AN INVESTIGATION OF RELATIONSHIPS BETWEEN METEOROLOGY AND RADIOACTIVITY LEVELS IN RAIN

Thesis for the Degree of M. S.

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William Evan Norris

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THESIS

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"AN INVESTIGATION OF RELATIONSHIPS BETWEEN METEOROLOGY AND RADIOACTIVITY LEVELS IN RAIN"

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ABSTRACT

AN INVESTIGATION OF RELATIONSHIPS BETWEEN METEOROLOGY AND RADIOACTIVITY LEVELS IN RAIN

by William Evan Norris

This thesis proposes a laboratory procedure for the continuous fission product monitoring of air and rain, and presents the results of a brief sampling period.

Low volume air samples were taken through Millipore filters in contrast to the usual procedure of high volume sampling through fiber filters, and the specific activity of each 0.15 inch increment of rain was measured. Air activity at the time of the rain and rain activity were compared in terms of the meteorology describing the rain.

A strong relationship between air and rain activities was observed. It was also observed that rain activity may be related to wind speed and overhead pressure systems, and an equation is proposed incorporating the former. In addition, it is proposed that summer rains, which are areally small, need not decrease in specific activity with time.

AN INVESTIGATION OF RELATIONSHIPS BETWEEN METEOROLOGY AND RADIOACTIVITY LEVELS IN RAIN

By

William Evan Norris

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I. INTRODUCTION

This is one of a series of theses investigating the fundamental processes of radioactive pollution in surface and subsurface waters. The pollutional sources are fallout fission products, radioisotope wastes, and naturally occurring alpha emitters.

This thesis is primarily concerned with the development of a laboratory procedure for the measurement, by gross beta counting, of fission product radioactivity concentrations in the atmosphere and rain. Such measurements, correlated with the work of other researchers, should lead to relationships yielding the amount of fallout deposited by rain in terms of the concentration of radioactivity in the atmosphere and the meteorology describing the rains.

Although correlation between atmospheric radioactivity and radioactivity in rainwater exists (1), and some excellent research has been presented in recent months, no relationship is known which combines all the variables in one model.

The fallout studied is defined by the U. S. Department of Defense as "delayed fallout," that is; the very fine particles resulting from nuclear detonations, with radii of a few microns or less, which fall extremely slowly under the influence of gravity. These particles remain suspended in the atmosphere for a considerable time and are carried over great distances as the result of the movements of air. They

are removed primarily by the scavenging effect of rain and snow, producing the delayed fallout (2).

The presence of fission product radioactivity has been reported in streams and waterways (3), foodstuffs (4,5), and sewage treatment plants (6). This world-wide problem does not represent an immediate health hazard, but it may result in long-term injury from internal deposition and possible genetic effects from low-level irradiation (7). For these reasons, and because dangerous levels of atmospheric radioactivity could be attained in all-out nuclear conflict, studies such as this are justified.

This thesis embodies (a) a literature study which abstracts some of the most recent publications on atmospheric fission product radioactivity, (b) the laboratory setup, and (c) the results of a brief sampling period.

The atmospheric radioactivity levels measured in the laboratory were compared with the same measurements provided by The Division of Occupational Health of the Michigan Department of Health. Weather data were obtained from the United States Weather Bureau at Lansing, Michigan.

II. LITERATURE REVIEW

The purpose of this chapter is to outline some of the current thought on the migration of fission product debris from the stratosphere to the earth's surface, insofar as it is deposited by rain.

Except for a continual leakage from the stratosphere, the introduction of fission product debris into the troposphere is apparently the result of the violent mixing of the tropospheric and stratospheric interface. Miyake, et al (8,9) have correlated remarkable increases in the specific activities of air and rain with the simultaneous occurrence of troughs at the 500mb isobaric surface and jet streams.

Sotobayashi and Koyama (10) have investigated the movement of debris from the upper troposphere to rain producing layers. They observed that troughs in the 300 and 500mb isobaric surfaces were accompanied by cold front type rains which were characterized by generally higher levels of specific radioactivity. The air currents associated with these troughs have a sinking motion and serve as a conveyer which transports the debris into the rain producing layer. By assuming that this moving air mass neither receives nor loses debris in its descent, the content of debris per unit volume in the lower troposphere increases in proportion to the magnitude of the downward component of the upper air current.

Greenfield (11) made a theoretical analysis of the rain scavenging of radioactive particulate matter from the atmosphere. The following four mechanisms were considered: (1) direct collision of falling raindrops with particles, (2) sweeping up of particles by water-cloud droplets which would subsequently reach the ground in rain, (3) rain-induced downdrafts, and (4) interaction of electrostatic forces between particles and raindrops. The fourth possibility was rejected in consideration of a previous paper of Greenfield's (12), and the third was considered an unlikely contributor of significant amounts of deposition. Mechanisms (1) and (2) were analyzed in detail.

Greenfield assumed that any interaction between rain and a radioactive cloud would involve a rain extensive in comparison with the cloud. In the first analysis it was further assumed that the percentage of particles of a particular size removed from a vertical cylinder (with the diameter of the largest raindrop) through the cloud, would be equal to the percentage of the same size particles removed from the entire cloud. It was concluded that, as the result of the collision of falling raindrops with particles, particles with diameters less than 2μ would be removed in only minute amounts, depending on the amount of rainfall.

A two-step process was involved in the analysis for the collection of particles by water-cloud droplets and subsequent deposition by rainfall. It was considered that (1) a certain percentage of particles would be captured by water-

cloud droplets, and (2) a certain percentage of the droplets would be subsequently captured by falling raindrops. The conclusions were that the probability was very small that a water droplet would pick up significant quantities of particles with diameters greater than 0.08μ, and that the mechanism of droplet capture by raindrops would occur in a manner similar to the direct capture of radioactive particles.

Assuming the parameters which would produce the maximum realistic contamination, including a mean particle diameter of 2μ, Greenfield showed that over 97 percent of the radioactivity is scavenged by direct interaction with raindrops. For all practical purposes then, the removal is by direct impingment of falling raindrops. Reviewing his equations for the direct interaction of raindrops with particulate matter, Greenfield says "the fraction scavenged can be considered to be independent of rainfall intensity and very dependent on the actual depth of rainfall experienced."

Greenfield's analysis was interpreted by Itagaki and Koenuma (13) to mean that in the process of descent a raindrop or snowflake would collide with a radioactive particle and carry it to earth, and that this was the predominant transfer mechanism. They measured the radioactivity of seven rains and nine snows at five different elevations and found that the specific activity decreased with altitude. In the case of snow the extrapolated altitudes of zero activity coincided almost exactly with the measured altitudes of the cloud tops.

Salter, et al (14) measured the concentration of Sr-90

in precipitation resulting from large-scale uplift. They interpreted Greenfield's analysis as meaning that the predominant deposition mechanism was by cloud-droplet capture and subsequent rainout, in apparent contradiction to the above. By examining three rains they found evidence of correlation between the cloud ceiling and the concentration of Sr-90 in the rainwater. The cloud ceiling was interpreted to be a measure of the concentration of radioactivity resulting from raindrop evaporation while falling through the unsaturated layer of air near the ground.

Miyake, et al (8,15) have proposed an equation for the deposition of radioactivity in terms of the concentration in the air and the amount of precipitation, in accordance with Greenfield's analysis. Their data are accurately represented by the equation when atmospheric activity is taken as constant over an extended period of time. It is pointed out that fluctuations of rain activity may be mainly due to fluctuations in the concentration of radioactive dust in the air.

The author proposes that there will be significant fluctuations in rain activity as the result of the meteorology surrounding and defining the rains. With this in mind, the following laboratory setup was devised.

III. LABORATORY INSTRUMENTATION AND PROCEDURE

The primary goal of this thesis was to provide a laboratory setup and procedure for the continuous fission product monitoring of atmospheric radioactivity and rainfall radioactivity. The criteria were that the system must be simple in design and readily adaptable to any laboratory, yet complete in its scope and capable of statistically reliable data. It should produce data which would lead not only to straightforward correlation analyses, but also to an explanation of the transfer process. The system should be such that all rains would be sampled without requiring the presence of a laboratory technician at other than normal working hours.

Following are descriptions of the air sampling, rain sampling, and radioactivity counting setups and procedures. The apparatus is shown in figure 1, Appendix I. Unless otherwise indicated, all references to figures are to Appendix I.

Air Sampling

This technique differed significantly from what may be regarded as "standard" for measuring fission product radioactivity. Commonly, as high as 1500 cubic meters of air are sampled in twenty-four hours through a fibrous filter media, as has been the procedure at the Taft Sanitary Engineering Center (16). This large volume has the distinct advantage that counting statistics are highly reliable. However,

error is introduced as the result of particulate matter which penetrates and is stored within the filter matrix. The result of deeply imbedded particulate matter is that its emitted radioactivity is absorbed in the filter matrix and is never counted. According to Cotton (17) this effect "cannot be accurately compensated by calculations."

In the following procedure approximately thirty cubic meters of air were sampled every twenty-four hours through Millipore filters. The smaller sample volume results in slightly reduced reliability due to counting statistics. However, Millipore filters--which are cellulose plastic, uniformly porous membranes-- have the particularly useful characteristic that they retain on the surface all particles which exceed in dimension the specified pore size. Moreover, the electrostatic charge generated at the filter surface inhibits the penetration of colloidal size particles into the pore structure. This means that there is negligible absorption of radioactivity by the filter matrix. Finally, Millipore filters are available with precisely the diameter necessary to fit the counting apparatus.

The following procedure was used:

1. Air samples were routinely taken on a twenty-four hour basis, at a flow reading of 25 liters per minute. The flowmeter was calibrated at 70° Fahrenheit and no correction was made for temperature differentials. Pressure drop was corrected for and the flow rate was considered to be the average of the flows at the start and finish of the sampling

period. The sample volume was reported at sea level pressure. Calculations were facilitated by the use of the graph shown in figure 2.

2. The amount of dust collected was determined by weighing the filters before and after sampling. Before weighing, the filters were dried in an incubator at approximately 100° Fahrenheit for at least eight hours.

3. The filters were mounted on sample mounts and stored in a dessicator for counting.

The following equipment was used:

1. Type AA, 47mm diameter Millipore filters and holder
2. Rotometer, 30 liters per minute capacity
3. Manometer
4. Building vacuum system
5. Precise balance

Excellent control of the flow rate was attained with the building vacuum system. It can be stated with a high degree of certainty that the variation in flow rate did not exceed ±0.7 liters per minute.

Rain Sampling

It was assumed that knowledge of the distribution of radioactivity in rain would be revealing. A collection basin was situated on the roof of the engineering building and was connected to a series of five sampling cans in the laboratory by ¼-inch copper tubing. The basin was designed with an area such that 1080 milliliters of rain were collected with a rain-

fall of 0.15 inches. The rain was transferred to the laboratory through the copper tubing and collected in the cans, each successive 1080 milliliters being collected in a different can. An overflow was provided so that all rain in excess of the five cans, or in excess of 0.75 inches, would be collected in one container.

A recording rain gage was also placed on the roof, approximately 30 feet from the collection basin.

The rain from each can was processed so that suspended and dissolved radioactivity could be counted separately. This method did not differ significantly from accepted procedures, with the exception that one liter samples were processed instead of 250 or 500 milliliters. The procedure used was in all essentials the same as that described by Setter, Hagee, and Straub (18). Techniques for preparing radioactive samples on planchets and filters are described by the Nuclear-Chicago Corporation (19), and by Vaughn, et al (20).

The following procedure was used:

1. The rain gage was maintained every four or five days, and after each rain.

2. Rain samples were acidified, in the collection cans, by the addition of approximately 5 milliliters of 1N HCl, in order to reduce activity adsorption on the container surfaces, and to prevent precipitation of the dissolved solids.

3. One liter of each sample was siphoned off into a marked one-liter beaker immediately after being vigorously stirred.

4. The suspended solids were deposited on a preweighed Millipore filter and the filter was dried, weighed, mounted on a sample mount and stored in a dessicator for counting.

5. The filtrate was returned to the one-liter beaker and evaporated under an infra-red lamp to a volume of approximately 20 milliliters. The beaker was then tilted so that the remainder of the filtrate covered a minimal area and the sample was transferred to a copper planchet with a clean medicine dropper and evaporated to dryness under an infra-red lamp. The side of the beaker was washed down with a minimum of 0.1N HCl and this also was transferred to the copper planchet. Care was taken in the transfer process to insure that the dissolved solids were contained within the outer annular ring impressed in the bottom of the planchet. The planchet was then flame-dried with a Bunsen burner, placed in a sample mount and stored in a dessicator for counting.

The following apparatus was used:

1. Rain collection basin and standard
2. Collection cans with valve assembly
3. Belfort rain gage, weighing type
4. 250-watt infra-red lamps
5. Chimney funnel and suction apparatus

The cans were calibrated by pouring water through the system and observing the filling process. As a can filled, the rising water lifted a float which, by leverage, pressed a rubber tee against the orifice. The flow was substantially and quickly reduced as the design volume was approached, and

the final volume ordinarily fell between 1070 and 1150 milliliters.

Counting

All counting was performed with a gas-flow proportional counter at a sensitivity of 2.5 millivolts and a preamplifier gain of 1. The procedure followed was fundamentally the same as that described by Setter and Coats (16), with the exception that a blanket counting efficiency of 50 percent was assumed for all samples.

50 percent closely approximates the efficiency of gas-flow counters of this type, and was assumed so that the results could be compared, at the same order of magnitude, with other data. In the interest of obtaining "absolute disintegration rates" separate efficiencies would have to be determined for the samples on filters and the samples in planchets.

The following procedure was used:

1. A background count of 500 counts was taken before and after each counting session, with a sample mount in place. The background count was considered to be the average of these counts and was subtracted from the total counting rate for each sample.

2. 1000 counts were taken on each sample 4-5 days after sampling. The time lag allowed for the decay of natural radioactivity. Two or three additional counts were taken at 4-5 day intervals.

3. The sample counting rates were plotted against time on semi-log paper and extrapolated back to the time of sample collection. The extrapolated counting rates were converted to micromicrocuries per cubic meter of air and micromicrocuries per liter of water according to

$$\mu\mu c/m^3 = \frac{\text{counts per minute}}{(2.22)(\text{counting efficiency})(\text{volume of air})}, \text{ and}$$

$$\mu\mu c/l = \frac{\text{counts per minute}}{(2.22)(\text{counting efficiency})(\text{relative counting rate})}$$

where 2.22 disintegrations per minute is one micromicrocurie. The conversion of air activity counts to $\mu\mu c/m^3$ was facilitated by the use of the graph in figure 3.

4. Throughout the sampling period the counter and scalar were tested at regular intervals, according to the manufacturer's directions, for plateau constancy and sporadic counting.

The following apparatus was used:

1. Gas-flow proportional counter with preamplifier and micromil window, and scalar (Nuclear-Chicago Models D-47, D47F, and 186)

2. Automatic sample changer and printing timer (Nuclear-Chicago Models C-110B and C-111B)

3. Sample mounts and copper sample pans (planchets)

Setter, et al (18) show a self-absorption curve for mixed fission products. This is a plot of relative counting rate versus sample thickness in milligrams per square centimeter. It shows that for a sample thickness up to $0.8 mg/cm^2$ the relative counting rate is unity. With this procedure the

maximum thickness for air samples was 0.3mg/cm^2, indicating that self-absorption could be neglected. The maximum sample thickness from rainwater was 2.5mg/cm^2 and corresponded to a relative counting rate of 0.8.

Support for the Air Monitoring Technique

Support for the air monitoring technique is derived from a comparison of data obtained in the laboratory with data from the Michigan State Department of Health. These sets of data are plotted against time on figure 4. In spite of the differences in sampling techniques, and the 5.5 miles separating the sampling stations, there is a one-to-one correspondence between all major peaks and troughs. For the above reasons, and because the air activity is continuously changing, even on an hourly basis as shown in figure 5, complete agreement was not expected. The F-test shows, however, that one can be 99 percent confident that these values are related. This is summarized in figure 6, where the two sets of data are plotted against each other. The line, representing the linear regression equation (21), very nearly passes through the origin, as would be expected. This agreement shows that measurements are reasonably reliable within a 5.5 mile radius of the sampling station.

The usefulness of the rain sampling cans is demonstrated in figure 9a, Appendix II. Here, the 0.15 inch increments of rain are plotted against their respective activities for four rains. Considerable variation in activity is demonstrated in three rains.

IV. ANALYSIS OF DATA

It was anticipated that when rain activity is compared with the air activity at the time of the rain, the effects of meteorology would be evident.

Two empirical equations for the specific radioactivity of rain were generated from eleven summer rains. The first equation assumed a direct relationship between rain activity and air activity alone, and the analysis for the second equation considered the influences of rain intensity, cloud height, and wind speed.

The equations were then compared, and the possible significance of wind speed was discussed from which a more comprehensive equation was proposed. In addition, an explanation of increasing specific activity of rain with time was proposed.

The data is tabulated on page 40. Unless otherwise indicated, all references to figures are to Appendix II.

Analysis I

Figure 1 is a plot of the air activity versus the rain activity of each 0.15 inch increment. Figure 2 is the same plot for the average activity of each rain. The latter corresponds to one type of analysis made by Colpetzer (1), in which air activity in $\mu\mu c/m^3$ was plotted against rain activity in $\mu c/ml$. This examination was based on weekly averages,

and significant correlation was shown to exist, especially at higher levels of activity.

Such levels of activity are of the magnitude obtained in this experiment, and figures 1 and 2 indicate a definite relationship between air and rain activities. If this relationship is assumed to be given by the straight line in figures 1 and 2, the rain activity (C_R) is related to the air activity (C_A) by the equation

$$C_R = 223C_A. \qquad (1)$$

The ability of this equation to predict rain activity is shown by figures 5a and 6a for the activity of each 0.15 inch increment of rain and the average rain activity, respectively. Here, values from equation (1) are plotted against laboratory values.

Analysis II

It was originally assumed, by the author, that the concentration of rain activity would be some function of rain intensity, wind speed, cloud height, and atmospheric activity.

It was observed in figure 1 that higher rain activities were associated with greater wind speeds. This is demonstrated in figures 3a, 3b, 3c, and 3d--the same points plotted in figure 1--where wind speed intervals are parameters. No relationships involving rain intensity or cloud height were observed.

For each of the above figures the geometric mean wind

speed, $\bar{V}$, was determined, and an equation of the form

$$C_R = KC_A \qquad (2)$$

was written. Assuming $K = k\bar{V}^n$, logarithms of K values were plotted against logarithms of $\bar{V}$ values, figure 4, from which the constants k and n were resolved. The resulting prediction equation is

$$C_R = 60.3V^{0.534}C_A. \qquad (3)$$

The ability of this equation to predict rain activity is shown by figures 5b and 6b for the activity of each 0.15 inch increment of rain and the average rain activity, respectively. The activity of each rain is the average of the activities of all 0.15 inch increments.

Comparison of Prediction Equations

Throughout these analyses rains 6 and 11 were not accounted for. Figures 1 and 5 show that rain 6a (the first 0.15 inches of rain 6) aligns properly, but 6b never aligns. Rain 6 lasted approximately two days. An investigation of weather maps showed that the second portion of the rain occurred nearly under the center of a high pressure system, and that this occurrence was unique with respect to all other rains. Figure 7 shows the weather maps related to this rain. The movement of air was downward, and the rain-bearing cloud was presumably intensely seeded with radioactive particles from the upper atmosphere.

Rain 11 occurred at a critical time as far as air activity is concerned. Examination of figure 4, Appendix I, shows

that the air activity underwent a rapid rate of increase at that time, and could have been as high as six or seven $\mu\mu c/m^3$ when the rain occurred. This rain was associated with the passage of a cold front, and the high activity is undoubtedly associated with that mass of air. Figure 8 shows the passage of the cold front.

This behavior of rains 6 and 11, with respect to fronts and pressure systems, agrees with observations made by Sotobayashi and Koyama (10).

Figures 5 and 6 show a definite improvement in the alignment of points according to equation 3. Excluding rains 6 and 11, equation 3 accounts for all of the points whereas equation 1 does not.

The variation of activity within the same rain is shown in figure 9a by the laboratory values for each 0.15 inch increment. Figures 9b and 9c show the predicted values according to equations 1 and 3, respectively. Variations in specific activity are apparently explained by variations in wind speed.

Interpretation of the Wind Speed Effect

The literature review describes theoretical and empirical evidence that the principal mechanism of particle capture is by direct collision with raindrops or snowflakes.

If this is true, the amount of particulate matter captured must then depend on the amount presented to the raindrop in the descent. If, as in the case of this data, rain

intensity and cloud height have no significant effect, the average time of descent is constant and the particulate matter swept out is proportional to the atmospheric concentration. Additionally, particulate matter impinges on the raindrop with the horizontal speed of the wind. The amount presented in this manner is proportional to the atmospheric concentration and the wind speed. This interpretation suggests that, for summer precipitation, an equation of the form

$$C_R = K_{C_A} + kV^n C_A, \qquad (4)$$

for each increment of rain, might be more descriptive. In this equation K_{C_A} is a constant for a specific atmospheric activity, and V is the wind speed.

Interpretation of Increasing Specific Activity of Rain

Colpetzer (1) observed increasing specific activity with increasing depth of rainfall when weekly averages were considered. This effect is displayed in figure 9a, and might be explained by the deviation from Greenfield's model and the horizontal impingment of radioactive particulate matter as the result of wind speed.

Under the Greenfield model (a rain extensive in comparison with the radioactive cloud) a decrease in the specific radioactivity of rain as the result of rainout would be noticeable. However, the inverse of this model describes summer rains; that is, radioactive clouds are extensive in comparison with summer rains. This being the case, the air

within the volume occupied by a summer rain may be replenished with radioactive particulate matter at the rate of rainout, and a resultant decrease in the specific activity of rain with time need not occur.

The variation of atmospheric radioactivity with time is shown in figure 4, Appendix I. An examination of weather maps and U. S. Weather Bureau data at Lansing, Michigan disclosed that positive slopes are invariably associated with clear weather conditions, and negative slopes are associated with generally cloudy or precipitous conditions. The effect of a particular rain on the activity level is, therefore, considered to be negligible.

V. CONCLUSIONS

The following general conclusions are drawn from this study:

1. The laboratory setup meets the criteria of simplicity and adaptability. It produces data which are quantitatively useful, statistically reliable, and which lend insight into the transfer mechanism.

2. Atmospheric radioactivity measurements are reliable within a radius of at least 5.5 miles of the sampling station.

3. The relationship between the specific activity of rain and the specific activity of air at the time of the rain is reasonably well described by the equation, $C_R = 223C_A$.

4. Some relationship between the specific activity of rain and wind speed at the time of the rain appears evident, and an equation of the form $C_R = KC_A + kV^nC_A$, for each increment of rain, might be more descriptive.

5. In the case of summer rains Greenfield's model does not apply. No rain observed was extensive to the degree that the percentage of particles removed from a vertical cylinder was equal to the percentage of particles removed from the entire radioactive cloud. It may, therefore, be possible to predict increasing specific activity with time for summer rains.

6. The specific activity of rain did not appear to be related to either cloud height or rain intensity. The latter tends to support Greenfield's proposition that the fraction scavenged may be considered to be independent of rainfall intensity.

VI. FUTURE RESEARCH

There are many avenues of exploration to be considered in addition to those suggested in this thesis.

From this data it is evident that some parameter is needed to account for the effect of overhead pressure systems.

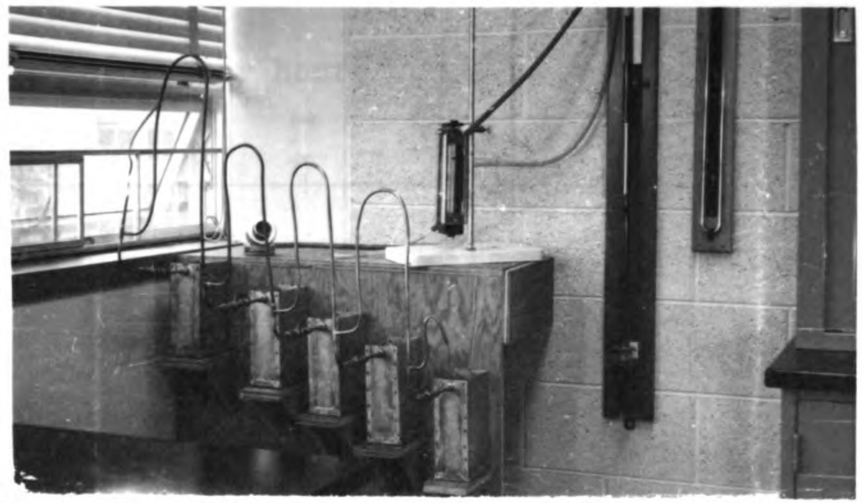
It is anticipated that there will be differences between the behaviors of air and rain activities with different seasons, and that different equations will apply.

An examination of Greenfield's analysis with respect to the mechanics of rainout of different size particles is needed. The type of capture, by cloud droplets or raindrops, may be selective of radioactive isotopes.

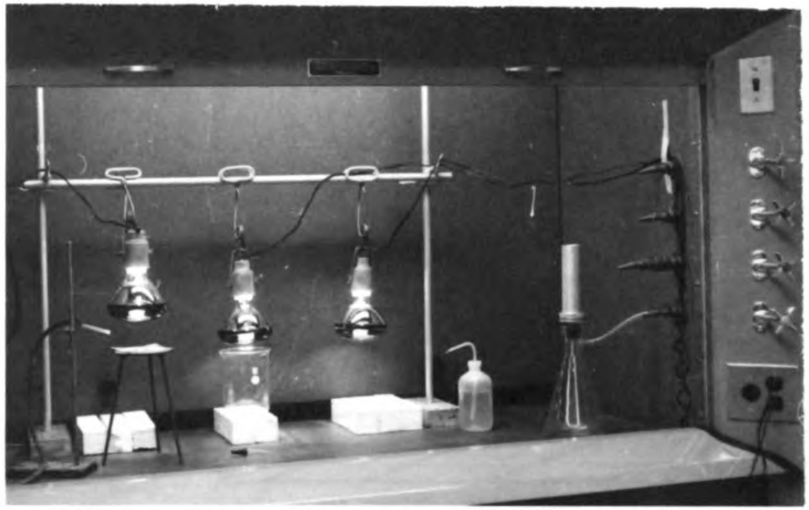
The immediate history of an air mass may yield parameters relating to particulate matter rainout, and a study of thermal gradient effects could explain the rapid build-up of atmospheric radioactivity.

APPENDIX I - LABORATORY

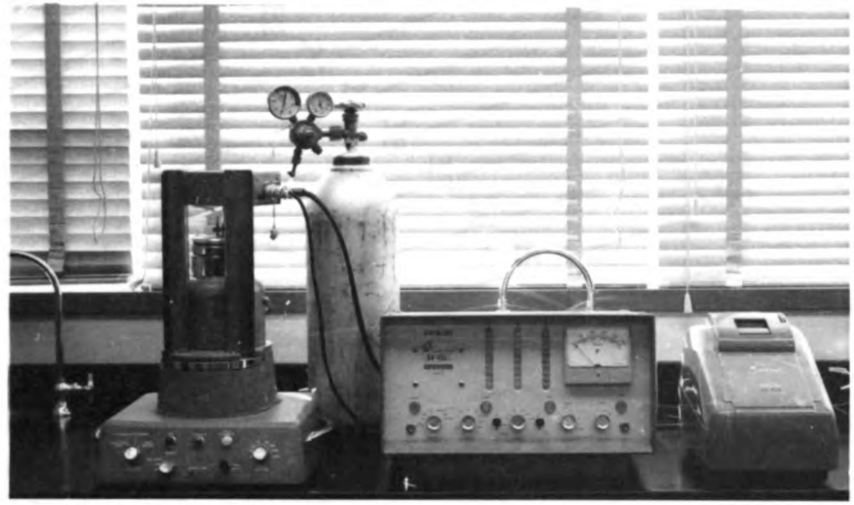
a. Rain sampling basin and recording rain gage.



b. Rain sampling cans and air sampling apparatus.



c. Rain processing apparatus.



d. Radioactivity counting apparatus.

Figure 1. Laboratory apparatus.

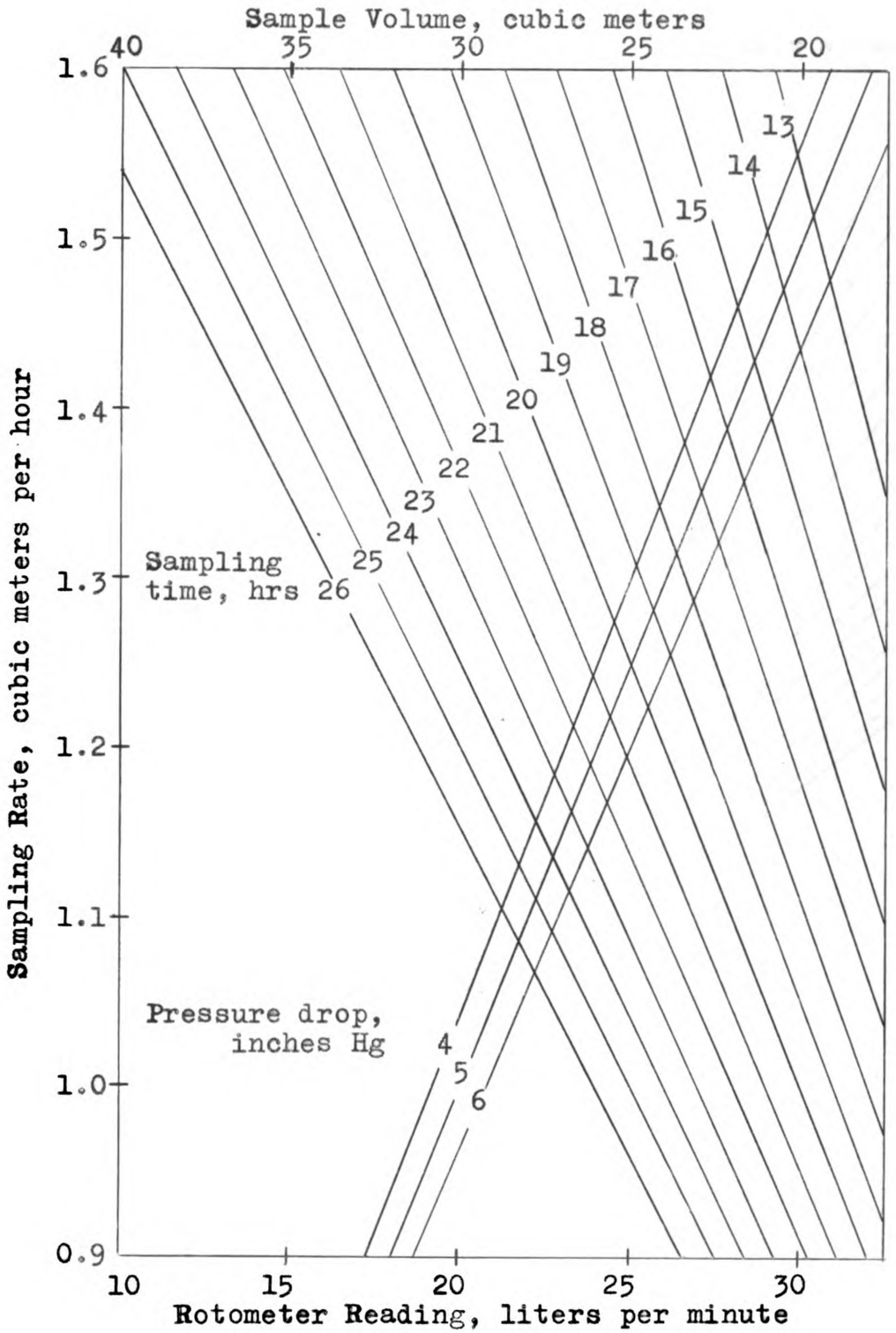


Figure 2. Sample volume graph

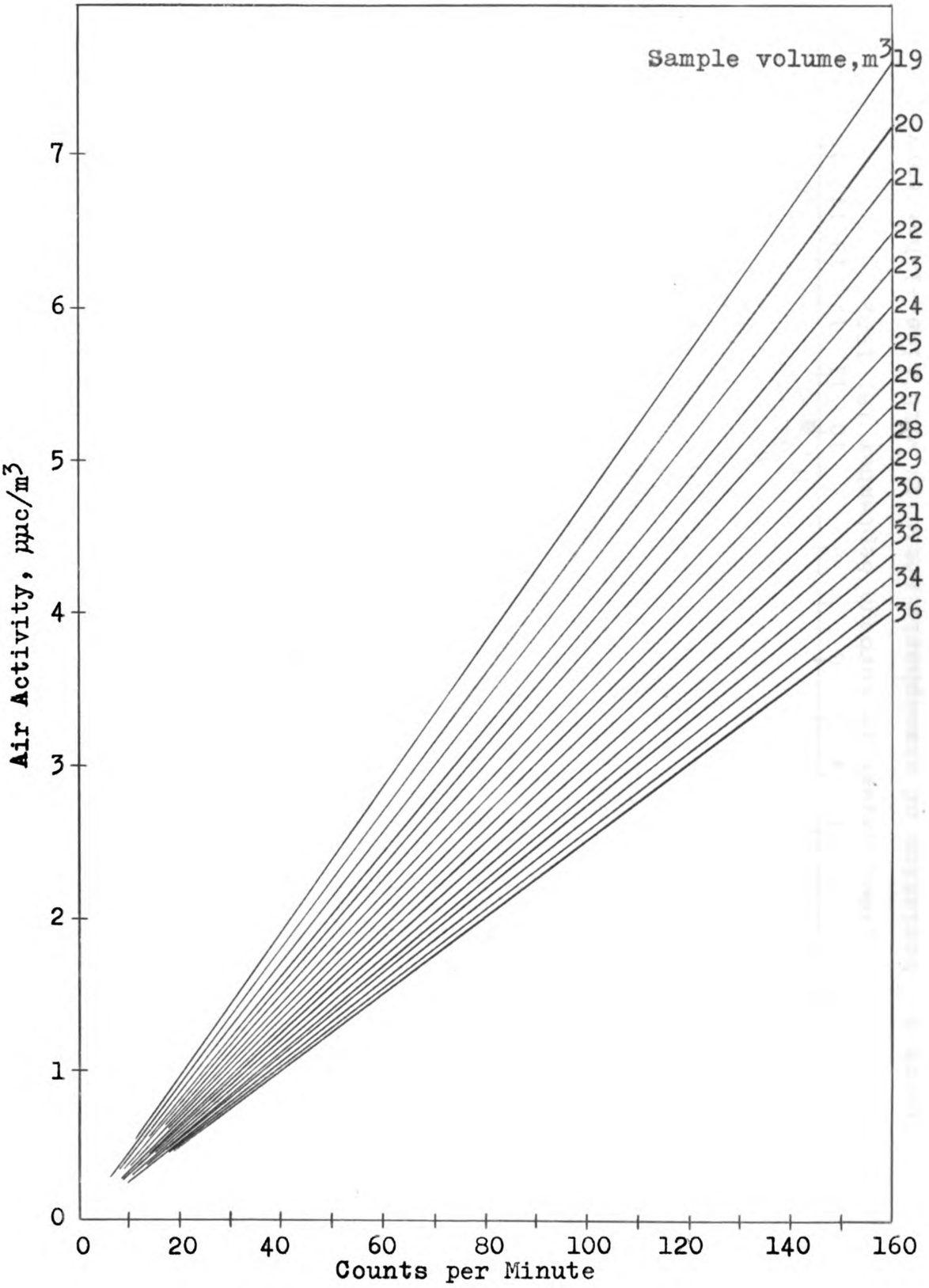


Figure 3. Air activity graph

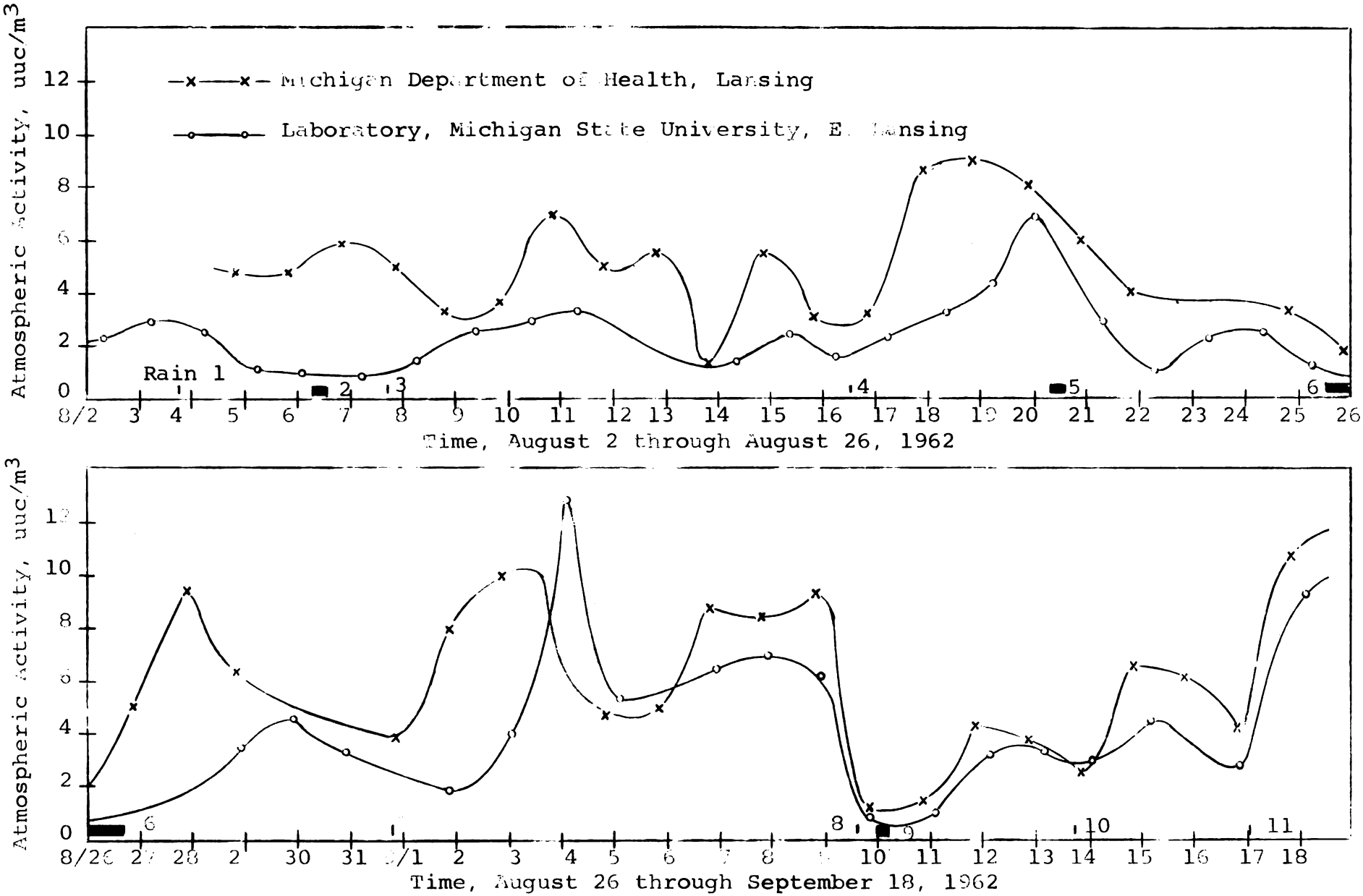


Figure 4. Variation of atmospheric activity with time--laboratory and Michigan Department of Health data. Periods of rains.

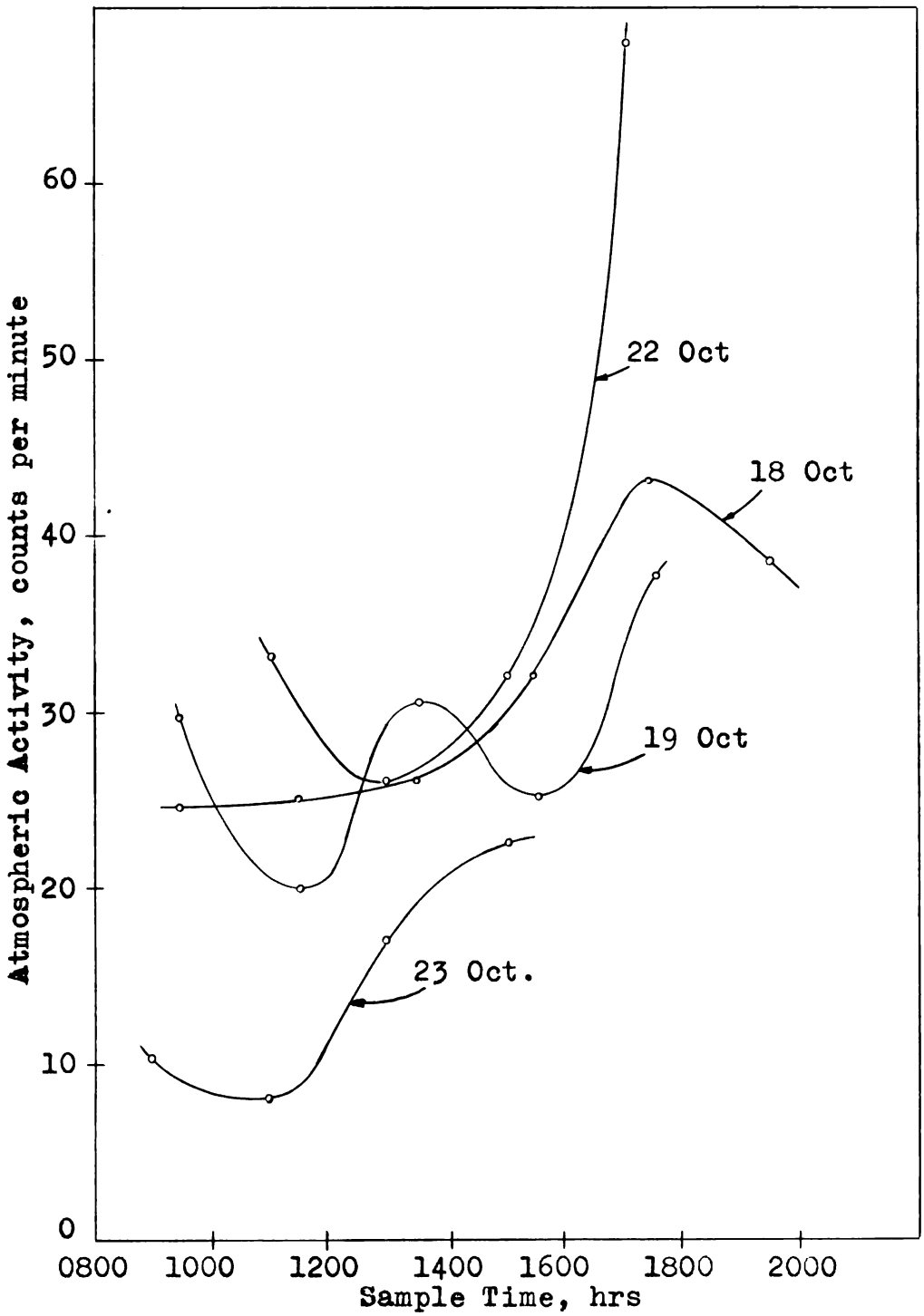


Figure 5. Hourly variation in atmospheric activity

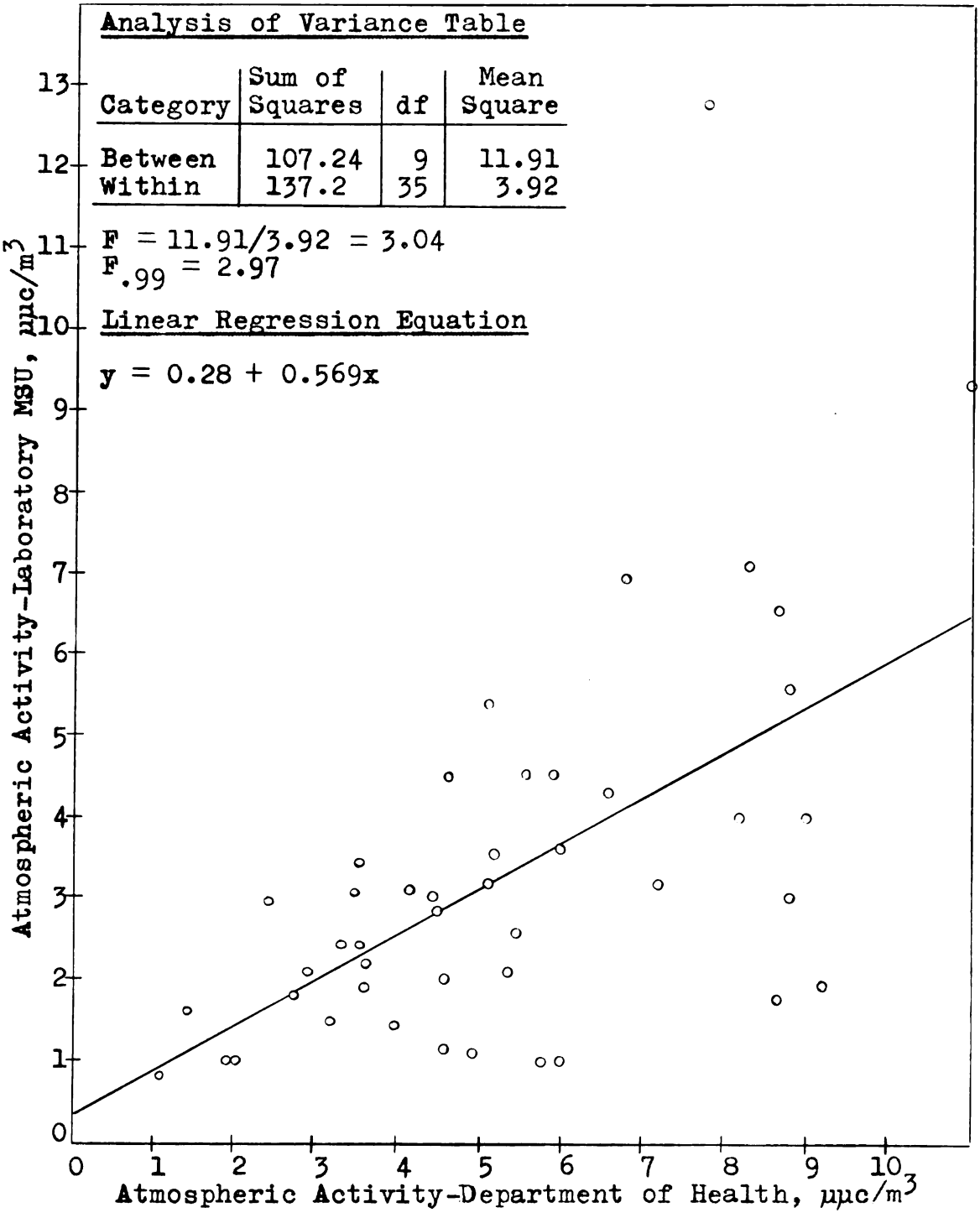


Figure 6. Comparison of atmospheric activity values--MSU laboratory data versus Michigan Department of Health data.

APPENDIX II - ANALYSIS OF DATA

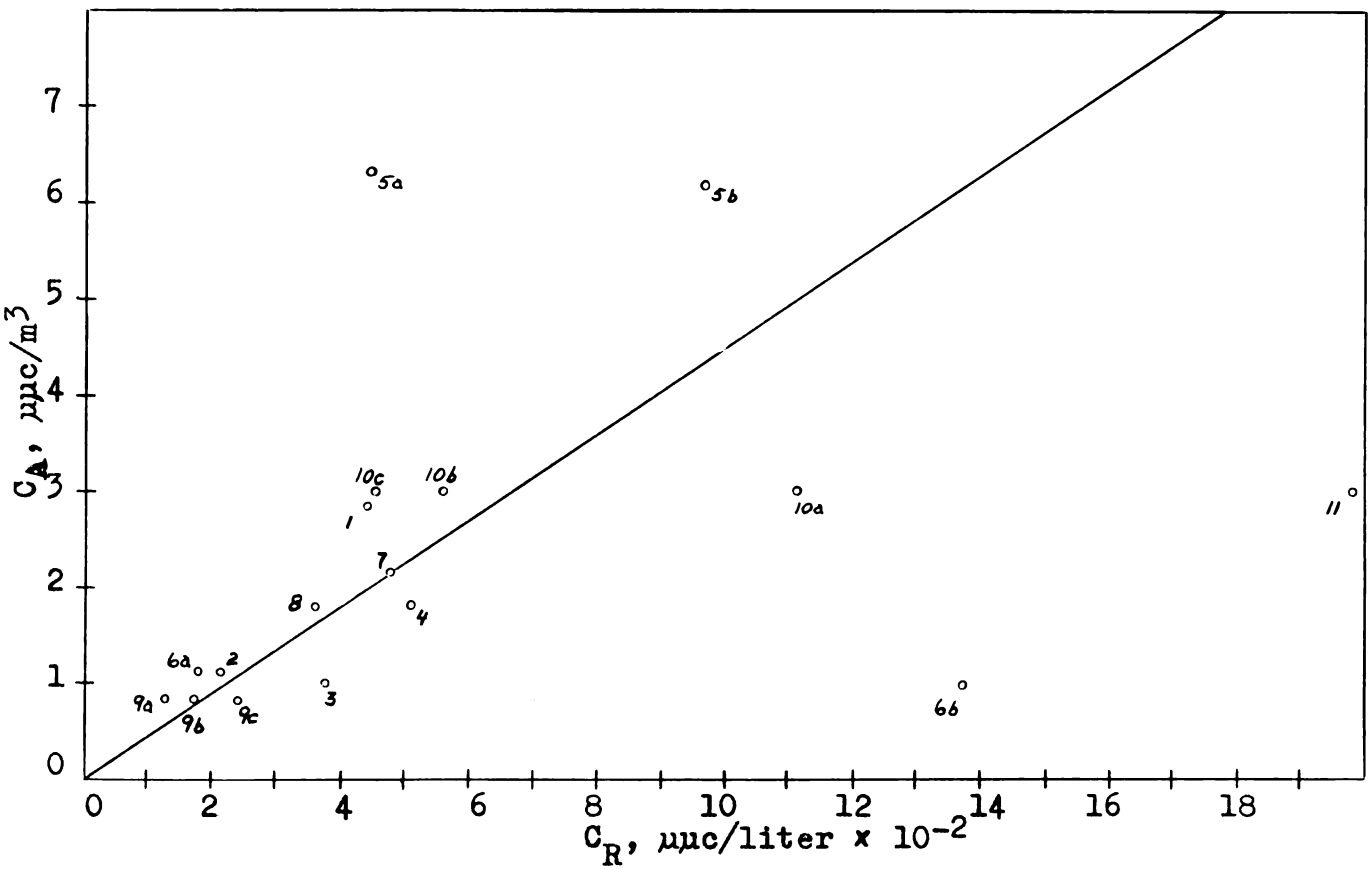


Figure 1. Atmospheric activity (C_A) versus rain activity (C_R) for each 0.15 inch increment of rain.

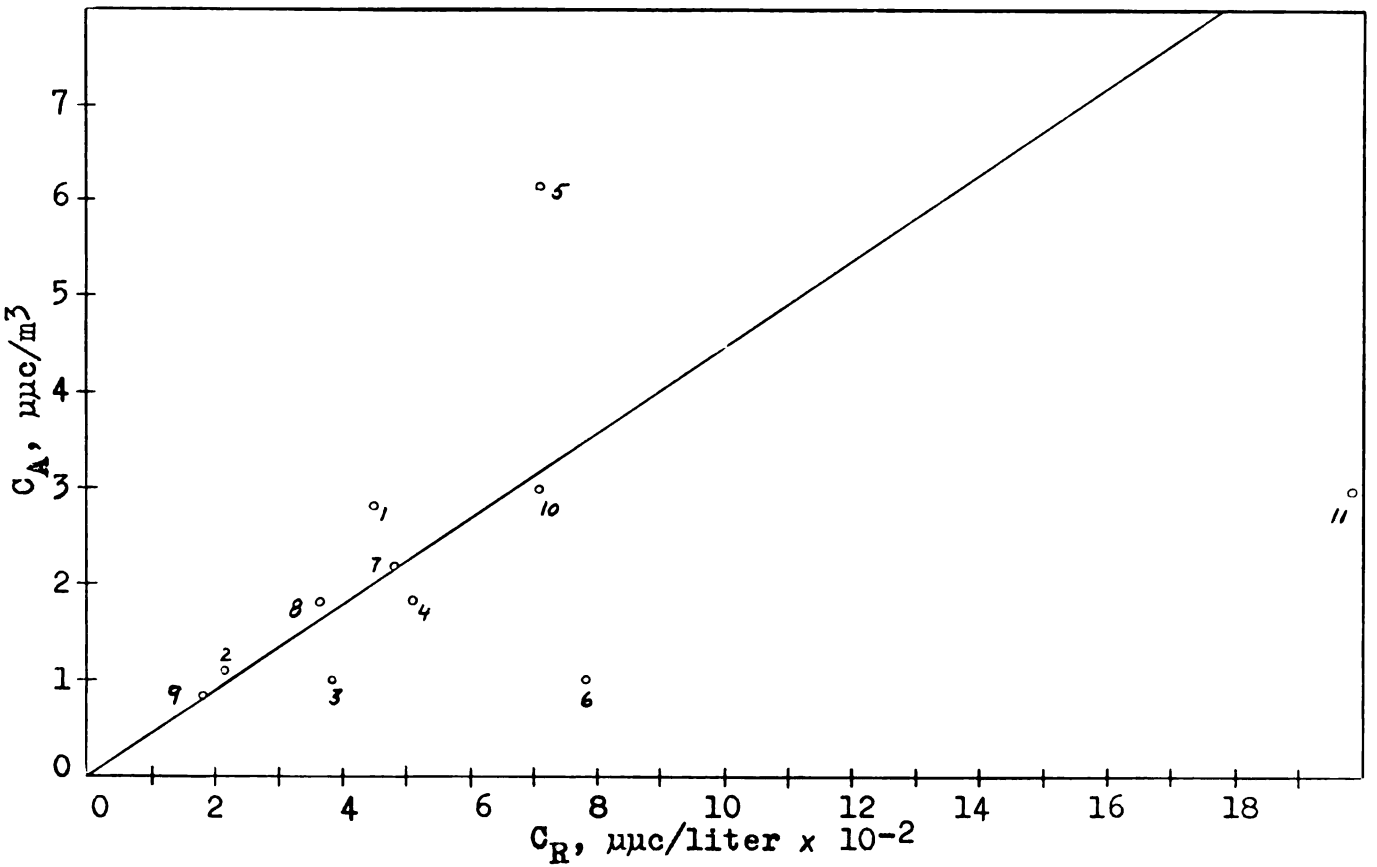


Figure 2. Atmospheric activity (C_A) versus average rain activity (C_R)

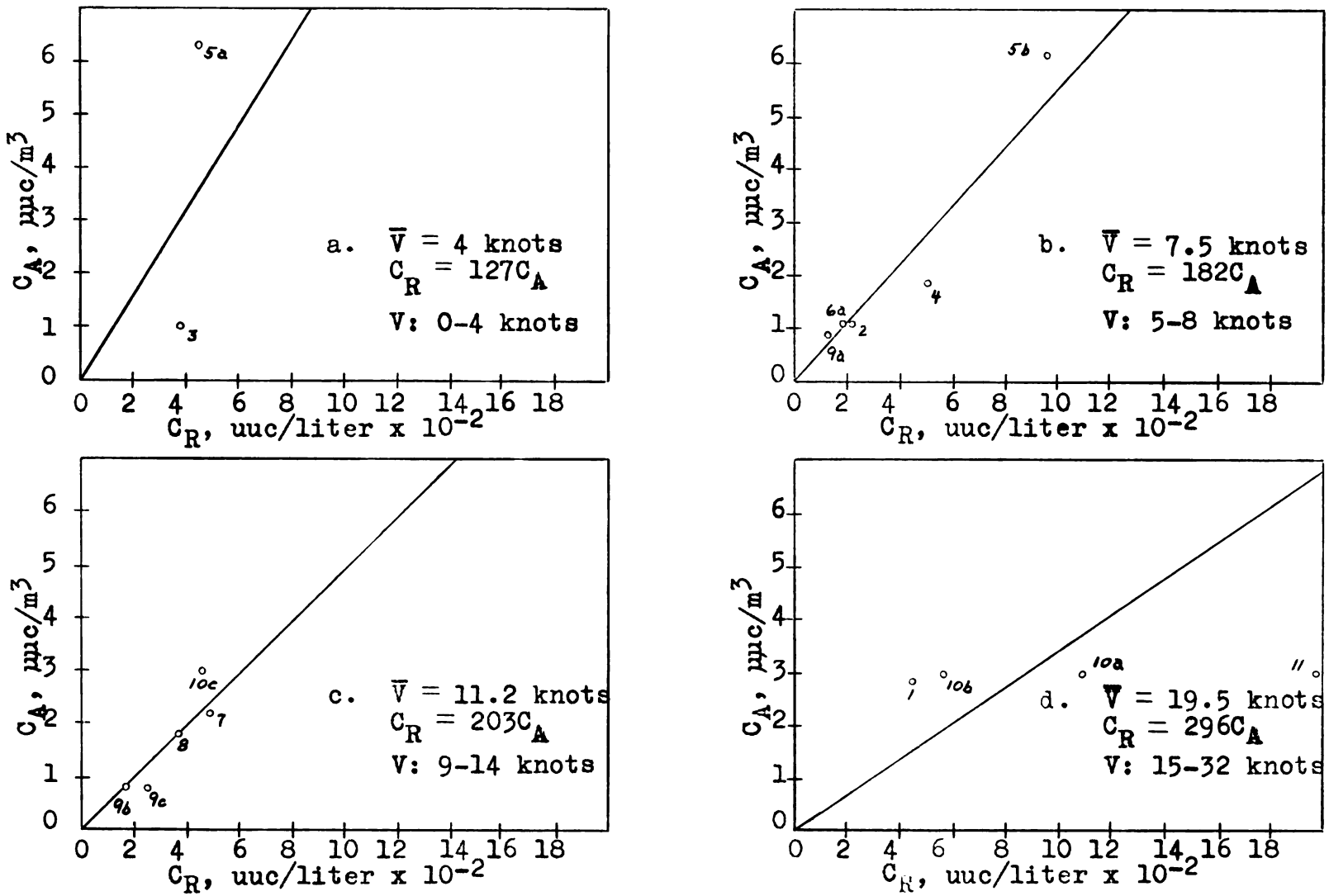


Figure 3. Atmospheric activity (C_A) versus rain activity (C_R) for each 0.15 inch increment of rain, with wind speed intervals as parameters.

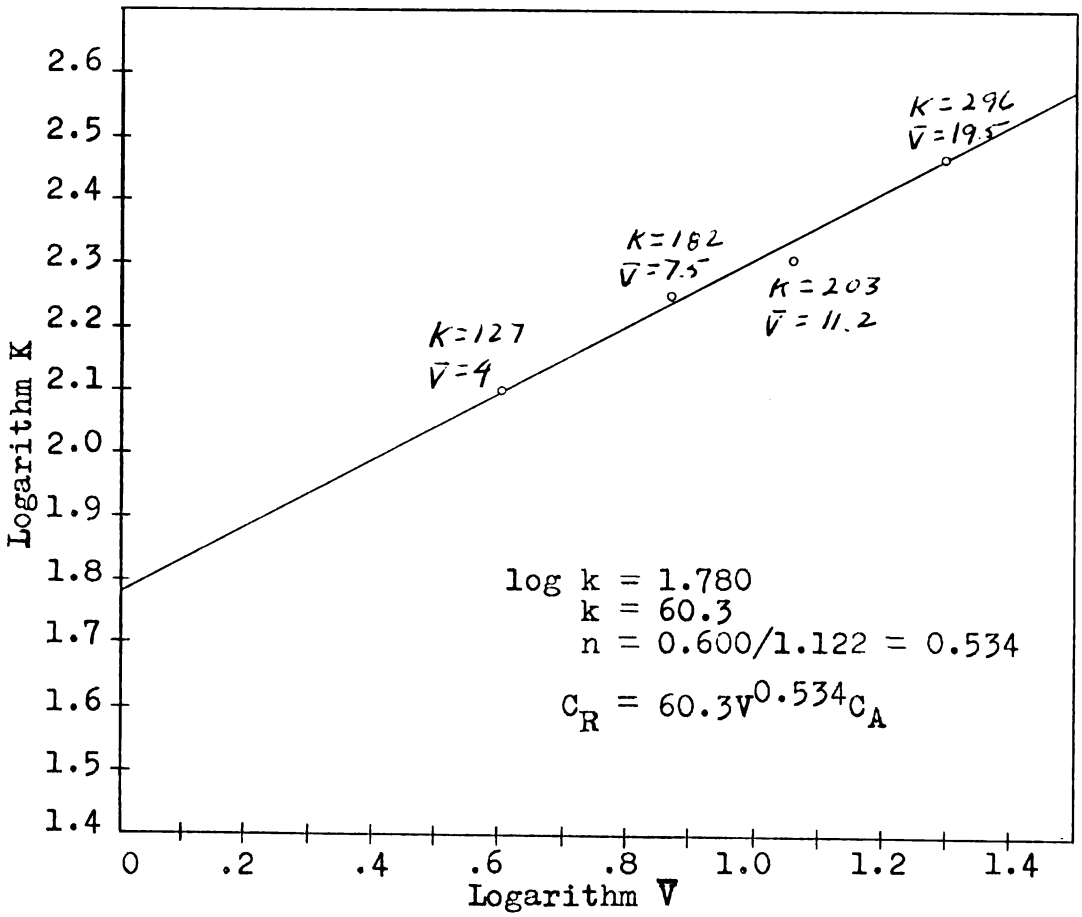


Figure 4. Logarithms of K values versus logarithms of $\bar{V}$ values.

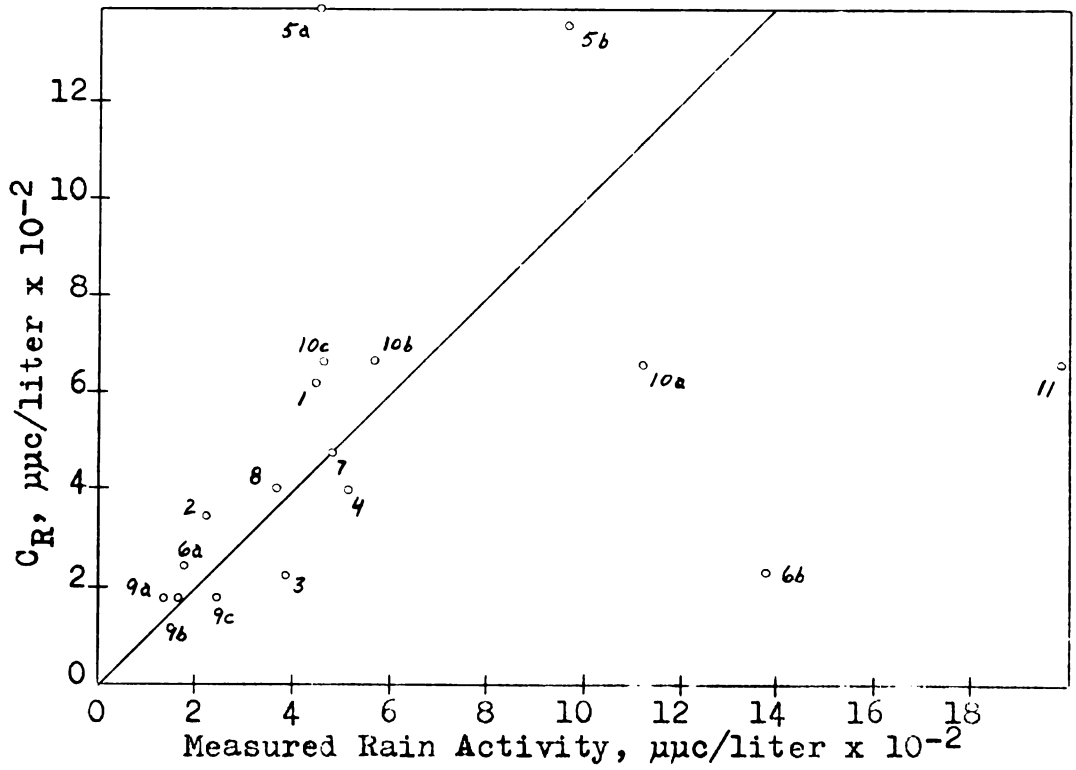


Figure 5a. Predicted versus measured rain activities for each increment. $C_R = 223C_A$.

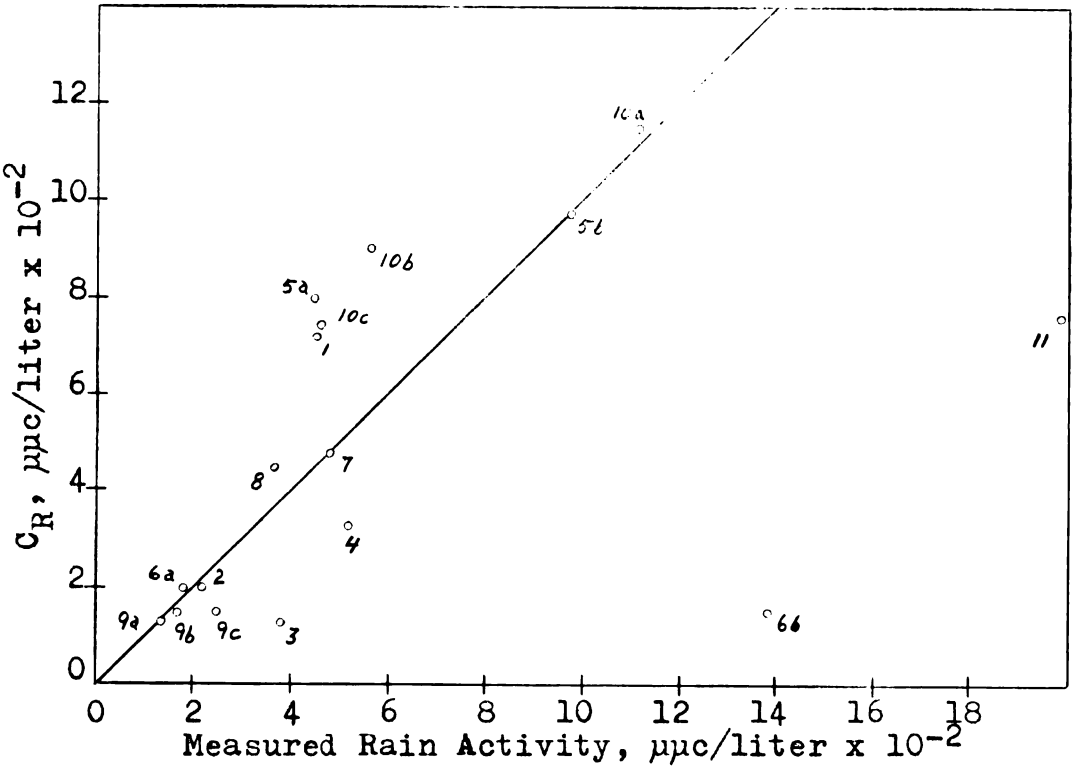


Figure 5b. Predicted versus measured rain activities for each increment. $C_R = 60.3V^{0.534}C_A$.

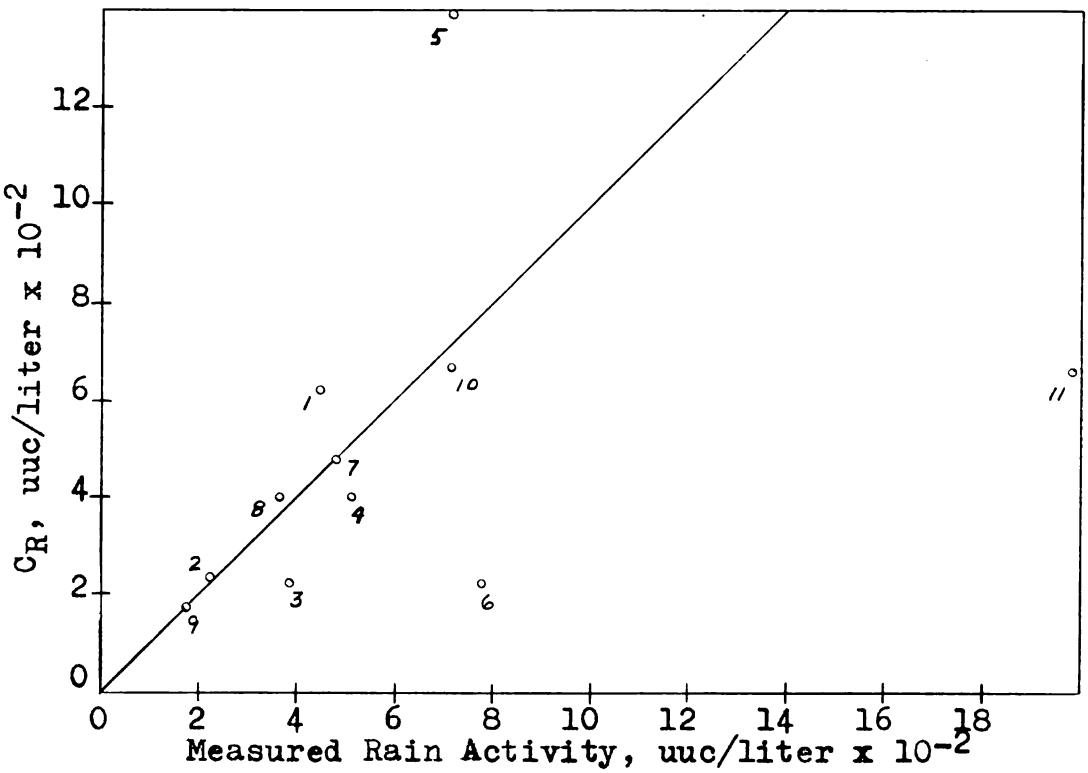


Figure 6a. Predicted versus measured average rain activities. $C_R = 223C_A$.

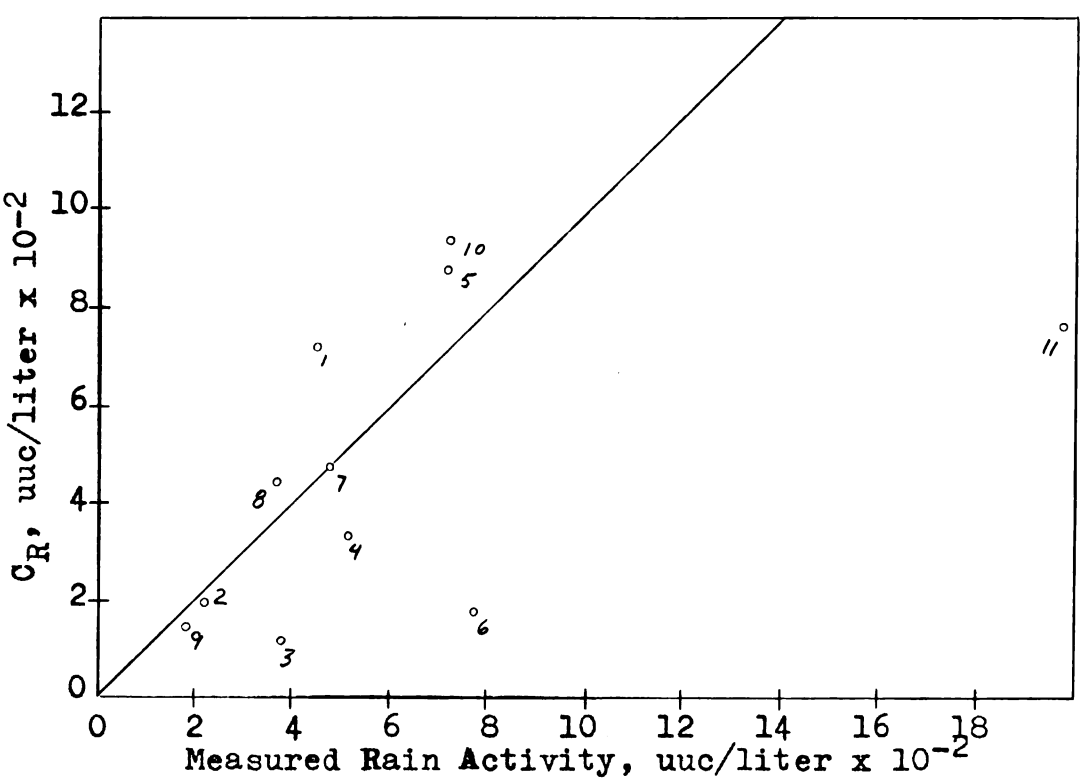


Figure 6b. Predicted versus measured average rain activities. $C_R = 60.3V^{0.534}C_A$.

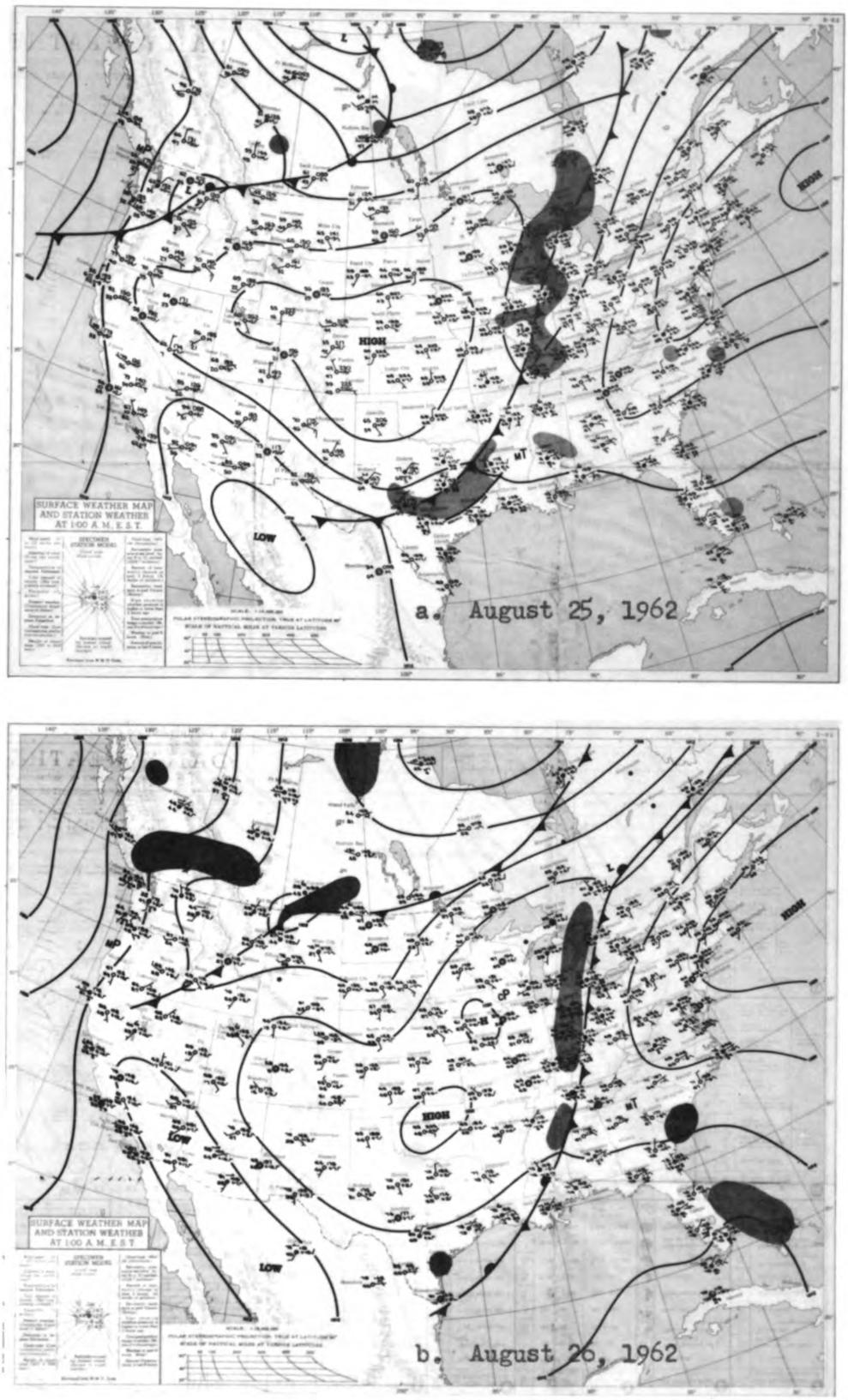


Figure 7. U. S. Department of Commerce Weather Maps, related to rain 6, showing the development of a high pressure system.

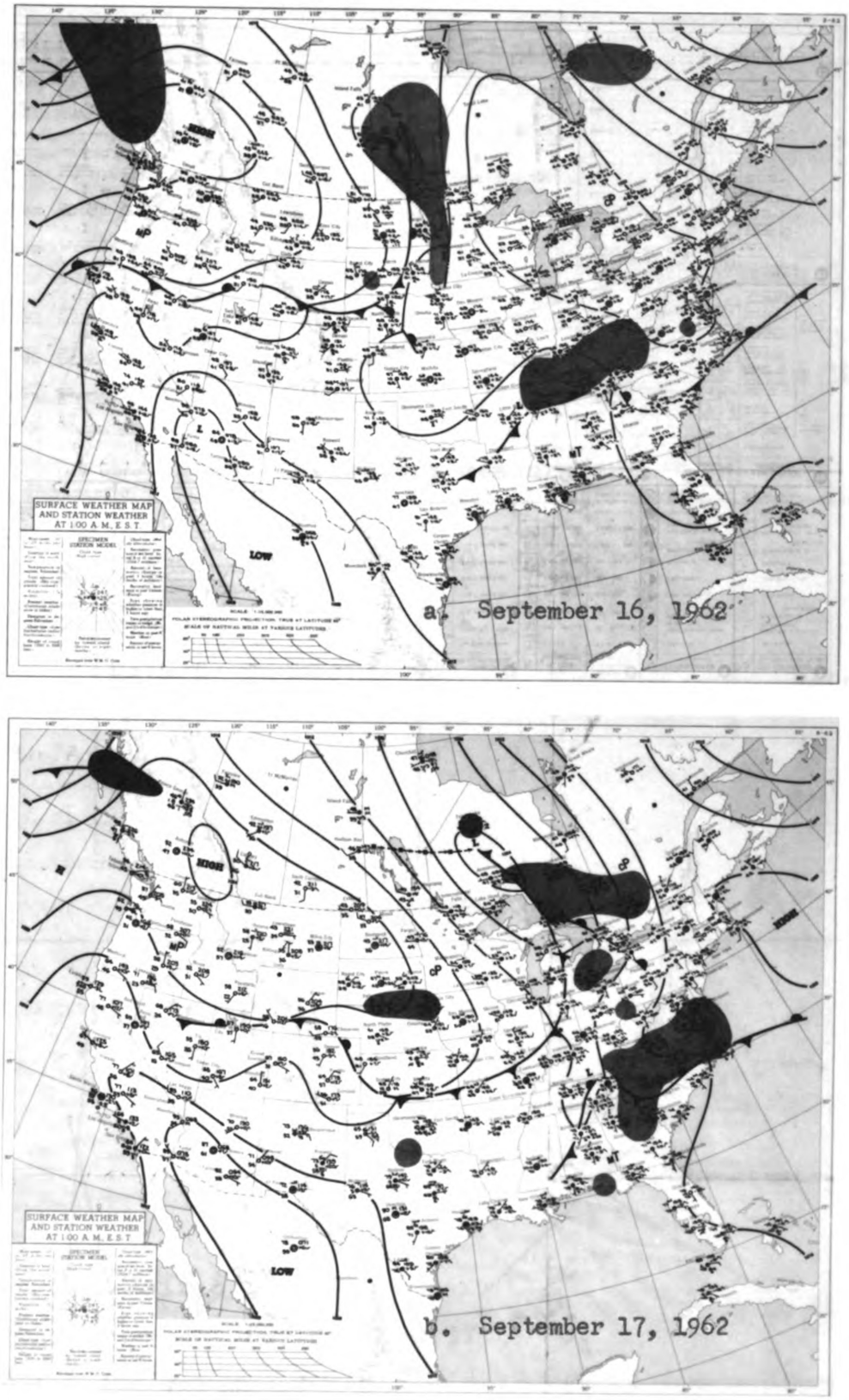


Figure 8. U. S. Department of Commerce Weather Maps, related to rain 11, showing the passage of a cold front.

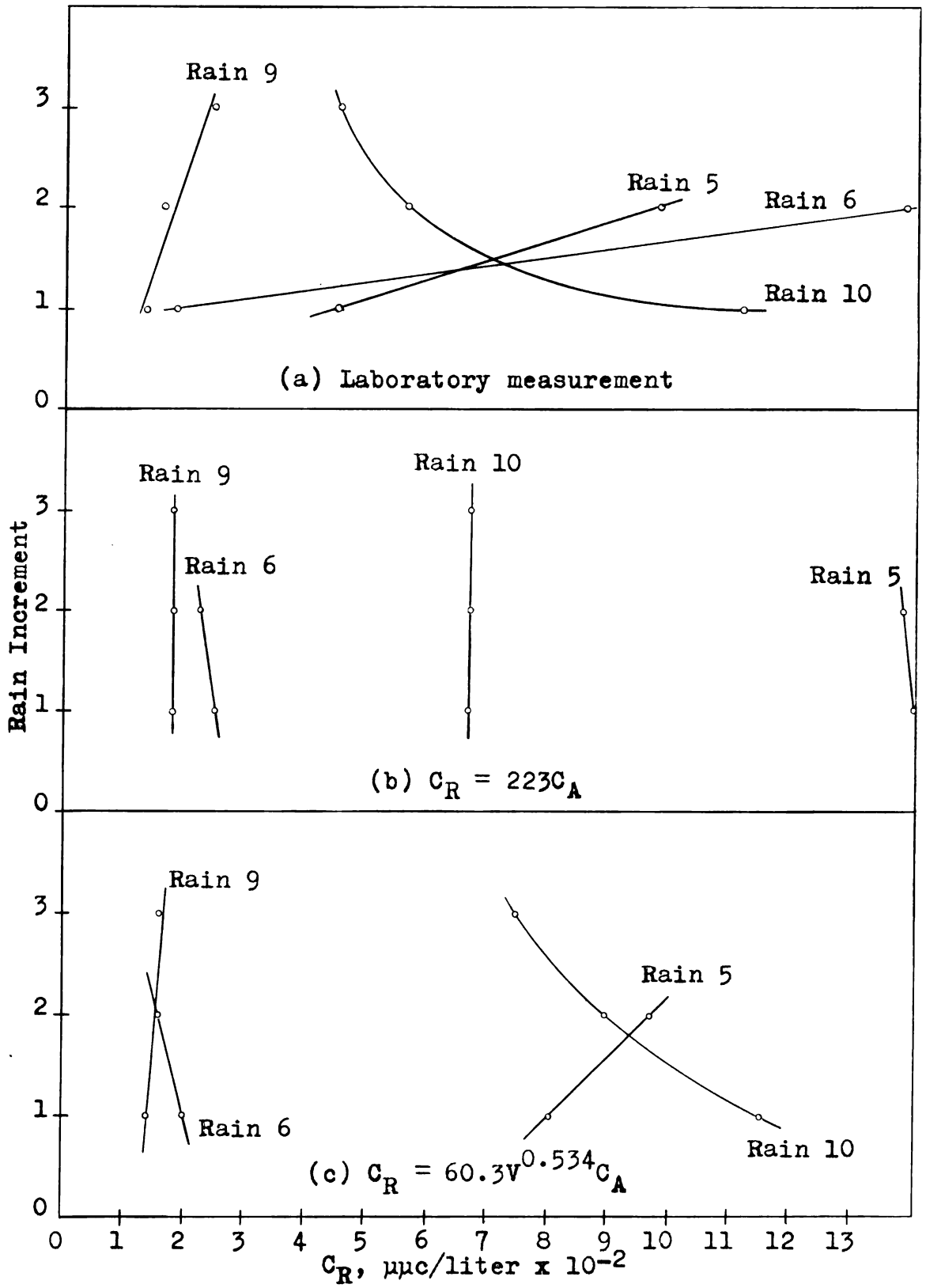


Figure 9. Variation of specific activity within each rain according to (a) laboratory measurement, (b) equation 1, and (c) equation 3.

Tabulation of data.

Rain	Date	Rain Intensity (in/hr)	Wind Speed* V (knots)	Cloud* Height (feet)	Air Activity C_A (uuc/m^3)	Rain Activity, C_R, uuc/liter		
						Measured	$223C_A$	$60.3V^{.534}C_A$
1	8/3	1.8	15	5000	2.8	446	625	717
2	8/6	.022	8	-	1.1	219	245	201
3	8/7	1.1	4	5000	1.0	387	223	126
4	8/16	1.8	8	5000	1.8	509	401	330
5a	8/20	.06	4	4000	6.3	447 } 710	1400 } 1390	796 } 885
5b	8/20	.28	6	5000	6.2	974 }	1380 }	973 }
6a	8/25	.013	8	700	1.1	182 } 781	245 } 234	202 } 179
6b	8/26	.009	6	500	1.0	1380 }	223 }	157 }
7	8/31	.45	11	2400	2.2	479	481	478
8	9/9	.60	14	1000	1.8	372	401	445
9a	9/10	.043	7	1000	0.8	135)	178)	136)
9b	9/10	.45	9	1000	0.8	168 } 181	178 } 178	156 } 149
9c	9/10	1.8	9	800	0.8	241)	178)	156)
10a	9/13	4.5	32	4000	3.0	1115)	669)	1150)
10b	9/13	3.0	20	4000	3.0	568 } 713	669 } 669	896 } 929
10c	9/13	.6	14	4000	3.0	456)	669)	740)
11	9/17	.2	15	3000	3.0	1982	669	770

*Wind speed data from the U.S. Weather Bureau, Lansing, Michigan.

BIBLIOGRAPHY

1. Colpetzer, Thomas R. "A Correlation Study of Environmental Radioactivity." Unpublished Master's thesis, Michigan State University, East Lansing, 1962.

2. "Residual Nuclear Radiation and Fallout." The Effects of Nuclear Weapons. Department of the Army Pamphlet No. 39-3. Washington: Government Printing Office, 1962.

3. Setter, Lloyd R., John E. Regnier, and Edward A. Diephaus. "Radioactivity of Surface Waters in the United States," Journal American Water Works Association, 51:1377-92, 1959.

4. Campbell, J. E., et al. "The Occurrence of Strontium-90, Iodine-131, and Other Radionuclides in Milk - May, 1957, Through April, 1958." American Journal of Public Health, 49:2, February, 1959.

5. Langham, Wright, and E. C. Anderson. "Biospheric Contamination from Nuclear Weapons Tests Through 1958." Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, Congress of the United States, May 5, 6, 7, and 8, 1959, Vol. 2. Washington: Government Printing Office, 1959.

6. Folsom, T. R., and G. J. Mohanrao. The Study of the Behavior and Significance of Certain Radioactive Isotopes Found in the Sewage Treatment Plants of Selected Cities. A Research and Development Report Prepared for the United States Atomic Energy Commission. San Francisco Operations Office, 1961.

7. Conard, Robert A. "The Effects of Fallout Radiation on the Skin." The Shorter-Term Biological Hazards of a Fallout Field. A Symposium Sponsored by the Atomic Energy Commission. Washington: Government Printing Office, 1956.

8. Miyake, Y., K. Saruhashi, Y. Katsuragi, and T. Kanazawa. "Radioactive Fallout in Japan and Its Bearings on Meteorological Conditions," Papers in Meteorology and Geophysics, XI:1, 1960.

9. Miyake, Y., K. Saruhashi, Y. Katsuragi, and T. Kanazawa. "Seasonal Variation of Radioactive Fallout," Journal of Geophysical Research, 67:1, 1962.

10. Sotobayashi, Takeshi, and Seitaro Koyama. "Deposition of Nuclear Debris and Atmospheric Conditions," Journal of the Faculty of Science, Niigata University, Japan, 3:2, 1962.

11. Greenfield, S. M. "Rain Scavenging of Radioactive Particulate Matter from the Atmosphere," Journal of Meteorology, 14:1957.

12. Greenfield, S. M. "Ionization of Radioactive Particles in the Free Air," Journal of Geophysical Research, 61:1956.

13. Itagaki, K., and S. Koenuma. "Altitude Distribution of Fallout Contained in Rain and Snow," Journal of Geophysical Research, 67:10, 1962.

14. Salter, L. P., P. Kruger, and C. L. Hosler. "Sr^{90} Concentration in Precipitation Resulting from Large-Scale Uplift," Journal of Applied Meteorology, 1:1962.

15. Miyake, Y., Y. Sugiura, Miss K. Saruhashi, and Mrs. T. Kanazawa. "The Estimation of the Amount of Sr-90 Deposition and the External Infinite Dose in Japan Due to Man-made Radioactivity," UN Scientific Committee on the Effects of Atomic Radiation Document, A/AC, 82/G/R, No.172. Pap. Met. Geophys., Tokyo.

16. Setter, L. R., and G. I. Coats. "The Determination of Airborne Radioactivity," (paper presented at the Twenty-first Annual Meeting of the American Industrial Hygiene Association, Rochester, New York, April 28, 1960).

17. Cotton, Richard A., Research Director, Millipore Filter Corporation, in a letter to the author, October 15, 1962.

18. Setter, L. R., G. R. Hagee, and C. P. Straub. "Analysis of Radioactivity in Surface Waters - Practical Laboratory Methods." American Society for Testing Materials Bulletin No. 227, January 1958.

19. Nuclear-Chicago Corporation. "How to Prepare Radioactive Samples for Counting on Planchets," Technical Bulletin No. 7 and 7B, 1960.

20. Vaughn, James C., Robert W. Schmidt, Alfred Tenny, and Arthur Shor. "Studies of Radioactivity in the Chicago Water Supply," Journal American Water Works Association, 50:581, 1958.

21. Dixon, Wilfrid J., and Frank J. Massey, Jr. Introduction to Statistical Analysis. New York: McGraw-Hill Book Company, Inc., 1957. 488pp.

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