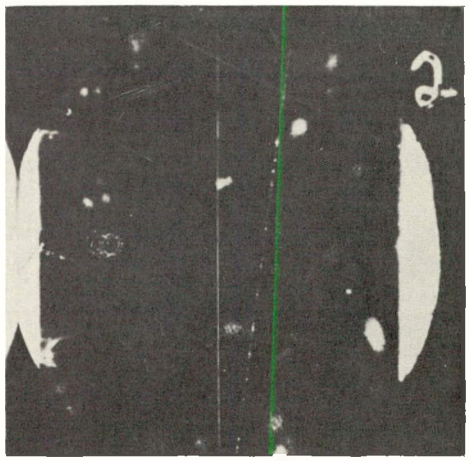
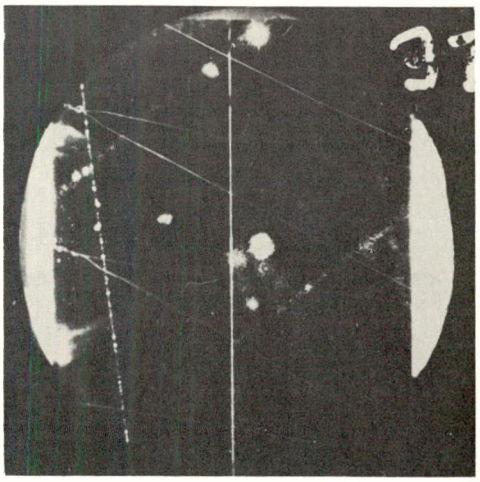


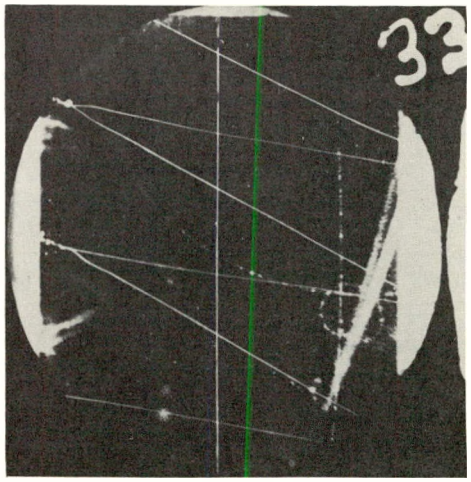
April 7, 1953



April 8, 1953



April 14, 1953



May 12, 1953

Figure 38

Photographs of a Wilson Cloud Chamber before and After the Large Fallout on April 7-8, 1953 at Cambridge, Massachusetts

tracks during the period of large fallout. The identifying dates were covered, the films mixed, and the counts recorded according to a code number to minimize the bias in counting. Figure 39 shows the mean count rate of electron-type spots per exposure on each day photographs were taken. The vertical bars show the 68% confidence zone of the mean count rate.

If the mean of the counts of electron-track spots per frame for the photographs taken from April 1 through April 7 is subtracted from each of the means of the daily readings from April 8 through April 17, an estimate of the rate of decrease in spot formation and thus of external gamma radiation may be made. These net rates are plotted against the number of days after the April 6 detonation on double logarithmic paper (Figure 40). A straight line of best fit (by eye) was passed through these points and agrees with the "1.2" law.

The rainstorm of April 7-8, 1953, started about 1:00 hours on April 7 in the western part of the sample collection area (Figure 2, Chapter III) and ended about 9:00 hours (western portion) to 8:00 hours (eastern portion) on April 8. Photographs of the cosmic ray chamber were started about 12:10 hours on April 7 and by 17:20 hours fifty-eight exposures had been taken. As can be seen in Figure 39, the mean electron spot count for that day was the lowest of the means of the five groups of measurements taken since April 1. The mean of the electron track counts of the last five pairs of photographs taken on April 7 was not significantly different from the mean of the other forty-eight photographs taken on this date. Also, as can be seen in Figure 39 the

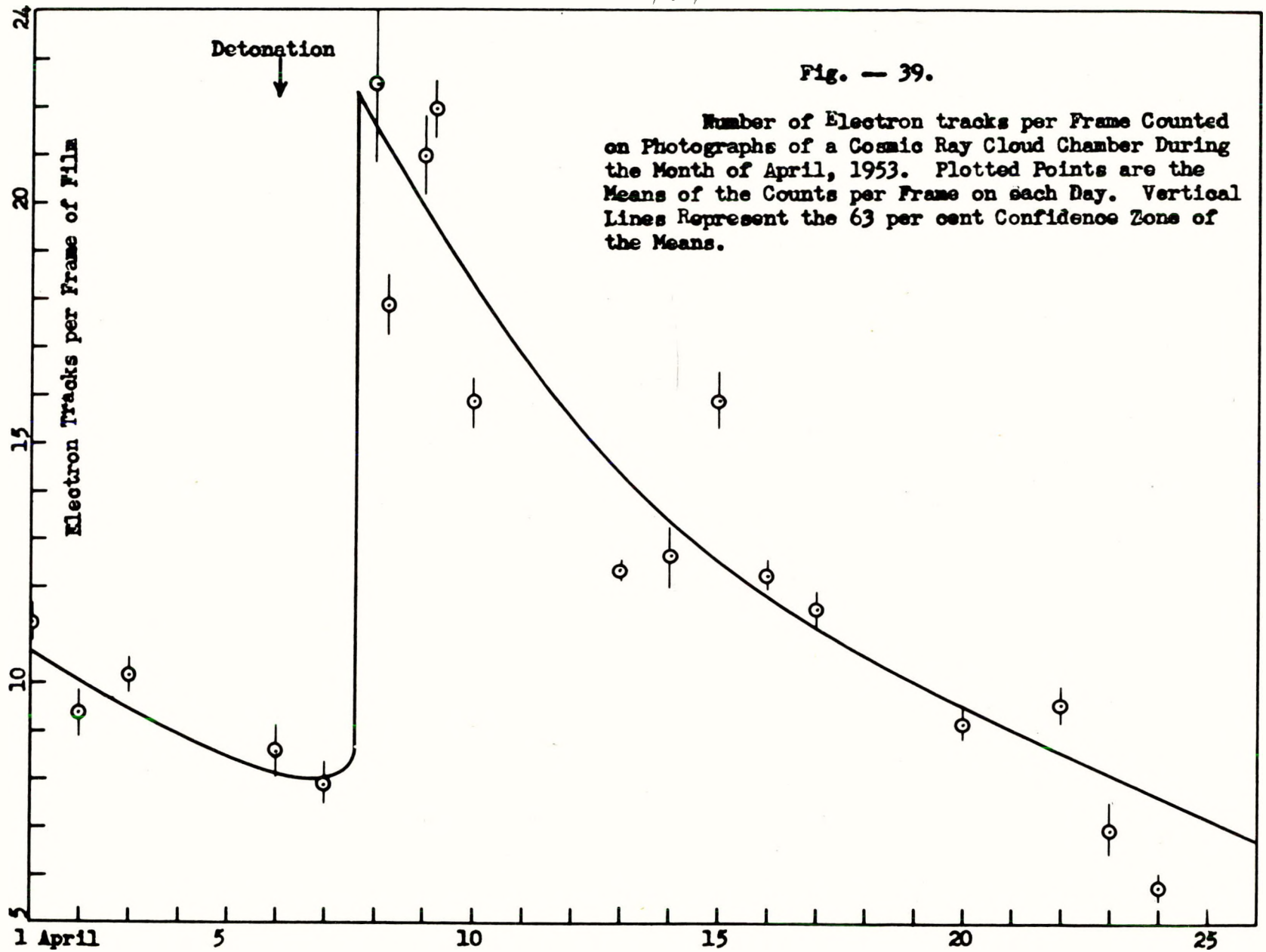


Fig. — 39.

Number of Electron tracks per Frame Counted on Photographs of a Cosmic Ray Cloud Chamber During the Month of April, 1953. Plotted Points are the Means of the Counts per Frame on each Day. Vertical Lines Represent the 63 per cent Confidence Zone of the Means.

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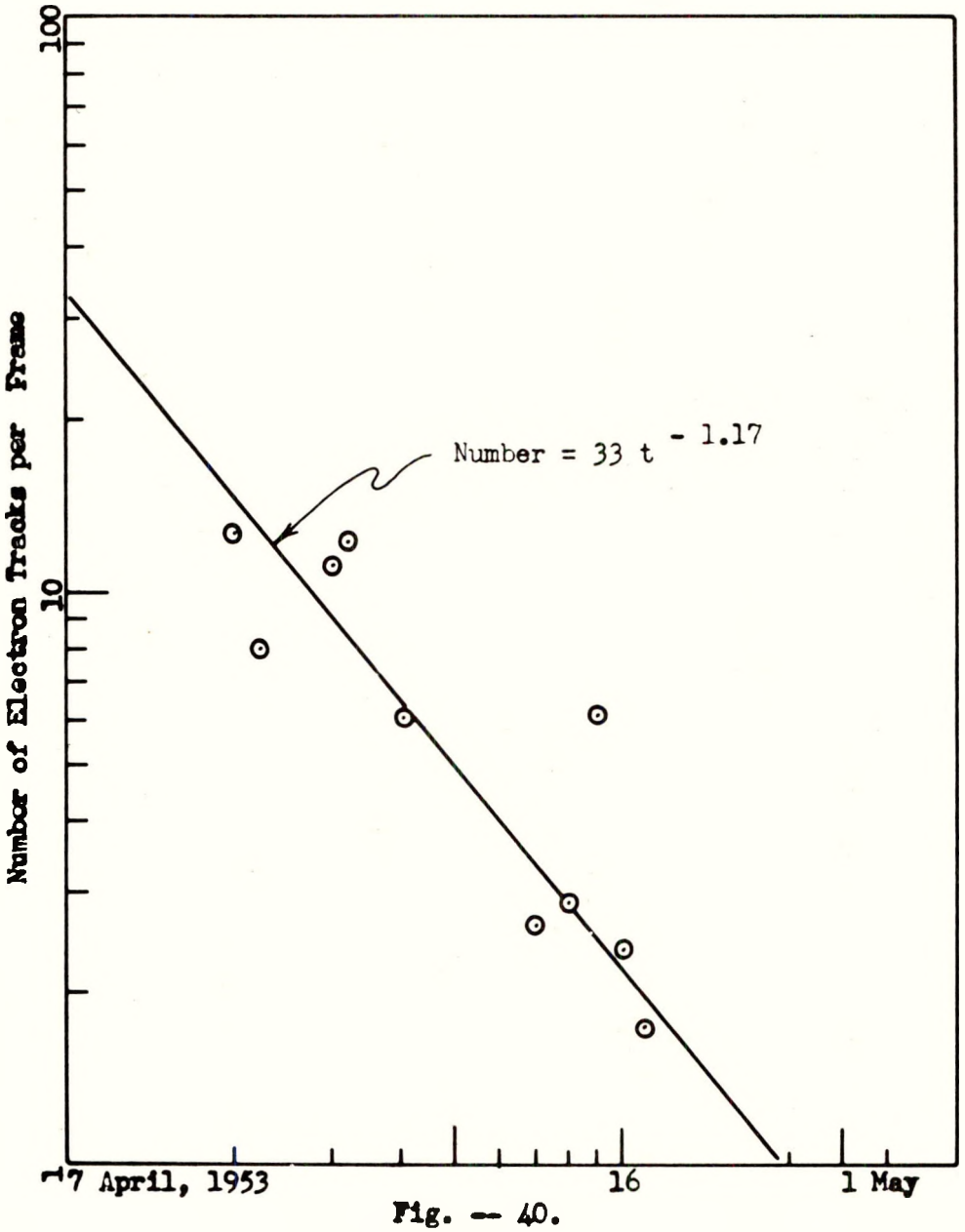

Fig. — 40.

Decrease in the Number of Ionization Tracks of Electrons Ejected from Atoms in a Wilson Cloud Chamber by Long Range Fallout Gamma Radiation.

count rate of the bank of ten large cosmic ray Geiger-Müller tubes that was taken on April 7 at approximately 18:00 hours was quite close to the mean of the count rates taken on April 1, 2, and 3. Therefore, the fallout must not have started at the first of the rainstorm but during the night of April 7-8. If the estimates made from meterological data (Figure 1, Chapter I) are approximately correct, then the major portion of the deposition (94) must have taken place between midnight and 8:00 hours on April 8. No rain fell after 8:00 hours on April 8 at Lawrence, Massachusetts, and the count rate for the integrated April 7-8 sample, when measured on the afternoon of April 8, was 99,000 counts per minute per liter. The United States Weather Bureau estimates that about half of the rain that fell at Lawrence on April 7-8 fell between midnight and 8:00 hours on April 8. An examination of the data for all stations in eastern Massachusetts shows that the major portion of the fallout count rate could not have come from dry fallout or rain following the morning of April 8.

E. FALLOUT RADIOACTIVITY AND THE WORCESTER TORNADO

On June 9, 1953, a cold front with attending squall line moved into eastern Massachusetts and produced the most violent tornado in the recorded history of New England, as well as one of the largest tornados ever observed in the United States (95). If a comparison of the location of the sample stations (Figure 2) with the approximate path of the storm (the dashed line on this map) is made, it can be seen that the samples

of rain and surface water collected on June 10 and 12 probably gave representative samples of the rain accompanying the tornado. Collections had been made on June 5 and 6, so that only the rains of June 7 and 8 were mixed in the samples with the fallout that occurred on June 9.

The count and decay rates of the fourteen rain samples furnish a unique opportunity to speculate on the relationship between huge tornados and long-range carriage of fission debris. Figure 41 and Figure 42 give plots of decay measurements and the curves that were fitted by eye through these plotted points. The assumption was made that the fission material originated in the June 4 detonation. The mean count rate of these samples was 157 counts per minute per liter on July 14; the mean of the decay parameters (n values) was found to be 1.12 ± 0.04. If this mean of these n values is used to extrapolate the mean count rate back to the time of deposition on June 9, a value of 1,620 counts per minute per liter is obtained. When the decay curve of the sample from Station Number 6 , which had the highest count rate per liter of the group, is extrapolated back to June 9, it is seen that it then had a count rate of about 6,000 counts per minute per liter. An examination of the master plot for eastern Massachusetts (Figure 26) shows that the concentration of radioactivity in the precipitation often exceeded this value during both the 1951 and 1952 Nevada test series and that the order of magnitude of this value was reached following both the 1952 Eniwetok and the 1953 Siberian experiments. The highest concentration of fallout radioactivity in rains measured at Harvard from

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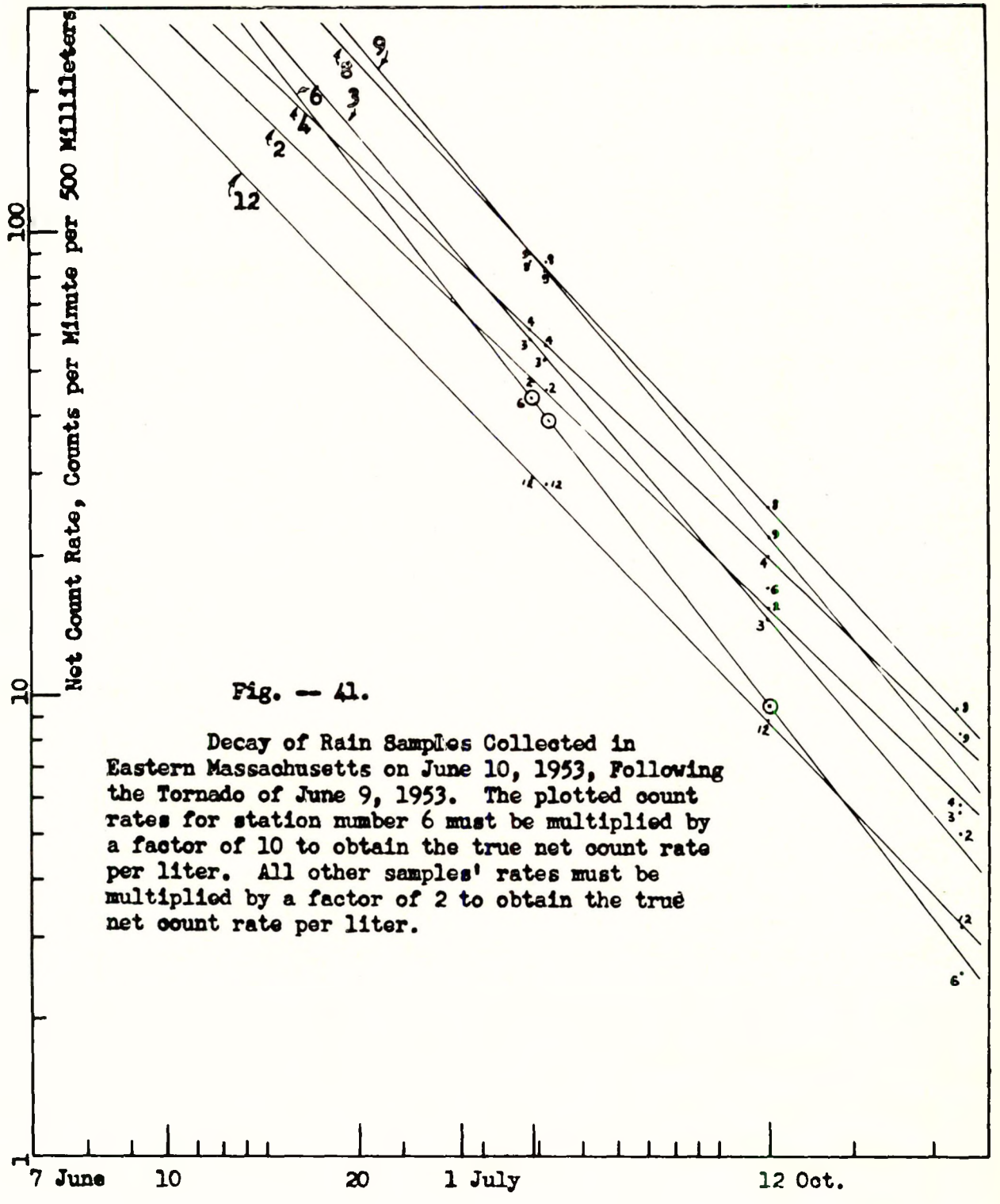

Fig. — 41.

Decay of Rain Samples Collected in Eastern Massachusetts on June 10, 1953, Following the Tornado of June 9, 1953. The plotted count rates for station number 6 must be multiplied by a factor of 10 to obtain the true net count rate per liter. All other samples' rates must be multiplied by a factor of 2 to obtain the true net count rate per liter.

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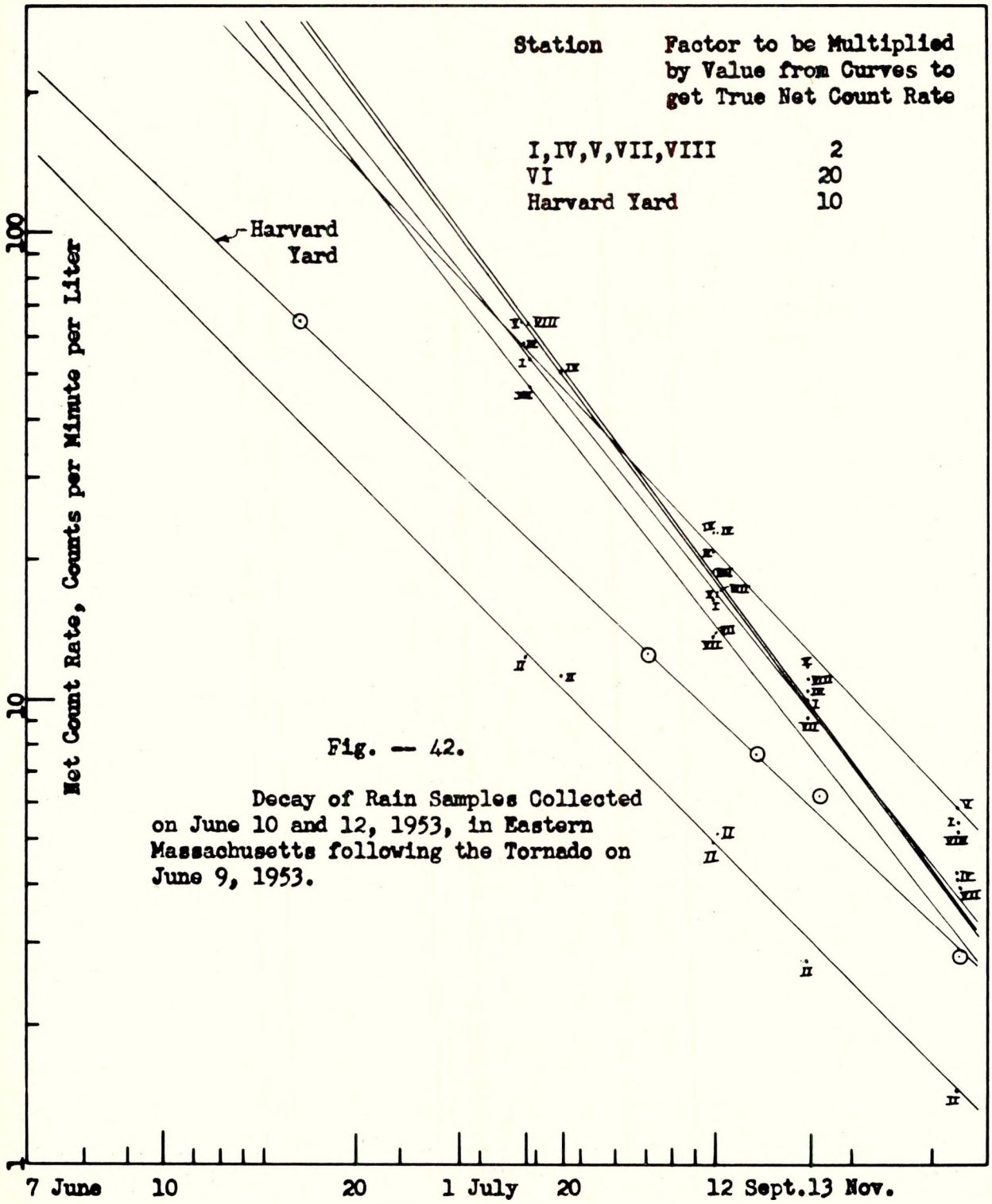

Fig. — 42.

Decay of Rain Samples Collected on June 10 and 12, 1953, in Eastern Massachusetts following the Tornado on June 9, 1953.

May, 1952, until December, 1954, was that collected at Quabbin Reservoir just after the April 7-8 fallout, where the estimated count rate at the time of fallout was about 1.6×10^6 counts per minute per liter or about two hundred and seventy times that of the station that received the maximum count rate concentration during the Worcester tornado. The ratio of the means of the count rates of all the precipitation samples collected just after the April 7-8 and June 9 fallouts is $1.4 \times 10^5 \div 1.6 \times 10^3$ or about 87. Tap samples from the city of Worcester following this tornado were measured by the usual procedure and registered only 4.1 counts per minute per liter.

By use of the decay curves of the fourteen different rain samples (Figure 41 and Figure 42) taken after the tornado, all count rate concentrations were placed at a common date (July 14) for comparison. The count rates (counts per minute per liter) for the stations are shown in Table 23.

Since the tornado first touched the ground at Petersham, Massachusetts, and three of the four most radioactive samples of the collection were taken near this point (Stations No. 6, No. 8, and No. 9), a "Student" t-test (65) was made for significance of the difference between the mean of this group of four count rates and the mean of the count rates of the remaining ten samples. This yielded a t-value of 2.82 , which indicates that the probability that the means of these two groups coming from the same normal population is less than 1%. This value suggests scavenging action of fission radioactivity from the air by the rain as the tornado progressed. No rain sample in the path of

the tornado was collected. The storm caused the loss of the sample at Station Number 10 (near Barre Plains) and the rain collector at Station Number 5 (Worcester) had been removed prior to this time by vandals. A station near the end of the path of the tornado (No. 8, at Cordaville) had a count rate less than the average of all of the other rains. Of the twenty-eight surface water samples collected from the twenty-two sampling stations on June 10 and 12, twenty-five gave an average count rate at first counting of 4.6 counts per minute per liter; however, three of them (samples on June 12 at Stations No. 11, No. I, and No. 14) gave an average of 40.7 counts per minute per liter. Samples taken at Stations Number I and Number 14 on June 10 had count rates that averaged only 5.8 counts per minute per liter, although none of the Massachusetts weather stations that record hourly rainfalls reported any rain on June 10, 11, or 12. Thus, this increase must have been due either to dry fallout or to showers not registered by the United States Weather Bureau.

In an article (3) in "Comptes Rendus" of 1951, M. Hubert states that soon after nuclear detonation a large part of the radioactive cloud pierces the tropopause and spreads into the stratosphere. The "Ivy" detonation at Eniwetok on November 1, 1952, sent a column of debris, coral, and water ten miles into the air. The "steam" began to churn horizontally at twenty-five miles elevation to form a cloud 100 miles across (96). Since the Worcester tornado rose to 61,000 feet, which is well above the elevation of the tropopause, one can speculate whether or not it was able to scavenge debris from "Ivy".

It is interesting that the decay curves of several of the rain samples collected following the Worcester tornado had decay parameters

(calculated with the assumption that the radioactivity was created in the June 4, 1953, detonation) considerably lower than any of those of samples collected during the large fallout of April 7-8, 1953. The decay measurements on the rain accompanying the tornado, including the one made on the June 9 Harvard rain sample 345 days after detonation, followed the hyperbolic formulation well. The lower decay parameters could not have been due to statistical errors of counting alone but could have been caused by some of the older, more slowly decaying debris from the "Ivy" detonation in the sample. Unfortunately, no chemical separations have been made on the radioactive material in these samples.

TABLE 23

NET COUNT RATES OF RAIN SAMPLES COLLECTED FOLLOWING THE LARGE NEW ENGLAND TORNADO OF JUNE 9, 1953

Station Number *	Sample Net Count Rate (cpm/l)
2	94.3
3	114.8
4	121.5
6	430.9
8	172.0
9	178.0
12	58.8
II	247.0
IV	113.4
V	127.5
I	104.4
VII	93.9
VIII	126.5
Harvard Yard	157.3

* No rain fell on this date at the Lawrence, Massachusetts, station. Samples were collected on June 10-12 but counted first only on July 12-13.

F. DECAY OF RADIOACTIVITY IN SURFACE WATER SAMPLES

1. The Nevada test series of the spring of 1952

The same hyperbolic relationship (1, 55) used in calculating the decrease in radioactivity in precipitation also was applied in the study of the radioactivity decay of surface water samples. In the sampling program in eastern Massachusetts from April, 1952, until July, 1953, it was found that during each of the Nevada test series the concentration of radioactivity in the precipitation averaged approximately one hundred times the concentration in the surface waters. For the major portion of the sample collection period the count rate levels of the surface waters were so low that the naturally radioactive species and probably the long-deposited fission radioactivity interfered to an extent that no fallout decay formulation was attempted.

The fallout at the end of the Nevada series of 1952 raised the radioactivity in the surface waters of eastern Massachusetts to a level that was high enough to make possible by the techniques used an investigation of the decrease in the mean count rate on repeated measurement of the fourteen samples collected each week during the month following the fallout. Figure 43 shows the points that represent these mean count rates on repeated measurement. Although it was assumed that all of the radioactivity was formed in the detonation of June 1, 1952, it is quite probable that the samples also contained important portions of debris from other detonations. The decay curves were fitted to the plotted points by eye and terminated on their left at the times of sample collection. A curve was then drawn through these termination points to

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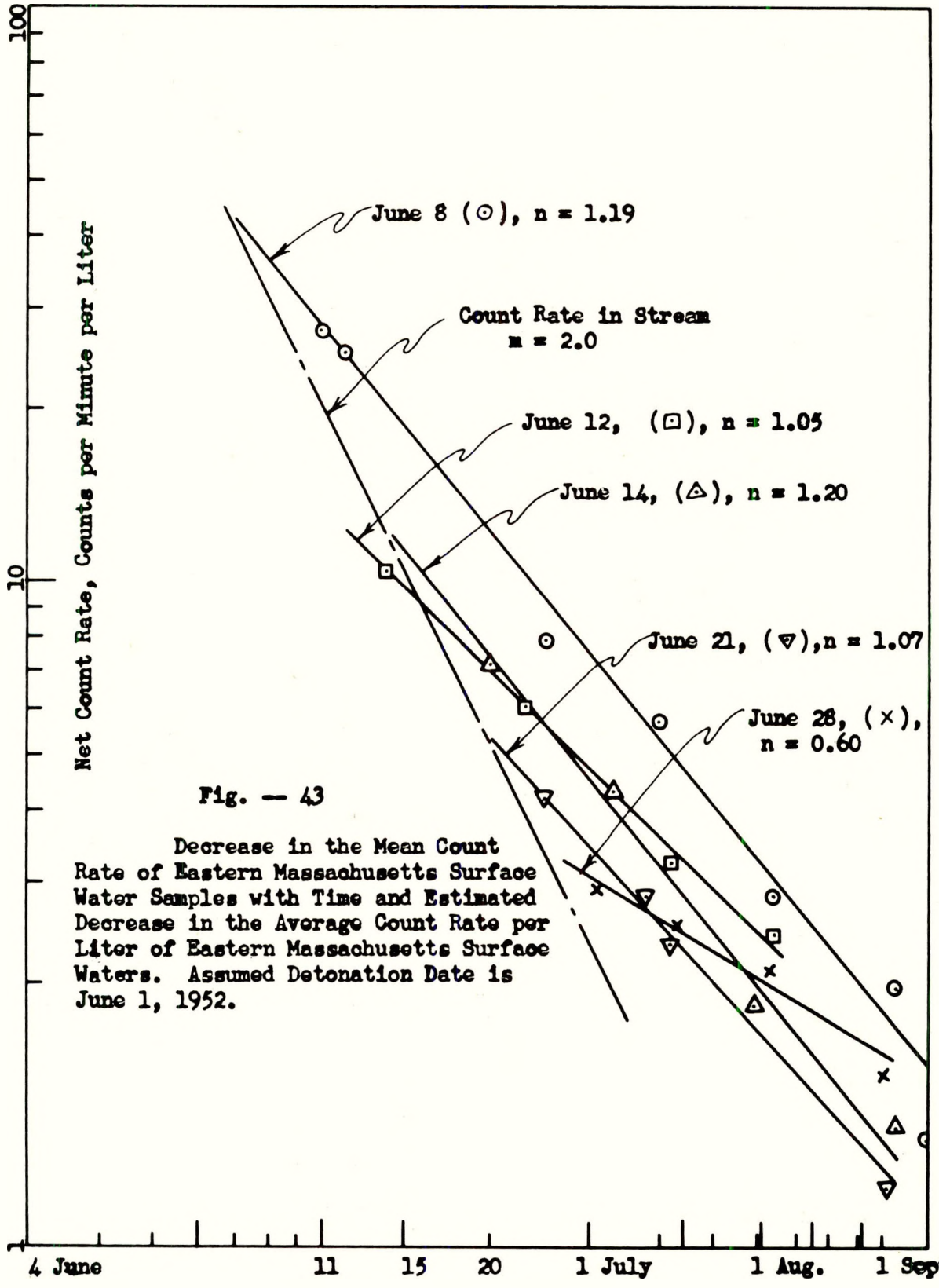

Fig. — 43

Decrease in the Mean Count Rate of Eastern Massachusetts Surface Water Samples with Time and Estimated Decrease in the Average Count Rate per Liter of Eastern Massachusetts Surface Waters. Assumed Detonation Date is June 1, 1952.

give an estimate of the rate at which the count rate decreased with time in the stream. The curve through these points will be discussed in detail in the next section. Despite the low count rate of the surface water samples collected during this series, those decays permit considerations that are not so conveniently calculated from the higher concentrations of radioactivity caused by the other Nevada series (that of the spring of 1953) that was also monitored for this thesis. The fallout that occurred in Massachusetts during the first half of June, 1952, in all probability was the major fallout caused by the series; therefore, one can assume for the purposes of calculation that all of the radioactivity in these streams originated in the last detonations. With the relatively high proportion of natural radioactivity present, the antecedent fission radioactivity would make the calculated fission debris decay parameters small. The average decay parameter for the collections of June 8 through June 21 is $n = 1.1$. On the assumption that all of this measured radioactivity was created in the detonation of June 1, the decay curves shown on Figure 43 were plotted. Figure 44 shows decay curves based on the same count rate data with the assumption that the radioactivity was created in the June 6 detonation. In this case, for the collections of June 8 to June 21, the decay perameters (n) have a mean value of 0.9 .

2. Surface water radioactivity due to the 1952 Eniwetok tests

The highest average count rate reached by samples from the fourteen eastern Massachusetts stations during and immediately following the

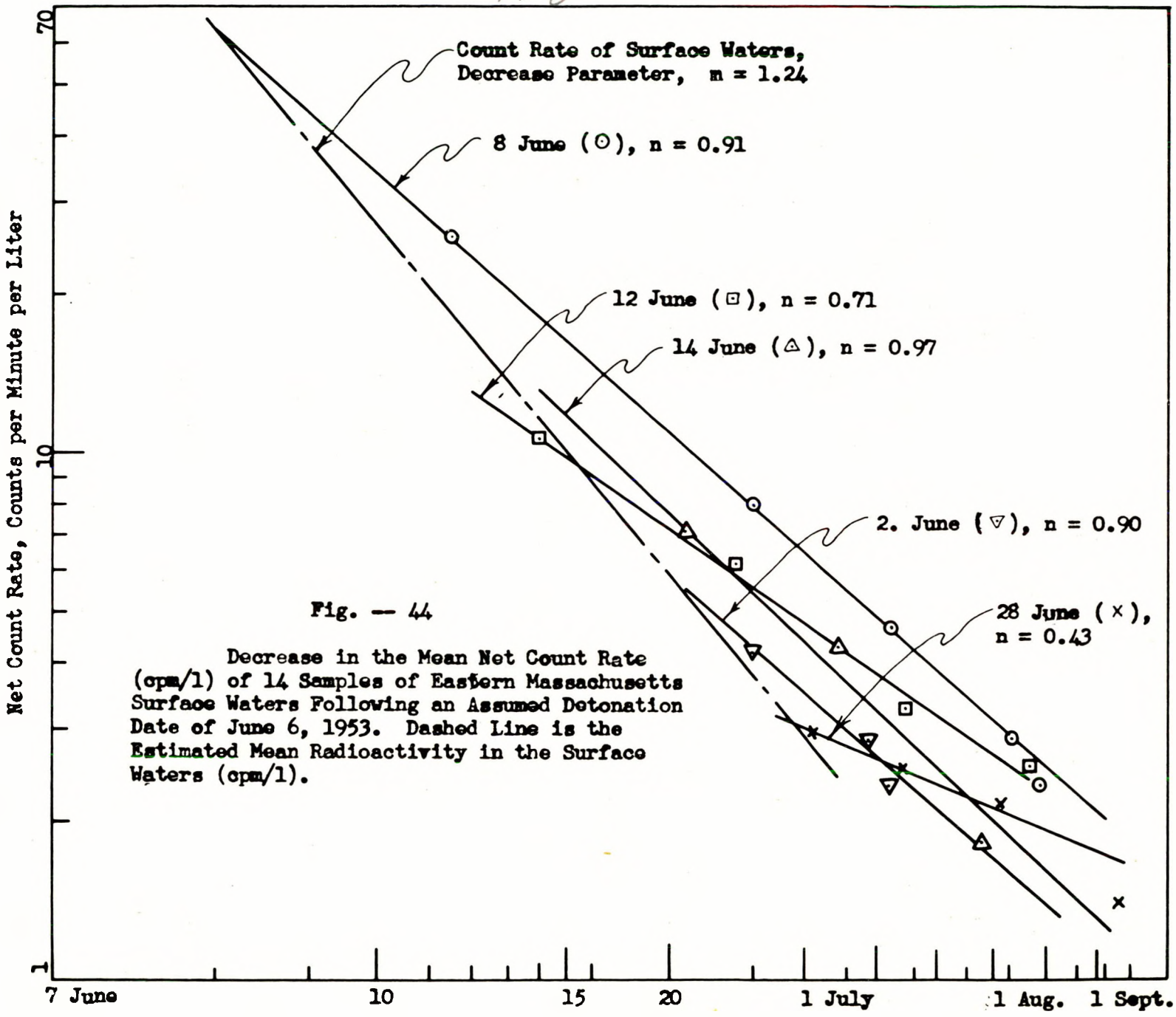

Fig. — 44

Decrease in the Mean Net Count Rate (cpm/l) of 14 Samples of Eastern Massachusetts Surface Waters Following an Assumed Detonation Date of June 6, 1953. Dashed Line is the Estimated Mean Radioactivity in the Surface Waters (cpm/l).

Eniwetok tests in 1952 was 6.9 counts per minute per liter at first count. This value is only about six times the average of the mean of the count rates at first count of water samples at these same stations for the weekly collections in August, September, and October of that year. There were no announced detonations for more than four months preceding these Eniwetok tests. Since the date of the second experiment in this Eniwetok series is not given, no data or analysis of the surface water decay rates are presented.

3. The fallout from the Nevada tests of the spring of 1953

As can be seen on the survey plot of the radioactivity concentrations measured for eastern Massachusetts, there were several depositions of fission debris in the spring, 1953, series that raised the levels of count rates high enough for decay study. The mixing of fission material from two or more blasts makes analysis for the decay values difficult, except in the case of the April 6 detonation, which produced a fallout so large that it probably obscured the effects in the surface waters of most of the subsequent depositions from this series.

As illustrations of the plots made for the decay determinations, Figure 45 and Figure 46 represent the measurements on the surface waters collected on April 8 and April 9. The decay parameters (n values) were obtained from the slopes of the lines of best fit as placed by eye through the plotted values of the repeated measurements of the individual samples. Table 24 summarizes these values for the surface water samples collected for the three weeks following the large fallout and Figure 47

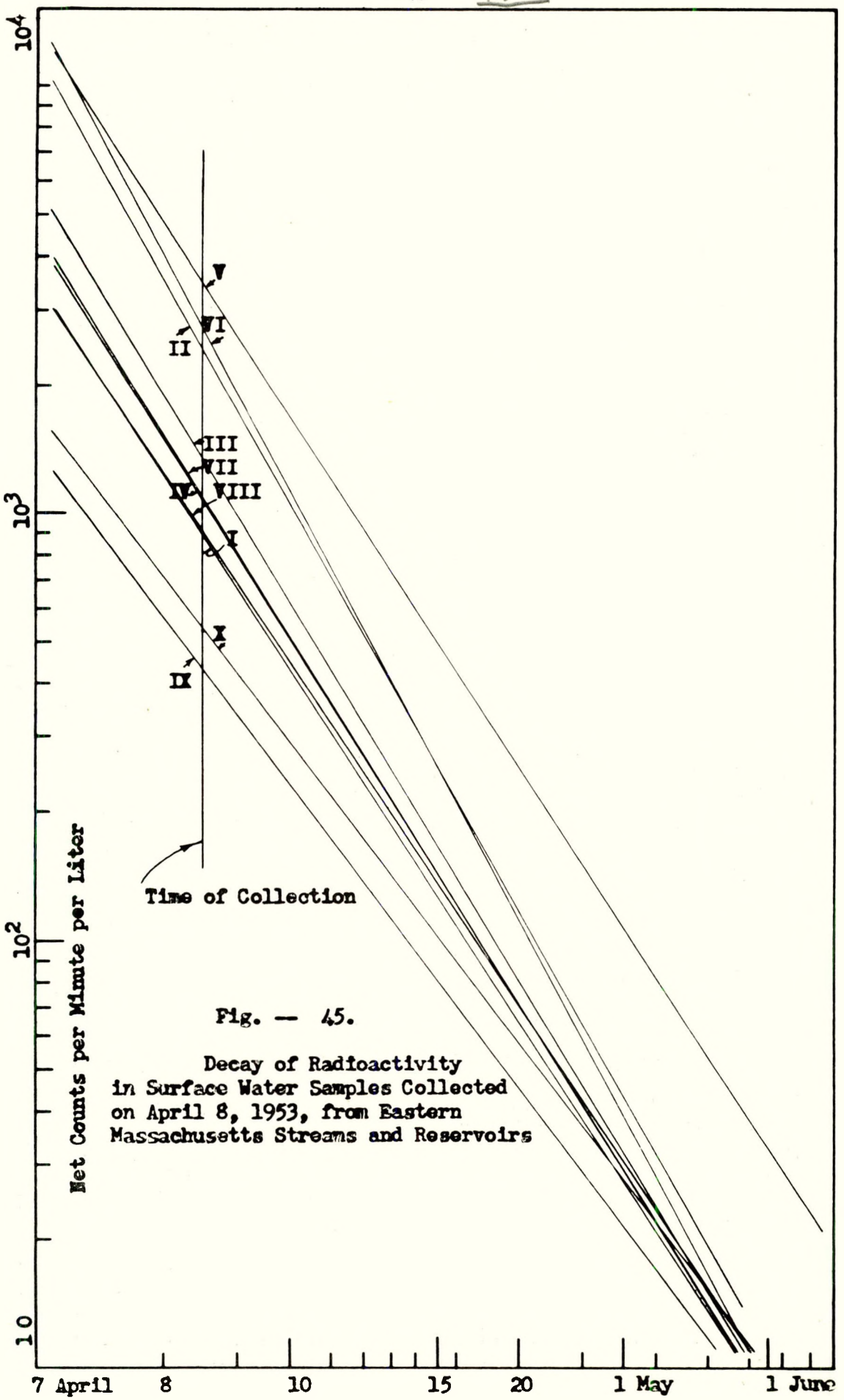

Fig. — 45.

Decay of Radioactivity in Surface Water Samples Collected on April 8, 1953, from Eastern Massachusetts Streams and Reservoirs

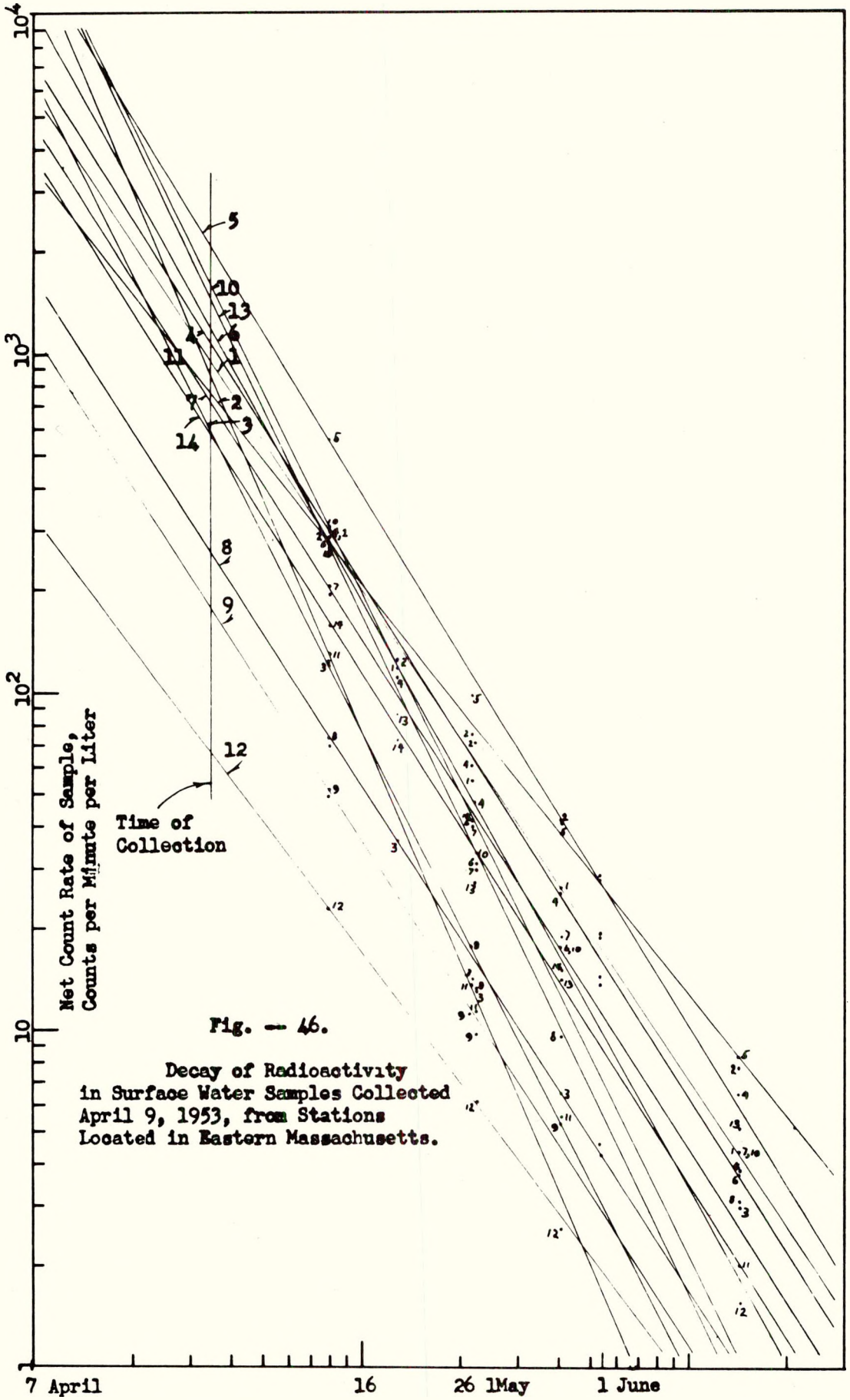

Fig. — 46.

Decay of Radioactivity in Surface Water Samples Collected April 9, 1953, from Stations Located in Eastern Massachusetts.

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TABLE 24

DECAY PARAMETERS (n VALUES) FOR THE SURFACE WATERS OF EASTERN MASSACHUSETTS FOLLOWING THE APRIL 6, 1953, NEVADA DETONATIONS

Station	April 8	April 9	April 13	April 20	April 28
1	-	1.45	1.92	1.51	1.35
2	-	1.21	1.78	1.44	1.59
3	-	1.90	1.89	1.41	1.92
4	-	1.53	1.64	1.36	1.32
5	-	1.59	1.81	1.32	1.48
6	-	1.74	1.94	1.16	1.43
7	-	1.50	2.05	1.35	0.56
8	-	1.50	1.73	1.16	1.13
9	-	1.49	2.25	1.64	0.71
10	-	1.96	2.56	1.67	0.72
11	-	2.26	2.08	1.47	1.67
12	-	1.28	1.60	1.19	1.30
13(V)	1.49	2.01	-	1.00	1.46
14(IV)	1.55	1.52	1.78	1.05	0.92
I	1.46	-	0.92	1.65	0.98
II	1.73	-	1.54	0.97	0.90
III	1.61	-	1.03	-	0.85
VI	1.83	-	1.20	-	0.96
VII	1.59	-	1.18	0.63	0.90
VIII	1.48	-	0.63	0.61	-
IX	1.29	-	1.38	-	0.33
X	1.28	-	1.38	0.73	0.70
m n	1.53	1.61	1.63	1.23	1.10
m log n	0.133	0.208	0.113	0.098	0.010
SD log n	-	-	0.147	-	-

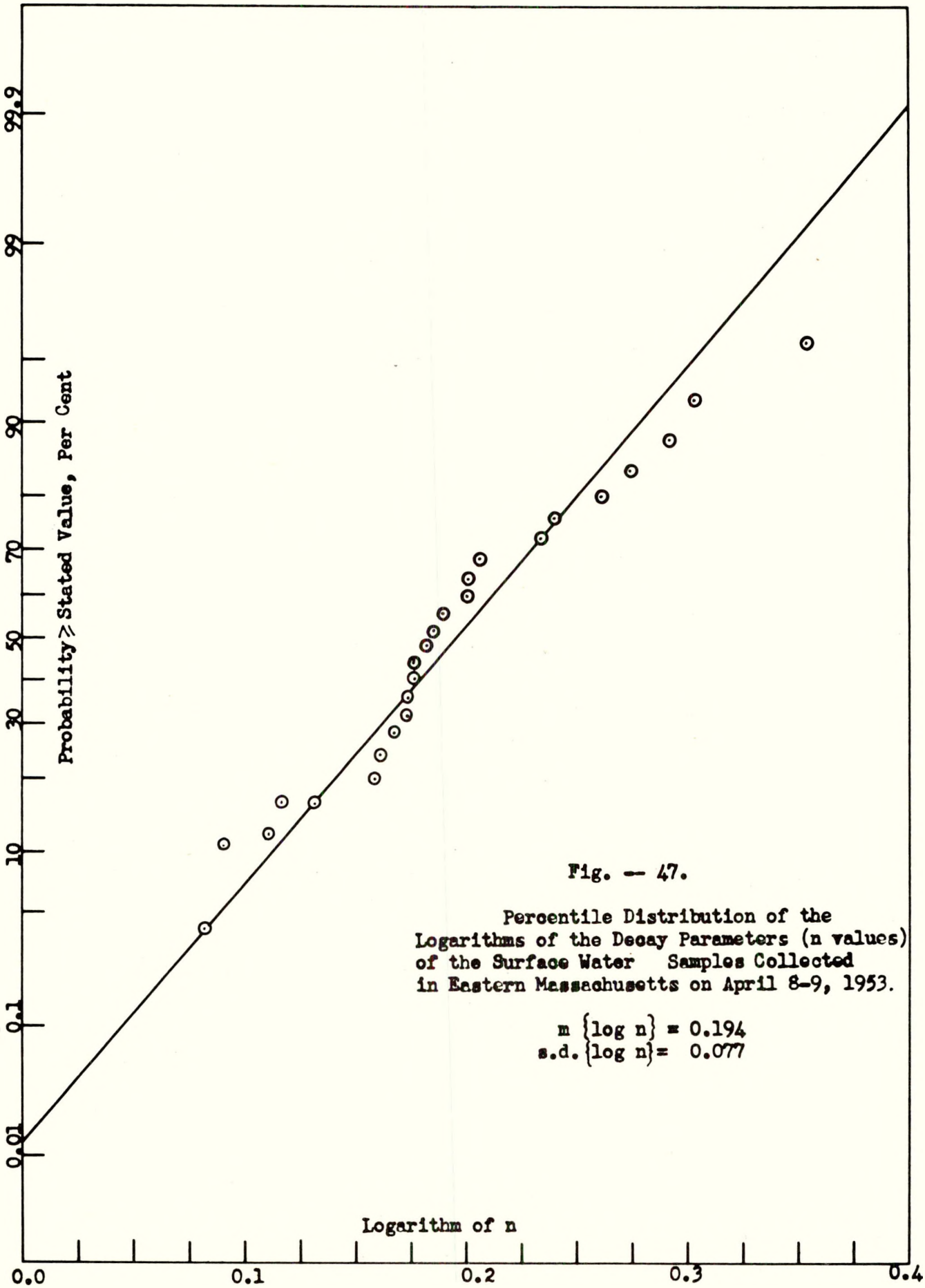

Fig. — 47.

Percentile Distribution of the Logarithms of the Decay Parameters (n values) of the Surface Water Samples Collected in Eastern Massachusetts on April 8-9, 1953.

$m\{\log n\} = 0.194$
$s.d.\{\log n\} = 0.077$

shows the percentile distribution of the logarithms of the decay parameter of the samples collected from the surface waters on April 8 and 9.

The change in the decay rate with time after this detonation is shown by a curve (Figure 48) drawn through the geometric mean values of the decay parameters obtained from the measurements of the samples taken between April 8 and April 28. The single points represent decay parameters measured on the Cambridge rain samples and plotted as on the date the samples were collected. The fluctuations in the decay parameters could not be due to statistical errors of counting alone.

Some of the relationships between the mean decay parameters of the eastern Massachusetts surface waters following the April 6, 1953, detonation were compared statistically by means of the "Student" t-test (65). In each case the pertinent distributions were plotted on probability paper and it was found that they were sufficiently close to arithmetic normality or logarithmic normality for distribution comparisons. At a probability level of at least 95% the following statements are true: (a) the mean of the decay parameters of the surface water samples taken on April 13 are different from the means of the respective decay parameters of those samples collected on April 8, 9, and 20 and (b) the mean of the decay parameters of the surface water samples collected on April 8 and 9 differs from the mean of the decay parameters of the rain samples collected on the same dates. Thus, there is reason to believe that there is selective removal of some of the radioactive nuclear species when long-range fallout enters the surface waters.

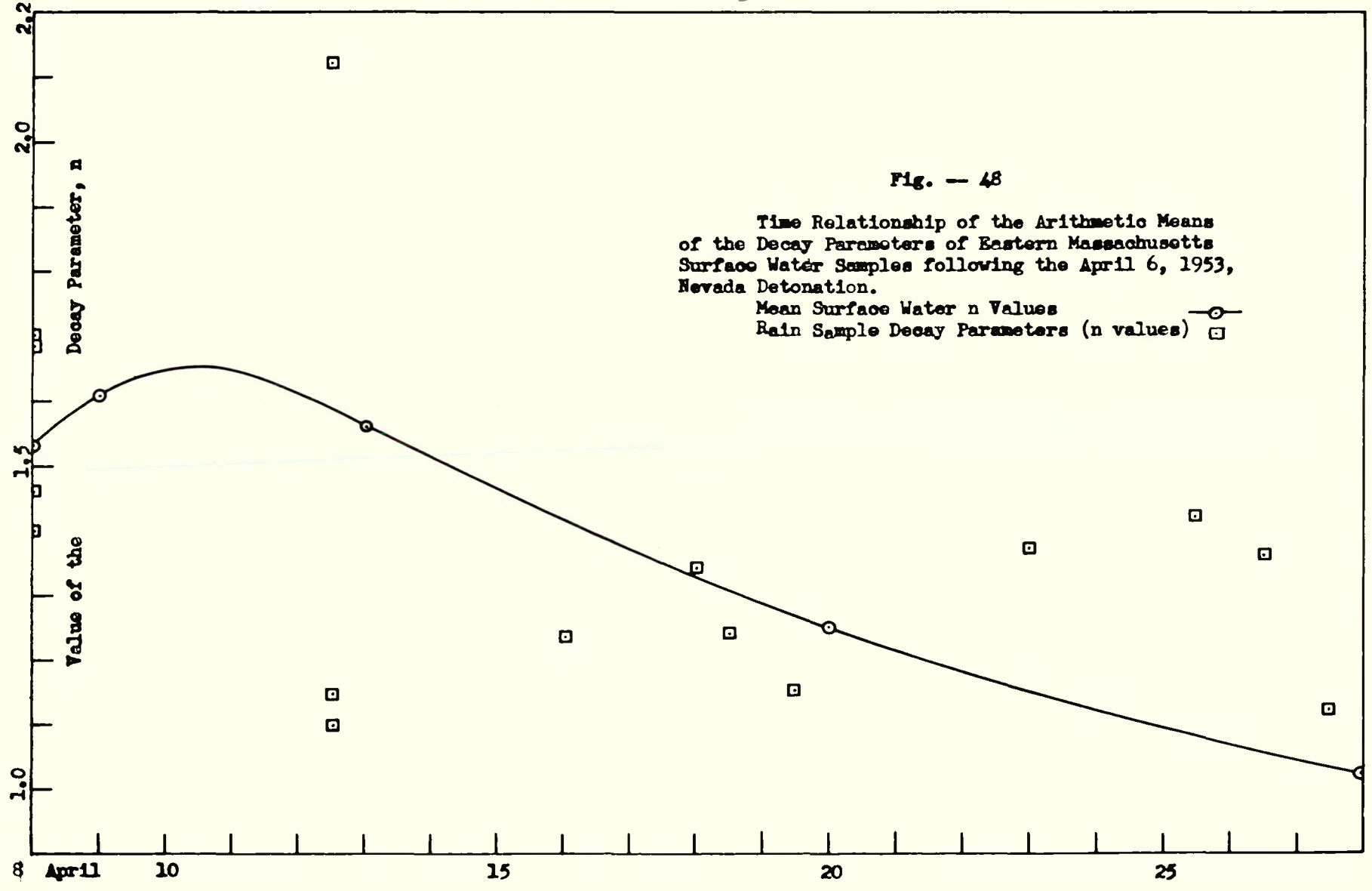

Fig. — 48

Time Relationship of the Arithmetic Means of the Decay Parameters of Eastern Massachusetts Surface Water Samples following the April 6, 1953, Nevada Detonation.

Mean Surface Water n Values —⊙—

Rain Sample Decay Parameters (n values) ⊡

G. DECREASE IN RADIOACTIVITY OF SURFACE WATERS AFTER FALLOUT

In the study of the decrease of radioactivity (counts per minute per liter) of streems and reservoirs, the same hyperbolic type of formulation that is used for the decay of mixed fission products (1) was found (55) to be practical for determining the perameters of decrease in count rate of successive samples taken on the same stream after fallout. For distinction, this parameter will be designated "m", the decrease parameter. The curves of best fit through the points representing the estimated count rates of the successive groups of samples at their time of collection are shown in Figure 43 and Figure 44. It can be seen that it makes a large difference in this decrease parameter whether it is assumed that the radioactivity was formed in the detonation of June 1 or that of June 6.

Table 25 summarizes the decrease parameters for the various stations following the early June, 1952, and the April 8, 1953, fallout.

H. SEDIMENTATION OF RADIOACTIVITY IN WATER

The first experiments conducted on the sedimentation of long-range fallout in water (55) indicated that it could be an important factor in the removal of this material by nature. Following the increase in radioactivity of the rainfall resulting from the Eniwetok detonations in November, 1952, a series of experiments was conducted to determine more exactly the extent of this type of natural purification. On December 11, eight 500-milliliter graduated cylinders were filled with samples from

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TABLE 25

PARAMETERS FOR THE DECREASE IN COUNT RATE OF SURFACE WATERS FOLLOWING THE JUNE 1, 1952, AND APRIL 6, 1953, NEVADA DETONATIONS

Eastern Massachusetts Station No.	Decrease Parameter (m)	
	June 1, 1952	April 6, 1953
1	1.63	1.48
2	1.36	2.05
3	1.08	1.72
4	1.36	1.91
5	1.51	2.14
6	2.74	1.62
7	1.97	1.89
8	2.75	1.28
9	2.69	0.90
10	2.68	2.05
11	2.48	1.59
12	2.00	0.87
13 (V)	1.71	1.77
14 (IV)	1.60	1.63
I	-	1.46
II	-	1.90
III	-	1.64
VI	-	2.38
VII	-	1.63
VIII	-	2.41
IX	-	1.38
X	-	1.54

the rain of November 22, then left standing in a room maintained at 20° centigrade. At the end of one day, five duplicate 100-milliliter samples were collected at various depths from each of two cylinders of rainwater by slow withdrawal of the water at the surface using a light floating siphon device. At the end of three, five, and eight days this procedure was repeated. Fifty samples were collected and evaporated for this experiment, and all samples were counted on December 27, 1952. Results are graphed in Figure 49. Each plotted point represents the mean of duplicate samples. Lines were fitted to the plotted points for each depth at various ages by least squares methods in an attempt to get an indication of sedimentation. No sedimentation was indicated.

In another experiment to measure sedimentation of the fallout that occurred following the November, 1952, detonations at Eniwetok, tests were run on rainwater that had been left standing undisturbed in 50-gallon crocks for about two months. One-liter samples were carefully siphoned from the tops and from the bottoms of the crocks, evaporated, and counted by the usual techniques. Table 26 summarizes the results, which also indicate that no large portion of the radioactivity in this rainwater settled.

TABLE 26

TESTS FOR SEDIMENTATION OF RADIOACTIVITY IN RAINWATER

Date of Rain	Date Collected and Measured	Count Rates Top of Crock		Count Rates Bottom of Crock	
		mean	S.D.	mean	S.D.
Dec. 5, 1952	Feb. 5, 1953	37.7 cpm/l	1.8 cpm/l	47.6 cpm/l	2.1 cpm/l
Dec. 14, 1952	Feb. 5, 1953	21.0 cpm/l	1.3 cpm/l	18.0 cpm/l	1.2 cpm/l
Nov. 22, 1952	Feb. 6, 1953	9.24 cpm/l	.92 cpm/l	7.34 cpm/l	.92 cpm/l

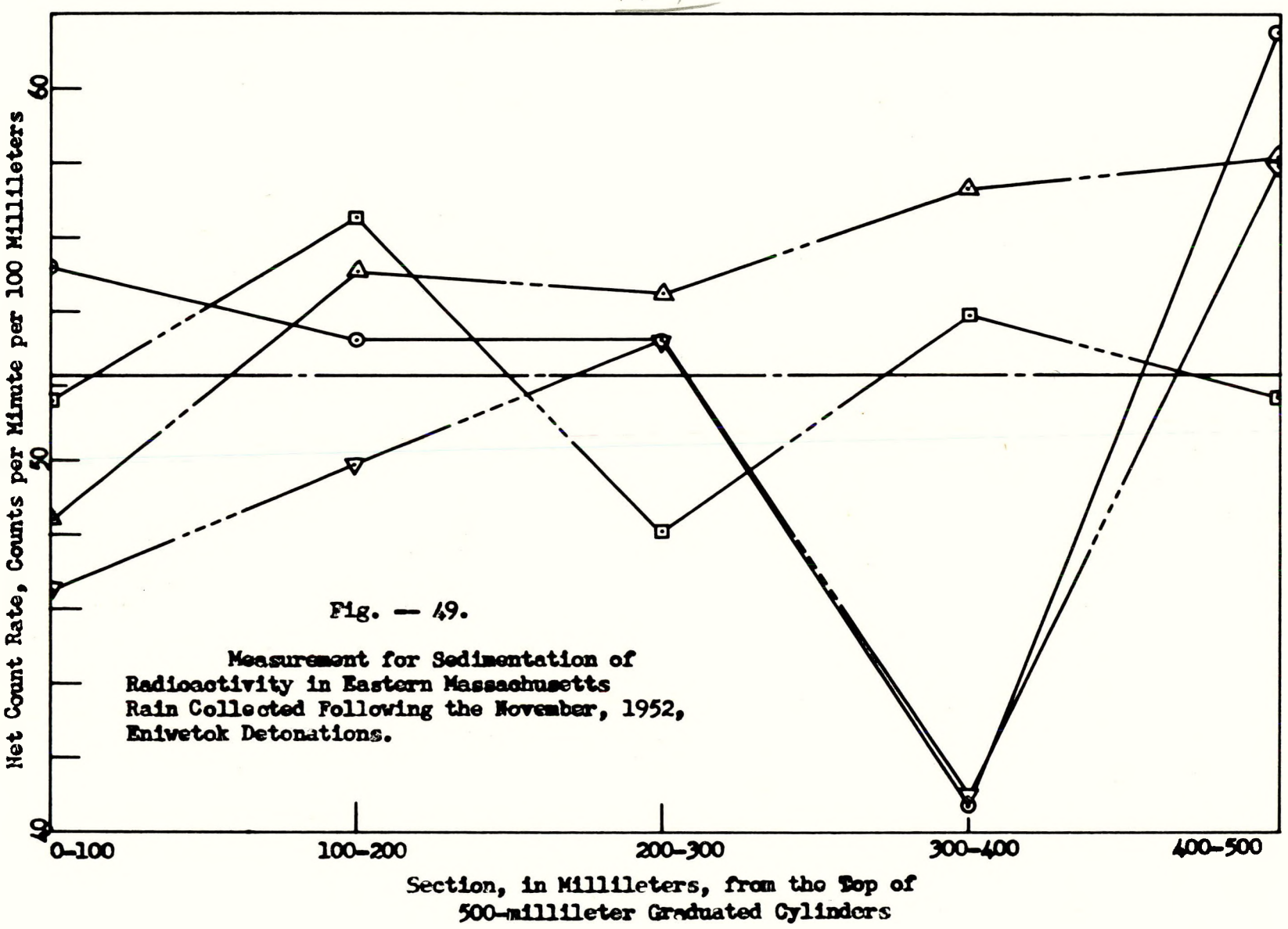

Fig. — 49.

Measurement for Sedimentation of Radioactivity in Eastern Massachusetts Rain Collected Following the November, 1952, Eniwetok Detonations.

I. RETENTION OF LONG-RANGE FALLOUT BY AN ION EXCHANGE COLUMN

On February 4, 1953, an experiment was begun in which rainwater that fell on Cambridge, Massachusetts, on December 5, 1952, was run slowly through an ion exchange column eight millimeters in diameter. The rainwater first passed through four grams of anionic resin (Amberlite) and then through four grams of cationic resin (Dowex 50). On March 13, when nineteen liters of the rain water had passed through the column, the resin was carefully withdrawn from the columns in sections, placed in planchets, and counted on April 2. Table 27 summarizes the data obtained.

TABLE 27

ABSORPTION OF LONG-RANGE FALLOUT IN RAIN BY ION EXCHANGE RESINS

Millimeters from Bottom of Column	Net Count Rate of Resin Counts per Minute
213 - 207 (anionic resin)	21.32
207 - 201 "	7.59
201 - 191 "	0.85
191 - 181 "	3.86
181 - 158 "	2.72
158 - 138 "	1.53
138 - 117 "	3.75
117 - 95 "	5.65
95 - 85 (cationic resin)	125.10
85 - 72 "	22.64
72 - 63 "	11.07
63 - 42 "	9.98
42 - 21 "	2.32
21 - 0 "	1.63

It was found that about five times as much radioactivity was retained in the first layer of the cationic resin as in the corresponding layer of the anionic resin. The count rate at the top of the anionic resin section can be in part due to retention of suspended solids in the rain water, such as fly ash and pieces of leaves, which could have held some of the fallout. Ion exchange processes could be developed by sanitary engineers as useful methods for monitoring radioactivity in water treatment plants and systems.

J. A "RUNOFF COEFFICIENT" FOR LONG-RANGE FALLOUT RADIOACTIVITY

1. Eastern Massachusetts

During the Nevada tests in the spring of 1953, integrated samples of the rainfall were collected at eighteen eastern Massachusetts stations, at Harvard, and at the Lawrence Experiment Station. Seven of these sampling stations were located on streams gaged for discharge by the United States Geological Survey. As the quantity of daily rainfall is measured at many points on the collection area by the United States Weather Bureau and by the Massachusetts Department of Public Health, all of the necessary information is available for a calculation of the ratio of the total amount of radioactivity that fell on each of the seven watersheds to the total amount of radioactivity that ran off. All but one of these calculations are based on the radioactivities of the precipitations and streams just before and for about three months after the large fallout that occurred following the April 6, 1953, Nevada detonation.

Fallout calculations for each watershed were obtained by correcting each of the one-liter rain sample net count rates for fluctuations in sensitivity of the Geiger-Müller tubes and scalers, then averaging the rates obtained from the stations of each watershed, and finally multiplying the mean count rate by the quantity of rainfall that occurred (in liters per square mile) on the watershed during the period of days the integrated rain samples were collected. The measured count rates of the three weekly sets of rain samples collected following the April 7-8 fallout were corrected to the count rates at the time of fallout, using the decay rates obtained from the dieaway measurements of the samples. The net count rates per square mile at the time of precipitation were then summed for the period under consideration and the total count rate was converted to microcuries per square mile by the procedure given in Chapter V. The masses of the evaporated samples were usually so low that self-absorption corrections were not large.

The count rates of the stream samples were also corrected for instrument sensitivity fluctuation and those collected during the first month after the large fallout were corrected for the decay that occurred between collection and first counting. Each of these count rates was then plotted for its collection date, a smooth curve drawn through these points and count rates thereby estimated for all days between collections. Figure 50 shows these curves for six of the streams and a curve representing the averages of the six. These curves are placed in the same figure but displaced laterally for better illustration. In each curve, the peak is placed at 3:00 hours on April 8, the estimated mean time of this large fallout.

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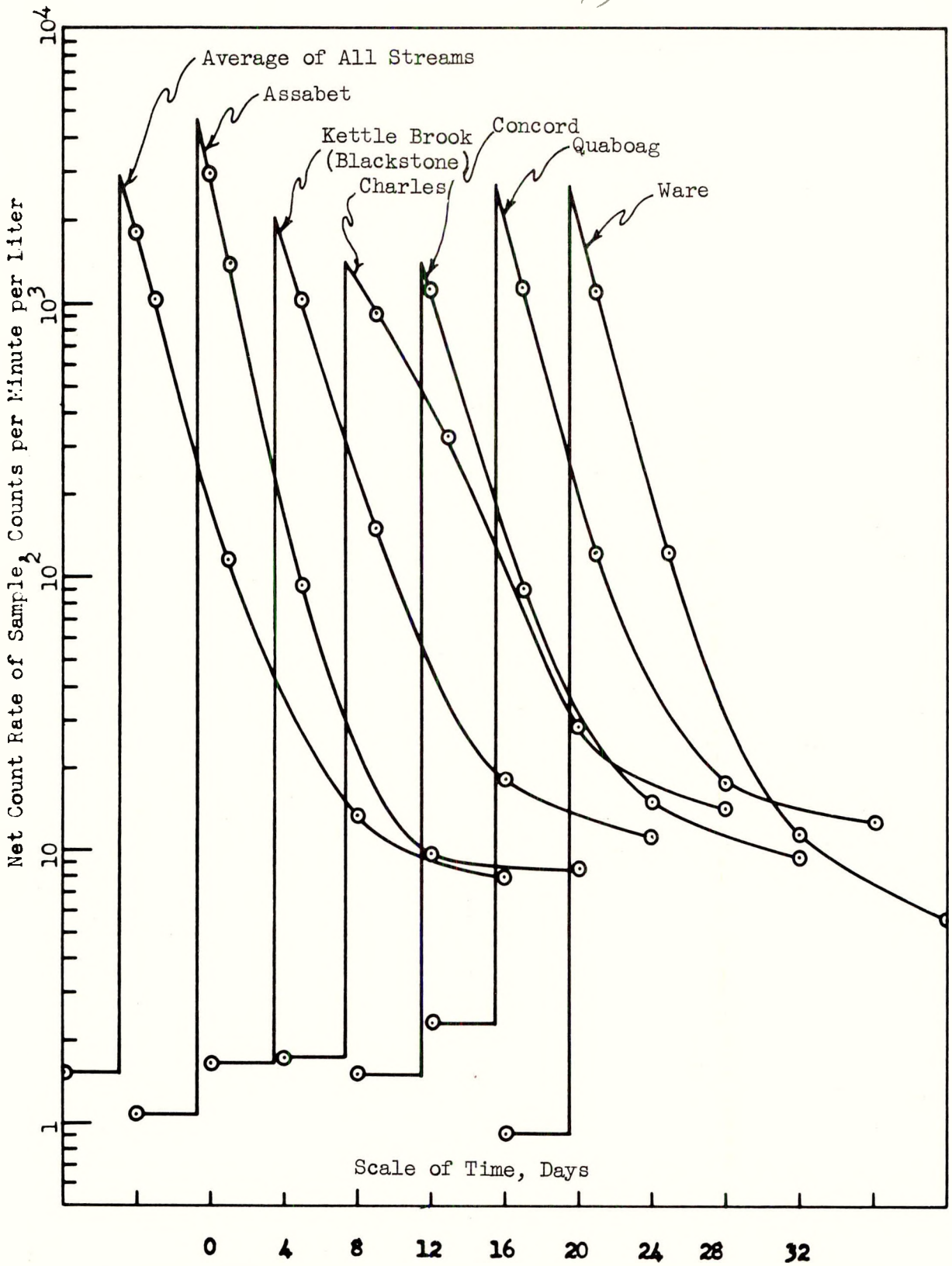

Fig. 50—Radioactivity in Six Eastern Massachusetts Streams Following the April 6, 1953, Nevada Detonation. The Vertical Portion of Each Curve Represents the Time of Fallout (Approximately 38 Hours after Detonation). Curves for Streams are Separated by Displacement of Abscissa Values.

The daily count rate values, when multiplied by the stream flow (liters per square mile per day), gave the radioactivity runoff in terms of (counts per minute) • (days per square mile). These were cumulated over the same period as the precipitation radioactivity values. All samples of evaporated residue used to measure the count rates in the streams were weighed, and the average weight for each stream was used in the conversion of the sums of runoff radioactivity to microcuries per square mile for the period in comparison.

If this sum of the radioactivity discharge values (microcuries per square mile on the day of discharge) is divided by the total radioactivity (microcuries per square mile on the day of deposition) over the same period after major fallout, an estimate is obtained for the proportion of the beta-ray-emitting long-range fallout radioactivity actually appearing in the streams. This quotient the author calls the "runoff coefficient" for fallout radioactivity. It reflects retention by the watershed, self-purification by the stream, and nuclear decay in the runoff period. Table 28 summarizes these results. The high values of the coefficient for the Merrimack River in 1951 and the Assabet and Concord Rivers in 1953 could be due to imprecise measurements of the concentration of radioactivity in the precipitation on the watersheds. The geometric mean of all of these measurements is 1.2% and the median value is 0.37%. The 95% confidence limits for the true geometric mean of the coefficients are 0.27% and 5.1%.

The calculation was repeated for the same time period, using instead the mean rainfall, the mean radioactivity concentration in the

rain, the mean areal discharge of the six streams, and the mean radioactivity concentrations in the streams. The runoff concentration on this basis was 0.44%. For these streams, the average water surface area is about 2.2% of the total area of the watersheds.

TABLE 28

"RUNOFF COEFFICIENTS" FOR GROSS BETA RADIOACTIVITY FROM EASTERN MASSACHUSETTS STREAMS

River	Time Period	Runoff Coefficient of Radioactivity
Assabet	April 1 - June 12, 1953	11.6%
Charles	April 1 - June 25, 1953	0.37%
Concord	April 1 - July 8, 1953	8.31%
Kettle Brook (Blackstone)	March 1 - July 8, 1953	1.03%
Merrimack	April 8 - May 19, 1953	0.11%
Merrimack	Oct. 28 - Nov. 26, 1951	6.26%
Ware	March 1 - July 8, 1953	0.359%
Overall calculation for the six streams	April 5 - April 28, 1953	0.439%

The more difficult problem of the runoff of the long-lived fission debris from watersheds was not investigated.

2. The Genessee River near Rochester, New York

The type of runoff coefficient calculation made for the eastern Massachusetts streams was also made for the Genessee River, which flows into Lake Ontario near Rochester, New York. In this case, data from only one radioactivity sampling station for the rain and one for the river water were available. Results are summarized in Table 29. The low

coefficient for the period from April 1 through October 31, 1952, was due to the radioactive debris that fell on June 4, 5, and 6, amounting to 92% of the total measured for the entire period. This large fallout had little effect on the count rate of the river samples collected in this period. Local fallout at the precipitation collector, but not into the river, could account for this.

TABLE 29

"RUNOFF COEFFICIENTS" FOR GROSS BETA RADIOACTIVITY
FROM GENESSEE RIVER NEAR ROCHESTER, NEW YORK

Time Period	Number of Samples		Runoff Coefficient of Radioactivity
	Precipitation	River	
Feb. 13 - Mar. 31, 1951	27	38	3.99%
Apr. 1 - June 11, 1951	80	33	0.92%
Oct. 16, 1951-Mar.31,1952	110	66	1.43%
Apr. 1 - Oct. 31, 1952	122	114	0.0573%
Nov. 1,1952-Mar. 17,1953	73	37	0.562%
Mar. 18 - Apr. 16, 1953	23	7	1.54%
			Median = 1.22%
Ratio: Total Runoff Radioactivity / Total Fallout Radioactivity	435	295	2.32%

CHAPTER VII

PASSAGE OF NUCLEAR DETONATION DEBRIS THROUGH MUNICIPAL WATER TREATMENT PLANTS

A. INTRODUCTION

Although there have been several pilot plant scale investigations of removal of radioactivity from water, little has been reported on the passage of long-range fallout through water treatment plants (55). Data are included in this thesis showing the removal efficiencies for radioactive fallout of three full-scale water filtration plants. These efficiencies are lower than those obtained in pilot plant studies (97) and far smaller than speculative estimates reported in other investigations (98, 99). Operational data pertaining to the filter plants are presented in Table 30.

B. SAMPLE PREPARATION AND MEASUREMENT

All measurements reported from three different laboratories were made by similar techniques and instruments. In all cases, samples of water were evaporated and the solids carefully transferred to planchets of the same size. Data were collected at rapid sand filtration plants located in Cambridge and Lawrence, Massachusetts, and in the Rochester, New York, area. The water samples from the Rochester, New York, area

TABLE 30

Filter Plant Data

Item	Cambridge, Mass.	Lawrence, Mass.	Rochester, N.Y., Area	
			Rapid Sand	Slow Sand
Quantity treated—*mgd*	17	8	34	4
Solids in influent—*ppm*	70*	76†	165†	165†
Solids in effluent—*ppm*	105*	84†		
Chemicals				
Alum—*ppm*	19	34	22	0
Lime—*ppm*	8.4	13	‡	0
Total chlorine—*ppm*	0.5	2.8	1.1	1.1
Detention period				
Flocculation—*min*	45	20	5	
Sedimentation—*hr*	2	5.4	1§	
Filter sand				
Depth—*in.*	27	30	25	24
Effective size—*mm*	0.50	0.46	0.42	0.20
Uniformity coefficient	1.5	1.9	1.5	1.5
Filtration rate—*gpm/sq ft*	2.3	2.24	2.0	0.18
Avg filter run—*hr*	36	59		8

* Dissolved solids. † Total solids. ‡ Soda ash added. § Solids-contact process basin.

plant were all four liters in volume, those in Cambridge one liter, and those taken at Lawrence 100 milliliters. All radioactivity count rates (principally of beta but also including some gamma radiation) of the evaporated residues were measured with lead-shielded Geiger-Müller tubes that had thin end-windows of approximately two milligrams per square centimeter of mica. Samples for all stages of the plants were taken simultaneously and nearly always counted in close succession. The differences in the total solids in the various stages of each plant were not enough to affect appreciably the relative values of the count rate readings, excepting the Rochester area plant sludge and backwash samples.

All evaporated samples from the Cambridge plant were weighed on an analytical balance and corrections were made for geometry, backscatter, and self-absorption. Factors for these corrections were obtained from calibration measurements made with five pure radioactive nuclear species having different energy levels whose disintegration rates had been established by the National Bureau of Standards. The corrected values obtained were then converted to micromicrocuries per liter. A decay correction was made by the "1.2 law" assuming that all the radioactivity measured was created in the Nevada detonation on April 6, 1953. These corrected values are given in Table 31 in micromicrocuries per liter in addition to the uncorrected count rates. Ratios of the radioactivity of the effluent of the various stages of the Cambridge plant to the radioactivity of the raw inlet are also given. The only important difference between the ratios for the counts and for the corrected values is seen in the tap samples taken soon after the large fallout. This was largely due

TABLE 31

Radioactivity at Cambridge Plant

Assumed Detonation Date	Sampling Period	Sampling Point*	No. of Samples	Observed Radioactivity				Relative Activity	
				Avg		σ_s†		From cpm/l	From μμc/l
				cpm/l	μμc/l	cpm/l	μμc/l		
	2/9/53–	R	24	1.27		0.14		100	
	4/7/53	S	14	1.34		0.19		105	
		F	13	1.57		0.21		124	
		T	3	0.98		0.43		77	
4/6/53	4/8/53–	R	10	20.70	187.0	0.36	3.9	100	100
	4/21/53	S	9	13.08	117.5	0.30	3.0	63‡	63
		F	9	10.43	90.1	0.27	2.4	50‡	48
		T	2	6.75	30.0	0.62	2.1	33‡	16
4/6/53	4/22/53–	R	24	4.30	14.52	0.16	0.55	100	100
	5/18/53	S	25	3.07	10.39	0.14	0.51	71‡	72
		F	25	2.63	8.99	0.15	0.53	61‡	62
		T	18	3.36	11.08	0.17	0.56	78‡	70
4/6/53	5/19/53–	R	31	2.85	9.19	0.13	0.42	100	100
	7/3/53	S	31	2.24	7.31	0.13	0.43	79‡	80
		F	30	1.91	6.08	0.13	0.43	67‡	66
		T	32	2.37	6.30	0.13	0.39	83‡	69

* Key: R—raw water; S—settling-basin effluent; F—filter effluent; T—tap water.

† Standard deviation, from Eq 1 (page 979).

‡ These count rates are significantly different from that of raw water at the 2σ level. For the test period Apr. 8-21, for example:

$$\frac{20.70 - 13.08}{[(0.36)^2 + (0.30)^2]^{\frac{1}{2}}} = 16.8 > 2$$

Hence the difference in observed activities cannot be attributed to random fluctuation in counting rates.

to the decay correction, for the collection-measurement time interval was shorter for the laboratory tap samples than for the plant samples.

No conversion of count rates to disintegration rates has been made for the period preceding the April 6, 1953, detonation, since the radioactivity during this time represented the residual effects of several antecedent bomb blasts, as well as natural radioactivity, and an exact decay correction could not be made. Calibration measurements for the counting equipment used at Lawrence and Rochester were not available; therefore, no conversion of the count rates to disintegration rates has been made with these data. The count rates reported refer to radioactivity at the time of measurement. The time lag between collection and counting was usually less than forty-eight hours.

Also, as another check, most of the Cambridge plant samples were measured a second time on a low-background anti-coincidence instrument and the uncorrected net count rate data grouped as before. No important difference in relative passage values was observed.

In all cases sufficient time elapsed between collection and counting for the count rate of naturally-occurring radon (Em^{222}) and thoron (Em^{220}) and their daughters to decay to a low value. A large number of measurements was made on various surface waters in eastern Massachusetts in the late summer and early fall of 1952, a period of no reported detonations and of little measured fallout. These measurements gave, on the average, approximately one count per minute per liter. It is presumed that this radioactivity was predominately radium (Ra D [Pb^{210}] + E [Bi^{210}]) and radio-potassium (K^{40}). No correction has been made for natural radioactivity in the net count rates reported.

C. MEASUREMENTS AT THE CAMBRIDGE, MASSACHUSETTS, PLANT

The Cambridge, Massachusetts, plant treats 17 million gallons daily of water drawn from a reservoir system having a surface area of 790 acres and a capacity of 3,540 million gallons. Alum, lime, and chlorine are used in this plant, which has flocculation, sedimentation, and rapid sand filtration. There is a 43 million gallon, open, treated water distribution reservoir with a surface area of 6.6 acres that furnishes water to the distribution system for about sixteen hours per week when the plant is inoperative. Table 31 gives the measurements made on this plant during the Nevada test series in the spring and early summer of 1953. The average net count rate recorded in the period from February 9 through April 7 was caused by the small amount of fallout that occurred in this period and by the natural radioactivity. It is interesting to note that measurements made in successive periods following the April 7-8 fallout indicated an increasing ratio of the radioactivity of the effluent to the radioactivity of the influent of the plant. Discrepancies between the disintegration rates in the filter plant effluent and tap samples perhaps reflect the new radioactivity entering as rain in the open distribution reservoir.

Figure 51 shows the radioactivity changes with time for the various stages in the Cambridge plant. The data are plotted on logarithmic scales, so that in accordance with the hyperbolic formulation (1, 55) the decay trends are approximately linearized. The vertical bars through each plotted point represent the 95% confidence zones of the mean of the grouped net counting rates as based on the counting error.

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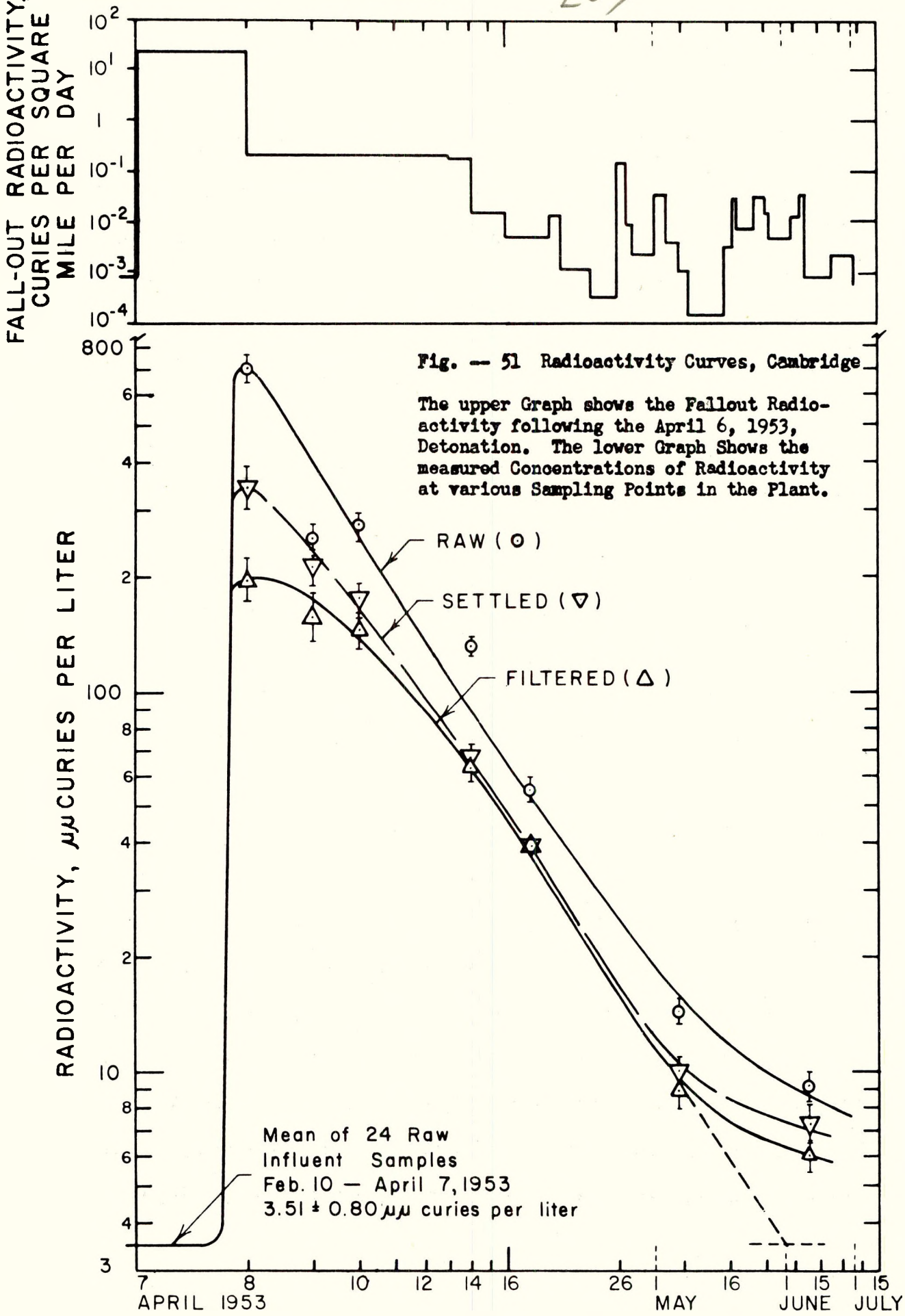

Fig. — 51 Radioactivity Curves, Cambridge

The upper Graph shows the Fallout Radioactivity following the April 6, 1953, Detonation. The lower Graph Shows the measured Concentrations of Radioactivity at various Sampling Points in the Plant.

At the top of this figure is a representation of the estimated radioactive fallout in curies per square mile during the period in which the measurements were made. The rain gage station used in this calculation (Waltham, Mass.) is located near the center of the Cambridge reservoir system and the radioactivity concentration measurements were made on rain collected at the Harvard campus in Cambridge. The fallout graph shown is not nearly so accurate as are the data for the relative counts measured in the various stages of the plant during the period of high fallout radioactivity. It is significant to note that the radioactivity in various stages of the plant had not returned to the pre-April 7 level two months after the large fallout. Part of this effect was due to fallout after April 8, as shown in Figure 51. It is also interesting to note that after the third day following the large April 7-8 fallout, the Cambridge rapid sand filters were able to remove very little of the radioactivity coming from the sedimentation basin.

D. MEASUREMENTS AT THE LAWRENCE, MASSACHUSETTS, PLANT

The watershed of the Lawrence, Massachusetts, plant also received a large fallout on April 7-8, 1953. This plant treats 8 million gallons per day of Merrimack River water in much the same fashion as the Cambridge plant. Table 32 gives the radioactivity of the water passing the various stages of the plant in counts per minute per liter above background.

It is pertinent to note that the count rates in the Lawrence plant following the April 6, 1953, detonation were markedly higher than those at Cambridge. The difference can in part be attributed to the

TABLE 32

Radioactivity at Lawrence Plant

Detonation Date	Sampling Period	Sampling Point*	No. of Samples	Observed Radioactivity		Relative Activity
				Avg *cpm/l*	σ_e† *cpm/l*	
11/1/51	11/6/51	R	1	369	18	100
		S	1	282	16	77‡
		F	1	92	12	25‡
		Res	1	66	12	18‡
		T	1	71	11	19‡
4/6/53	4/8/53–4/12/53	R	6	410.1	7.0	100
		S	6	226.0	4.9	55‡
		F	6	169.3	4.6	41‡
		Res	6	551.7	5.6	135‡
		T	6	168.0	3.8	41‡
4/6/53	4/13/53–4/17/53	R	5	49.6	3.2	100
		S	5	41.5	3.1	83
		F	5	43.3	3.2	87
		Res	5	35.2	3.1	71‡
		T	5	29.6	3.0	60‡
4/6/53§	4/21/53–6/30/53	R	37	13.0	1.1	100
		S	37	5.7	1.0	43‡
		F	37	6.6	1.0	50‡
		Res	37	5.7	1.0	43‡
		T	36	6.8	1.0	52‡

* Key: R—raw water; S—settling-basin effluent; F—filter effluent; Res—reservoir; T—tap water.
† Standard deviation, from Eq 1 (page 979).
‡ Significant difference from raw-water rate at 2σ level. (See Table 2. last footnote.)
§ Assumed.

greater time lag between collection and measurement of the Cambridge samples. However, it has been found that during a general rain storm over a large area a remarkably non-uniform distribution of radioactive fallout can occur. Immediately following the precipitation of April 7-8, 1953, in eastern Massachusetts, the radioactivity in surface waters as measured at twenty-five sampling stations ranged from 21 to 1,077 counts per minute per liter with a mean of 299 counts per minute per liter.

All treated water at Lawrence passes through a 42 million gallon open reservoir, which has a surface area of seven acres. Four daily sets of samples taken following April 7 showed a higher radioactivity for the effluent of this reservoir than for the plant effluent or influent. The decay curves of the set collected on April 9 are shown in Figure 52. The vertical bars through the plotted points represent the 95% confidence zones of the net count rates.

E. MEASUREMENT AT A PLANT IN THE ROCHESTER, NEW YORK, AREA

The 38-million-gallon-per-day Rochester, New York, area plant treats Lake Ontario water taken 1.5 miles from shore and 55 feet below the surface. Alum, soda ash, bentonitic clay, and chlorine (to breakpoint) are used in the plant, which has both unique mechanically-backwashed Blaisdell slow sand filters and conventional rapid sand filters in parallel. No chemicals are added for coagulation before the slow sand filters. There is no open reservoir for treated water as there is at Lawrence and Cambridge. Table 33 gives the counts per minute per

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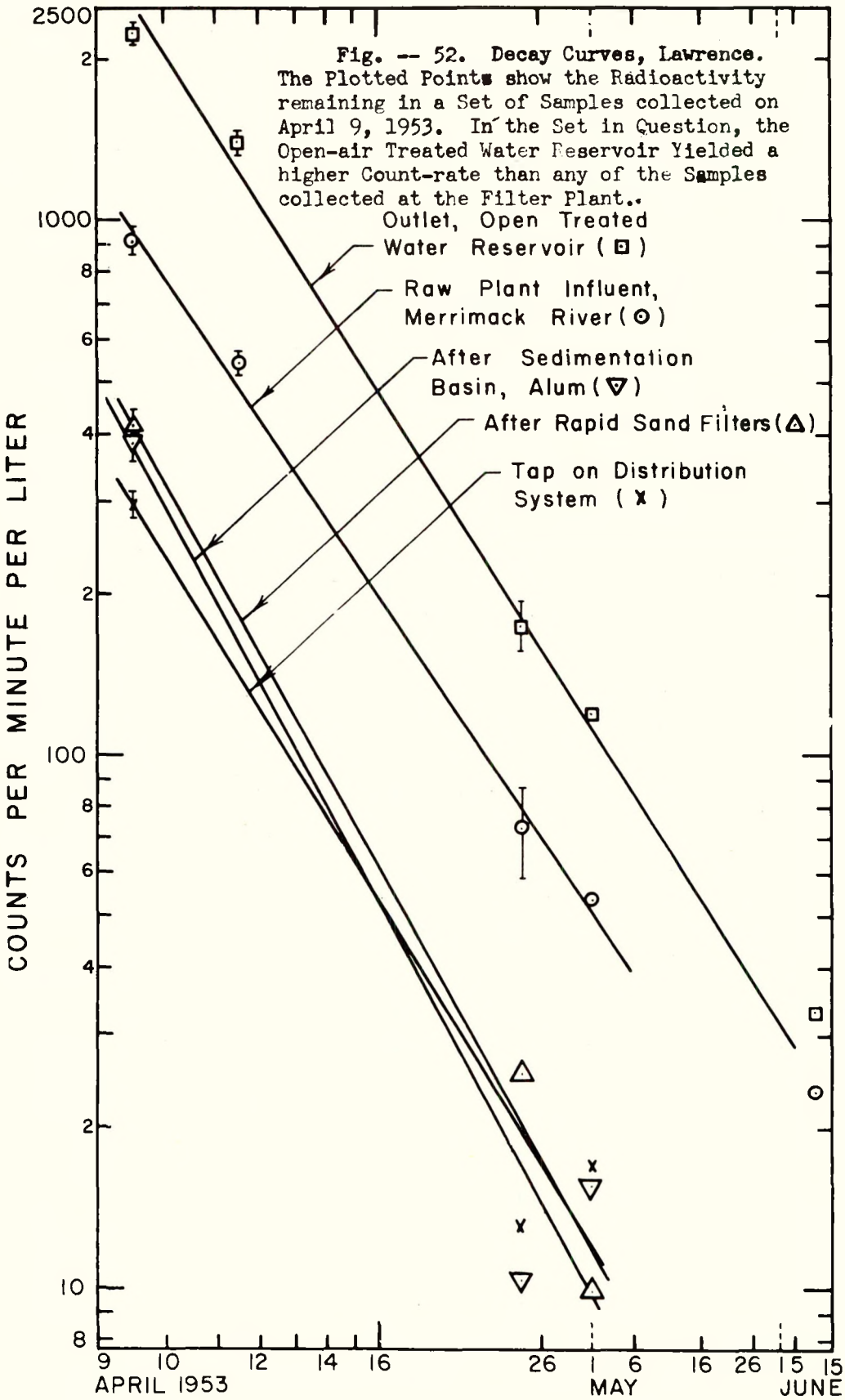

Fig. -- 52. Decay Curves, Lawrence. The Plotted Points show the Radioactivity remaining in a Set of Samples collected on April 9, 1953. In the Set in Question, the Open-air Treated Water Reservoir Yielded a higher Count-rate than any of the Samples collected at the Filter Plant..

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TABLE 33

Radioactivity at Rochester Area Plant

Sampling Period	Sampling Point*	No. of Samples	Observed Radioactivity		Relative Activity
			Avg cpm/l	σ† cpm/l	
3/16/51–10/31/51	R	39	0.70	0.076	100
	Sl	37	31.5	1.3	4,500‡
	B(SS)	37	3.75	0.32	535‡
	F(SS)	39	0.44	0.074	63‡
	F(RS)	38	0.40	0.073	57‡
	T	38	0.55	0.076	79‡
11/1/51–11/15/51	R	12	12.3	0.040	100
	Sl	11	1,390	17	11,400‡
	B(SS)	12	205	2.8	1,660‡
	F(SS)	12	4.53	0.30	37‡
	F(RS)	11	4.32	0.32	35‡
	T	12	5.33	0.30	43‡
11/16/51–1/1/52	R	31	0.89	0.087	100
	Sl	24	65.0	2.56	7,300‡
	B(SS)	30	22.1	0.64	2,470‡
	F(SS)	30	0.51	0.11	57‡
	F(RS)	24	0.38	0.13	43‡
	T	31	0.71	0.11	80
1/2/52–6/3/52	R	81	0.44	0.057	100
	Sl	58	24.2	1.2	5,500‡
	B(SS)	81	3.20	0.24	728‡
	F(SS)	78	0.45	0.06	102
	F(RS)	59	0.32	0.068	73
	T	81	0.35	0.058	80
6/4/52–6/20/52	R	15	3.72	0.17	100
	Sl	11	387	6.0	10,400‡
	B(SS)	14	48.8	1.00	1,310‡
	F(SS)	15	1.45	0.15	39‡
	F(RS)	12	1.76	0.17	47‡
	T	15	1.26	0.17	33‡
6/21/52–7/20/52	R	7	1.29	0.21	100
	Sl	6	63.8	4.3	4,950‡
	B(SS)	7	13.6	0.98	1,060‡
	F(SS)	7	1.08	0.20	84
	F(RS)	6	0.61	0.21	47‡
	T	7	0.88	0.20	68‡
7/21/52–11/1/52	R	29	0.352	0.10	100
	Sl	25	20.4	1.8	5,790‡
	B(SS)	29	2.40	0.29	682‡
	F(SS)	29	0.47	0.10	134
	F(RS)	23	0.47	0.11	134
	T	29	0.50	0.10	142
11/2/52–2/1/53	R	26	0.26	0.10	100
	Sl	23	22.0	2.0	8,500‡
	B(SS)	26	2.25	0.43	870‡
	F(SS)	26	0.31	0.10	119
	F(RS)	23	0.39	0.11	150
	T	26	0.40	0.11	154
2/3/53–3/20/53	R	14	0.54	0.14	100
	Sl	14	21.2	2.5	3,920‡
	B(SS)	14	2.60	0.48	482‡
	F(SS)	14	0.26	0.14	48
	F(RS)	14	0.66	0.15	122
	T	14	0.37	0.14	69
1/2/53–5/12/53§	R	37	0.398	0.072	100
	Sl	36	10.2	0.62	2,560‡
	B(SS)	37	2.53	0.30	635‡
	F(SS)	36	0.293	0.075	74
	F(RS)	36	0.528	0.077	133
	T	37	0.302	0.075	76
5/15/53–6/12/53§	R	8	0.53	0.16	100
	Sl	8	40.1	1.62	7,560‡
	B(SS)	9	7.62	1.06	1,440‡
	F(SS)	8	0.59	0.16	112
	F(RS)	8	0.69	0.16	130
	T	8	0.59	0.16	111

* Key: R—raw water; Sl—sludge blanket of solids-contact process basin; B(SS)—slow sand filter backwash water; F(SS)—slow sand filter effluent; F(RS)—rapid sand filter effluent; T—tap water.

† Standard deviation, from Eq 1.

‡ Significant difference from raw-water rate at 2σ level. (See Table 2, last footnote.)

§ Measured at different laboratory from other Rochester sample groups.

liter at the various stages in the plant for periods after an appreciable fallout in the lake (91). The dates of fallout causing the increases in lake water radioactivity were November 1, 1951, and June 4-5, 1952, and were due to the Nevada detonations of October 28 or 30, 1951, and June 1, 1952, respectively. The count rates of the sludge and the backwash samples are significantly greater than those of the water samples and indicate the extent to which radioactivity is accumulated and concentrated in coagulation and filtration. Sludge samples should yield valuable information in routine monitoring operations.

F. ERROR ANALYSIS

Precision of measurement of count rates is limited by several sources of error which, for convenience, can be classified in three groups: collection, sample preparation, and counting. The counting procedure provided the most important source of variance. The cumulative effect of the errors is such that little reliance can be placed on results from single sets of samples from a plant. However, when several sets are averaged as in Tables 31, 32, and 33, the precision is adequate to delineate trends.

The counting error was evaluated by the following formula:

$$\delta_c = \frac{1}{n}\left[\sum_{1}^{n}\frac{r_{s+b}}{t_{s+b}} + \frac{r_b}{t_b}\right]^{1/2} \qquad (7\text{-}1)$$

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where δ_c is the standard counting error of the mean of the n count rates in each group

$r_s + b$ is the counting rate of the sample with background

$t_s + b$ is the total time the sample with background was counted

r_b is the counting rate of the background alone

t_b is the total time the background alone was counted.

At both Lawrence and Cambridge the measurements were alternated with background measurements. Automatic sample-changing equipment was used and at least 1,024 counts were cumulated for each sample and each background. The samples from the plant in the Rochester, New York, area were changed manually and background was counted each day usually for a half hour or more. The standard deviations given in Table 31, 32, and 33 were computed by Equation (7-1) and pertain only to counting error.

The raw inlet count rates from each of the three plants for periods having no appreciable trends in the radioactivity of the samples were used to get the ratio of all the errors to the counting error alone. The standard error of each of these groups was computed by the formula

$$s_t = \sqrt{\frac{\sum (r - \bar{r})^2}{n(n-1)}} \qquad (7\text{-}2)$$

where s_t is the standard error of the group mean due to all types of error

n is the number of samples

r is the net count rate of each sample

$\bar{r}$ is the mean of the net count rates of the group.

For these groups the standard counting error of the mean of the group was obtained using Equation (7-1). A Lexis-type ratio was obtained by dividing s_t by s_c.

It can be seen from Table 34 that the counting error was the major factor limiting reproducibility. The Cambridge data had the smallest errors. It is believed that in following radioactivity trends in soft eastern waters at levels 1 to 100 times those due to natural radioactivity, the technique used at Harvard (one-liter samples; 1,024 counts per sample) represents a reasonable balance between the time and effort expended in sample preparation and counting and the precision attained. With larger samples, such as used at Rochester, self-absorption of radiation in the solid residue becomes a limiting factor; with smaller samples such as used at Lawrence, reproducibility is limited by the low disintegration rates obtained. The measurements at Rochester were made primarily for monitoring, and with the relatively short counting times used the precision attained was satisfactory for the purpose.

TABLE 34 212

ERROR ANALYSIS

Item	Cambridge Plant	Lawrence Plant	Rochester Area Plant
Sampling Period	April 21, 1953-June 30, 1953	April 21, 1953 - June 30, 1953	November 4, 1952-March 20, 1953
Number of samples	36	29	40
Standard error Total, s_t (Eq. 7-2)	0.180	1.75	0.302
Counting, s_c (Eq. 7-1)	0.135	1.070	0.321
s_t/s_c	1.33	1.64	0.942

G. OTHER INVESTIGATIONS OF PLANT PASSAGE OF RADIOACTIVITY

An investigation reported by Straub (97) at the Oak Ridge National Laboratory showed a passage of only 27-30% of influent radioactivity at a pilot plant having coagulation, sedimentation, and filtration facilities and using alum, lime, and sodium silicate. Instead of actual fallout material, the radioactivity source was pile-produced "fission products mixed to simulate conditions after a bomb blast, but with increased percentages of radioactive ruthenium, iodine, and strontium, which are hard to remove."

Other studies (98, 99) based largely on theoretical considerations have resulted in estimates of treatment plant efficiencies much larger than those found in this investigation.

It is of significance to note that very few of the passage percentages in Tables 31, 32, and 33 are as low as those reported by Straub. This difference is not surprising, for it is very unlikely that the ages of the radioactive material used at the Oak Ridge National Laboratory were like those of the detonation debris investigated for this thesis. Fallout measurements were made at the Cambridge plant from approximately two days to three months after fission. During this period those nuclear species causing most of the radioactivity of fission products change often because of different decay rates and radioactive daughter production. Moreover, decay measurements reported in Chapter VI indicate that during runoff from land surfaces and storage in reservoirs, self-purification results in a selective removal of certain radioisotopes from the fallout material, so that the response of the mixture to treatment is altered.

Other differences between the fallout and the pile-produced materials could derive from differences in fission rate and temperature in the processes in which they are created. Whereas the Oak Ridge National Laboratory fission products were probably in ionic or radiocolloidal form, there is ample evidence to show that long-range fallout is, at least partially, in the form of discrete particles. The particulate composition of the fallout is illustrated in Figure 53, which is a print of an autoradiograph of the solids from twenty-two liters of rain collected at Harvard. The solids were spread on an aluminum pan 0.020 inches thick. K-type X-ray film was used with a six-week period of exposure. Another piece of the same sheet of film was placed in

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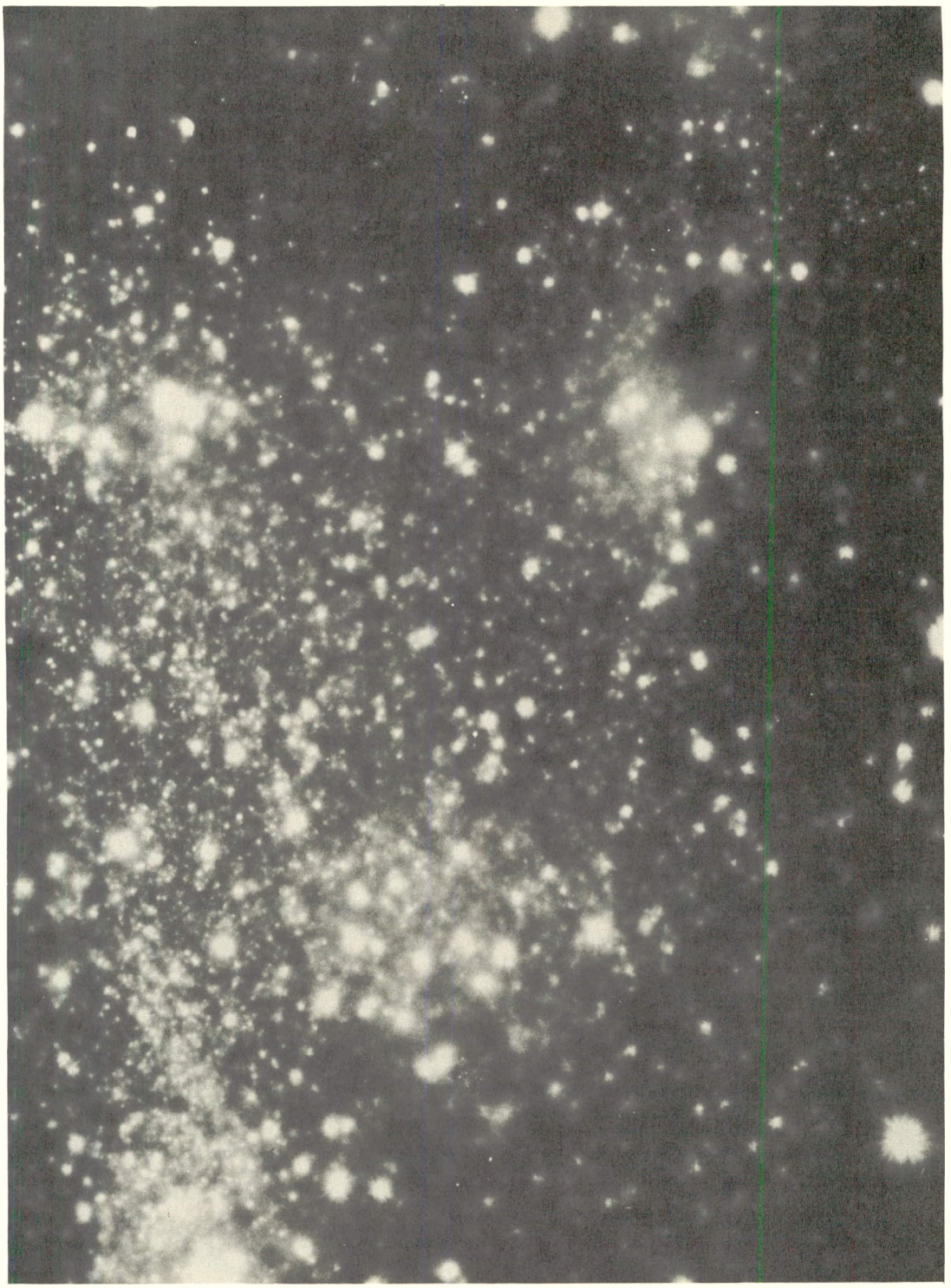

FIGURE 53 AUTORADIOGRAPH OF THE FALLOUT AT HARVARD UNIVERSITY. 22 LITERS OF RAIN WATER WAS COLLECTED AND EVAPORATED. THE RESIDUE WAS KEPT IN CONTACT WITH TYPE K X-RAY FILM FOR 6 WEEKS. THE RESULTING AUTORADIOGRAPH ILLUSTRATES THE PARTICULATE COMPOSITION OF THE FALLOUT.

contact with the bottom of the pan and given the same exposure and development. Figure 54 is a print of this autoradiograph. The difference in the size of the spots (representing exposure) is principally due to absorption of the beta radiation of the fallout by the 0.020 inches of aluminum.

Hursh (100) measured the natural radium (Ra^{226}) content of raw and tap water samples from forty-two public water supplies in the United States. For twenty-nine plants with flocculation, sedimentation, and filtration, the mean radium passage (ratio of concentration in tap water to that in plant influent) was 58%, with a standard deviation of 31%. The product moment coefficient of correlation between the percentage passage of radium and the concentration of radium (Ra^{226}) in the raw water is -0.72. The probability is 95% that the true coefficient lies between -0.47 and -0.84. This indicates that a high radium content in the raw water is associated with high removal in treatment, which, in turn, suggests that the radium (Ra^{226}) is associated with suspended matter in the water. For eleven supplies with no treatment other than chlorination, the mean ratio of the radium concentration in the tap water to that in the raw water was 72%, with a standard deviation of 47%. Although the same factors did not necessarily operate, this value is in accord with those results in Tables 31, 32, and 33 that indicate low removal efficiencies during periods remote in time from bomb tests.

FIG. 54 REVERSE AUTORADIOGRAPH OF SAME FALLOUT SAMPLE USED FOR FIG. 53 BUT WITH A 0.020 INCH INTERVENING ALUMINUM ABSORBER BETWEEN THE FALLOUT SAMPLE AND TYPE K X-RAY FILM. THE UPPER THREE INCHES OF THIS FIGURE WERE NOT EXPOSED AND THEREFORE SERVE AS A CONTROL.

H. COMPARISON WITH RADIOACTIVITY TOLERANCE LEVELS

As an operational guide in evaluation of health hazards due to dispersal of radioactivity from continental weapons tests, the Atomic Energy Commission has used the following criterion: to determine whether radioactivity measured in water at any time after fallout is safe, the radioactivity measurement is extrapolated to three days' age by the "1.2" decay law; if the extrapolated value is less than 5,000 micromicrocuries per milliliter, then the water can be used safely for any period of time.

For the Cambridge data the plant effluent radioactivity at three days (Figure 51) is 0.175 micromicrocuries per liter or 1/29,000 of the foregoing estimated safe upper limit of concentration. The safety factor is increased to 34,000 if a comparison is made of the total exposure in the time interval between thirty-eight hours (the travel time from Nevada) and fifty-five days following the blast, when radioactivity levels had fallen close to the natural level. The cumulative exposure for this period calculated from the plant effluent curve of Figure 51 was 1.24 micromicrocurie days per milliliter. The safe exposure by the Atomic Energy Commission criterion can be calculated from the integral

$$5000\ (3)^{1.2} \int_{1.67}^{55} t^{-1.2}\, dt = 42{,}800\ \mu\mu c \text{ days/ml.}$$

The safety factor accordingly is 42,800/1.24 = 34,000. The higher safety factor of the latter computation stems from the fact that

the actual disappearance of radioactivity in the water supply proceeds at a faster rate than given by the "1.2" law. The safety factor for the Lawrence effluent during the same period is estimated at 1,500 to 2,000.

I. CONCLUSIONS

Approximately 2,200 samples taken in the various treatment stages of three water plants were measured for beta-gamma radioactivity. All plants had alum coagulation and rapid sand filtration. The water sources were a system of small reservoirs, a river, and Lake Ontario.

From these measurements, which are summarized in Table 35, it was found that within about two weeks after a nuclear detonation, about 45% of the gross long-range fallout radioactivity completely passed through rapid sand filtration plants. From two weeks to ten weeks after detonation, about 53% passed through the plants. For periods further than ten weeks from detonation, practically all the radioactivity (partially of natural origin) passed through the plants. In every case the removal efficiencies were lower than those reported in pilot scale plants with simulated fission product mixtures.

No measurement of radioactivity made in the investigation to date has exceeded the emergency levels set for radioactivity in water containing bomb fission debris.

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TABLE 35

Summary of Filter Plant Radioactivity

Sampling Point*	Cambridge, Mass. (Reservoir System)			Lawrence, Mass. (Merrimack R.)			Rochester, N.Y., Area (L. Ontario)		
	No. of Sample Sets	Net Count Rate *cpm/l*	Relative Activity	No. of Sample Sets	Net Count Rate *cpm/l*	Relative Activity	No. of Sample Sets	Net Count Rate *cpm/l*	Relative Activity
First 2 wk After Large Fallout									
R	9	20.7	100	11	246	100	24	7.55	100
S	9	13.1	63	11	142	58			
F(SS)							24	2.82	37
F(RS)	9	10.4	50	11	112	45	24	2.98	40
Res				11	317	129			
T				11	105	43	24	3.07	41
2–10 wk After Large Fallout									
R	50	3.48	100	36	13.0	100	30	0.96	100
S	50	2.61	75	36	5.7	43			
F(SS)							30	0.60	62
F(RS)	50	2.24	64	36	6.6	50	30	0.44	44
Res				36	5.7	43			
T	50	2.73	78	36	6.8	52	30	0.74	77
More Than 10 wk After Large Fallout									
R	13	1.27	100				141	0.45	100
S	13	1.34	105						
F(SS)							141	0.43	96
F(RS)	13	1.57	124				141	0.38	85
Res									
T							141	0.43	96

* Key: R—raw water; S—settling-basin effluent; F(SS)—slow sand filter effluent; F(RS)—rapid sand filter effluent; Res—Reservoir; T—tap water.

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CHAPTER VIII

FUTURE RESEARCH IN THE SANITARY ENGINEERING ASPECTS OF LONG-RANGE FALLOUT

There are many paths that future research could follow in the sanitary engineering aspects of long-range fallout. The author believes that the practical worth of this thesis lies in the preliminary estimates of what the effects would be at great distances from detonations of nuclear devices if many were used in warfare. Fortunately, radioactive fallout at levels far below those that are dangerous can be analyzed, traced, and measured.

It would be worthwhile to make investigations at the Nevada Proving Grounds to check the runoff into any nearby streams or lakes, either large or small, intermittent or continuous, to find how the radioactivity is transported and deposited. The past radioactivity monitoring records for Lake Mead near the Nevada Proving Grounds could be reviewed as preparation for an intensive study should an ample fallout occur there following some future Nevada test. If such a study were planned, the water, plankton, and also benthal and littoral deposits should be measured beforehand for natural and long-deposited fallout radioactivity concentrations.

Possibly in a restricted zone such as the Nevada test site samples of the radioactive species that are the principal hazards to health could be placed on the ground and left to nature. Periodic checks and tests could be made to find how the radioactivity moves across the soil surface and penetrates the ground and also what the extents of carriage by runoff and wind are. On a model basis, the penetration of elements into soil, their progress through artificial streams, and their uptake by plankton and suspended solids could be measured. An attempt should be made to simulate the chemical and physical states in which these elements would be expected to be found following a nuclear detonation.

In anticipation of possible explosions of nuclear reactors, the same type of experiments as just described could be made, using instead synthetic materials made as similar as feasible to those likely to be produced in disasters of this kind. One would expect large variations in these explosions, but results from properly designed experiments should yield valuable information on which to base procedure in emergencies.

As far as the general study of low-level fission debris in nature is concerned, it would seem reasonable first to apply major effort on those nuclear species that are thought to be the most dangerous because of toxicity and abundance, such as strontium, barium, cesium, zirconium, and promethium. As these radioelements exist in important amounts in fission products, it seems likely that most could be easily measured at relatively low levels of radioactivity.

Preliminary studies along this line were conducted at Harvard for this thesis in the spring of 1953. Separations were attempted of radioactive strontium, barium, and ruthenium. It appeared that a small amount of fallout radio-barium was isolated from a sample of eastern Massachusetts river water collected on April 8, although the amount obtained was much too low for positive identification. Some of these separation studies are being continued. It seems important to conduct more studies on the most hazardous species of fission isotopes, both in laboratories and in field experiments, using reactor-produced materials, as well as the analysis of fallout.

If the equipment and procedure were established at a municipal water treatment plant before a fallout occurred, it would be possible to run partial radiochemical analyses on surface water samples collected at various stages in passage through the plant. It would be possible to make these analyses on samples collected at times relatively remote from fallout depositions. A study of the concentrations of the more important fission species in municipal water treatment plant sludge should also give valuable information.

In many parts of the United States, it is simple, if one is patient, to collect rain that has easily-measurable quantities of radioactivity formed in Nevada detonations. At Harvard, ample quantities of this rain have been collected for laboratory experiments by connections to the roof drains. With samples on hand, many tests can be run. As the time after fallout increases and the dominant species in the fission material change, repeated experiments with the same rain-

water could be conducted to measure the effects of fission debris age.

A surprisingly large number of investigators have collected information on long-range fallout, often incidental to their routine monitoring procedure. A review and comparison of these measurements based on the viewpoint of the sanitary engineer should yield useful conclusions on this aspect of the problem.

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