A CORRELATION STUDY OF ENVIRONMENTAL RADIOACTIVITY

Thesis for the Degree of M. S.

MICHIGAN STATE UNIVERSITY

Thomas Richard Colpetzer

1962

THESIS

This is to certify that the

thesis entitled

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Shosei Serata
Major professor

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OF

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By

Thomas Richard Colpetzer

AN ABSTRACT

Submitted to
Michigan State University
in partial fulfillment of the requirements
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THOMAS RICHARD COLPETZER ABSTRACT

This thesis reports an investigation of the relationship between air particulate radioactivity and rainfall radioactivity. The effects of the amount of rainfall upon rainfall activity levels were also studied.

Statistical tests were made to determine whether any association or correlation exists between air particulate activity and rainfall activity. In addition, inferences were made about how rainfall activity levels vary with amount of rainfall.

The analysis of data collected during periods of low activity levels indicated that rainfall activity in units of μc/ml tended to increase as the amount of rainfall increased. For the same period no relationship was found between rainfall and rain activity in units of $\mu c/cm^2/day$, and the correlation coefficient for these two variables was not significantly larger than zero.

Rainfall activity in units of μc/ml and $\mu c/cm^2/day$ was found to be correlated with air particulate activity when the radioactivity levels were relatively high. No correlation was found between the two variables when activity levels were low.

It was found that when activity data were analyzed on a basis of monthly averages rather than weekly averages, correlation was found between air particulate activity

and total deposition of rainfall activity ($\mu c/cm^2/day$) when the activity levels were low.

Statistical tests not used in this study were proposed for use in future investigations.

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I. INTRODUCTION

The increase in the background level of environmental radioactivity in recent years has prompted many investigators to study the sources and characteristics of radioactivity in our environment. Monitoring of the radioactivity in rain water, surface water, food, soil, and the atmosphere is being carried on continuously.

It has been the purpose of this study to investigate air particulate activity and rainfall activity to determine if any relationship exists between these two variables. Some degree of correlation was expected since the radioactive matter in rainfall originates from air particulate activity and high activity levels in the atmosphere would be expected to cause correspondingly high levels of rainfall activity.

The data used in this study were from three sources. The Division of Occupational Health of the Michigan Department of Health provided records of radioactivity levels at various stations near the nuclear reactor site at Monroe, Michigan. Precipitation records for the same period of time were obtained from climatological reports published by the U. S. Government. The final source of data was the U. S. Public Health Service which reported atmospheric and rainfall radioactivity levels for Lansing, Michigan.

II. LITERATURE REVIEW

Sources of Environmental Radioactivity

Setter and Russel (1)* discuss radioactive contamination of the environment in the Cincinnati, Ohio area. They indicate that the more important sources of man made radioactive wastes are from: use of radioisotopes in industry, medicine, and research institutions; mining and chemical processing of uranium ore; nuclear reactors for power production or research; chemical processing of spent reactor elements for the recovery of nuclear fuel; and fallout due to the use of nuclear testing devices. In their study of radioactivity in the surface waters of Texas; Gloyna, Drynan, and Smallhorst (2) state that the major potential sources of surface-water activity are fallout and washout, natural deposits of radioactive materials, and radioactive wastes.

Atmospheric Radioactivity

Recently, emphasis has been placed on the study of nuclear detonation fallout; whereas, it has long been established that significant amounts of radon (Ra^{222}) and thoron (Rn^{220}) in equilibrium with their daughter products are found existing naturally in the atmosphere. Davis, et al. (3) found that decay curves for air samples which they collected indicated an apparent half-life of

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* Numbers in parenthesis are references listed at the end of this paper.

about 30 minutes. Since radon in equilibrium with its daughter products exhibits an apparent half-life of approximately 30 minutes, it was assumed that radon was the primary active element in the air samples. On this basis, Davis, et al. (3) performed a correlation analysis on the relationship between weather variables and atmospheric radioactivity. They found statistically significant correlations between wind speed at the time of sampling and temperature, dew point, visibility, and the north-south component of the wind vector prior to the time of sampling.

Calculations were made by Dunning (4) on the contribution of shorter lived fission products in radioactive fallout to the world-wide external gamma levels. These shorter-lived isotopes represent a mixture of fallout debris from many nuclear detonations in the past and have been stored for different lengths of time in the stratosphere and troposphere.

Radioactivity in the atmosphere is associated with particulate matter to a greater or lesser degree. Setter and Russell (1) report that in 1957, the annual fallout in the Cincinnati, Ohio area amounted to 15,200 $m\mu c/m^2$ of which 56 per cent was attached to suspended matter. A large proportion of the fallout was young fission products (2 - 5 days old). The 1-year-old portion of this fallout was estimated to be 200 $m\mu c/m^2$.

Radioactivity in Precipitation

Without precipitation there is little fallout even though the particulate activity of surface air is abnormally high. Fallout measurements, largely as precipitation, have been made on a continuous basis at Cincinnati, Ohio since March 1953. Setter and Straub (5) state that a correlation study is now possible between air-particle concentration and rainout (washout by precipitation). Analysis of data by List (6) shows that a preponderence of the fallout occurs during precipitation.

The activity which is measured in rainfall is that of the long-lived fission products which are potentially the most hazardous. Morgan and Stanbury (7) state that the fission products derived from the testing of atomic and thermonuclear weapons are normally washed out of the lower atmosphere by rain after hold-up periods in the stratosphere during which time the shorter-lived nuclides decay.

Beta activity in rainfall reflects nuclear weapons testing. In a study of the radioactive fallout in the Cincinnati, Ohio area by Nader, et al. (8), it was found that a sharp rise in the beta activity of rain occurred two days after the first Atomic Energy Commission's nuclear-weapons test in March 1953. In addition, the radioactivity of subsequent rains during the test period was usually of greater intensity and varied widely. It was found that the activity of rains can generally be attributed to particular bomb bursts, on the basis of a comparison of the half-lives of rain activities with the theoretical decay of the fission product.

The units in which the activity of rainfall is reported are of two general forms. Many researchers (5,7,8,9) prefer to report rainfall activity in terms of activity per unit volume of rainfall such as micro-curies per liter (μc/l). Others (1,10,11) prefer to report rainfall activity in terms of activity per unit area of surface on which the rainfall is collected. Typical units would be milli-micro-curies per square meter per day ($m\mu c/m^2/day$). The first method of reporting is useful when one is interested in the variation in rainout of activity with time. When total radioactive rainout is being compared with radioactivity which appears in surface waters, the latter method appears more appropriate.

According to the Division of Radiological Health, Public Health Service (12), measurements have indicated that the bulk of deposited activity occurs through precipitation, but concentrations in surface air are not directly related to the amount deposited through precipitation.

Precipitation of radioactivity may be in the form of snow as well as rain. Observations on radioactive snows at Ann Arbor, Michigan (13) definitely established the presence of radioactive rare earth isotopes. The energy and half-life for these isotopes were such that it could be stated that the activities undoubtedly originated in the Las Vegas atomic test explosions. Activities as high as 100 times background were obtained in these samples.

Radioactivity in Surface Waters

Increasing attention has recently been paid to the possible contamination of drinking water supplies with radioactive materials. The measurements of Kahn and Reynolds (14) have shown that water from rivers and lakes contains detectable amounts of Sr^{90} and Cs^{137} which are potentially the most hazardous of the long-lived fission products. The quantities of pollution by natural radioactivity and from nuclear-energy operations are so small quantitatively that chemical separations, energy determinations, and half-life measurements are difficult (15).

Setter, et al. (10) found a linear relationship to exist between radioactivity and concentration of suspended solids in stream surface water. These findings provide circumstantial evidence that radioactive runoff is proportional to rain intensity, because the concentration of suspended matter is roughly proportional to rain intensity. Because radioactivity is associated with suspended matter, impounded waters have a high ratio of dissolved to total activity and a low concentration of suspended matter (10).

Thomas, et al. (16) found significant increases in activity in the surface waters in Massachusetts which were apparently due to fallout of fission products from Nevada nuclear-weapons tests. Previous testing did not cause large increases in the activity of surface waters in Massachusetts, but meteorological conditions were such that a portion of the air mass over the Nevada test site on June 1st moved rapidly eastward with only a small amount of rainout before reaching the Massachusetts area.

III. APPROACH TO PROBLEM

Objectives

The primary purpose of this study has been to determine if there is any measurable amount of correlation between radioactivity in rainfall and the level of radioactivity in the atmosphere. It would be desirable to ascertain what, if any, effect a rise in the atmospheric radioactivity level would have upon the amount of radioactivity which reaches the earth in the form of precipitation.

There is also interest in the levels of radioactivity in other media such as surface waters, soil, and biological specimens. It was hoped that on the basis of this statistical study, new light would be shed on the factors which influence these radioactivity levels.

The results of this study will provide a basis for re-evaluation of the present method and extent of data collection. A considerable amount of time, effort, and money has been, and will be, spent on the monitoring of radioactivity levels in various media. If some part of this data collection is found to be unnecessary, a considerable saving in effort and expenditure could be attained. An ideal development would be the elimination of the necessity for monitoring of rainfall radioactivity. This could be realized by finding a high degree of correlation between the levels of radioactivity in the atmosphere and in rainfall.

An additional objective has been to provide a somewhat systematic procedure which can be followed in the future study of correlation between radioactivity levels in other media. This thesis reports the first extensive study of correlation between levels of gross beta radioactivity in the atmosphere and in rainfall, and the statistical procedure presented will be found useful in future studies.

On the basis of the results of this study, it was hoped that some answers could be given as to the method of transfer of the radioactive particulate matter from the atmosphere to the earth in the form of rainfall. It was realized that this goal might be severely limited by the method of attack used in this study and the amount of data used in the analysis.

Finally, the purpose of this study was to provide an insight into the difficult problems involved and to provide a basis for further research in the field of environmental radioactivity.

Problems Encountered

One of the most serious problems encountered in this study was the limited amount of data which was available for analysis. Some of the possible methods of analysis which might have been used were precluded by the fact that, for various reasons, the weekly rainfall activity levels were not all reported. If a continuous series of weekly

observations had been available, it would have been possible to investigate the activity levels for the existence of a time lag between the atmospheric radioactivity and radioactivity of rainfall. In any future study of this type where data are being collected by the investigator, an effort should be made to assure that all the possible data are taken and that no unnecessary omissions are allowed.

Radioactivity levels are often reported on a weekly basis and the data used in this study were weekly averages. Analysis of this data did not permit insight into how air particulate activity is affected by daily rainfall. If records of daily activity levels had been available, it would have been possible to determine how efficiently various types of rains remove radioactive particles from the atmosphere. Future analysis should be based on daily activity levels if at all possible.

An inherent difficulty of the statistical analysis of data of this sort is that it is not possible to prove beyond doubt that correlation between weather variables does not exist. If correlation is found to exist between two variables, this result may be stated with a well defined degree of certainty and no special problems arise. On the other hand, if no correlation is found using a certain type of analysis, this does not preclude the possibity that the two variables being studied might be found to be highly correlated if some other type of analysis were used. This problem cannot be avoided, but an attempt has been made to conduct a thorough analysis reporting the procedures used so that future efforts by other analysts will not be wasted.

Low levels of radioactivity are a problem to a correlation study of this type. Such low levels are difficult to determine accurately and the standard errors of the results tend to be quite high. This normal variation of counting makes a correlation analysis very difficult and a large amount of data is required to show correlation even if the variables are indeed related.

Many weather variables have been found to influence the atmospheric radioactivity levels to a greater or lesser extent. An important factor is the movement of air due to wind from one area to another. Air moving from an area where a rain has just occurred is likely to have a low concentration of radioactive particulate matter. It is obvious that the activity of a given rainfall might be quite low if the air activity is low on that particular day while at the same time, the average air particulate activity for the week of record has been very high. If situations of this sort occur often, it will appear that there is no correlation between air and rain activity. Possibly the activity levels in rainfall are also affected by a number of undetermined weather parameters. It is obviously impossible to recognize and take into account all the possible variables which exert their influence

and this limitation should be recognized when evaluating the results of this study.

The radioactivity levels being analyzed in this study are composed of contributions from various sources. In addition to the nuclear testing debris in the atmosphere, there is secondary radiation from cosmic rays, activity initiated at nuclear power reactors, and natural background radiation. For a strictly accurate correlation analysis it is necessary that there be comparability among data for different periods. If any of the variables are varying with time the data are said to be non-homogeneous. This was actually one problem faced in this study, and the assumption was made that this non-homogeneity would have no adverse effect upon the comparability of the data for different periods. The validity of this assumption determines the validity of the results of this study to some extent.

Air particulate activity as reported in the data used in this study is actually the activity of air which is near the surface of the earth. Thunder storms which occur during the summer months originate as ice crystals at altitudes ranging from 30,000 to 50,000 feet (21). These ice crystals are formed around particulate matter some of which is radioactive. Therefore, particulate activity from very high altitudes as well as from surface air is rained out during the summer months. Particulate activity at the higher altitudes is not necessarily the same as surface activity and it is possible that no correlation exists between surface air activity and the rainfall activity.

Winter rainfall is of a different nature than that which occurs during the summer months. Winter rains are caused by the movement of a warm front into a lower temperature area. The warm air is forced to rise and at altitudes of 3,000 to 8,000 feet the temperature falls below the dew point of the air and precipitation results. Therefore, during the winter months the activity in rainfall may be quite different than the activity found during the summer, and comparison of the data without regard to the seasons is not entirely justified. Seasonal variation in activity was not taken into account in this study because of the limited amount of data available for analysis.

Future Study

For the purpose of future study a block diagram is presented in Figure 1. The steps shown in the diagram are essentially the ones followed in the approach to the problem of this study. Future correlation studies of environmental radioactivity may use this approach with some modifications.

IV. ANALYSIS OF DATA

Description of Data

The data used for analysis in this study were obtained from the Division of Occupational Health of the Michigan Department of Health (11) and from the U. S. Public Health Service (17). Supplementary data on precipitation at Monroe, Michigan were obtained from U. S. Weather Bureau publications (18).

The data supplied by the Michigan Department of Health were observations of radioactivity levels in rainfall and air particulate matter. These observations were originally made to determine any changes in background radiation which might have occurred as a result of activation of the Enrico Fermi nuclear power reactor near Monroe, Michigan. This analysis used data which the Michigan Public Health Department collected in the period from June 15, 1959 to December 19, 1960. The U. S. Public Health Service data covered the period from October 29, 1961 through May 20, 1962 and was collected by the Lansing, Michigan station of the Radiation Surveillance Network.

Radioactivity of air particulate matter was reported as gross beta activity one day after collection. Weekly average values of activity were reported in units of micro-micro-curies per cubic meter of air sampled.

The radioactivity of rain samples was also expressed as gross beta activity which did not include the contribution of naturally occurring radon and thoron and their daughter products. The data were tabulated in two separate units; micro-curies per milliliter, and micro-curies per square centimeter per day. As with the air particulate activity, the results were reported in terms of weekly averages. Certain weekly data were missing where no samples were collected and where the net count rate of the samples was less than or equal to the background count rate of the counting equipment, the results were reported as, "Trace".

The precipitation data obtained from U. S. Weather Bureau publications were reported in inches of rainfall. Rainfall data were only available at the Monroe station and daily rainfall records were condensed to weekly totals corresponding to the weekly sampling periods for radioactivity of the rain samples.

Radioactivity of Rain Samples

Data from the Michigan Department of Health were used to investigate how the radioactivity of rain varies with rainfall. Rainfall activity expressed in units of micro-curies per square centimeter per day ($\mu c/cm^2/day$) was plotted against the corresponding volume of rainfall in inches. These plots are shown in Figures 2 and 3. It was expected that some trend would be observed in these plots if average activity precipitated per day was:

(1) independent of the atmospheric activity levels, and

(2) independent of the frequency, intensity, and duration of the various rains contributing to the total weekly rainfall.

It was further expected that the trend would be for an increase in average rain activity to occur if the rainfall increased.

Another approach used in the investigation of rain activity was to plot activity, expressed in units of microcuries per milliliter (μc/ml), versus rainfall in inches. It is to be noted that each value plotted was a paired observation of rainfall and rain activity for a certain week. A trend in the plots was expected if the average activity per unit volume of rainfall was:

(1) dependent upon the total amount of rain which fell during the week, and

(2) independent of the atmospheric activity levels.

These plots appear in Figures 4 and 5.

The third relationship studied was a plot of rainfall activity in μc/ml versus a measure of the total deposition of activity. The measure of total deposition was obtained by multiplying the rainfall activity expressed as μc/ml by the corresponding total weekly rainfall in inches. Again, each of the points plotted represented a paired observation. For convenience, the plot was made on semi-logarithmic paper and is shown in Figure 6.

Atmospheric Activity Versus Rainfall Activity

In an effort to determine if there was any relationship between atmospheric radioactivity and radioactivity of rainfall, several different approaches were taken. Figures 7 and 8 show air particulate activity and rainfall activity plotted on the same time scale. These figures show data reported by the Michigan Department of Health. The time scale is not continuous because of the discontinuity of the data, and therefore the rain activity and air particulate activity can only be compared on a same week basis. Figure 7 shows rain activity in units of µc/ml and Figure 8 shows rain activity in units of µc/cm^2/day.

Using the data reported by the Michigan Department of Health, air particulate activity was plotted versus rainfall activity in Figures 9 through 12. The points on these scatter diagrams each represent air particulate activity and rainfall activity for a given week.

Application of Corner Test

For preliminary investigation for correlation, Figures 9 through 12 were used to make a statistical test called the Corner Test (19). The Corner Test was made to test the hypothesis that the two variables were independent. The non-parametric test statistic was computed in the following manner. A vertical line was passed through each scatter diagram which divided the data with 50 per cent of the observations on the right and 50 per cent on the left. A similar horizontal line divided the observations with 50 per cent above and 50 per cent below. Positive and negative signs were assigned to the various quadrants as follows: upper right positive, upper left negative, lower left positive, and lower right negative. Starting at the extreme right and moving toward the vertical median line, the number of observations encountered before an observation was found across the horizontal median were counted. In Figure 9 this number was +1. This number was given the sign of the quadrant in which the extreme observations were found. Proceeding similarly from the top, left, and bottom, four numbers were obtained. The algebraic sum of these four numbers (+6 in Figure 9) was taken as the test statistic. If this sum was found to be too large at the 5 per cent level of significance, the hypothesis that the variables were unrelated was rejected. The test statistic was judged to be too large when its value exceeded a computed value found in an appropriate table (19). The chance that this computed value would be exceeded was only 5 per cent if the variables were not associated.

Application of F-Test

Air and rain activity data from the Michigan Department of Health and from the U. S. Public Health Service were used to test for the existence of correlation. The

Department of Health data were analyzed on a monthly and weekly basis to see if the same results would be obtained in both cases. The Public Health Service Data were analyzed only on a weekly basis. All rainfall activity was expressed in units of μc/ml and μc/cm^2/day. To simplify the analysis, the rain activity and air activity observations were classified into various categories according to the magnitude of the activity. The F-test was applied by comparing, (1) the total variation in the rainfall activity for specified levels of air activity with, (2) the total variation between the mean rainfall activities for the various categories and the overall mean rainfall activity. The test statistic, F, was the ratio of the variation between the categories to the variation within categories. If F was found to be larger than the critical F value at the 5 per cent level of significance, the hypothesis that there was no correlation between the variables was rejected. The critical F value was selected so that the chance of it being exceeded was only 5 per cent if there was truly no correlation between the variables. A sample calculation for the test statistic, F, is shown in the appendix.

V. RESULTS AND EVALUATION

Radioactivity of Rain Samples Versus Rainfall

Radioactivity of rainfall, in units of $\mu c/cm^2/day$, was plotted versus rainfall in inches in Figures 2 and 3 to determine if there was any relationship between the two variables. It was predicted that radioactivity would increase with rainfall if the rain activity is independent of the corresponding air particulate activity and independent of the frequency, intensity, and duration of the various rains. By examining Figures 2 and 3 it was seen that no such trend was present. This result does not indicate that rainfall activity is dependent upon the characteristics of the various rains and upon the air particulate activity. However, it has been shown that the possibility of dependence of the two variables does exist.

A similar approach was made in Figures 4 and 5 where rain activity, expressed in units of $\mu c/ml$ was plotted versus rainfall. A downward trend in rain activity was expected if the rain activity in these units is independent of air particulate activity levels and dependent upon the total amount of rain which falls. This trend was observed to some degree in Figure 5, although the trend was not definite and unquestionable. Because of the dispersion of the observations in these Figures, the strict independence of rainfall activity and air particulate activity was not verified. The fact that rainfall activity in units of $\mu c/ml$ will decrease with increasing rainfall if air particulate activity remains essentially constant was not definitely proven and further investigation of these variables was made.

In Figure 6, radioactivity of rainfall in $\mu c/ml$ was plotted versus a measure of the total deposition of activity. The measure of total deposition was obtained by taking the product of rain activity in $\mu c/ml$ and total weekly rainfall in inches. It was expected that the concentration of activity in rainfall would decrease as the total amount of deposition increased. The result, however, was in direct disagreement with this prediction. The observations, when plotted on a semi-logarithmic graph, indicated an increase of the intensity with an increase of total deposition. The data used in this particular analysis were those collected by the Michigan Department of Health and the activity levels in rainfall at the time of collection were very low. These low activity levels which are very difficult to measure may be the reason that the prediction was not verified. In very small amounts of rainfall where concentrations of activity are expected to be high, low activity levels would be more difficult than usual to measure and counting errors as high as 50 or 100 per cent of the observed count rate might be present. A similar situation might be found when the total precipitation is high. The rained-out activity would be very highly diluted in this case and again difficult to measure. It

is not impossible that a complete reversal of the results could occur within the limits of the counting errors.
It is concluded that investigation of the variation of activity concentration with total deposition of activity should be made when activity is at a much higher level. No definite statement can be made about the results of this particular analysis.

Atmospheric Activity Versus Rainfall Activity

A statistical test called the Corner Test was applied to the scatter diagrams of air particulate activity and rainfall activity shown in Figures 9 through 12. The data were those taken by the Michigan Department of Health at two separate stations. The purpose was to test the hypothesis that the two variables were not associated. The results of the analysis were as follows:

Figure	Station Number	Units of Rainfall Activity	S = Test Statistic	Critical S
9	1	μc/cm²/day	6	10
10	2	μc/cm²/day	5	10
11	1	μc/ml	13	10
12	2	μc/ml	10	10

With rain activity expressed in μc/cm²/day, the hypothesis that there was no association between the variables was not rejected at the 5 per cent level of significance for either Station 1 (Carleton) or Station 2 (Monroe).
However, when the rain activity was expressed in units of

μc/ml, the hypothesis of no association could be rejected for the data from Station 1 and Station 2. This means that the test statistic, S, could have taken values as high as those observed no more than 5 per cent of the time if the variables were truly not associated. In summary, it has been found that weekly rainfall activity in units of μc/ml was associated with air particulate activity for the same week. Weekly rainfall activity in units of μc/cm^2/day was not associated with air particulate activity.

The F-test was used to test the hypothesis that there was no correlation between air particulate activity and rainfall activity. The Michigan Department of Health data were tested both on a monthly and weekly basis to determine if results would be influenced by the period of record. The results are shown below.

(1) Monthly averages of radioactivity levels reported by the Michigan Department of Health gave the following results:

Station Number	Units of Rainfall Activity	F	Critical F
1	μc/cm^2/day	5.87	3.48
2	μc/cm^2/day	3.69	3.58
1	μc/ml	0.03	3.63
2	μc/ml	0.80	3.58

(2) Weekly averages of radioactivity levels reported by the Michigan Department of Health yielded:

Station Number	Units of Rainfall Activity	F	Critical F
1	$\mu c/cm^2/day$	1.03	2.64
2	$\mu c/cm^2/day$	0.73	2.64
1	$\mu c/ml$	2.39	2.36
2	$\mu c/ml$	0.52	2.37

Using the weekly averages it is seen that the F-ratios were less than the critical values with only one exception and in this case the F-ratio was only very slightly larger than the critical level. In general it could be stated that there was not sufficient evidence to reject the hypothesis of no correlation between air particulate activity and rainfall activity in $\mu c/ml$ or $\mu c/cm^2/day$. However, when the monthly averages were tested for correlation, it was found that rainfall activity in units of $\mu c/cm^2/day$ appeared to be correlated with the average air particulate activity for the same month. This result can be explained by the fact that total deposition of radioactivity as $\mu c/cm^2/day$ depends upon the amount of rain which falls. Even if air particulate activity remained at a constant level for a series of weeks, the total deposition would be highly variable because of the large variation in weekly rainfall. When monthly averages are considered, the fluctuation in amount of rainfall is much smaller. As a consequence, a correlation between total deposition of activity and air particulate activity is more easily discovered when monthly averages are considered.

The results of the F-tests applied to the data collected at Lansing, Michigan are shown below.

Units of Rainfall Activity	F	Critical F
μc/cm²/day	6.72	2.83
μc/ml	3.47	2.83

It is possible to reject the hypothesis that there is no correlation between air particulate activity and rainfall activity in units of μc/cm²/day and μc/ml. This disagrees with the results of analysis of the data collected by the Michigan Department of Health where it was concluded that no correlation existed between air particulate activity and rainfall activity. The activity levels reported by the U. S. Public Health Service were approximately ten times as high as those reported by the Michigan Department of Health. The higher activity levels were the result of recent nuclear testing by the USSR. The higher levels were determined more accurately because the counting errors were greatly reduced, and the results of analysis of the higher activity levels are more reliable. In addition, it is obvious that the components of gross beta activity are different for fission products of different ages. It is quite possible, although not verified by experimental results, that correlation exists between air and rain activity for some nuclides and not others. At present it is only possible to state that there appears to be definite correlation between rainfall activity and air particulate activity when the activity levels are high and consist of young fission products, and no correlation is found for the low levels of activity.

Because of the dispersion of the observations on the scatter siagrams, it was decided that development of prediction equations for the data was unwarranted. For the same reasons, correlation coefficients were not computed. If the correlation coefficients had been computed they would have been quite small and would not have been especially useful in this study.

VI. CONCLUSIONS

From the results of this study the following general conclusions may be drawn:

(1) Radioactivity of rainfall in units of μc/cm^2/day is apparently not related directly to the amount of rainfall in inches. Two conditions would have to be satisfied for for these variables to be so related that an increase in rainfall activity as μc/cm^2/day would result from an increase in rainfall. First, it would be necessary that the level of air particulate activity remain constant or have no effect upon rainfall activity. Second, the concentration of rainfall activity (μc/ml) would have to remain nearly constant for any amount of rainfall. Since concentration of activity varies independently from amount of rainfall then no relation would be expected between rainfall and rainfall activity as μc/cm^2/day.

(2) The average weekly concentration of rainfall activity as μc/ml has been found to increase as the total amount of rainfall increases. This result disagrees with original predictions that rain activity concentrations would decrease with increasing rainfall. These results were interpreted as being inconclusive because the low levels of activity being analyzed were highly variable. It would be necessary to analyze data taken at a time when the activity levels were much higher to obtain reliable and conclusive results.

(3) When environmental radioactivity levels were low,

rainfall activity in units of $\mu c/cm^2/day$ appeared to be unrelated to air particulate activity when average weekly levels were considered. However, the monthly average deposition of activity appeared to be highly correlated with the corresponding monthly average air particulate activity. The explanation for the monthly correlation and not weekly correlation was that total deposition of activity depends upon the number of rains which occur during the period of record. Since weekly rainfall is much more variable in intensity, frequency, and duration than monthly rainfall, a better correlation is expected if monthly averages are considered rather than weekly averages.

With high environmental radioactivity levels, there appeared to be highly significant correlation between air particulate activity and rainfall activity as $\mu c/cm^2/day$. Analysis of the high activity levels was made only on a weekly basis.

(4) The concentration of gross beta activity in rainfall was found to be unrelated to air particulate activity levels when the activity levels were generally low. Both weekly and monthly averages were considered. Significant correlation was found between air particulate activity and rainfall activity as $\mu c/ml$ when the activity levels were high. It was suggested that different fission products were contributing to the gross beta activity depending upon the relative ages of the activity. Further study of specific nuclides and their relation to gross beta activity will be necessary to provide reliable answers to these questions.

VII. FUTURE INVESTIGATIONS

The Public Health Service Radiation Surveillance Network was established in 1956 in cooperation with the Atomic Energy Commission to provide a means of promptly determining increases in levels of radioactivity in air and precipitation due to fallout from nuclear weapons tests. Since the resumption of nuclear testing in September 1961, the network has been expanded to 62 stations. Gross beta activity of air and rain are reported daily by these stations. It is suggested that the data collected by the Radiation Surveillance Network stations should be used in the future study of gross beta radioactivity levels in air and rain. The advantages of using these data for analysis are: (1) the data are readily available, (2) the activity levels observed since September 1961 are relatively high, and (3) the results of such an analysis would be applicable to the entire continental United States.

In addition to the statistical procedures used in this study, more refined and extensive analysis can be made on the Public Health Service data. The following systematic approach is suggested and is fully described by Mills in Statistical Methods (20). To provide a rational attack upon the problem the following questions are to be asked:

(1) Do the available observations provide sufficient evidence that the two variables being studied are related?

(2) If the existence of true correlation is assumed, will a straight line acceptably define the regression?

(3) If correlation exists and a straight line is not appropriate as a regression function, will a given second degree function provide an acceptable measure of regression? If such a function is not suitable, will a different function with the same number of constants, or a polynomial of higher degree, give an acceptable fit?

To obtain the answer to the first question, the procedure followed in this study is adopted. If the answer to the first question is no, the investigator should go no further. If it is yes, the testing of regression functions should be carried out until an acceptable function is found.

In the second step a straight line is fitted to observations by the method of least squares. Computed values of the dependent variable are compared to the observed values by the technique of analysis of variance. The variation of the dependent variable due to the influence of random forces is compared to the variation between computed and observed values by the F-test. If the latter variation is significantly larger than the random variation, it is concluded that a linear relationship does not adequately predict the results.

A search is made for a polynomial of higher order

which defines the relation between the two variables if a straight line is not acceptable. The procedure is the same as described above where the method of least squares is used to determine the best values of the constants in an equation of the desired form.

To make such an extensive investigation feasible, it is suggested that the analysis be done on an electronic computer. The application of computer analysis is desirable to save time and labor and to obtain accurate results.

After the analysis of gross beta activity levels in air and rain, an investigation should be made of the correlation between specific nuclide concentrations in air and rain. Potential health hazards such as strontium-90, iodine-131, and cesium-137 contribute to gross beta activity and research is required to determine if any correlation exists between concentrations of these isotopes in air and rain. Study is also required to determine how various individual nuclides are related to gross beta activity levels. At the present time no relationship has been discovered between gross beta activity and concentrations of various radioactive isotopes, and great deal of study is required in this particular area.

APPENDIX

SAMPLE CALCULATION

The formulas to compute the F-ratio used in this study are shown below. An example of the computation of the F-ratio for monthly averages of activity levels at Monroe, Michigan is also included.

Analysis of Variance Table

Category	Sum of Squares	df	Mean Square
Between	$\sum_{i=1}^{k} n_i(\bar{X}_{i.} - \bar{X}_{..})^2$	$k - 1$	$s_m^2 = \frac{\sum_{i=1}^{k} n_i(\bar{X}_{i.} - \bar{X}_{..})^2}{k - 1}$
Within	$\sum_i \sum_j (X_{ij} - \bar{X}_{i.})^2$	$N - k$	$s_p^2 = \frac{\sum\sum(X_{ij} - \bar{X}_{i.})^2}{N - k}$

$$F = s_m^2 / s_p^2$$

where; k = number of categories into which the air particulate activity levels are divided.

n_i = number of rain activity observations in the ith category of air activity.

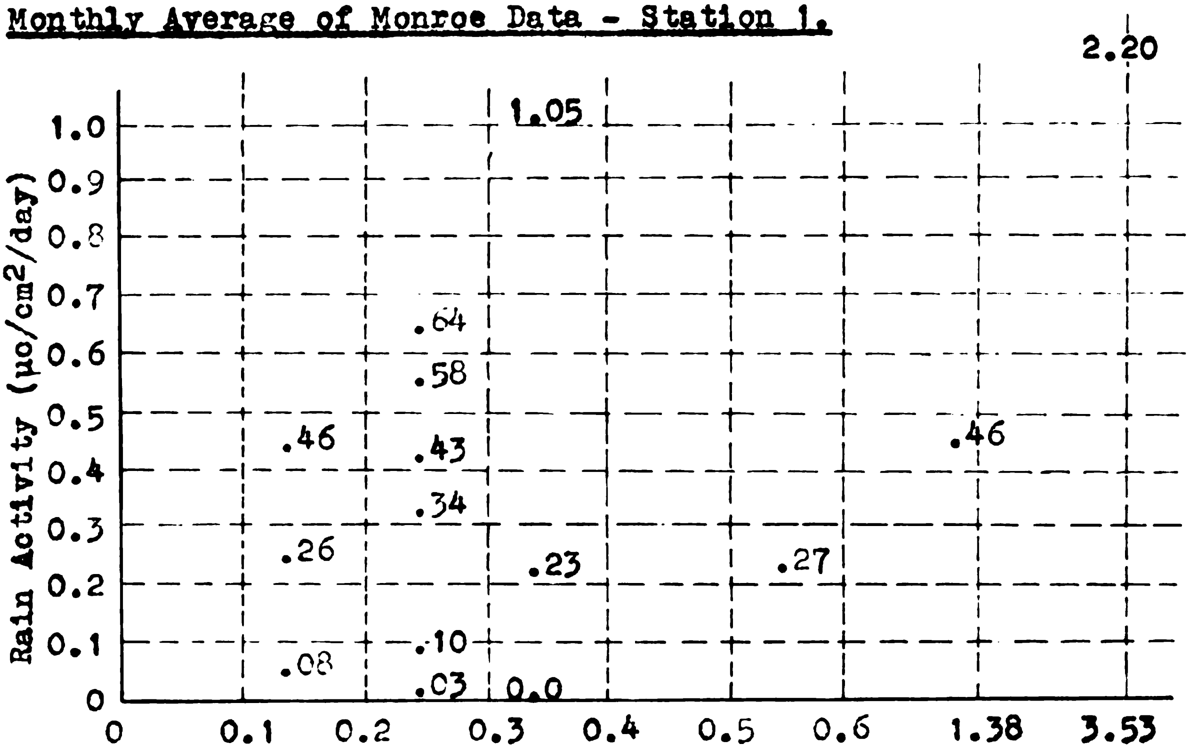
N = total number of rainfall activity observations.

X_{ij} = the measurement of the jth rainfall activity observation found in the ith category of air activity.

$\bar{X}_{i.}$ = the average level of rainfall activity within the ith category of air activity.

$\bar{X}_{..}$ = the overall mean rainfall activity obtained by summing all the rainfall activity observations and dividing by the number of total observations.

Monthly Average of Monroe Data - Station 1.



$n_i =$	3	6	3	1	1	1
$\sum_{j=1}^{n_i} X_{ij} =$	.80	2.12	1.28	.27	.46	2.2
$\bar{X}_{i.} =$	.27	.35	.43	.27	.46	2.2

$$N = \sum_{i=1}^{k} n_i = 15$$

$$\sum_i \sum_j X_{ij} = 7.13$$

$$\bar{X}_{..} = \text{Grand Mean} = 7.13/15 = 0.48$$

Within Sum of Squares:

$(.46 - .27)^2 = .036$
$(.26 - .27)^2 = .000$
$(.08 - .27)^2 = .036$
$(.64 - .35)^2 = .084$
$(.58 - .35)^2 = .053$
$(.43 - .35)^2 = .006$

$(.34 - .35)^2 = .000$
$(.10 - .35)^2 = .062$
$(.03 - .35)^2 = .102$
$(1.05 - .43)^2 = .384$
$(.23 - .43)^2 = .040$
$(.00 - .43)^2 = .185$

0.988

Between Sum of Squares:

$$3(.27 - .48)^2 = 0.132$$
$$6(.35 - .48)^2 = 0.101$$
$$3(.43 - .48)^2 = 0.008$$
$$1(.27 - .48)^2 = 0.132$$
$$1(.46 - .48)^2 = 0.000$$
$$1(2.2 - .48)^2 = 2.860$$

$$3.233$$

Summary:

Analysis of Variance Table

Category	Sum of Squares	df	Mean Square
Between	3.233	5	0.647
Within	0.988	9	0.110

$F = 0.647/0.110 = 5.87$

$F_{.95}(5,9) = 3.48$

Conclusion:

Since the F-ratio is greater than the critical F-ratio for 5 and 9 degrees of freedom, we reject the hypothesis that there is no correlation between air particulate activity and rain activity in units of $\mu c/cm^2/day$. The value of 2.20 for rain activity contributed heavily to the high value obtained for the F-ratio. It is quite probable that the F-ratio would be significantly reduced if this observation were not used in the analysis, and the strength of the conclusion obtained is not great.

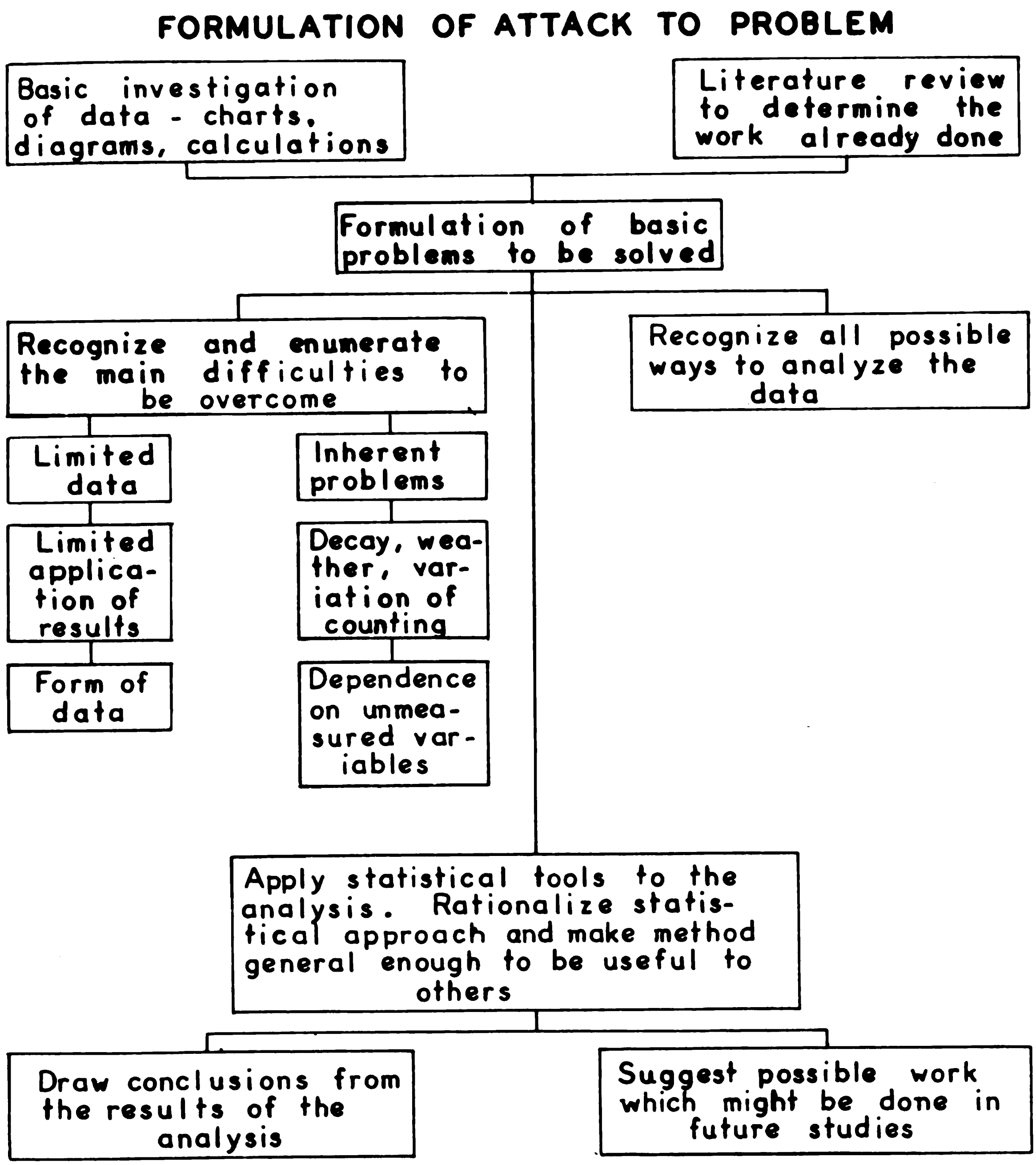

FORMULATION OF ATTACK TO PROBLEM
Basic investigation of data - charts, diagrams, calculations
Literature review to determine the work already done
Formulation of basic problems to be solved
Recognize and enumerate the main difficulties to be overcome
Recognize all possible ways to analyze the data
Limited data
Limited application of results
Form of data
Inherent problems
Decay, weather, variation of counting
Dependence on unmeasured variables
Apply statistical tools to the analysis. Rationalize statistical approach and make method general enough to be useful to others
Draw conclusions from the results of the analysis
Suggest possible work which might be done in future studies

Figure 1

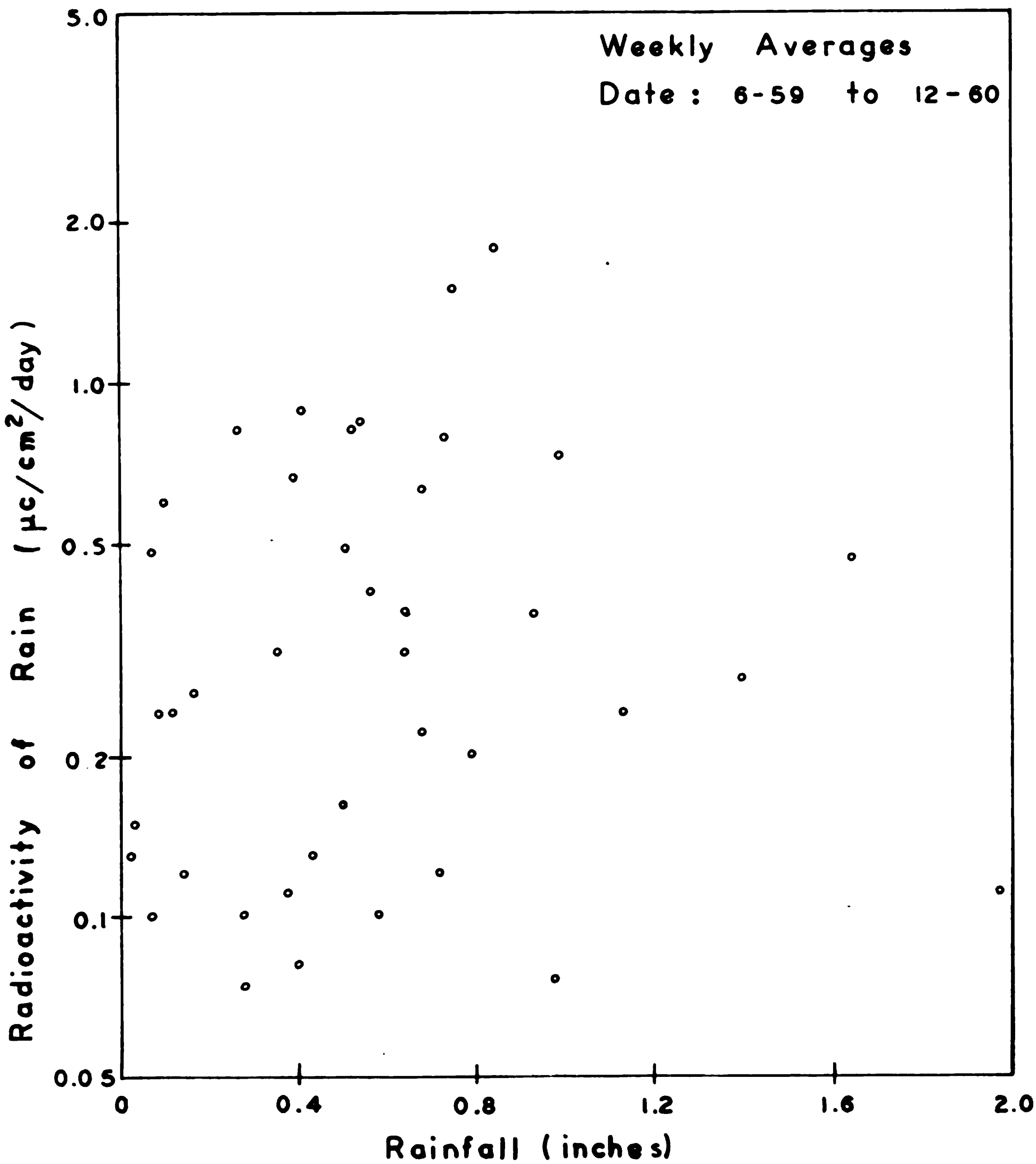


Figure 2. Radioactivity of rainfall in $\mu c/cm^2/day$ versus rainfall for Station 2 at Monroe, Michigan.

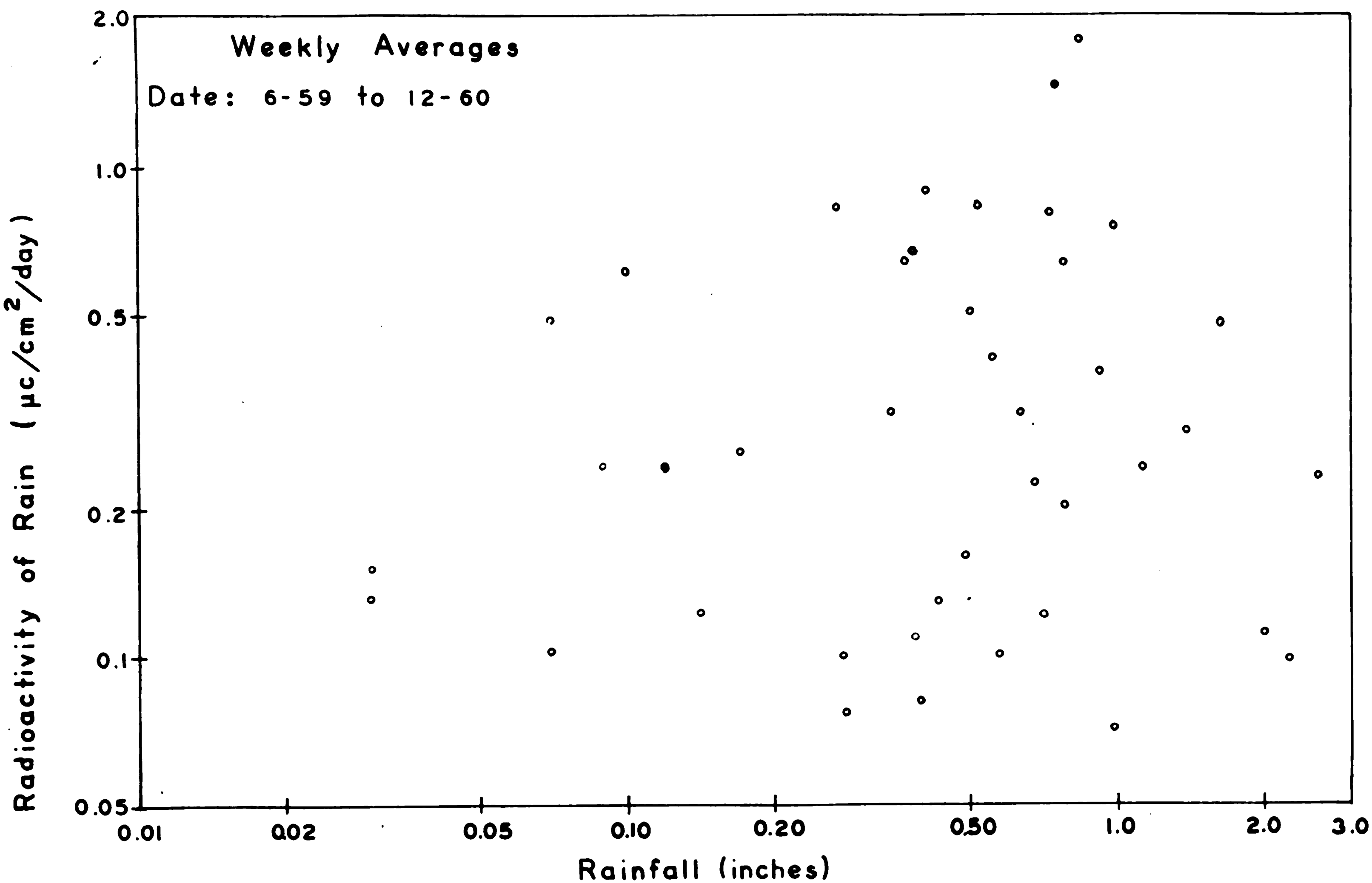


Figure 3. Radioactivity of rainfall in $\mu c/cm^2/day$ versus rainfall for Station 2 at Monroe, Michigan.

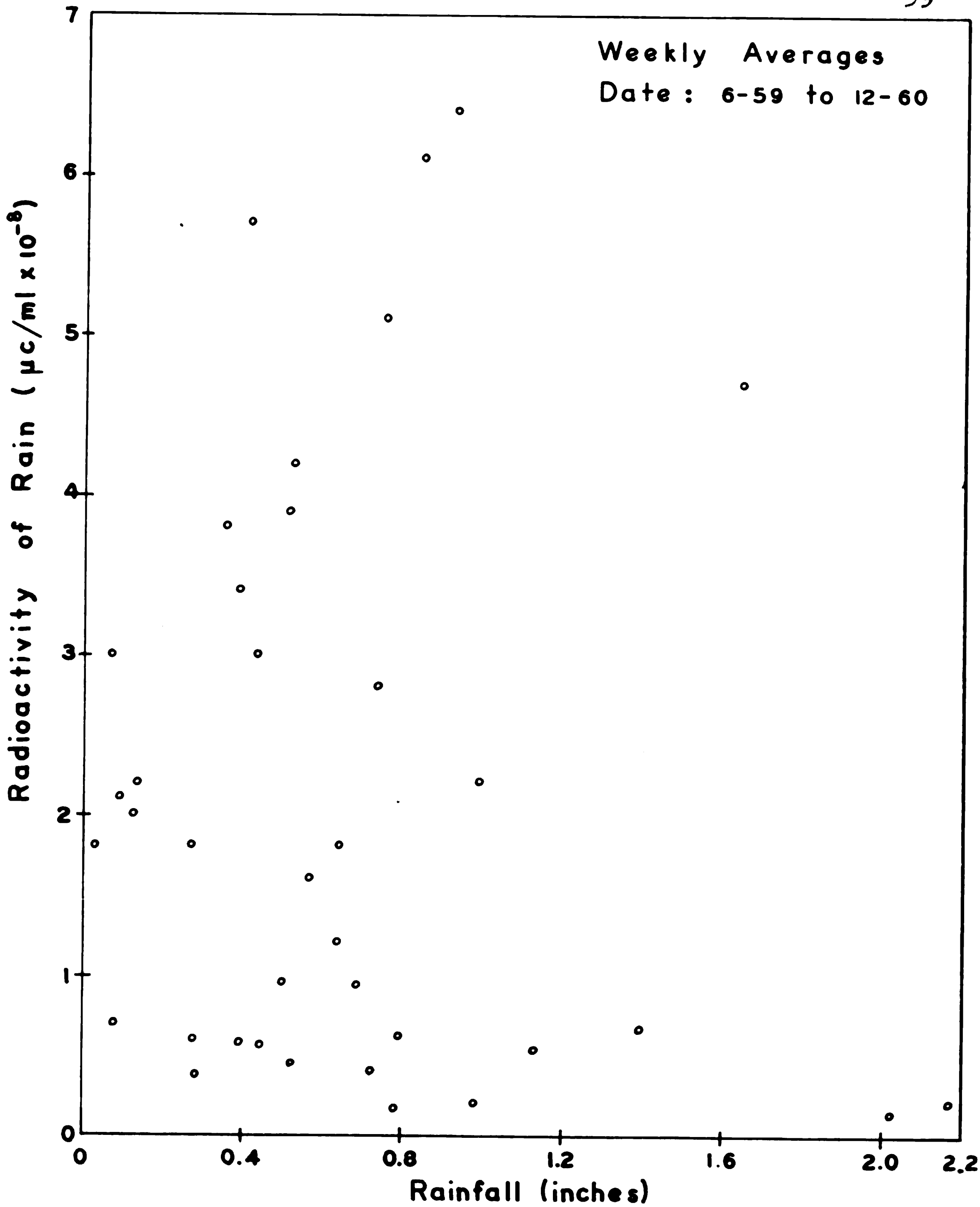


Figure 4. Radioactivity of rainfall in $\mu c/ml \times 10^{-8}$ versus rainfall for Station 2 at Monroe, Michigan.

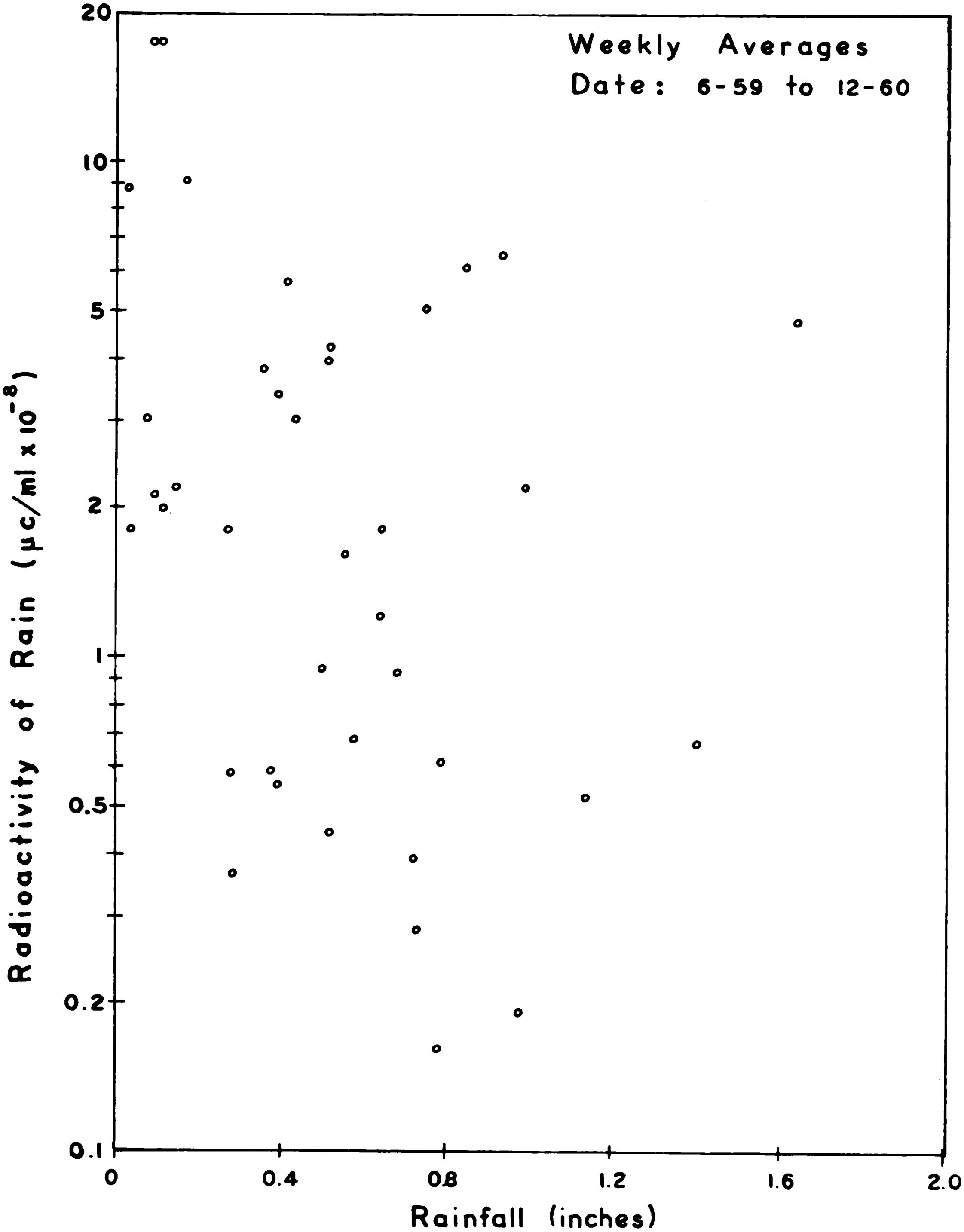


Figure 5. Radioactivity of rainfall in $\mu c/ml \times 10^{-8}$ versus rainfall for Station 2 at Monroe, Michigan.

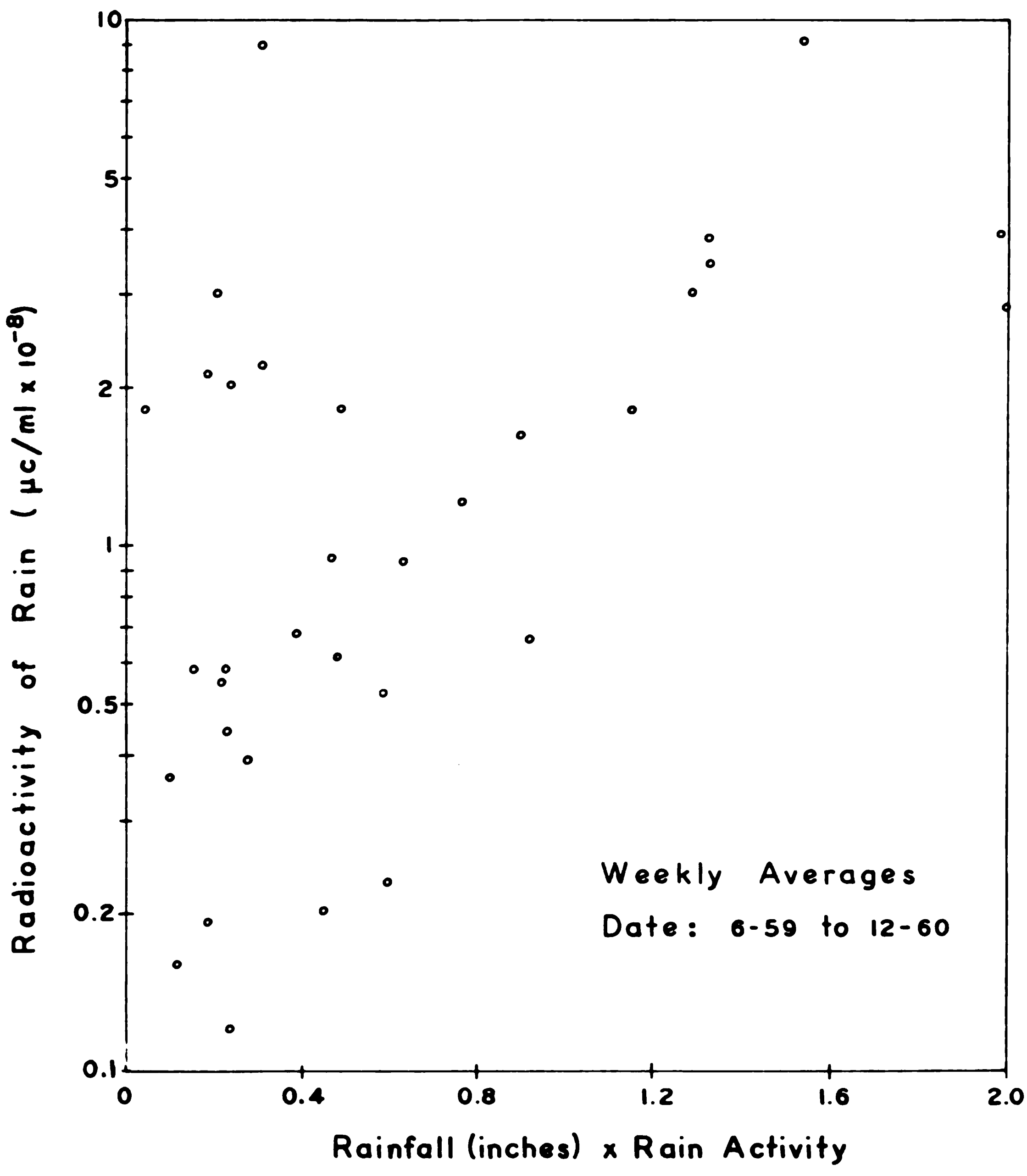


Figure 6. Radioactivity of rainfall in μc/ml versus the product of rainfall and rain activity for Station 2 at Monroe, Michigan.

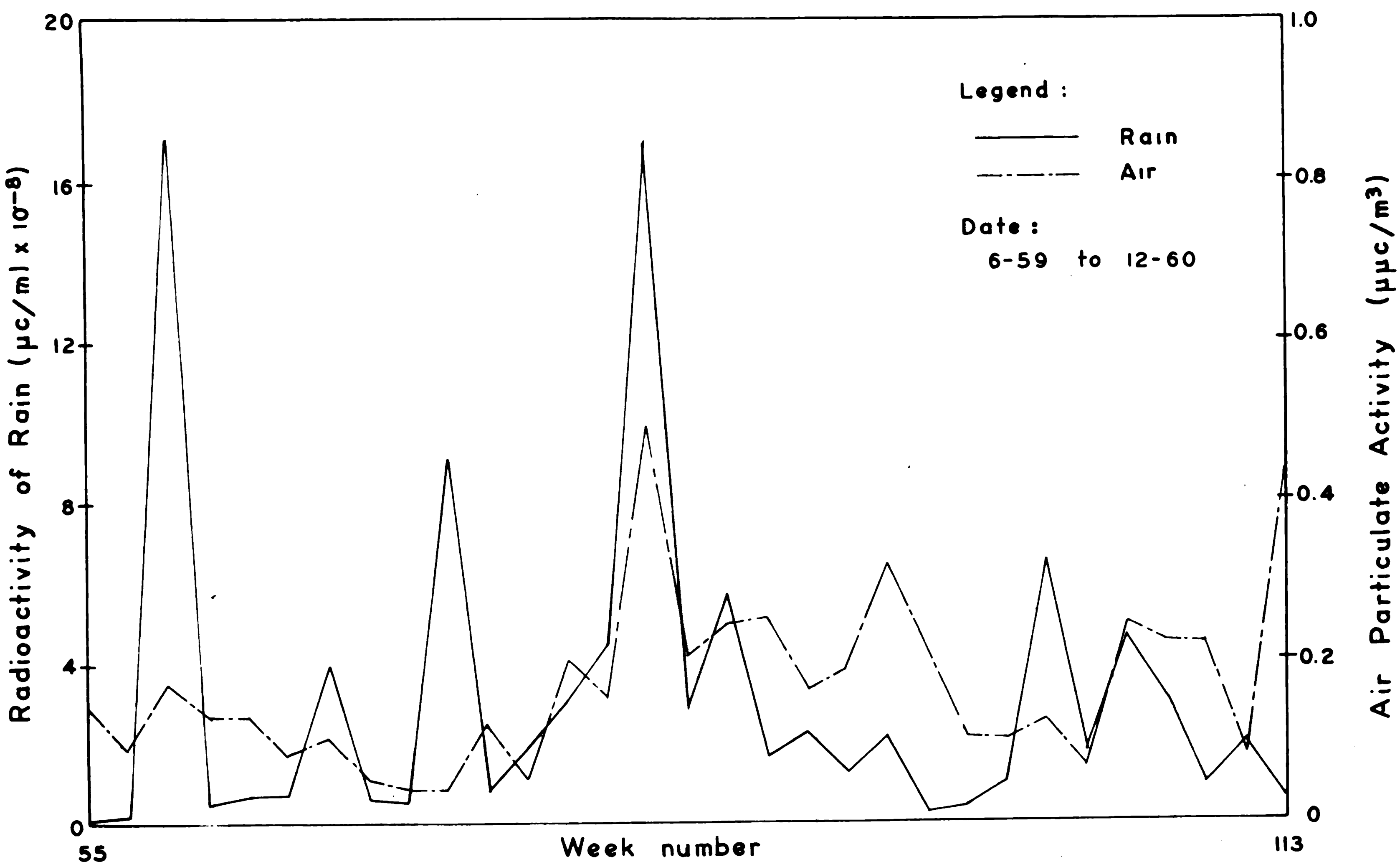


Figure 7. Variation of rain and air activity with time for Station 2 at Monroe, Michigan.

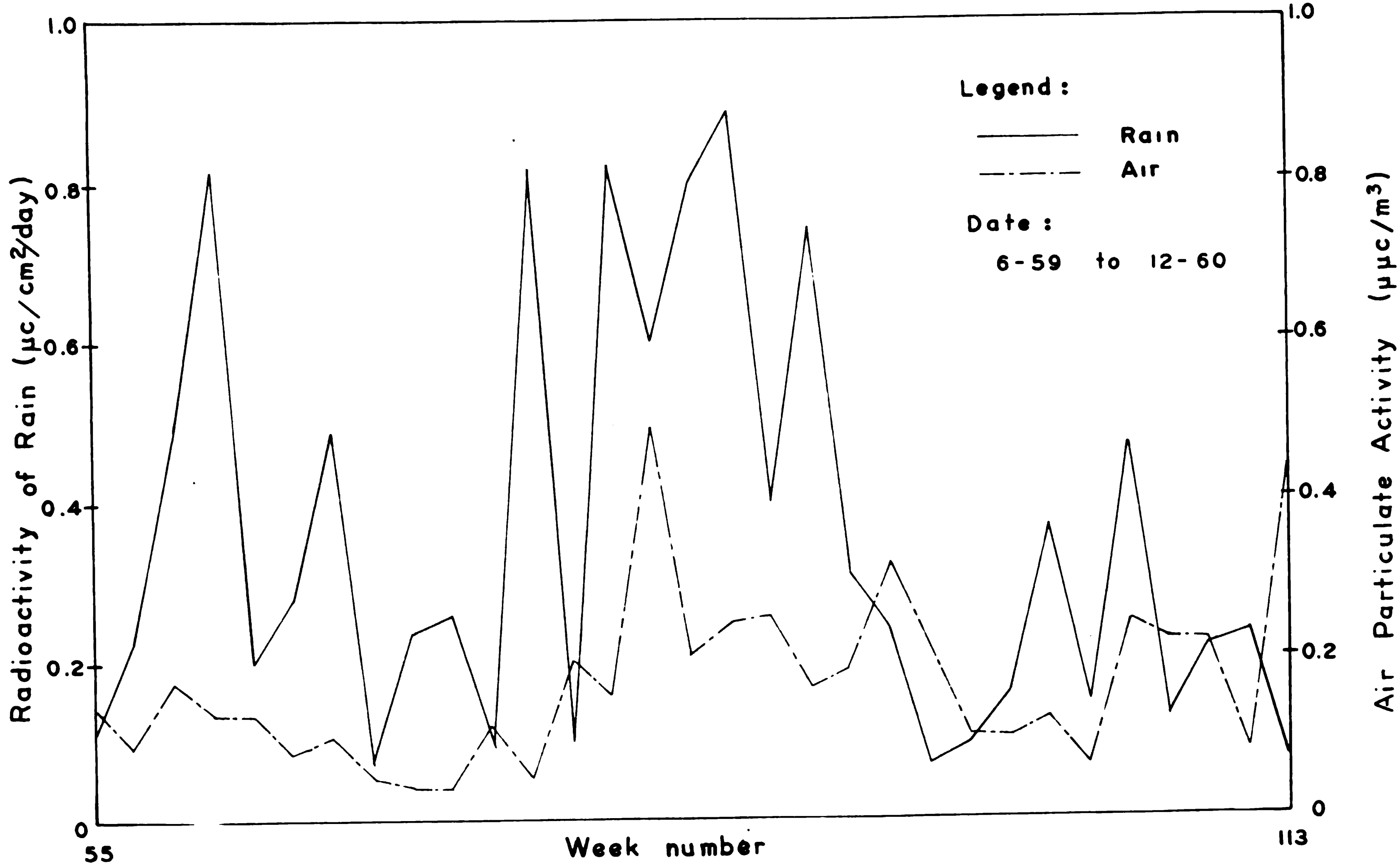


Figure 8. Variation of rain and air activity with time for Station 2 at Monroe, Michigan.

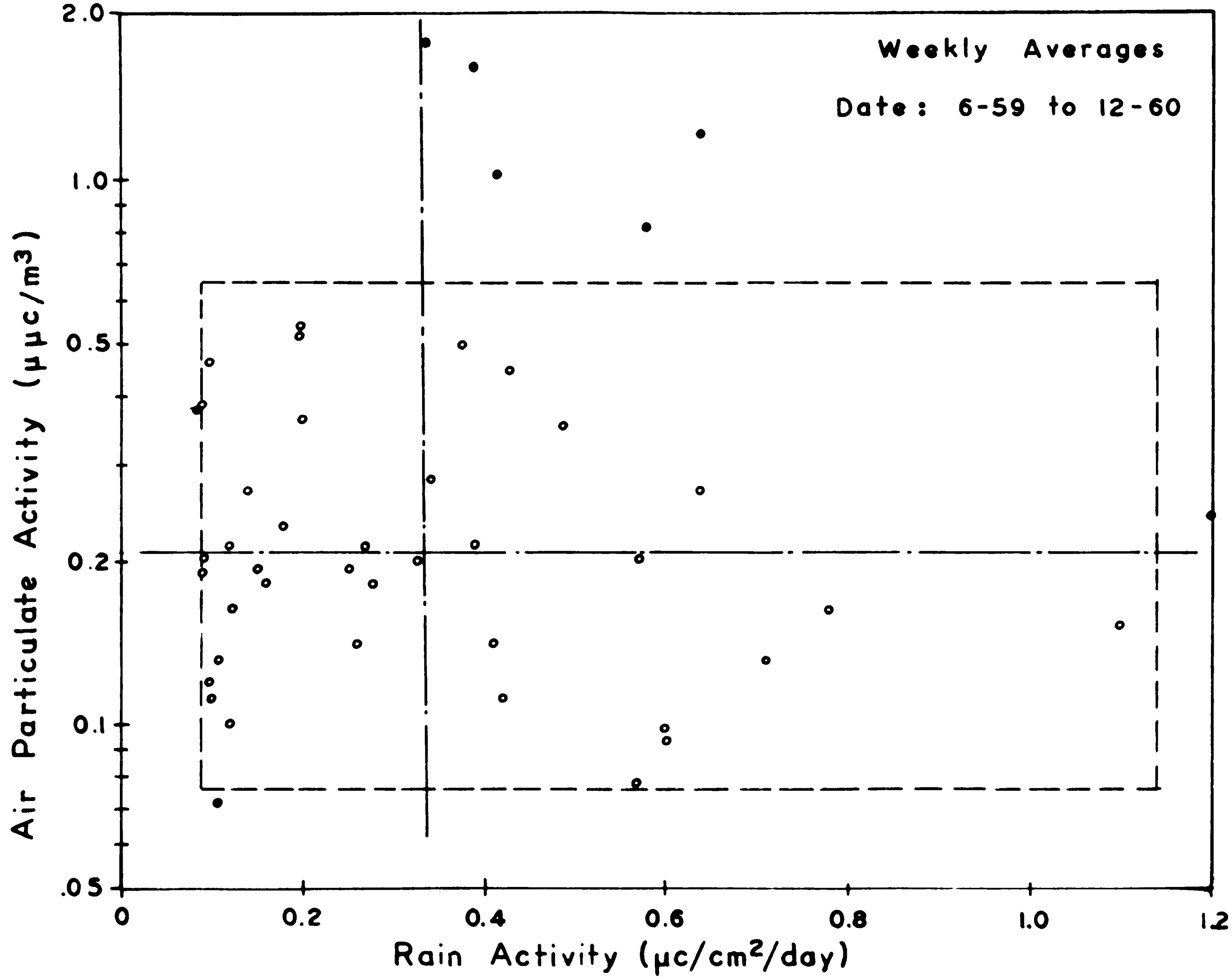


Figure 9. Scatter diagram of air particulate activity versus rainfall activity for Station I at Carleton, Michigan.

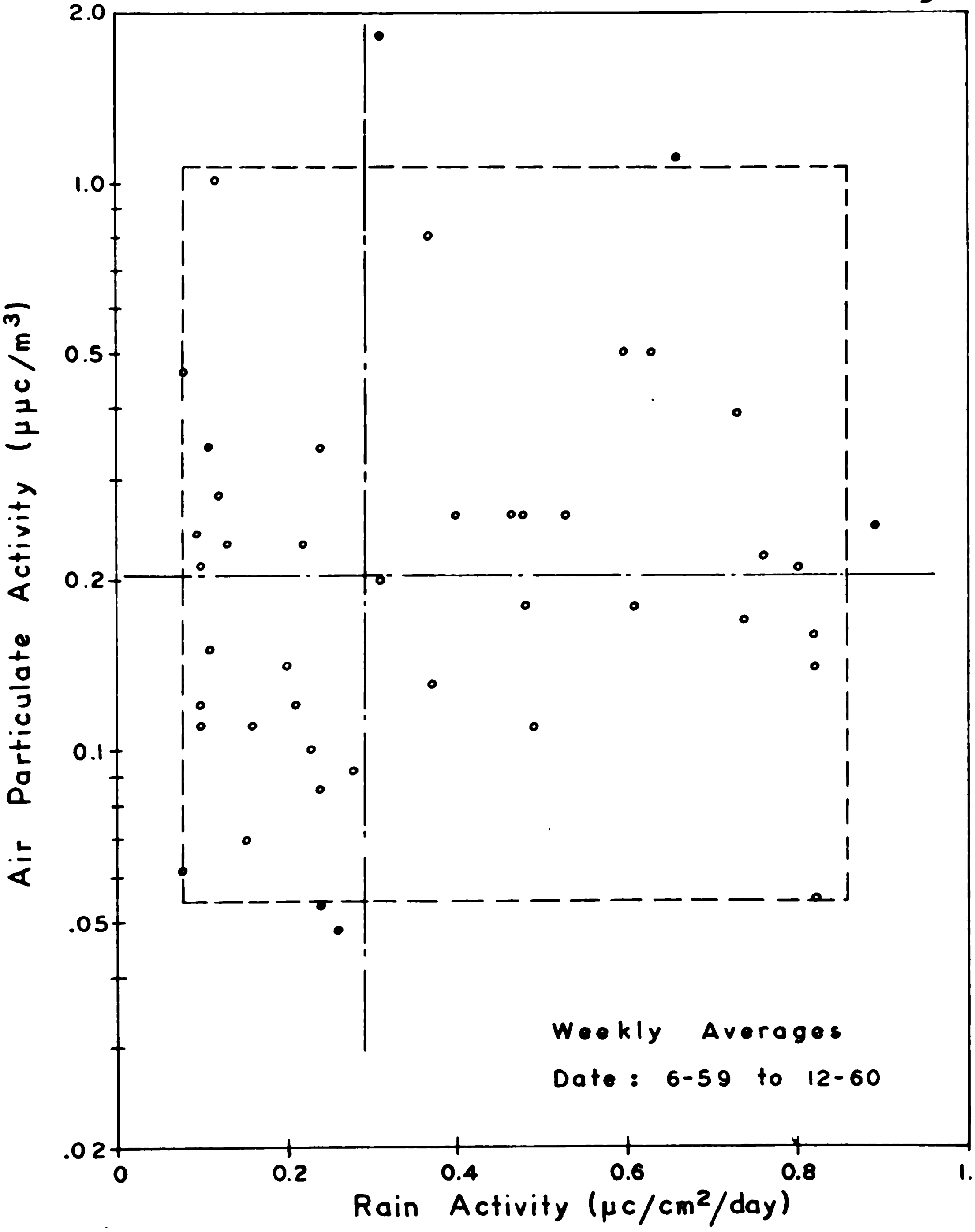


Figure 10. Scatter diagram of air particulate activity versus rainfall activity for Station .2 at Monroe, Michigan.

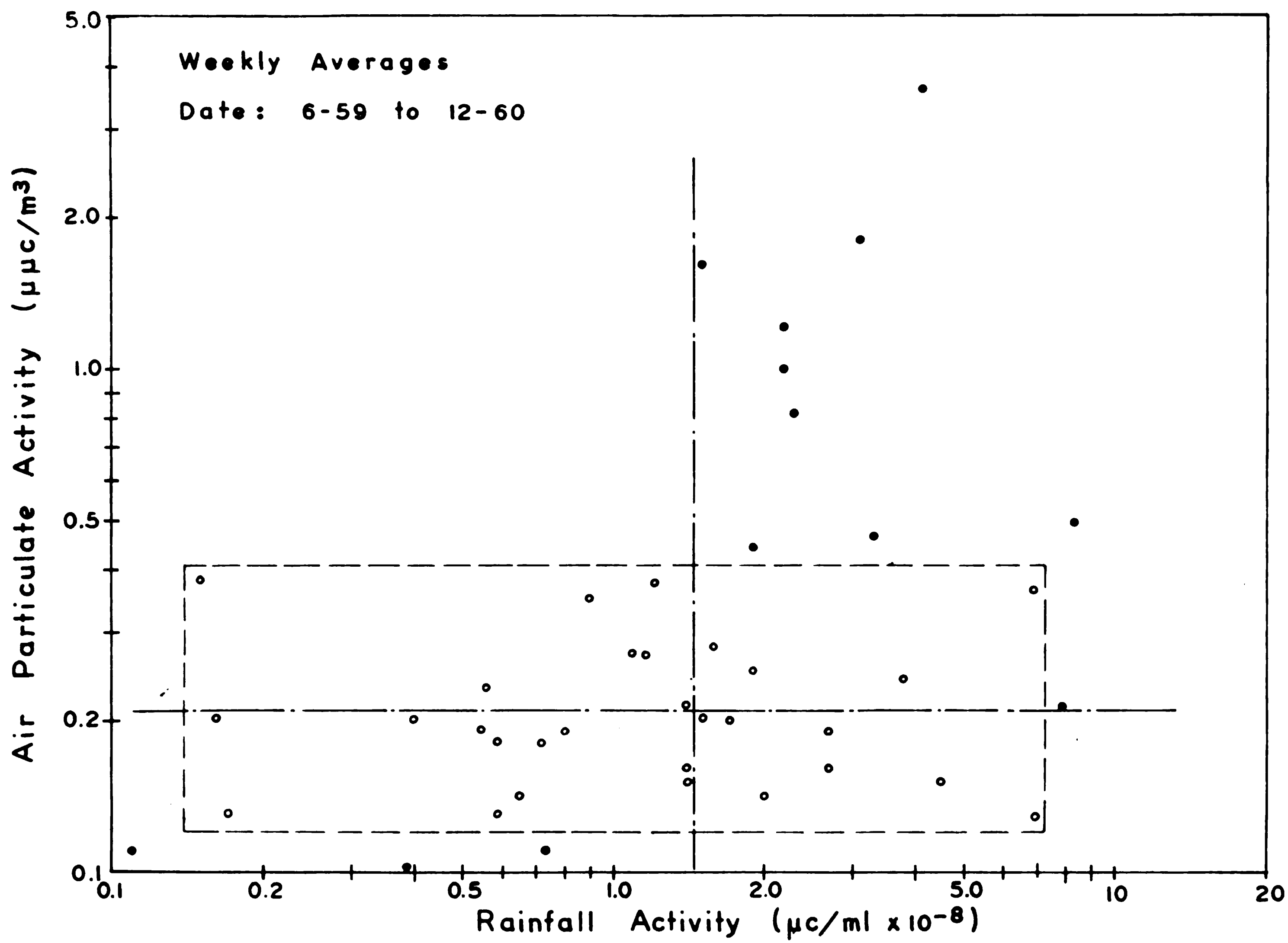


Figure II. Scatter diagram of air particulate activity versus rainfall activity for Station I at Carleton, Michigan.

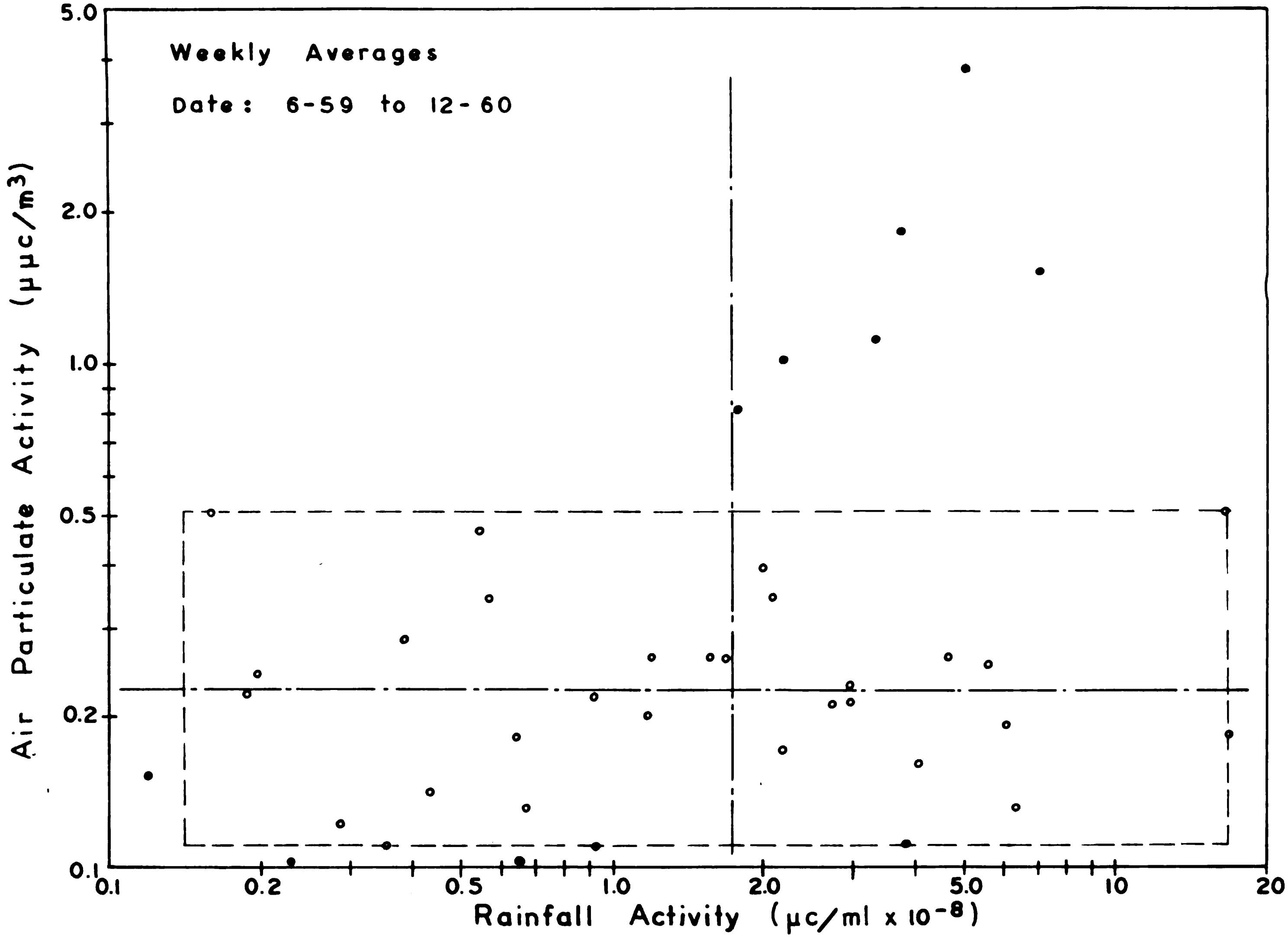


Figure 12. Scatter diagram of air particulate activity versus rainfall activity for Station 2 at Monroe, Michigan.

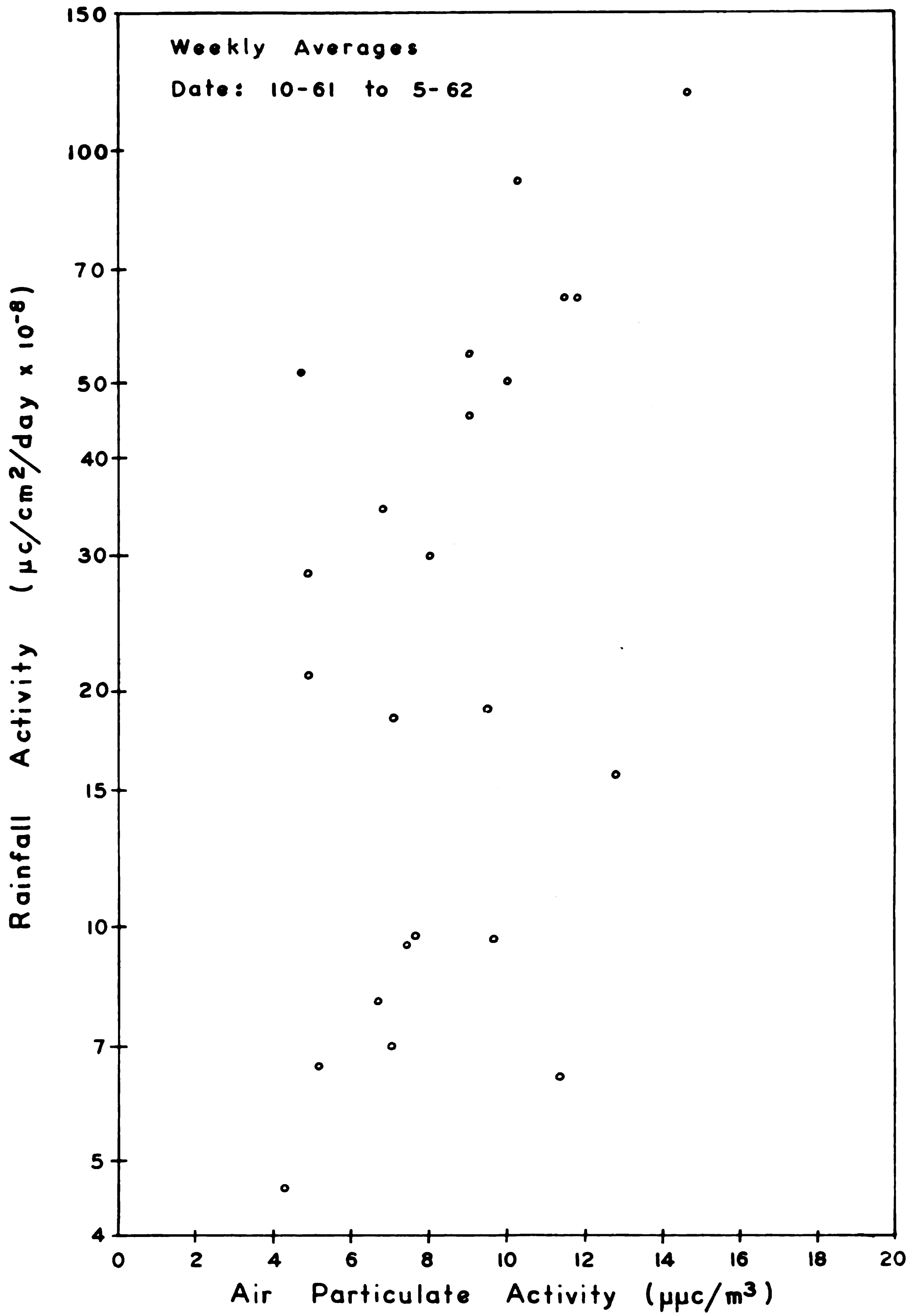


Figure 13. Scatter diagram of air particulate activity versus rainfall activity for Lansing, Michigan.

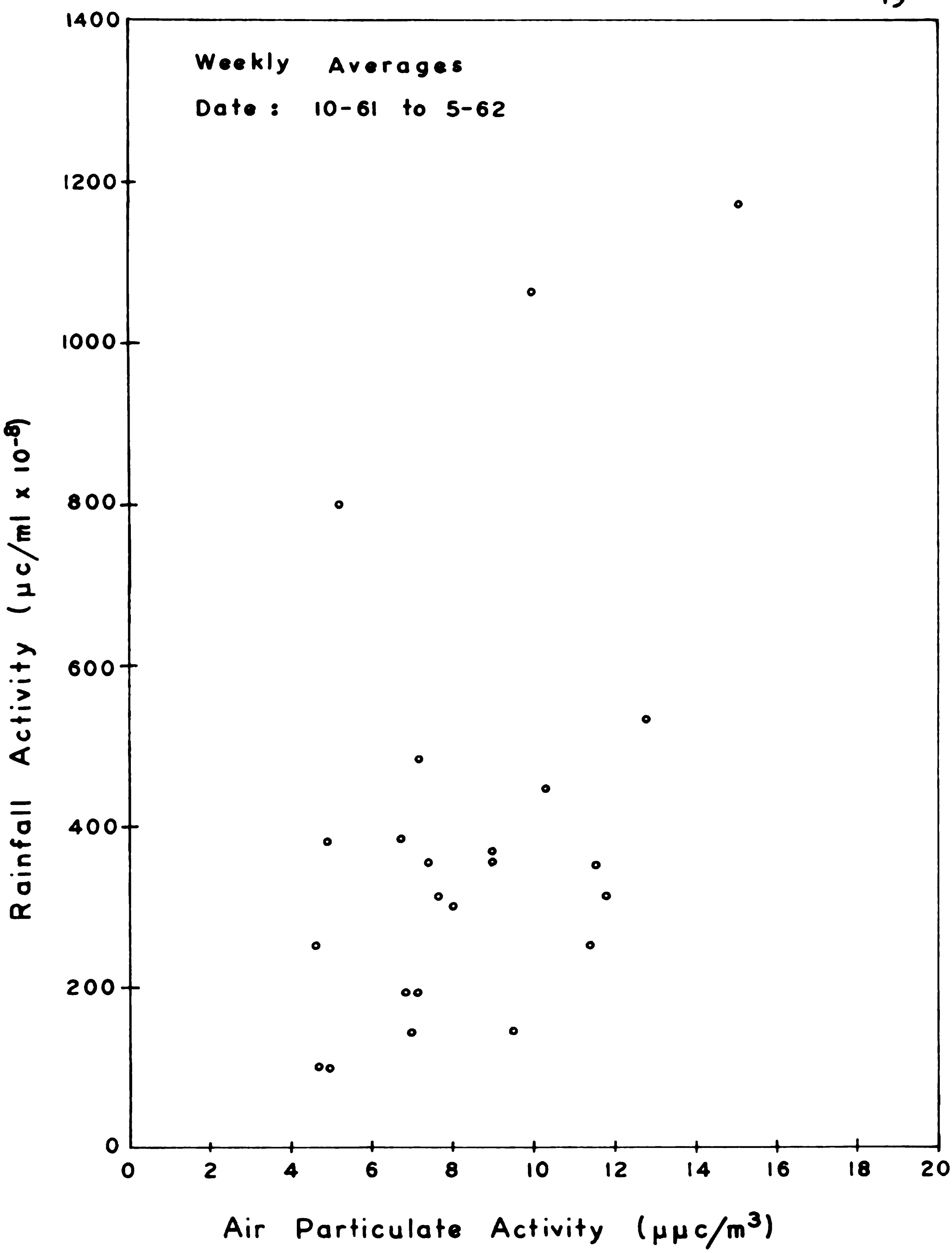


Figure 14. Scatter diagram of air particulate activity versus rainfall activity for Lansing, Michigan.

Table 1. Radioactivity data used in analysis from Station 1 at Carleton, Michigan.

Sampling Week	Air Particulate Activity (μμc/m³)	Rainfall Activity (μc/cm²/day)	Rainfall Activity (μc/ml)
39	3.6	2.7	4.10
40		2.3	53.0
41		1.6	8.0
42	1.6	0.39	1.4
43	1.8	0.34	3.1
44	1.2	0.64	2.2
45	0.94		
46	1.0	0.42	2.2
47	0.81	0.58	2.3
48	0.46	0.10	3.3
49	0.20	0.09	0.16
50	0.23	0.18	0.56
51	0.56		
52	0.36	0.20	6.9
53	0.37		
54	0.24	1.9	3.8
55	0.16	0.12	0.14
56	0.11	0.10	0.11
57			
58	0.12	0.10	0.59
59	0.14	0.41	2.0
60	0.18	0.28	0.72
61	0.11	0.42	0.73
62	0.19		
63	0.13	0.71	7.0
64	0.07	0.11	0.85
65	0.14	0.26	0.65
66	0.08	0.57	14.0
67	0.10	0.12	0.39
82	0.21	0.27	7.9
83	0.15	1.10	4.5
84	0.49	0.38	8.3
85	0.20	0.57	1.7
86	0.27	0.14	1.1
87	0.20	0.33	1.5
88	0.16	0.78	2.7
89	0.28	0.34	1.6
90	0.21	0.39	1.4

Table 1. (continued)

Sampling Week	Air Particulate Activity ($\mu\mu c/m^3$)	Rainfall Activity ($\mu c/cm^2/day$)	Rainfall Activity ($\mu c/ml$)
91	0.27	0.64	1.10
92	0.15	1.10	1.40
93	0.44	0.43	1.90
94	0.13	0.11	0.17
95	0.41		
96	0.38	0.09	0.15
97	0.35	0.49	0.90
98	0.37		
99	0.18		
100	0.18	0.16	0.59
101	0.19	0.25	0.80
102	0.25		
103	0.19	0.09	2.7
104	0.37	0.08	1.2
105	0.26		
106	0.25	1.20	1.9
107	0.20		
108	0.45		
109	0.21	0.12	0.40
110	0.09		
111	0.19	0.15	0.55

Table 2. Radioactivity data used in analysis from Station 2 at Monroe, Michigan.

Sampling Week	Air Particulate Activity (μμc/m³)	Rainfall Activity (μc/cm²/day)	Rainfall Activity (μc/ml)
39	3.8	1.5	5.1
40	2.5		
41	1.5	2.2	7.2
42	1.6		
43	1.8	0.31	3.8
44	1.1	0.66	3.4
45	1.1		
46	1.0	0.12	2.2
47	0.8	0.37	1.8
48	0.5	0.63	0.16
49	0.24	0.10	0.20
50	0.28	0.12	0.39
51	0.43		
52	0.40		
53	0.35		
54	0.19	1.8	6.1
55	0.15	0.11	0.12
56	0.10	0.23	0.23
57	0.18	0.48	17.0
58	0.12		
59	0.14	0.82	0.44
60	0.14	0.20	0.61
61	0.09	0.28	0.66
62			
63	0.11	0.49	3.9
64	0.06	0.08	0.58
65	0.05	0.24	0.52
66	0.05	0.26	9.1
67	0.13	0.10	0.68
68	0.06	0.82	1.8
82	0.21	0.10	3.0
83	0.16	0.82	4.2
84	0.50	0.60	17.0
85	0.21	0.80	2.8
86	0.25	0.89	5.7
87	0.26	0.40	1.6
88	0.17	0.74	2.2
89	0.20	0.31	1.2
90	0.26	0.48	1.7

Table 2. (continued)

Sampling Week	Air Particulate Activity (μμc/m³)	Rainfall Activity (μc/cm²/day)	Rainfall Activity (μc/ml)
91	0.26	0.53	1.2
92	0.18	0.61	0.65
93	0.39	0.73	2.0
94	0.12	0.21	0.29
95	0.44		
96	0.42		
97	0.34	0.24	2.1
98	0.40		
99	0.22	0.08	0.19
100	0.11	0.10	0.36
101	0.11	0.16	0.94
102	0.13	0.37	6.4
103	0.07	0.15	1.8
104	0.16		
105	0.26	0.47	4.7
106	0.23	0.13	3.0
107	0.23		
108		0.13	8.9
109	0.27		
110	0.10		
111	0.23	0.22	0.93
112	0.08	0.24	2.0
113	0.46	0.08	0.55
114	0.34	0.11	0.58

Table 3. Rainfall data used in the analysis from Monroe, Michigan.

Sampling Week	Rainfall (inches)	Sampling Week	Rainfall (inches)
39	0.75	83	0.52
40	0.02	84	0.10
41	2.97	85	0.73
42	0.99	86	0.41
43	0.35	87	0.56
44	0.39	88	0.99
45	0.33	89	0.64
46	0.14	90	--
47	0.64	91	--
48	0.78	92	--
49	2.24	93	--
50	0.72	94	--
51	2.22	95	Trace
52	Trace	96	0.36
53	0.04	97	0.09
54	0.84	98	0.04
55	2.02	99	0.98
56	2.60	100	0.28
57	0.07	101	0.50
58	0.50	102	0.93
59	0.52	103	0.03
60	0.79	104	Trace
61	1.40	105	1.64
62	0.04	106	0.43
63	0.51	107	0.15
64	0.28	108	0.03
65	1.13	109	0.05
66	0.17	110	0.16
67	0.58	111	0.68
68	0.27	112	0.12
		113	0.40
82	0.07	114	0.39

Table 4. Radioactivity data used in analysis from Lansing, Michigan.

Sampling Week	Air Particulate Activity (μμc/m³)	Rainfall Activity (μc/cm²/day)	Rainfall Activity (μc/ml)
10/29/61	10.3	90.2	444
11/ 5/61	8.8	--	--
11/12/61	15.1	116.8	1170
11/19/61	11.5	64.2	350
11/26/61	9.6	--	--
12/ 3/61	10.0	50.2	1060
12/10/61	5.3	--	--
12/17/61	8.2	--	--
12/24/61	6.7	8.0	380
12/31/61	9.0	54.2	350
1/ 7/62	9.0	45.5	364
1/14/62	11.8	64.2	310
1/21/62	9.5	19.0	143
1/28/62	7.0	7.0	140
2/ 4/62	8.5	--	--
2/11/62	12.8	15.7	530
2/18/62	6.8	34.2	190
2/25/62	7.1	18.5	190
3/ 4/62	8.0	--	--
3/11/62	5.2	6.6	798
3/18/62	7.4	9.4	350
3/25/62	11.4	6.4	250
4/ 1/62	8.0	30.0	300
4/ 8/62	4.9	21.0	98
4/15/62	4.6	4.3	250
4/22/62	7.2	9.6	480
4/29/62	4.7	51.4	99
5/ 6/62	4.9	28.2	380
5/13/62	7.6	9.7	310

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