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Final Report

RAIN SCAVENGING OF PARTICULATE MATTER FROM THE ATMOSPHERE

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ABSTRACT

Data relating to rain cleansing of the atmosphere have been gathered throughout six different rain-producing weather systems and analyzed with respect to the concentrations of radioactivity in successive samples. For some of the rains, analysis of the pollen content of sequential samples was also made. The variations of the respective contamination levels of rain samples within and between storms are examined in relation to several existing hypothetical relationships set forth in literature. Verification of the projected hypothesis fails in most cases, and appears in others to be fortuitous. It is concluded that more specific attention to the physical processes of scavenging during cloud- and rain-drop formation, and to the effects of accretion, coalescence and evaporation in the fall of the raindrops will be fruitful.

Three categories of identifiable contamination are noted (1) old radioactive debris; (2) fresh radioactive debris and (3) airborne pollens. These are observed to have differing source regions, and their study is expected to clarify details of the scavenging processes and of rainstorm dynamics. A model for rain cleansing by convective systems is proposed after consideration of the convective storm models of Newton (1950) and of Browning and Ludlam (1962) in relation to the observed variations of the various kinds of contamination.

I. INTRODUCTION

Since radioactive fallout became a problem, a large number of people (Bleichrodt, et al., 1959; Storebö, 1959; Murayama, 1961; Salter, et al., 1962; Huff, 1963) have assayed samples of various kinds and attempted to relate their findings to meteorological parameters. Another group of people (Chamberlain, 1953; Greenfield, 1957; McCully, 1956; Pemberton, 1960; Goldsmith, et al., 1963) have attempted to devise mathematical models of the physical processes deemed important in bringing the radioactive contaminants to earth. In general, a number of hypotheses have been projected from both the experimental and theoretical approaches, and relatively few data suitable to test these hypotheses have been collected.

Whereas the work reported herein was most particularly designed to observe and evaluate the scavenging effectiveness of rains of different character, the data we have gathered in the course of our work appear to give new insights into other aspects of the problem as well.

II. OBSERVATIONAL PROGRAM

The entire theme of our research to data has been that more specific, better defined, and new kinds of data are needed to improve understanding of the rain scavenging function. A basic premise has been that any information bearing on rain scavenging, whether of radioactive or of other material, is pertinent to the problem and probably useful in its solution. In this vein specifically, we have paid attention to the rain scavenging of airborne pollens whose source, distribution, and physical properties can be defined quite well.

A. SITE DESCRIPTION

The site of the Willow Run Meteorological Field Station is an open expanse of flat land providing a long wind fetch over grass. The resulting freedom from low level obstructions and the turbulence they cause in the surface wind field is considered important in terms of (1) the distribution of low level contaminants and (2) the effects of the surface winds upon the raindrop-size distribution and the transport of splash-generated spray droplets.

Figure 1 shows the distribution of instruments and buildings near the sampling site. The central location of the sampling units (raindrop sorter, large sampling pans) is clear. Three standard rain gages are located 100 yards away respectively to the north, south and east. A tipping bucket rain gage is placed 50 yards east of the sorter house, and its recorder is located in the small building adjacent to the sorter. Building 8, west of the sorter house, is a permanent structure which houses most of the laboratory and clerical functions of the project and the zenith-pointing radar (under development), and serves as a source of water for cleaning the sampling devices.

B. SPECIAL INSTRUMENTATION

The aerodynamic raindrop sorter has been described by Dingle (1961a). Photographs of the instrument as it was assembled at the Willow Run Field Site are shown in Figure 2. The design is essentially a low speed wind tunnel the working section of which slopes upward at an angle of 45°. The latter criterion was determined by design studies which have been presented by Dingle and Brock (1960). To prevent random air flow fluctuations through the working section, it is obviously necessary to isolate the air intake and exhaust from the free air stream. To do this the housing shown in Figure 3

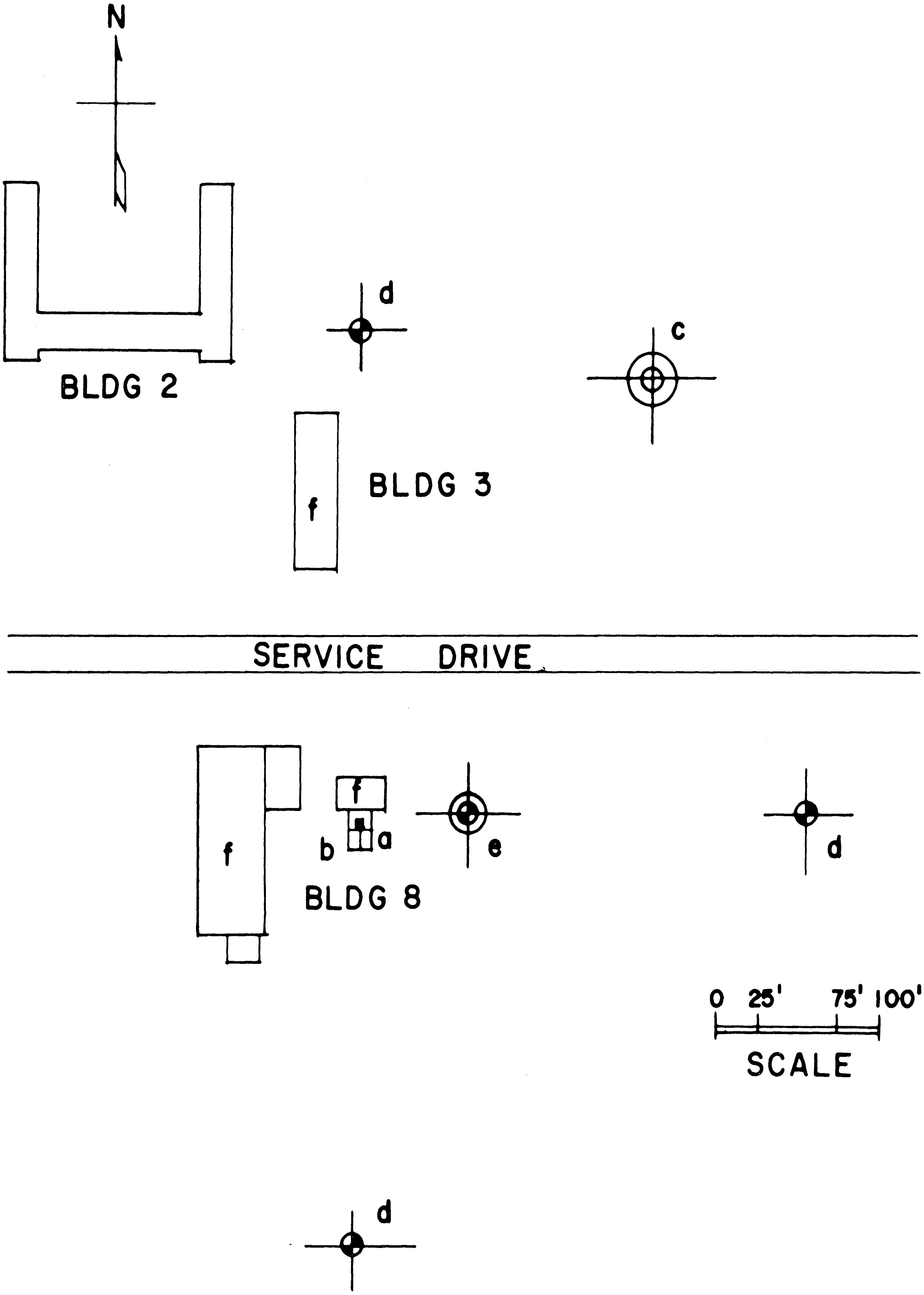

Figure 1. Schematic map showing relative locations of (a) the raindrop sorter, (b) the large sampling pans, (c) the raindrop-size spectrometer station, (d) the three standard rain gages, (e) the tipping bucket gage, and (f) the adjacent buildings.

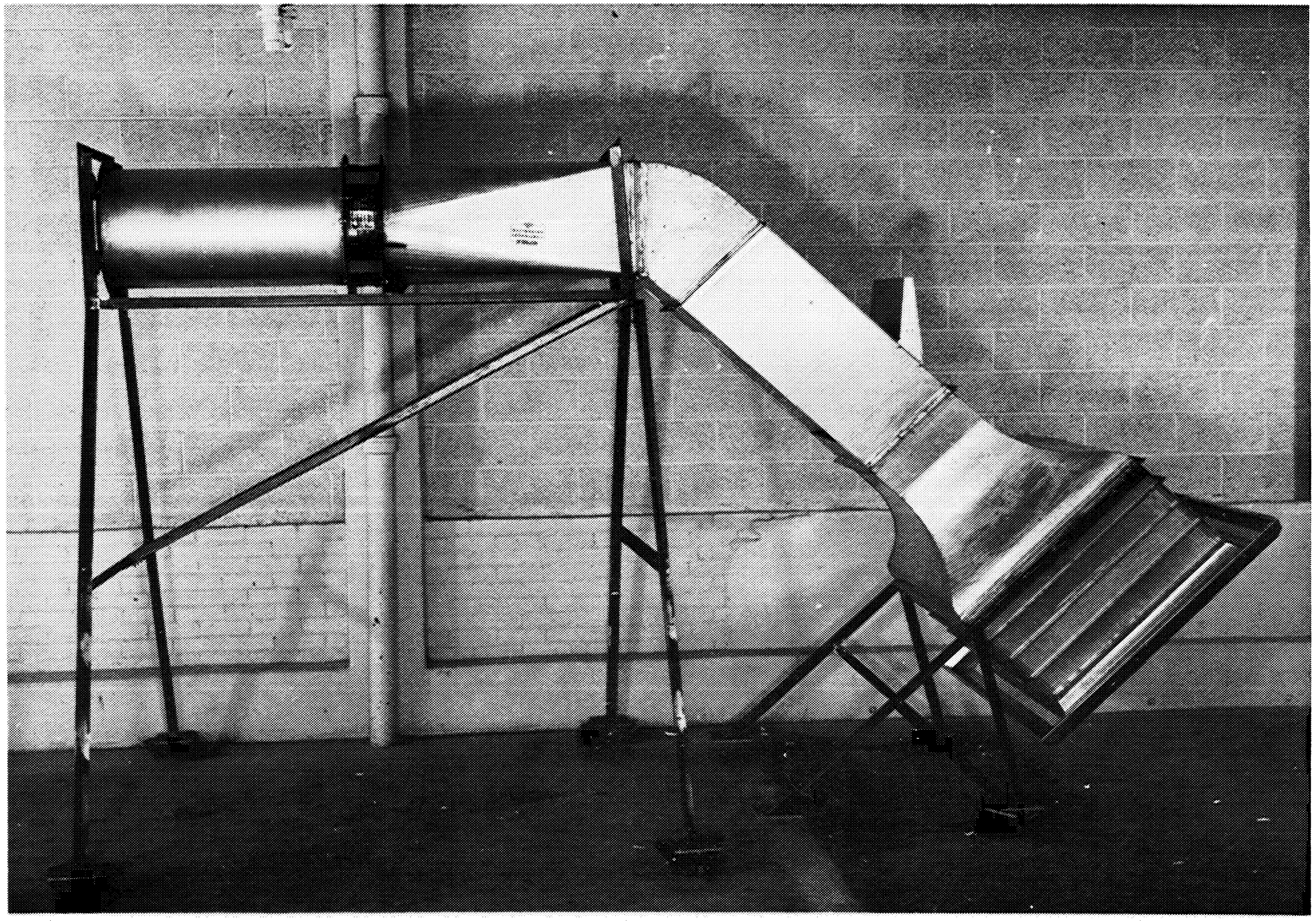

Figure 2. The aerodynamic raindrop sorter, assembled view.

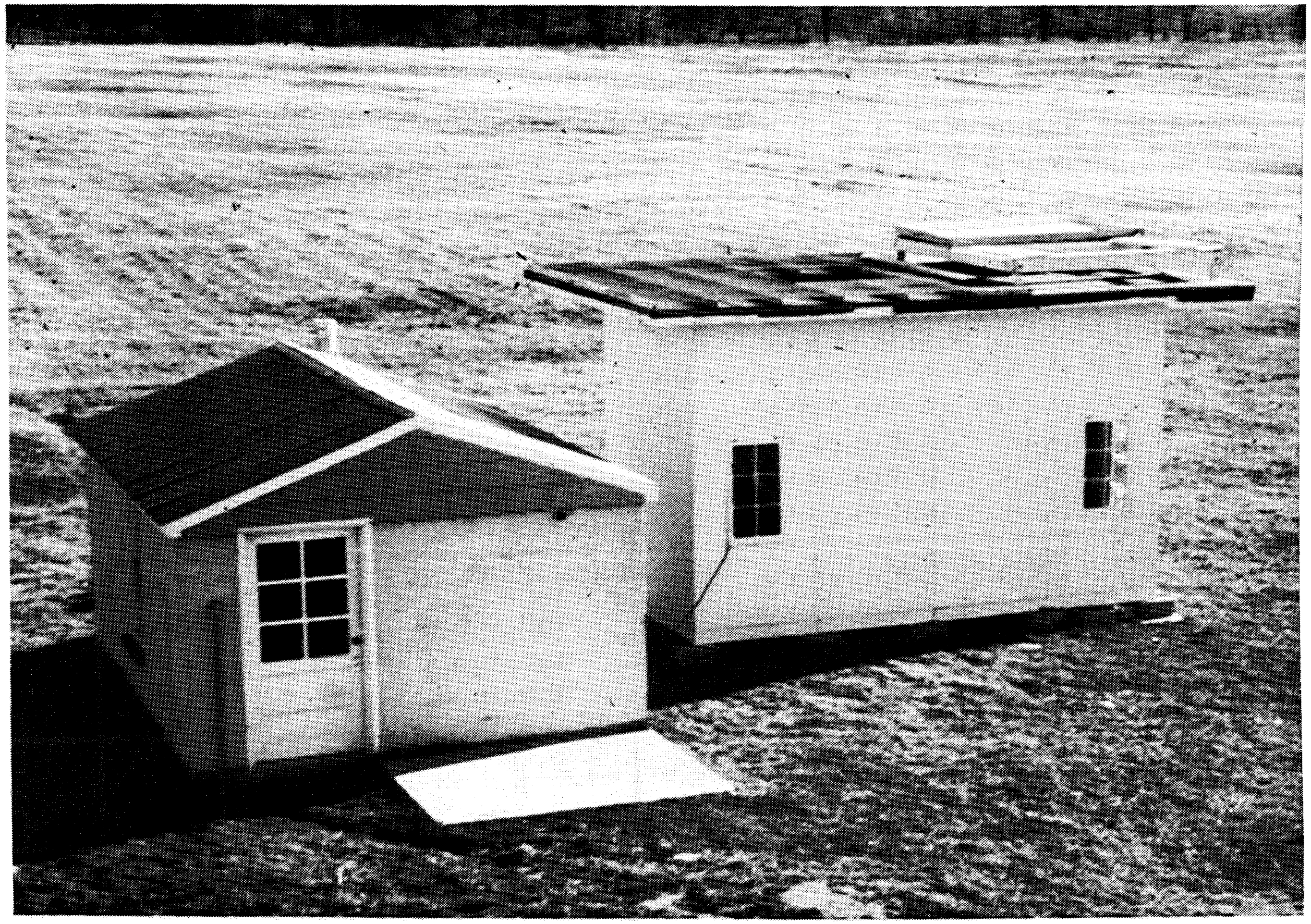

Figure 3. Housing for the raindrop sorter.

was constructed.

The large rain sampling pans, which together total 5.2m^2 in area, have also been described in the above reference (Dingle, 1961a). These are located on the roof of the raindrop sorter housing so as to be as near its rain intake as possible, and to provide for reasonable convenience of the personnel attending the sampling units.

C. SAMPLING PROCEDURE

1. The Raindrop Sorter

The inside surfaces of the sampling section of the raindrop sorter were scrubbed with soap and water and rinsed with tap water before each rain. In an effort to avoid errors in the measurement of sample volumes, the fan was run for several minutes to remove any excess rinse water. One-quart polyethylene sample bottles were placed under the drains from the collection troughs for the five drop-size fractions, and the fan was turned on when the first drops reached the site. Because the sorting is sensitive to changes of the wind, an attempt was made to change samples in accordance with significant changes in the wind direction or speed, so as to maintain a more or less constant distribution of drop sizes in each series of samples. The function of the sorter was to procure rain fractions separated according to drop size. The sample volume requirement and the intake limitation made it necessary to forego any time-based division of these samples.

2. The Large Sampling Pans

Before each rain, the pans were scrubbed to remove all dry deposition. This technique was found sufficient to remove nearly all the radioactive fallout from the pans, and it can be assumed to have had the same effect for pollen grains. For rains in which both radionuclides and pollens were determined, the usual procedure was to take alternate samples for radiochemical and pollen analysis, at least during the early and middle parts of each rain. In most, but not all, cases, 1-gallon samples were taken for radiochemical analysis and 1-quart samples were taken for pollen analysis. When radionuclides alone were to be determined, successive 1-gallon samples were taken.

In the absence of better guide lines, it was planned to sample as many rains as possible early in the program. The amount of each sample was determined by the level of atmospheric contamination by radioactive materials, and the sensitivity of the low background beta counter. In the summer of 1961 it was decided that a minimum of 1-gallon was necessary. After atmospheric tests were resumed, it became possible to reduce this amount or to continue the 1-gallon samples and analyze them in more detail. The latter course was chosen.

D. COMPLEMENTARY DATA

In addition to data obtained from the radiochemical analyses of rain samples, supporting data on the nature of the precipitation system which produced the rain were taken at the collection station and acquired elsewhere. These additional data include conventional synoptic weather data and detailed observations of height of cloud bases, and height of radar echo tops.

Rainfall rate data were obtained in two independent ways. The first of these was by the tipping-bucket recording raingage. This instrument records every 0.01 in. of rain as a mark on a strip chart recorder. Knowing the chart speed, the average rainfall rate between successive marks on the chart was computed and assigned to the time at the midpoint between the marks. A detailed graph of rainfall rate versus time was thus prepared for each rain. These are shown, together with other data for the individual rains, in Figures 4 through 10. In all but the lightest rains, the data points are sufficiently close together in time that the record may be thought of as continuous. The rain of May 2, 1962, was of very low intensity; since the rainfall amount marks were not sufficiently frequent to indicate continuous rain, the data are presented in Figure 7 in the form of a bar graph. Here the height of each bar represents the average intensity over the period indicated by the width of the bar.

Average rainfall rate was also computed from the volume and collection time for each sample analyzed. As a check upon these relatively crude measurements, average rainfall rates were computed for the same samples by averaging the intensities obtained from the tipping-bucket rain gage during the time the sample was collected. Except during periods of light or intermittent rain, the two methods were in good agreement. However, in order to eliminate the effects of the lag of rain water in the collection pans, the intensities derived from the sample data were used in nearly every case.

Rainfall rate data were used in two different ways. First was to provide the detailed time record of rainfall rate. The variations observed in these records indicate the basic character of the rain and help to resolve the fine structure of the rain system. Second was to study the relationship between average rainfall rate and radionuclide concentrations in samples taken in sequence during the passage of each rain system. This study is discussed later.

Data on cloud bases were obtained from the regular, special, and local observations taken at the U.S. Weather Bureau station at Willow Run Airport (YIP), located approximately 1 n mile west of the sampling station. The heights were measured with a ceilometer or estimated by Weather Bureau personnel. These data have been plotted as parts of Figures 4 through 10. The solid circles indicate the base of the lowest layer of cloud, whether it was scattered, broken, or unbroken. Whenever the height of the ceiling* was not identical with that of the lowest clouds, the ceiling height is indicated by an "x." Some error is to be expected in using measurements made 1 n mile away to portray conditions at the collection site. However, the error should be small during rains of the large scale uplift type, for which it has been suggested (Salter, Kruger, and Hosler, 1961, 1962), that the cloud base may be an index of the physical factors that control the concentration of radionuclides in rain water.

Maxiumum heights of radar echoes were obtained from the hourly reports of the operator of the WSR-57 radar at the U.S. Weather Bureau station at Detroit Metropolitan Airport (DTW) which is located about 8 n miles east of the sampling station.

Admittedly, radar observations of this type are not well-suited to give detailed information of the radar profile of the cloud producing precipitation at the collecting station. For systems exhibiting variability in the heights of echo tops, the average and maximum heights as given by the Weather Bureau are recorded (Figures 4 and 10). In one case (Figure 4) it appears very likely that a single intense cell noted in an area echo by the radar operator was the one that produced a heavy shower at the collection site. It was found that the Weather Bureau radar observations were best suited to a qualitative classification of the precipitation system (i.e., squall line, stratiform area, etc.), and this information has been incorporated into the description of each rain.

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*Defined by the Glossary of Meteorology as the height ascribed to the lowest layer of clouds or obscuring phenomena when it is reported as broken, overcast, or obscuration, and not classified as "thin" or "partial."

III. RADIOCHEMICAL ANALYSIS

The radiochemical analysis procedure consisted of the following steps. First, the sample was pumped through a graduated series of filters to remove insoluble particles. The filtrate was reduced by evaporation and the residue ultimately transferred to a stainless steel planchet, which was then counted for radioactivity. The filters were dried, placed into identical planchets, and similarly counted. Early in the program, the samples were counted for gross beta activity only using a low-background (1 cpm) beta counter. After the resumption of atmospheric testing by the USSR in September, 1961, activity levels rose sufficiently to permit determination of individual radionuclides by gamma-spectrometry.

A. FILTRATION

The filtering procedure has been described in an earlier report (Dingle, 1961b) and will not be discussed here. Basically, there were initially three reasons for filtering the rainwater. First of all, it was desired to reduce the amount of solids in the planchet so as to minimize the self-absorption of beta radiation by the sample. Secondly, it was thought that this procedure would yield information about the solubilities of some of the fission product radionuclides in rain water. This information was of interest to the National Sanitation Foundation, the agency through whose cooperation the radiochemical analyses were made. Thirdly, it was hoped that such a procedure would yield at least qualitative information regarding the size spectrum of the atmospheric particles collected by the rain water.

It became apparent during the course of the study that the filtration process would yield little or no information as to the size spectrum of the radioactive particles. Agglomeration of the particles with each other and attachment to the walls of the polyethylene sample bottles took place very rapidly, and it was evident that the size spectrum "seen" by the filter train was not the same as the one existing in the rain water immediately after collection. Some information on solubility was obtained but this, too, was limited by the discovery that there was appreciable adsorption of soluble zirconium on the filters. In a series of experiments in which a known amount of soluble zirconium-95 was filtered, it was found that an average of about 35% of the soluble zirconium-95 was adsorbed. Moreover, the variance of the percent adsorbed was such as to indicate that the process was not a reproducible one, so that no suitable correction for the effect was possible.

Although the filtering technique proved effective in reducing the amount of solids in the planchet, the value of obtaining gross beta activities in addition to activities of individual radionuclides (by gamma spectrometry) is somewhat questionable. The gross beta determinations could probably be eliminated. Self-absorption of gamma rays is insignificant for the amount of solid residue obtained from evaporating four liters of rain water; therefore filtration is not necessary in an analysis program in which determination of radioactivity is done by gamma spectrometry.

B. ANALYSIS

In each rain water sample, fission-product radionuclides were determined in three groups:

(1) Ce^{141}, $Ce^{144} + Pr^{144}$
(2) Ru^{103}, $Ru^{106} + Rh^{106}$
(3) $Zr^{95} + Nb^{95}$

In addition, $Ba^{140} + La^{140}$ were determined when fallout was fresh. In the groupings above, the plus signs indicate parent-daughter relationships, the parent being given first and the daughter second. The reasons for grouping the radionuclides are made clear in the discussion to follow.

A gamma scintillation spectrometer gives the counting rate (counts per unit time) as a function of the energy of the gamma photon lost to the scintillation crystal. The gamma spectrum of an individual gamma-emitting radionuclide shows peaks (called photo-peaks) corresponding to the energies of the several gamma rays emitted during decay of the radioactive atom. The energy at which the peaks occur is characteristic of each radionuclide and may be used in its identification. Quantitative determination is possible through the measurement of the area under the photo-peak after calibration with a standard of known activity.

Gamma spectrometry is especially well-suited to the determination of radionuclide mixtures since it makes separations unnecessary in many cases. In principle, each photopeak in the spectrum of an unknown may be assigned to a specific radionuclide, but in practive it is sometimes found that more than one nuclide contributes to an individual peak because the energies of gamma rays from several nuclides are too close together to be separately resolved by the spectrometer. It is this complication which leads to the grouping of several radionuclides, because several of the more plentiful fission-product radionuclides or their daughters have photopeaks which are not resolvable. Table I gives a list of the radionuclides determined and their gamma-ray energies. The figure in parentheses in the right-hand column is the number of gamma rays of the particular energy listed which are emitted per 100 disintegrations. When expressed as a percent, this figure is known as the "percent yield." The word "none" in the right-hand column merely indicates that no gamma rays from the particular radionuclide are counted, not necessarily that none are emitted.

TABLE I

RADIONUCLIDES DETERMINED BY GAMMA SPECTROMETRY
AND THEIR GAMMA ENERGY LEVELS

Radionuclide Group	Individual Radionuclide	Energy of Gamma Rays Counted (Mev)
$Zr^{95} + Nb^{95}$	Zr^{95} Nb^{95}	0.717 (98) 0.745 (100)
Ru^{103}, $Ru^{106} + Rh^{106}$	Ru^{103} Ru^{106} Rh^{106}	0.498 (90) none 0.516 (21)
Ce^{141}, $Ce^{144} + Pr^{144}$	Ce^{141} Ce^{144} Pr^{144}	0.145 (43) 0.134 (10.5) none
$Ba^{140} + La^{140}$	Ba^{140} La^{140}	none 1.596 (94)

Zirconium-95 and its daughter niobium-95 emit gamma rays too close together in energy to be resolved by the spectrometer. Furthermore, since the half-life of zirconium-95 is only about twice that of niobium-95 (65 days vs. 35 days), it is not possible to assume that parent and daughter are in equilibrium until many months after formation. In the general case, the percent yields of two gamma rays contributing to a single peak are not equal. In such a case one must know the relative abundances of the two nuclides contributing to the peak in order to compute the disintegrations per min (dpm) from counts per min (cpm). Fortunately, the counting yields of the zirconium and niobium gamma rays are nearly equal, and are also virtually 100% so that the number of cpm equals that of dpm, to a very close approximation. We are therefore able to determine the total activity of zirconium-95 plus niobium-95, but cannot determine the relative abundances with the gamma spectrometer.

In the group composed of ruthenium-103, ruthenium-106, and its daughter, rhodium-106, the 0.498 Mev gamma ray of ruthenium-103 and the 0.516 Mev gamma ray of rhodium-106 are not resolvable. The 30-sec half-life of rhodium-106 is very short compared to the 1-year half-life of ruthenium-106, so that parent and daughter achieve equilibrium promptly. However, the percent yields of the two gamma rays which contribute to the peak are not equal; thus it is necessary to know the relative abundance of ruthenium-103 with respect to the ruthenium-106, rhodium-106 pair to convert cpm to dpm and to picocuries (pc). To determine this relative abundance, the radiochemical separation method of Butler (1960) was applied to a few of the samples from each rain. The results then give an average relative abundance that can be applied to all samples from that rain.

A small sample-to-sample variation in the relative abundance was observed, so that some error is made by applying an average value to all samples. If the variations are indeed statistically significant, this observation raises interesting questions as to the physical mechanisms responsible for fractionation of isotopes of the same element. The answer very likely lies in the fact that the isotopes are products of different decay chains, but further research in this area will be needed to provide a definite answer. On the basis of known or assumed relative abundance values, it is possible to compute the activity of each radionuclide in this group separately. As mentioned, however, error is made because of uncertainties in the relative abundance values. The percent error is minimized by grouping the three radionuclides as we have done.

The same arguments hold for the group cerium-141 and cerium-144 plus praseodymium-144, where the 0.145 Mev gamma ray of cerium-141 and the 0.134 Mev gamma ray of cerium-144 are not resolvable. In this case the determination of the relative abundance values was done by following the decay of the photo-peak, according to the procedure given by Evans, <u>et</u> <u>al</u>., (1962). As in the case of the ruthenium isotopes, a small sample-to-sample variation of the relative abundance was observed.

For the barium-140, lanthanum-140 parent-daughter pair, the 1.596 Mev gamma ray of lanthanum-140 was counted. The 40.2-hr half-life of lanthanum-140 is short compared to the 12.8-day half-life of barium-140; therefore equilibrium may be assumed in most cases. Then the counting rate of barium-140 will be the same as that for lanthanium-140; for consistency the sum is reported.

C. ERRORS OF GAMMA SPECTROMETRIC DETERMINATIONS

In studies of the mechanisms by which bomb fission products are removed from the air by rain, the <u>precision</u> of the radiochemical analyses is of primary concern, whereas the <u>accuracy</u> is secondary. Precision is a measure of the reproducibility of determination: accuracy is a measure of how closely the determination approximates the "true" value. In a study of the cumulative deposition of a hazardous fission product, for example, one's analysis scheme must be accurate to avoid an incorrect picture of the potential hazard. However in the present case, in which the mechanisms whereby fission products are brought to earth by rain is being investigated, the <u>changes</u> in activity levels with time provide the best clues. Thus, one is more concerned with the magnitude of the random errors of the analysis procedure (precision), which affect each sample in a different way, than with the systematic errors (the term <u>accuracy</u> includes both random and systematic errors), which affect each sample in the same way.

D. FACTORS AFFECTING PRECISION

The error already mentioned, of applying an average relative abundance to each individual sample is a random error. Little is actually known regarding its magnitude, but a standard deviation of somewhat less than ±5% is estimated. Another random error is that due to the statistics of counting. Computation of the standard deviation of a count caused by statistical variations in the rate of decay is normally a simple operation, but the task increases in difficulty very rapidly when one attempts to include the statistical variations of the corrections for Compton scattering. Consequently, no complete statistical analysis of counting errors was done. It is estimated that the standard deviation due to counting statistics would be less than ±10%. Random errors in sample preparation might add another ±5%. Thus, the precision of sample preparation and counting is expected to be approximately ±15 to 20%. This is entirely acceptable in view of the magnitude of observed sample-to-sample variations. Furthermore, this is very likely a conservative estimate. Connally and Leboeuf (1953) found a precision of ±7% (95% confidence) for a series of analyses of a known mixture of three radionuclides. Adding ±5% for random errors of preparation would bring their value to ±12%, so that an estimate of ±15 to 20% is probably quite conservative.

It is shown later that time variations of the several radionuclide groups are largely parallel in nature. Nevertheless, it was felt that random errors would be minimized by computing the total of the measured radionuclides. Therefore, unless specified to the contrary, only the variations of this total are considered in the discussion. For convenience, the "concentration of total measured radionuclides" is sometimes merely referred to as the "concentration."

IV. RESULTS

Seven periods of rainfall sampled between September, 1961, and July, 1962, are described in this section. Of the five September, 1961, rains presented earlier (Dingle, 1961b) three yielded data sets sufficiently complete for further analysis in the present report. The rains occurring on May 2 and July 2-3, 1962, were of the large-scale uplift type. All the others were of the convective type. The rains are discussed in chronological order.

A. THE RAIN OF SEPTEMBER 1, 1961

Surface synoptic conditions at 1300E* and 500 mb conditions at 1900 on September 1, 1961, are shown in Figure 4a. The rain of nearly 2 in. collected on this data was associated with a weak low pressure area which had moved northeastward from the Gulf region and was located in Northwestern Ohio at the time of the rain. The rain took the form of an extended thunderstorm, undoubtedly composed of several individual cells, which began at 1030 and continued until 1312. The lower section of Figure 4 shows the pattern of rainfall rate during the storm. The first 30 min of the rain was from one, or possibly two strong cells. The remainder of the rain was of lower, but rather variable intensity. A clear definition of individual cells is not possible from the rain gage record during this period. No radar are available for this rain because the WSR-57 radar had not yet begun operation at DTW.

Nine samples were analysed radiochemically. The results, in the form of gross beta concentrations, are given in the upper section of Figure 4. This section of the figure also includes data on concentrations of ragweed (ambrosia artemisiifolia) pollen, which were determined for a number of samples. A brief general discussion of the implications regarding cloud dynamics connected with determining a tracer such as ragweed pollen in conjunction with studies of the scavenging of fission product radionuclides is given later. A further study of ragweed pollen concentrations in this particular rain has been published elsewhere (Gatz and Dingle, 1963). Time periods during which the individual samples were collected are shown immediately below the upper section of the figure. The sample numbers of the upper group are followed by a "P," indicating that these are the ones in which pollen was determined. The lower group includes all those samples analyzed radiochemically.

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*All times are Eastern Standard Time.

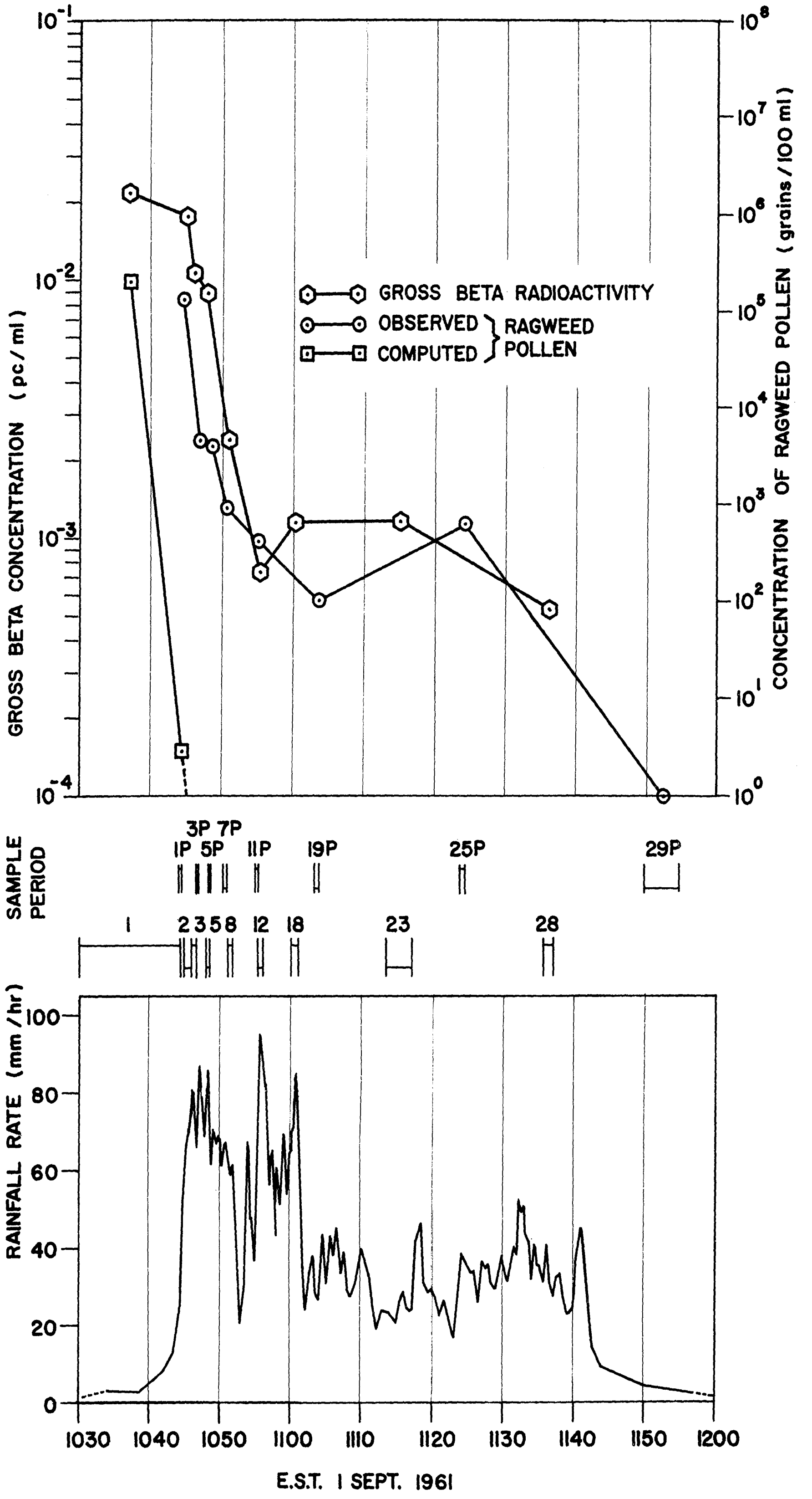

Figure 4. Data for rain of September 1, 1961.

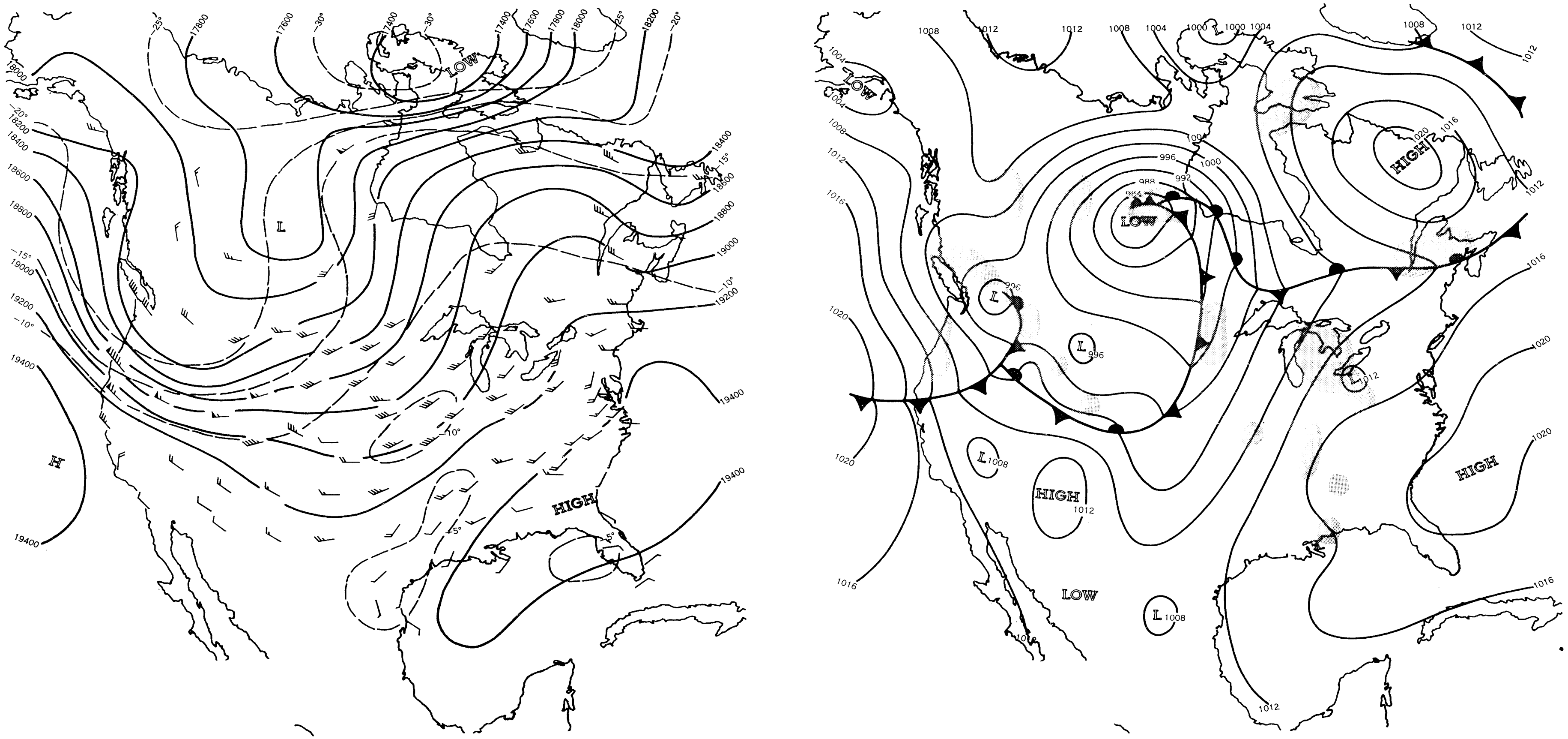

Figure 4a. Synoptic weather data for September 1, 1961.

Because of the lower environmental concentrations of fission product radioactivity prevalent at this time—Russian debris from the fall, 1961, tests did not reach the U.S. for two more weeks—only gross beta radioactivity was determined in this rain.

An outstanding feature of the time curve of gross beta radioactivity is the sharp decrease of concentration during the initial heavy shower. After this initial decrease, concentrations appear to have increased slightly, then remained constant for a time, and then decreased again. Because of the very low concentrations in the rain at this time, however, these variations may not be significantly greater than random errors in analysis and counting.

B. THE RAIN OF SEPTEMBER 23, 1961

Surface synoptic conditions at 1300 and 500 mb conditions at 1900 on September 23, 1961, are shown in Figure 5a. The 0.7 in. of rain collected at this time fell from air behind a rather strong cold front which had passed the collection station 9 to 10 hr before the rain began. The character of the rain was that of a short, heavy thunderstorm followed, after a momentary lull, by a period of light rain and drizzle. The thunderstorm began at 2038, and the drizzle ended about 0000 of September 24. The lower three sections of Figure 5 show the pertinent local meteorological conditions during the rain. The time curve of rainfall rate gives an immediate picture of the course of the rain, showing the initial heavy thunderstorm and the following less intense rain.

Hourly radar reports are obviously too infrequent to document adequately the variations in cloud top, but what information is available seems to show the expected variations. That is, echo-tops were higher during the intense rain and lower during the less intense period. It should be explained that the 40,000 ft height of the echo top marked "cell" at about 2042 represents the height measured at 2000 of what was very likely the same or a closely associated cell. At 2000, the radar at DTW located a strong unbroken precipitation area generally to the west of our station with a 40,000 ft top at 270° and 21 n miles from DTW, near the eastward edge of the area. This would put the strong cell about 13 n miles west of our station. The area including the cell was reported moving east at 25 kt. Moving at the stated speed and in the stated direction, the cell should have been in the vicinity of our station at 2031. YIP reported that thunder began at 2025 and rain began at 2032. Our log stated that rain began about 2038 (our station is 1 n mile east of YIP). The fact that the extrapolated movement of the strong cell would place it near us at very nearly the time when we received our heavy shower tends to identify this strong cell with the one that produced the heavy rain at our station. In addition, it is likely that only a very in-

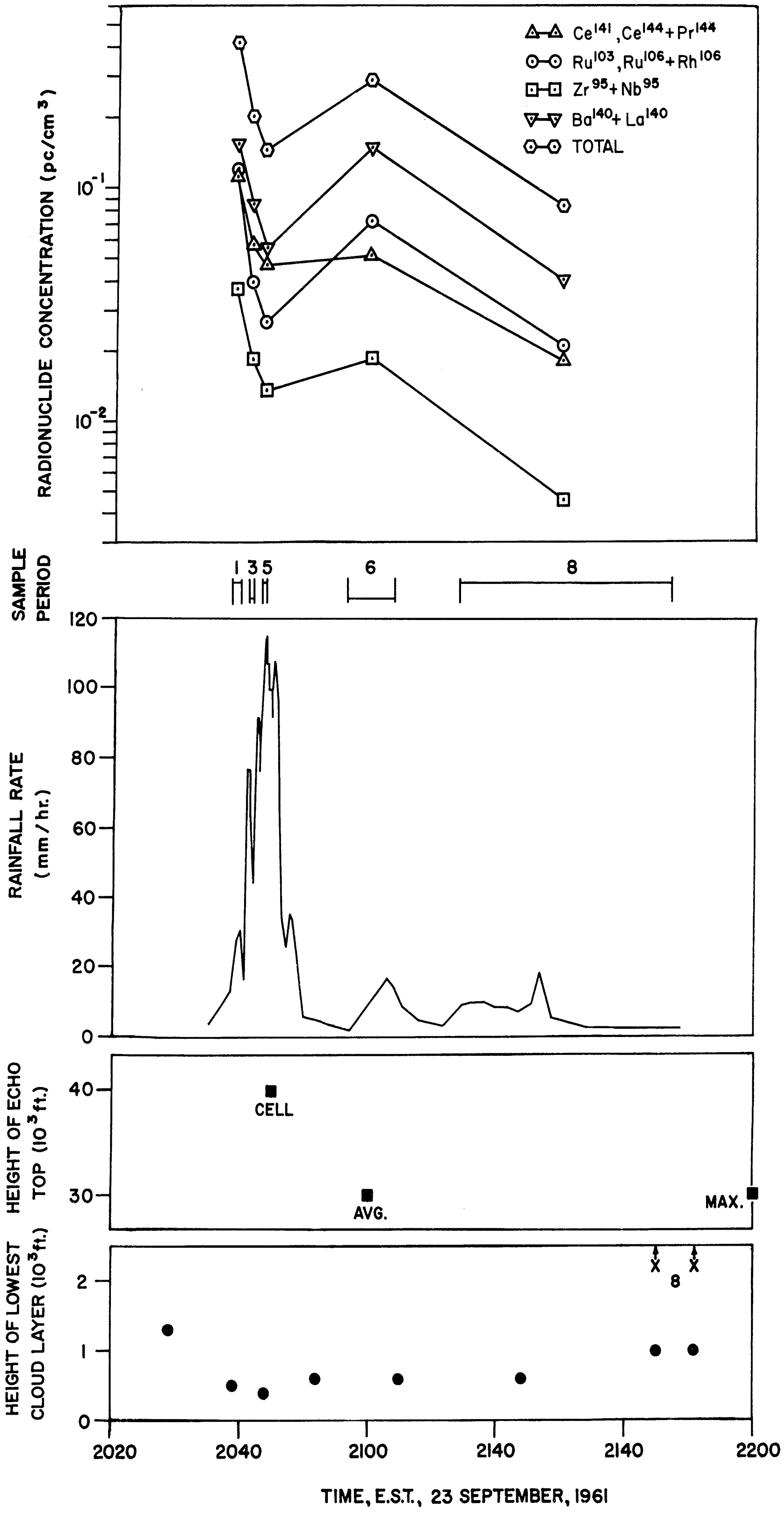

Figure 5. Data for rain of September 23, 1961.

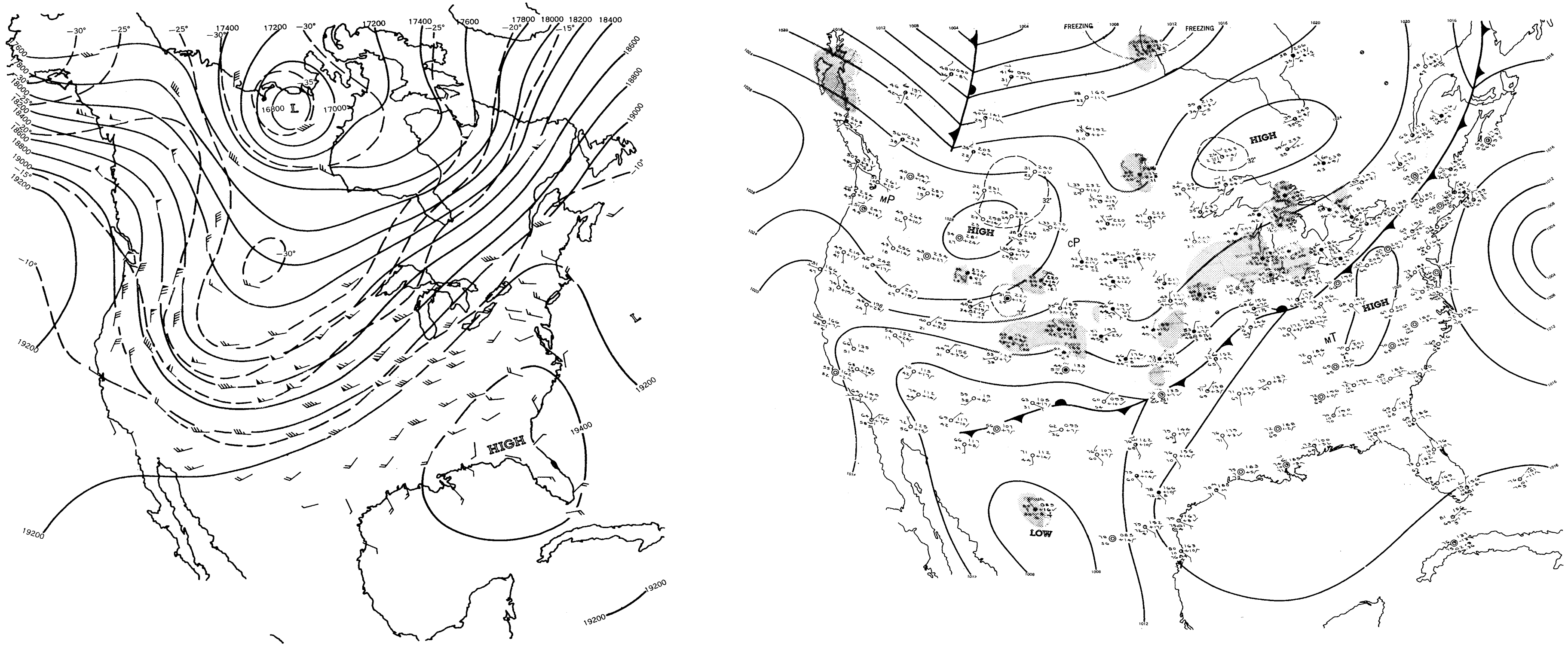

Figure 5a. Synoptic weather data for September 23, 1961.

tense cell, such as this one, could have produced rainfall rates of the magnitudes shown. The stage of development of the cell is not precisely known.

Data on bases of the lowest cloud layer and the ceiling height are given in the bottom section of Figure 5. Ceiling height corresponds to the base of the lowest layer in all but two measurements. During the collection of Samples 1, 3, and 5, the ceiling height fell from 500 to 400 ft. After the heavy shower, the ceiling height rose to 600 ft, where it remained for an hour or more. During this period Sample 6 was collected. An increase in concentration was observed in Sample 6. Between Samples 6 and 8, the concentration decreased rapidly, whereas the ceiling height increased to 8000 ft, and the base of the lowest layer rose to 1000 ft.

Five samples were analyzed radiochemically. The results in the form of concentrations of the radionuclide groups discussed earlier, are given in the upper section of Figure 5. Time periods over which the samples were collected are also shown. This rain marks the first time following the resumption of atmospheric nuclear testing by the USSR on September 1, 1961, that levels of radioactivity in rain collected at our station were high enough so that individual isotope groups could be determined by gamma spectrometry in four-liter samples. It is evident that fallout was quite fresh at this time, from the relatively high levels of barium-140 plus lanthanum-140. The nearly parallel variations of the several groups is quite striking. This characteristic has also been observed by others, as noted earlier, and is also quite evident in the data for the remaining rains presented in this report.

As can be seen from the figure, Samples 1, 2, and 5 were collected from the thunderstorm as the rainfall rate was approaching its peak. A sharp decrease with time from the maximum concentration was observed for each radionuclide group and for their total. Sample 6 included a small intensity peak which probably corresponded to a small cell in the trailing portion of the radar echo area. The concentration of the total measured radionuclides was observed to increase in this sample, followed by another decrease in Sample 8, which consisted of one small rain intensity peak in the midst of continuous light rain and drizzle.

Again, in this rain, as in the case of the rain of September 1, a sharp decrease of concentration with time was observed during a heavy shower. This case is not quite so well documented because no samples were collected during the second half of the intensity peak. Nevertheless, it appears that the sharp decrease of concentration with time may be characteristic of heavy rain from intense thunderstorm cells.

C. THE RAIN OF SEPTEMBER 30, 1961

Surface and 500 mb conditions at 1300 and 1900 respectively on September 30, are shown in Figure 6a. During the period from 1300 on September 30 to 0100 on October 1, a low pressure area moved from central Wisconsin northeastward to a position about halfway between Sault Ste. Marie and Moosonee, drawing a north-south trailing cold front from the Mississippi Valley nearly across Michigan. Our rain was collected from two squall lines, the first of which was approximately 90 n miles in advance of the cold front. The eastward movement of the cold front was accompanied by the eastward movement and deepening of a major long wave trough at 500 mb. A total of 0.38 in. of rain fell during this period.

The squall line yielding the first part of the rain was oriented in a NE-SW direction and moved eastward at 25 kt. Rain began at our station at about 2100. The recording rain gage data show that the rain which fell from this system was of rather widely varying intensity, indicating that this squall line was composed of a series of small weak cells. The showers were accompanied by strong gusty winds. At about 2130 the heavier showers ceased and a period of intermittent light rain was observed until approximately 2225. This rain appeared to originate in an area earlier characterized by radar as a weak stratiform area, decreasing in intensity, behind the first squall line. At 2225 another period of heavier rain began. This rain was from another squall line, noted by radar at 2155 as an unbroken line of moderate intensity, with no change in intensity, approaching the collection station from the southwest at 50 kt. This shower built up to peak intensity within about 4 min of its beginning at the station, then decreased to moderate intensity for 15 min, and then to very light intensity for about 20 min, after which the rain stopped.

Radiochemical results and local meteorological conditions are shown in Figure 6. The highly variable character of the intensity pattern from the first squall line plus the fact that no thunder or lightning was observed at either the collection station or YIP indicate that the first line was not a well developed system. The second line was similarly an association of weak cells and did not yield heavy rain comparable to that observed in the rains of September 1 and 23. Data on variations of the echo tops are again sufficient only to show that tops were generally higher during the heavier rain and lower during the light rain. Ceiling heights remained near 5000 ft except for one observation during the passage of the second squall line. A lower scattered layer was present during the passage of the first line and after passage of the second one.

Radionuclide concentrations, in contrast to those in the two previous rains, remained remarkably constant for most of the period. In this case, the rain was too light to permit resolution of concentration changes during individual intensity peaks. It is apparent from the figure that among

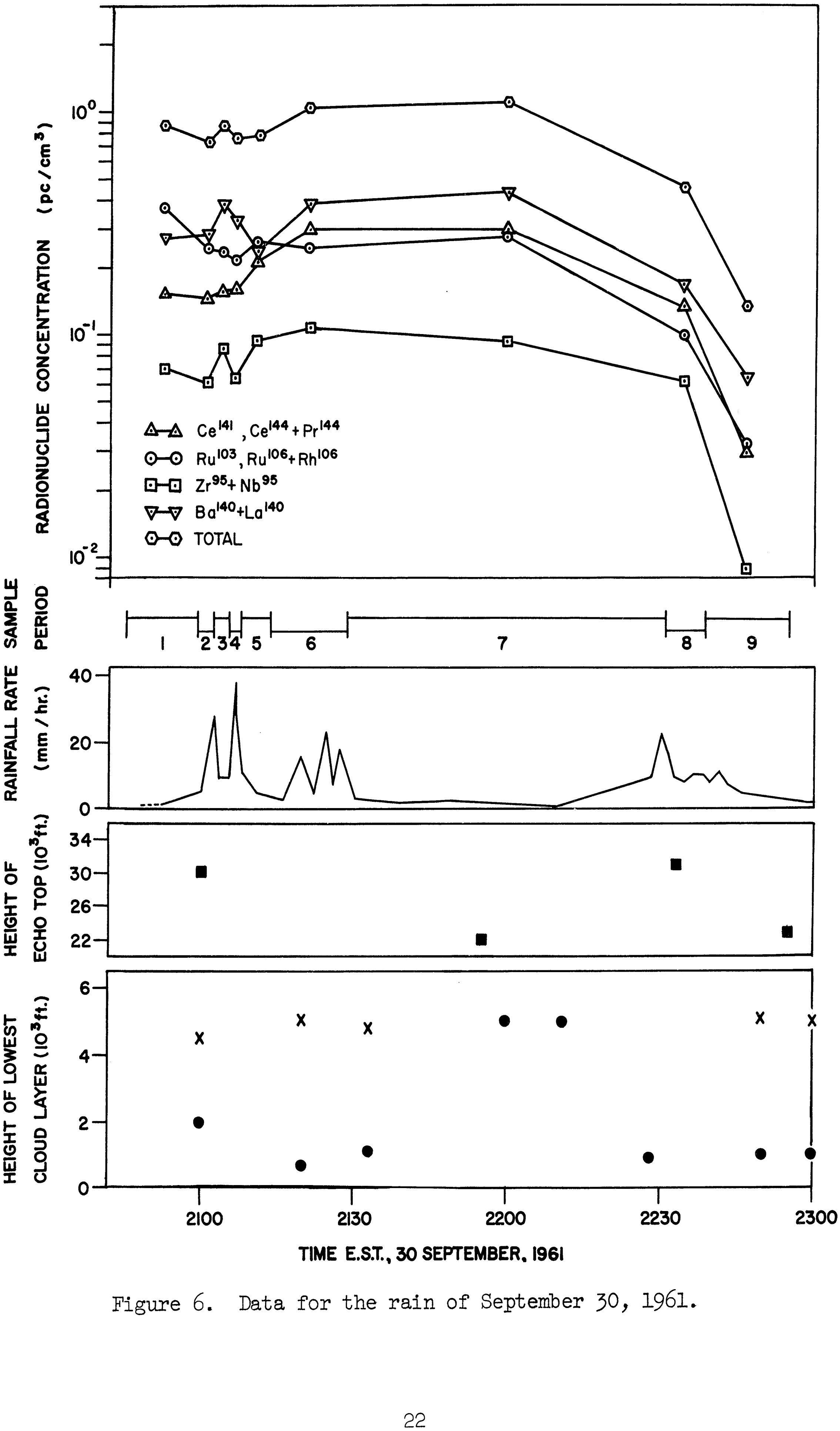

Figure 6. Data for the rain of September 30, 1961.

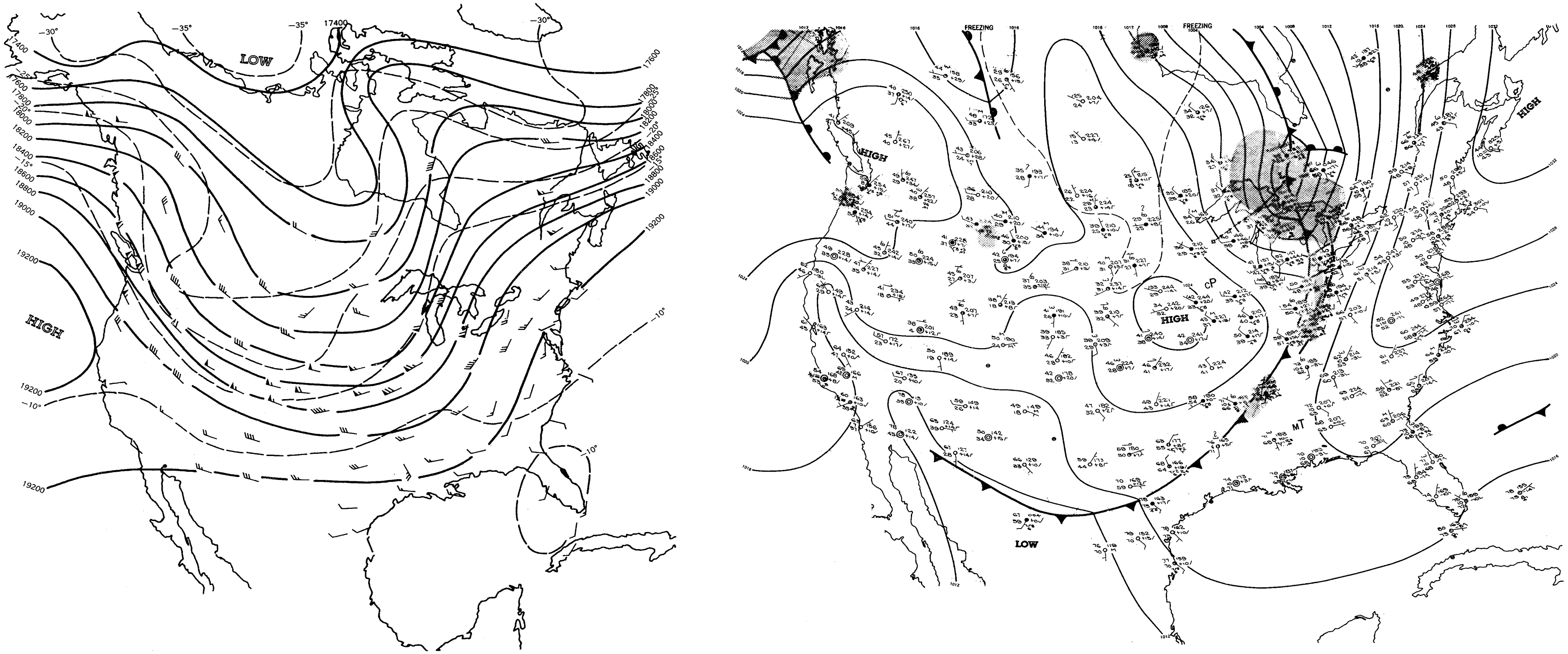

Figure 6a. Synoptic weather data for September 30, 1961.

Samples 1-5, the even-numbered samples represent rain collected during intensity peaks and the odd-numbered samples represent rain collected during periods of low intensity adjacent to the peaks. It is therefore apparent that little change of concentration occurred between cells (in the mean, at least). In addition, concentrations of the individual radionuclide groups appeared to vary independently of each other in Samples 1-5, while exhibiting their usual parallelism in the remaining samples. It is felt, however, that most if not all of the apparently independent behavior is due to random errors of preparation and counting.

Although a substantial portion of Sample 7 came from the second squall line, only Samples 8 and 9 can be considered to contain rain exclusively from this system. Unfortunately these samples cannot be considered representative of individual cells.

Again, echo top data are infrequent.

In summary, rain during this period originated from two separate squall lines, separated by an intervening period of very light rain or no rain at all. Rain during the first squall line was highly variable in intensity, suggesting a poorly organized system of several weak cells. Radionuclide concentrations remained essentially constant during passage of the first line, and dropped substantially during the passage of the second line.

D. THE RAIN OF MAY 2, 1962

Surface and 500 mb synoptic conditions are shown in Figure 7a. The rain collected during this period was associated with a low pressure center located over extreme southern Lake Huron, less than 100 n miles northeast of the collection station. A broad almond-shaped precipitation area was located west of the low, and produced the 0.11 in. of rain collected on this date. At 500 mb, a moderate trough had advanced eastward and developed a cut-off low center over lower Michigan during the 24 hr preceding the beginning of the rain collection.

This rain was definitely of the large scale uplift variety. This is indicated by the very low and uniform rainfall rate and the low echo tops shown in Figure 7. The arrow on one of the echo top observations indicates that maximum tops were at 20,000 ft at that time. During the period of collection of Sample 1, the collection station was under the influence of a precipitation echo characterized by radar as a broken area which was slowly increasing in intensity. As shown in the precipitation rate, the rain stopped for a time, and began again at about 1600. At this time a period of very light uniform rain commenced, lasting until about 2100 when the intensity diminished somewhat. Samples 2 through 6 were collected from

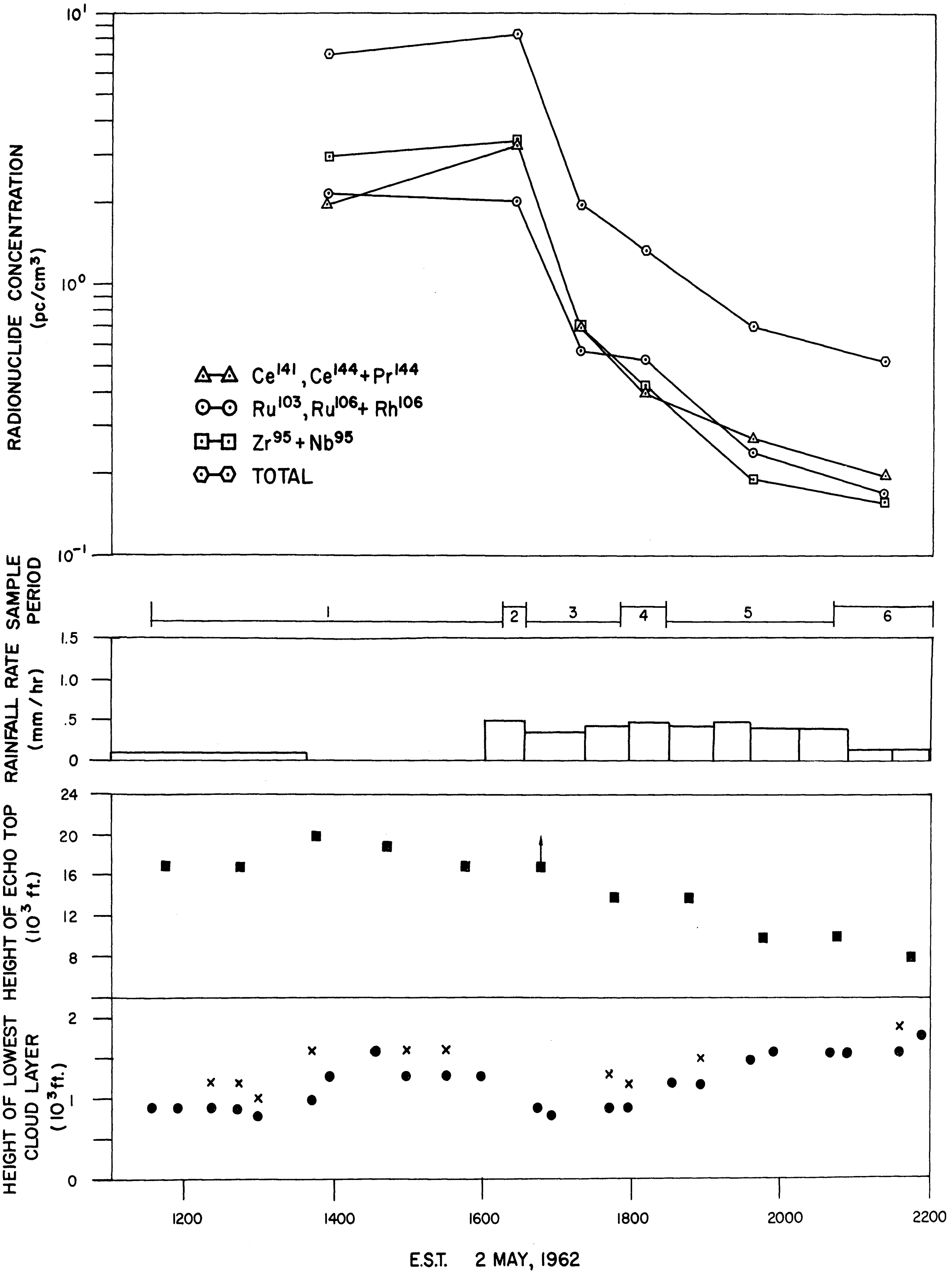

Figure 7. Data for the rain of May 2, 1962.

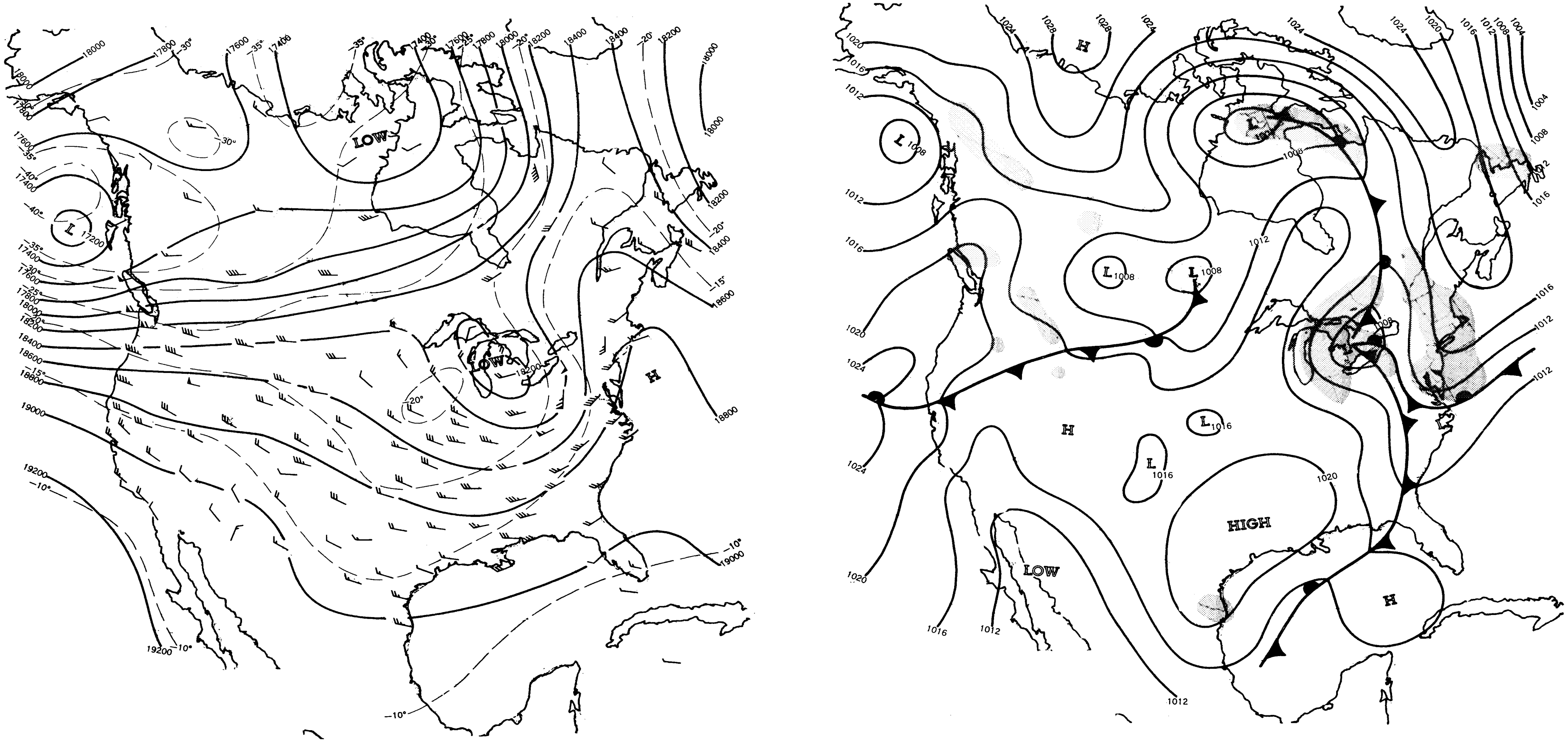

Figure 7a. Synoptic weather data for May 2, 1962.

a precipitation area identified by radar as a broken weak stratiform area, not changing in intensity, except near the end of the rain when the echo was called very weak, decreasing in intensity.

Sample 1 contained rain from two periods of rainfall separated by a 2-hr period of no rain. Since consideration of this sample would further complicate an already complex situation, and add little, Sample 1 has been excluded from the following discussion. After 1600, a general decrease in heights of echo tops was observed, whereas ceiling heights rose during most of this period. No significant difference in behavior was observed between the ceiling height and the base of the lowest cloud layer. Radionuclide concentrations were observed to decrease during the period, at first rather rapidly, and then more slowly.

E. THE RAIN OF MAY 19, 1962

The surface and 500 mb synoptic conditions on May 19 are shown in Figure 8a. During the sampling period our station was south of a stationary front which extended generally eastward from near Menominee, Michigan, and also southwestward from that point. A large, high pressure area was located over the southeastern U.S., with centers over Louisiana and Ohio. The rain which fell during the period came from a very small localized precipitation area. A total of 0.27 in. was recorded at the collection site. An apparently unrelated squall line was present in the Ohio valley. Evidently there was no air mass change during the sampling period. At 500 mb, rather a deep short wave trough moved northeastward from western Nebraska to northern Minnesota.

The rainfall intensity recorded at the collection station, as shown in Figure 8, never exceeded 15 mm/hr. Therefore, it is concluded that the rain collected came mostly from weak cells or the fringes of one or more of the other cells observed to be in the area at the time of sampling. It is quite evident again in this case that no intense, highly organized cells traversed the sampling station.

As before, this rain may be divided into parts on the basis of the pattern of rainfall rate and corresponding radar observations. The first small intensity maximum (near 0745) was apparently associated with a broken line of echoes reported as weak to moderate with no change in intensity. The line was 40 n miles wide, had tops of 36,000 ft, and was moving from the west-southwest at 15 kt. The second slight intensity maximum at about 0900 occurred under an area of weak scattered echoes, slowly decreasing in intensity. Echo tops were at 25,000 ft, and the individual cells moved from the west-southwest at 15 kt. The third part of the rain was composed of the two stronger intensity maxima centered about 1000. The echo at this time was reported as an area of scattered weak to moderate cells moving from

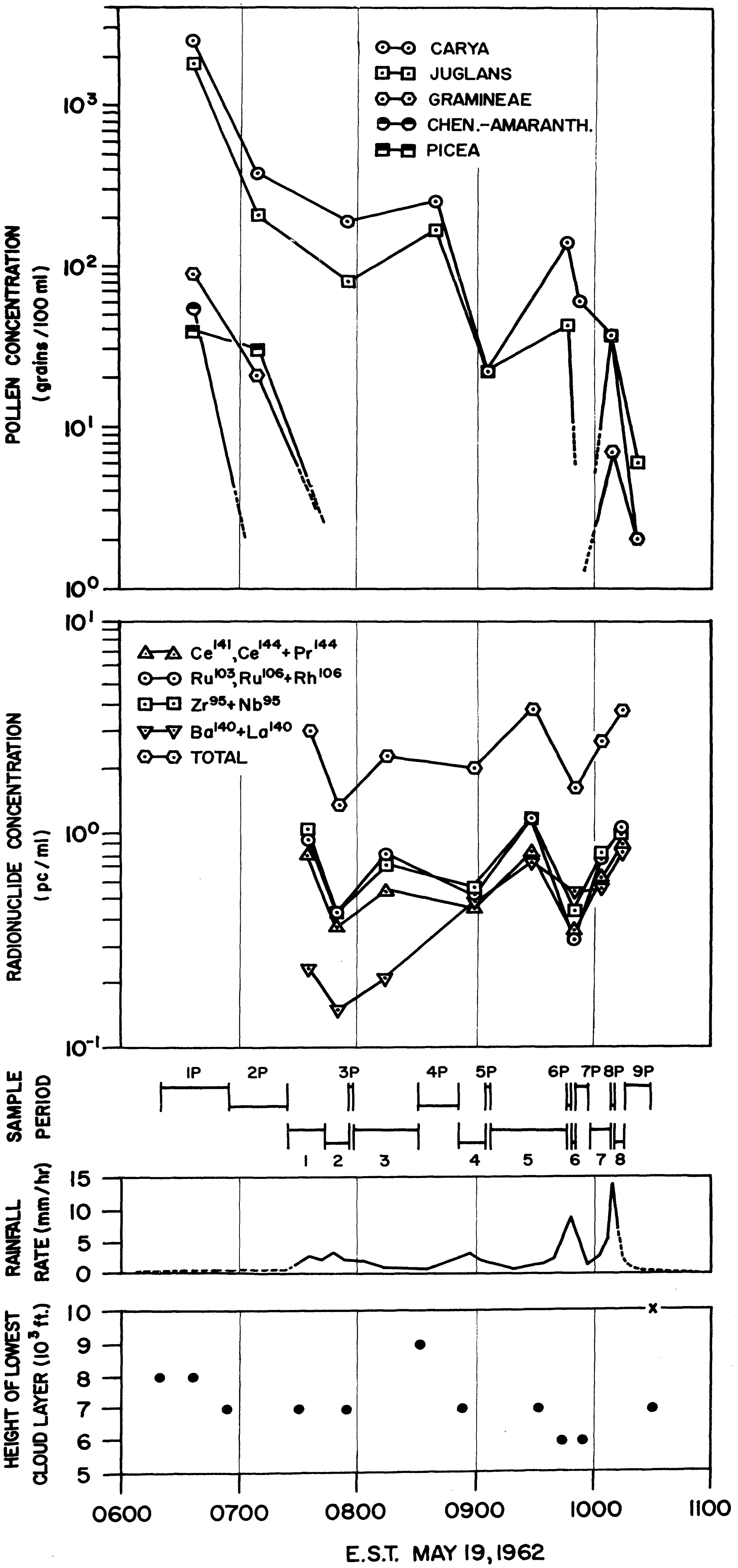

Figure 8. Data for the rain of May 19, 1962.

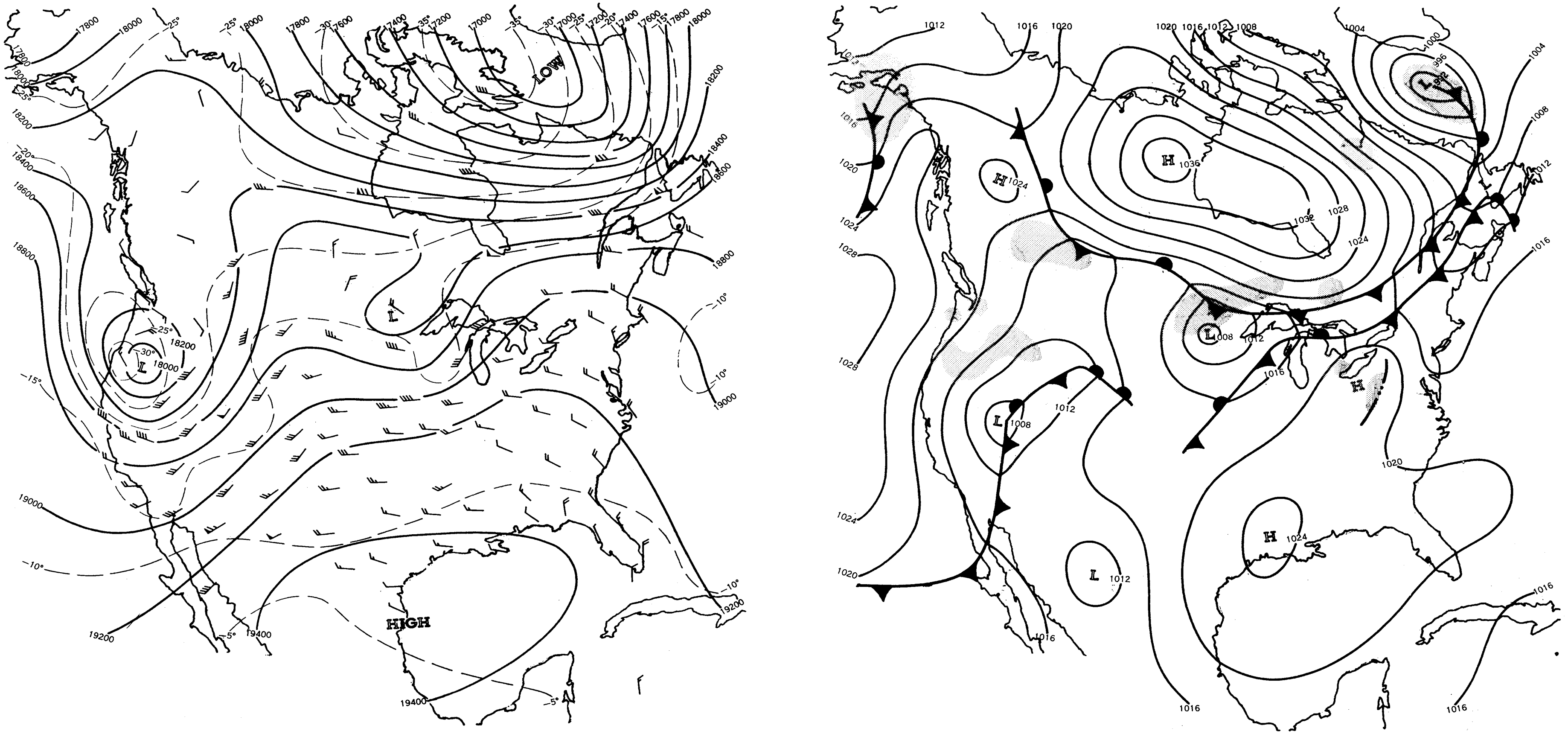

Figure 8a. Synoptic weather data for May 19, 1962.

the west-southwest, unchanging in intensity. Average tops were at 23,000 ft with maximum tops at 32,000 ft. The two intensity maxima undoubtedly resulted from passage of two of the individual cells in the area. Thus it is likely that the rain collected on this date came from a succession of individual convective cells. The absence of heavy rain indicates that the rain was from weak cells or from the edges of moderate cells. In any event, there was a clear absence of any strong, highly developed cells.

The heights of the lowest cloud layer shown in Figure 8 are identical in every case but one (at 1030) to the ceiling height. All heights were estimated by YIP personnel for this rain. Radar data indicated that echo tops were extremely variable. Since these data could not be considered representative of conditions at the sampling station, they were not plotted.

The variations of the concentration of total measured radionuclides were significant but seemingly quite random. Samples 1 and 2 were collected from the first broken line of cells mentioned above. A rather sudden decrease, quite similar to those noted earlier, was observed, followed by a concentration increase in Sample 3, which mostly represents an intervening period of light rain. The increase in concentration from Sample 7 to Sample 8 is interesting in that it opposes the expected and previously observed trend toward decreases during the passage of heavy showers. The remaining rainfall peaks yielded insufficient rain to give an adequate resolution of concentration changes.

This rain was the only one of those collected during the spring and summer of 1962 that contained debris from the U.S. atmospheric nuclear tests begun in the Pacific late in April, 1962. This statement is based upon the fact that barium-140, detectable in large amounts only in fresh fallout owing to its 12.8 day half-life, was observed in this rain. The last previous reported Russian test was conducted on February 2, 1962. Moreover, the barium-140 plus lanthanum-140 concentrations exhibited a clear deviation from the parallel behavior of the other radionuclide groups. This may be interpreted as evidence for a distinct source region for the U.S. debris. Contributions of U.S. debris to the other radionuclide groups were evidently hidden among the greater concentrations present from the Russian tests.

It is interesting to note the concentrations of barium-140 in the weekly sample of ground level air and rain taken between May 18 and May 25 by the National Sanitation Foundation in Ann Arbor. Concentrations in air showed no significant changes from the low environmental levels present from the Russian tests of the previous autumn. Concentrations in rain, however, showed a sudden increase. This indicates that the source of the U.S. debris was the upper troposphere or lower stratosphere. One observation that seems pertinent at this time is that the concentration of the barium-140 group increased with time with respect to the other radionuclide groups. If the source level of the U.S. debris can be ascertained, further investigation of these unusual data may yield important information concerning the mechanisms by which airborne radioactive particulate matter is incorporated into precipitation systems.

Again in this rain, alternate samples were analyzed for pollen content. The results are included as part of Figure 8. Zero concentrations are indicated by a broken line leading off the graph. The species counted were <u>Carya</u> (hickory, 47μ diam), <u>Juglans</u> (walnut, 42μ), <u>Gramineae</u> (grasses, 29μ), <u>Chenopodiaceae-Amaranthaceae</u> (goosefoots-pigweed, 25μ), and <u>Picea</u> (spruce, 75μ). These species were chosen because the pollen are spherical or very nearly so, and because they represent a range of sizes. Thus in this rain we have simultaneously determined tracers representing three distinct source regions of the atmosphere. The source of the pollens is near the ground; therefore they may be considered to be tracers of low-level air. The source of the U.S. debris is not known at present, but it may be of the nature of a thin lamina such as those observed by Anderson (1962). If the older bomb debris is considered to be well mixed in the middle latitude troposphere, the source of this tracer is the entire troposphere. (The maximum echo tops reported were at 36,000 ft, and the tropopause was at 39,500 ft, at 0700, so it is probable that no penetration of the stratosphere was made. A stratospheric source was therefore eliminated from consideration.) Although full investigation of these interesting data has been limited, preliminary investigations have followed the lines established for the earlier convective rains.

F. THE RAIN OF JUNE 25, 1962

Surface and 500 mb synoptic conditions are given in Figure 9a. The rain of 0.64 in. from which samples were collected on this data came from a thunderstorm associated with the passage of a southward-moving cold front. The front was associated with a low pressure system, centered over the lower east coast of Canada, and was oriented in an east-west direction across Michigan. The main shower was quite heavy, as shown by the rainfall rate of Figure 9. This shower was of rather short duration and appeared to develop very rapidly just prior to the onset of the rain at our station, so that it was not observed by radar during the regular hourly observation at 1845. Hence, it was not possible to plot echo tops for this rain. The following observations by personnel at the collection station offer the best description available.

The wind was calm before the passage of the cold front, which occurred shortly before the first rain fell at 1853. The rain intensity increased very slowly until about 1906, when it began to climb more rapidly. Behind the front the wind was gusty and from the north. It appeared, from observations in Ann Arbor and at our station at Willow Run, that rain fell ini-

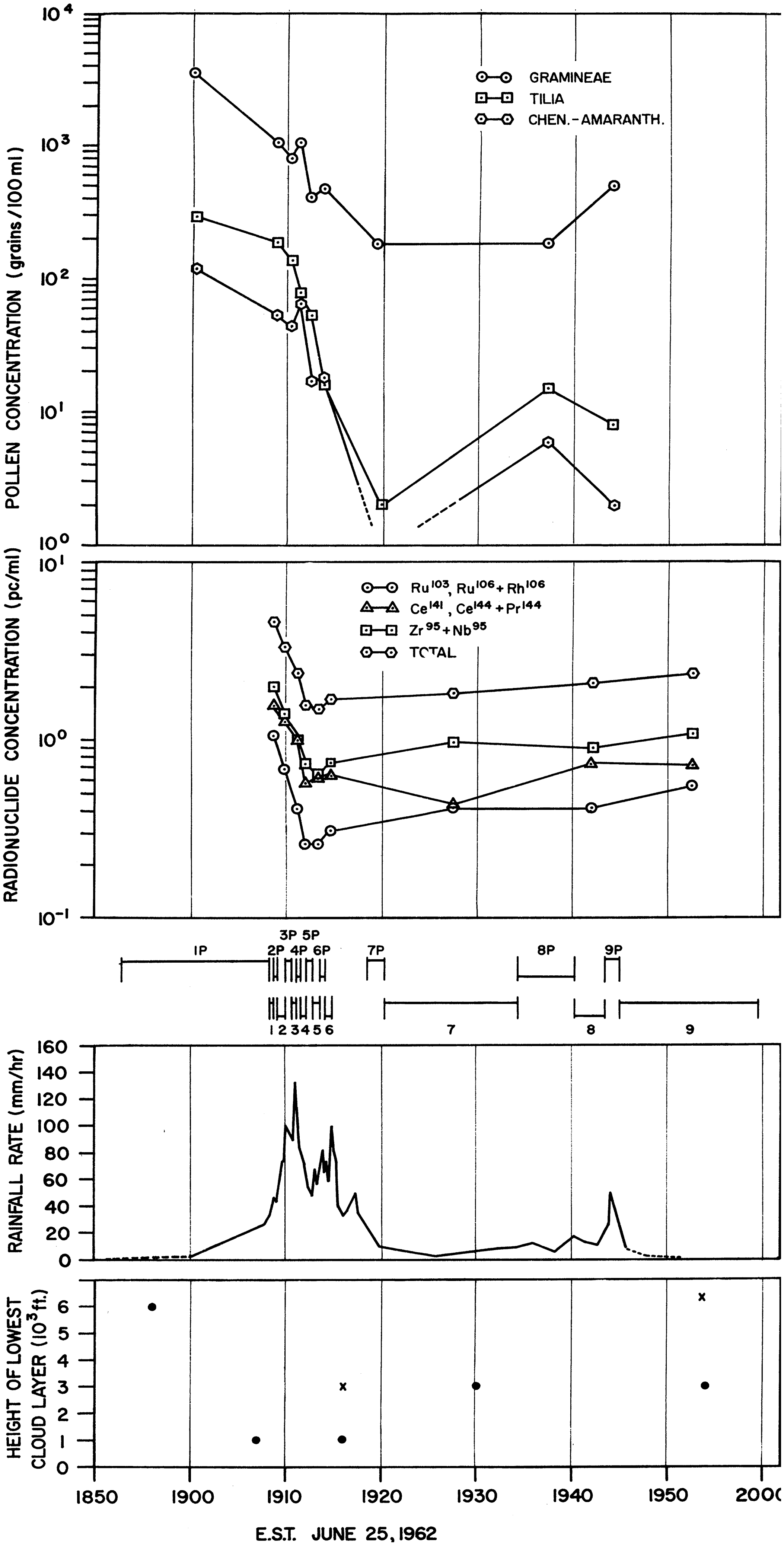

Figure 9. Data for the rain of June 25, 1962.

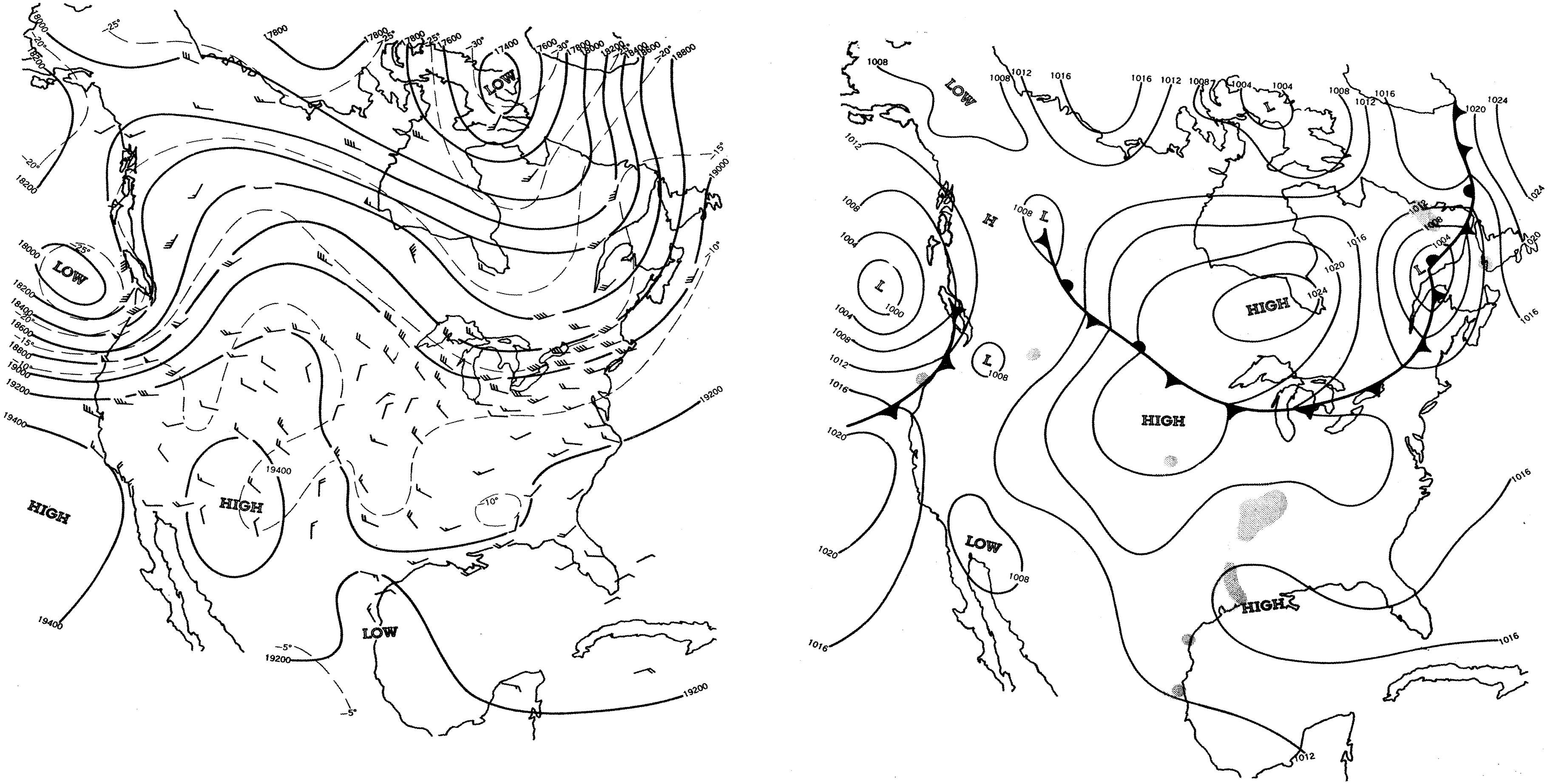

Figure 9a. Synoptic weather data for June 25, 1962.

tially only in rather small shafts. The cells appeared to be moving from west to east along or just behind the front. A small cell to the west-northwest suddenly increased in both size and intensity. It was this cell, or group of cells, which apparently produced the first heavy shower. The pattern of rainfall rate would suggest two or possibly three cells. The smaller intensity peak at about 1945 was from a broken precipitation area behind the first line and described in the radar report at 1945 as weak to moderate, with no change in intensity.

In three of the five cloud-height measurements plotted for this rain, the ceiling height and height of the lowest cloud base are identical. In the remaining two measurements, the increase in height of the lowest layer seems to show a time lag behind the increase in ceiling height. In general, however, both indices decrease to a minimum during the heavy shower and generally rise thereafter.

Alternate samples for pollen analysis were also taken during this rain. The results are plotted as part of Figure 9, but no further discussion is given at this time, except to note that again in this rain we have a specific tracer for the lower levels to compare with the well-mixed radionuclides assumed to represent the entire troposphere.

Radionuclide concentrations are plotted in Figure 9 and again they exhibit the sharp decrease in concentration which seems to be characteristic of heavy convective rains. Beginning with Sample 5, which was collected from the second major intensity peak in the main shower, the rapid decrease in concentration stopped abruptly, the concentration having reached an apparent minimum. Sample 6 maintained the same concentration. At this point sampling was discontinued for a short time to conserve sample bottles. The behavior of the radionuclide concentrations in Samples 1-6 of this rain is remarkably similar to that observed in another intense, multicelled storm—that of September 1, 1961. In both cases concentrations dropped rapidly with time after the onset of the heavy shower. A minimum was reached in the trailing portion of the first cell or the leading portion of the second. The minimum was apparently maintained for the duration of the heavy rain. Samples 7, 8, and 9 represent light rain almost exclusively, since most of the small peak at 1945 was collected in pollen Sample 9P. A gradual increase in radionuclide concentrations with time was observed.

To summarize, this is another case of a heavy convective storm in which radionuclide concentrations show marked and rapid decreases in time during the intense rain. Similarity to the September 1 rain was noted in the detailed behavior of the concentration changes in that an apparent minimum is reached after the initial sharp decrease.

G. THE RAIN OF JULY 2-3, 1962.

Surface synoptic conditions at 1300 on July 2, just as the rain began, and at 1300 on July 3, just before the rain ended, are shown in Figures 10a and 10b. 500 mb conditions at 1900 on each day are also given in the respective figures. Comparison of the two surface charts shows the development of a weak low on a stationary front through Iowa, Illinois, and Indiana, and the eastward progression of a broad band of precipitation located north of this front. A total of 1.24 in. of rain was received from this precipitation system as it passed over our collection station. Comparison of the 500 mb charts shows deepening and a slight eastward movement in both the east coast and west coast long wave troughs.

Radar reports from DTW provide a rather detailed account of the approach and passage of the precipitation area. Together with soundings at Flint, they show that this rain would be broadly classified as of the large scale uplift type. Although this rain contains several rather sharp intensity peaks, its classification as a large-scale uplift rain is apparently consistent with previous interpretation of the term. (See, for example, the rain of January 6-7, 1962, reported by Kruger, Miller, and Hosler, 1962.) Radar echoes were reported at two distinct levels at 0345. The lower echo area was scattered and weak at a distance of about 130 n miles west of DTW. The higher echo was also weak and indicated a stratum roughly of elliptical shape with its major axis extending from overhead to about 90 n miles west of the station. The base of this echo also sloped from 20,000 ft at its eastern end to 6,000 ft at its western end. Echo tops reached 23,000 ft.

By 0445 these echoes appeared to have merged and increased in their north-south dimension. The base of the echo overhead had lowered to 19,000 ft, but its western edge remained at 6,000 ft. The lower echo moved eastward and intensified to "broken" classification by 0645. The stratiform echo aloft continued to extend about 100 n miles east of the lower echo, and maintained its sloping base, which by 0745 reached the surface at its western edge where it merged with the other echo area.

Between 0845 and 1445, the system moved eastward, producing some very light rain at our station beginning about 1300. From 1545 to 2245 the echo over our station was reported as broken, containing mixed stratiform and cellular areas of weak intensity. There was little observable movement of the echo area, but elements moved from the west at 15 kt. From 1545 to 1945 there were some scattered moderate cells in portions of the echo. Some of these individual cells probably produced the intensity peaks observed after 2100. (See rainfall rate pattern plotted as part of Figure 10.) At 0045 on July 3, the echo area had diminished considerably in total size and broken into two parts. One of these parts still covered the collection station, but its passage or diminution probably accounts for the intensity minimum at about 0100. At 0145 mixed broken echoes were again

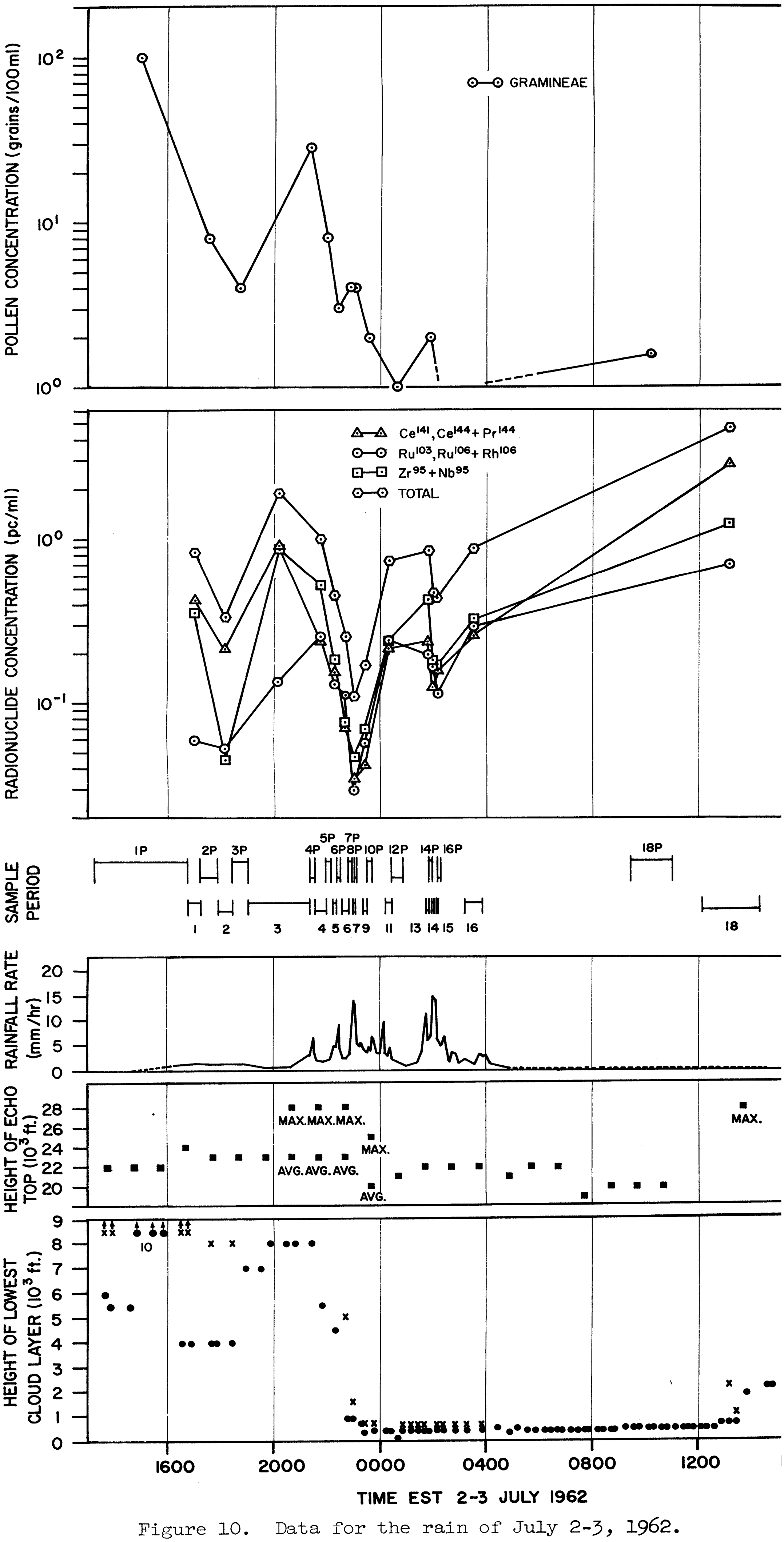

Figure 10. Data for the rain of July 2-3, 1962.

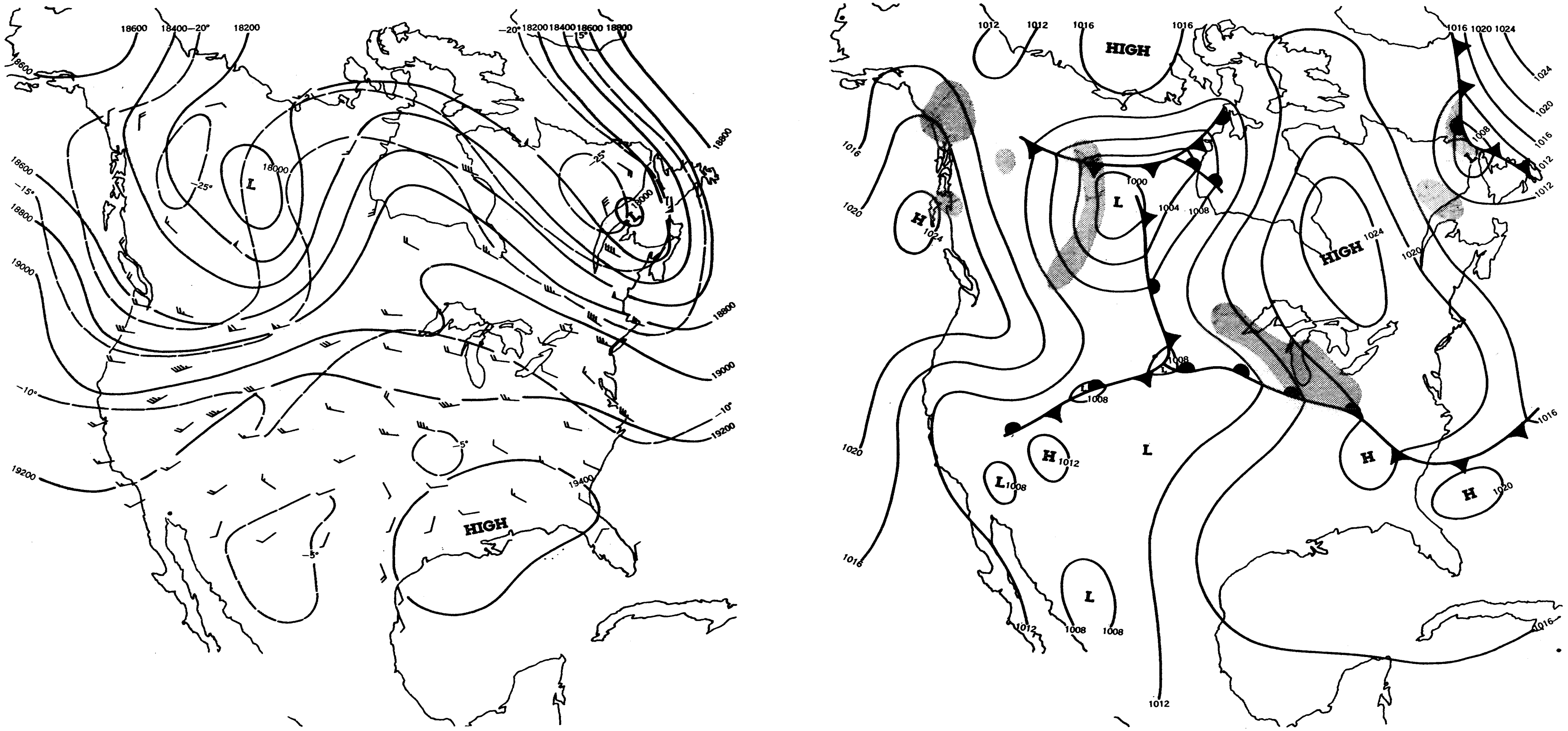

Figure 10a. Synoptic weather data for July 2, 1962.

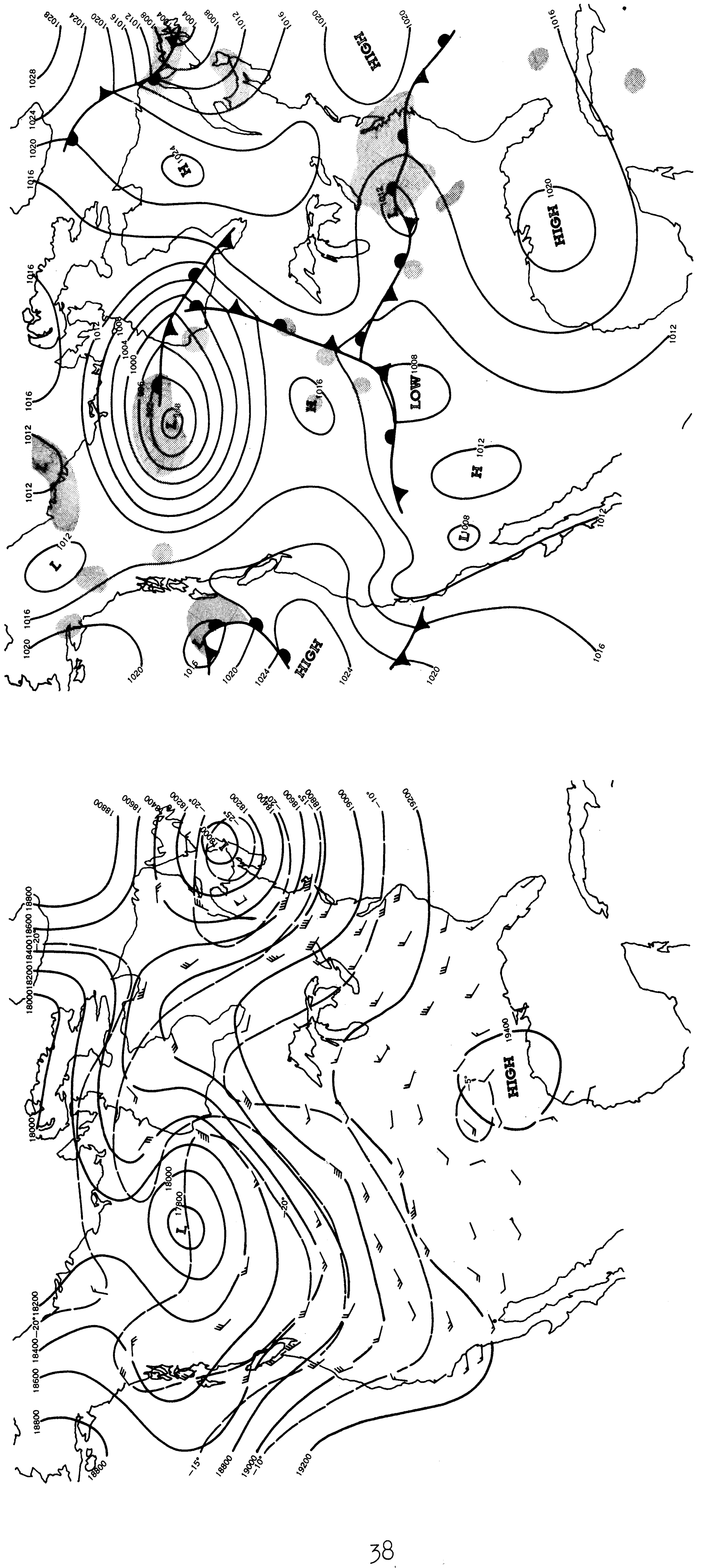

Figure 10b. Synoptic weather data for July 3, 1962.

present over the collection station. It is likely that several cells in this group produced the peaks observed at about 0200. Echoes had weakened considerably by 0345 and continued to weaken thereafter, although light rain or drizzle fell intermittently until after 1200.

Because of the long duration of this rain, it was possible to compile rather complete and representative data on echo tops and cloud base and ceiling heights (Figure 10). During the periods of mixed echoes, the tops were quite variable; therefore both average and maximum tops have been reported for these periods. Both figures represent conditions in the entire echo which covered our collection station at the particular time. Note that the arrows on the ceiling height and height of lowest base observations preceding 1700, July 2, indicate heights of 10,000 ft.

For the greatest part of the duration of this rain, the height of the lowest cloud base was not appreciably different from the ceiling height. Some considerable differences in these indices were observed before 1900 on the 2nd, however. Between 1600 and 1900 on the 2nd, the ceiling height decreased from 10,000 to 7,000 ft while the height of the lowest base remained constant at 4,000 ft. The two indices were virtually identical after 1900. A 1,000 ft increase was observed shortly before 2000. After about 1-1/2 hr at 8,000 ft, the ceiling fell quite rapidly during the next hour, and reached about 400 ft by 0000 on the 3rd. It remained essentially constant after this time until very late in the rain. Several pollen species were again determined in this rain. However, only Gramineae was sufficiently abundant to plot. The results are included in Figure 10.

This rain may be divided into several parts for analysis. During the first stage, which extends from the beginning of the rain until shortly after 2000 on July 2nd, continuous light rain is indicated. The generating level remained essentially constant between 1600 and 2000; furthermore it may be shown that the precipitation mechanism remained constant during this time, as well as for the remainder of the rain. The Flint soundings indicate that the 0°C isotherm remained near 12,000 ft during the entire period of rain. This is in good agreement with a measurement of the height of the bright band made by The University of Michigan APQ/40 radar which was in operation briefly during this rain. The bright band was observed at roughly 12,000-15,000 ft at 1528 on July 2. Since cloud tops were almost constantly higher than 20,000 ft, it is probable that an ice mechanism prevailed throughout the rain.

The second portion of the rain, between 2100 and 0400, is characterized by a succession of intensity peaks whose origin is not entirely clear. Soundings at Flint indicate that the atmosphere was stable during the period. One possible explanation which merits further investigation is the differential advection mechanism described by Miller (1955).

The third stage of the rain extends from 0400 until the end and is characterized by very light rain.

The radionuclide concentrations varied widely during the course of the rain. In particular, three instances are evident where concentrations decreased substantially and then increased by the same or a greater amount. The minima of the events referred to occurred roughly at 1800, 2300, and 0200.

V. DISCUSSION OF PROPOSED INDICES

Radiochemical analyses of samples of rain water collected at fixed ground stations from rain systems of all types have shown substantial time variation of the concentration of radioactive substances in the rain. These variations are present when comparing one rainfall with another (e.g., Walton, Fisher, and Drey, 1962) as well as from sample-to-sample within a single rainfall. Sample-to-sample variations have been presented for the seven rains discussed in this report and have also been reported by others (Salter, Kruger, and Hosler, 1961; Walton, Fisher, and Krey, 1962; Dingle, 1961). Because the variability within a system is pertinent to the understanding of that between systems, we have been especially concerned with the study of the variations within individual rainfalls. This is the principal focus of the present report.

Some consideration should perhaps be given to the fact that the fission product radionuclides used as tracers in the present work are not the same ones used by other investigators whose results and hypotheses are discussed in the light of the present data. Except for periods immediately after detonation of very powerful nuclear devices (see Mamuro, *et al.*, 1963), there is no evidence for fractionation of fission product radionuclides during individual rains. In addition, it has been our experience, as well as that of others, that concentration versus time curves for two or more radionuclides determined in the same series of samples from individual rains tend to be remarkably similar. This result has also been found by Walton, Fisher, and Krey (1962) for strontium-90 and cerium-144. In the following discussion, the assumption is made that concentrations of tracer radionuclides in rain vary in a nearly parallel manner.

Several hypotheses which relate concentration levels to raindrop evaporation have appeared in the literature. That is, it has been hypothesized that the evaporation occurring in the lowest layers of the atmosphere determines to what extent non-volatile matter in raindrops will be concentrated during the fall of the raindrops to the ground. This basic idea leads to the more detailed inference that the time variations of the atmospheric relative humidity field and the raindrop-size spectrum determine the time variations of concentration of radioactive substances in rain at the ground.

Experimental verification of the evaporation hypothesis would ideally require continuous monitoring of the relative humidity field in the region below a precipitating cloud, as well as observation of the raindrop-size spectrum. Because of the practical problems connected with obtaining these data at frequent intervals, to say nothing of doing it continuously, various indirect indices of the effect of evaporation upon concentration have been investigated.

A. THE RELATIONSHIP BETWEEN RAINFALL RATE AND CONCENTRATION

One indirect index, that has been proposed as an indicator of the evaporation of raindrops below the cloud is rainfall intensity (Bleichrodt, *et al.*, 1959). In general, more large drops are produced in rain of high intensity (Dingle and Hardy, 1962), and large drops lose relatively less mass by evaporation than smaller drops (Kinzer and Gunn, 1951; Hardy, 1962). As a result, rain falling at low rainfall rate is expected to contain a higher concentration of collected impurities when it reaches the ground than rain of a higher intensity which leaves the cloud base with the same initial concentration.

Bleichrodt, *et al.*, (1959) have plotted rainfall rate and concentration of gross beta radioactivity versus time for 11 individual rainfall occurrences. A crude inverse relationship is found, that is to say, increases in rainfall rate appear usually to be associated with decreases in concentration, and vice-versa. These data are limited to rainfall rates less than 20 mm/hr. Shirvaikar, *et al.*, (1960) found that gross beta concentrations in successive fractions of individual rain showers collected at Bombay during the 1958 monsoon rains were independent of rainfall rate in some cases, and observed an inverse relationship in a few other cases. Salter, *et al.*, (1961), reporting data gathered in three rains of the large scale uplift type, found no clear relationship between precipitation rate and strontium-90 concentrations.

The present data (Figures 4-10) show simultaneous time variations of rainfall rate and fission product concentrations. In addition, another representation of the relationship is given for some of these rains in Figures 11 through 16.

Figure 11 shows the variations of radionuclide concentrations with rainfall rate for the samples collected on September 23, 1961. This figure is an additional exhibit of the parallelism that exists among the several radionuclide groups when concentrations are plotted against another parameter.

Data for six rains have been plotted together in Figure 16. It is noted that while there is little over-all organization, the highest concen-

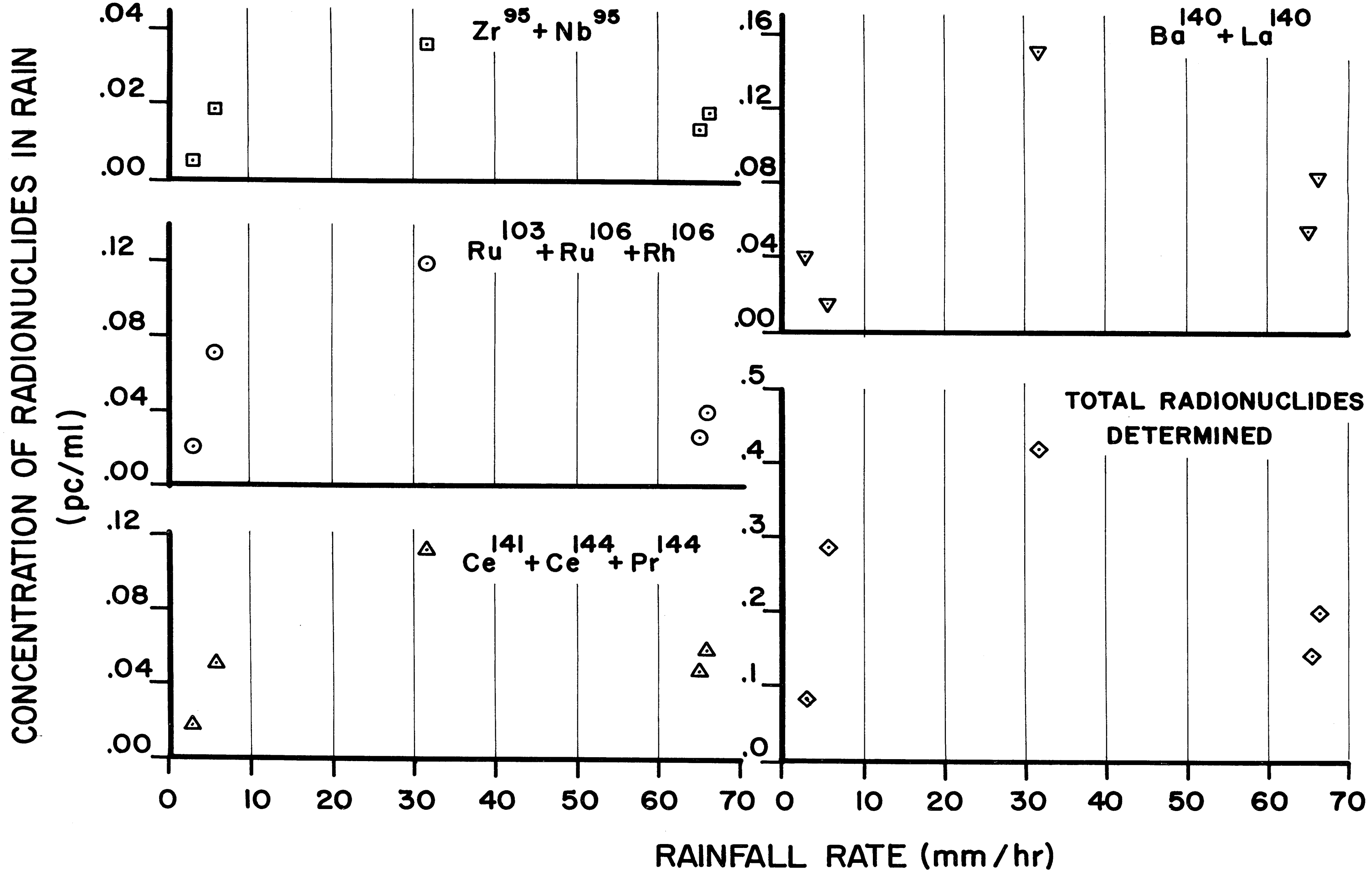

Figure 11. Graph of radionuclide concentration against rainfall rate for rain of September 23, 1961.

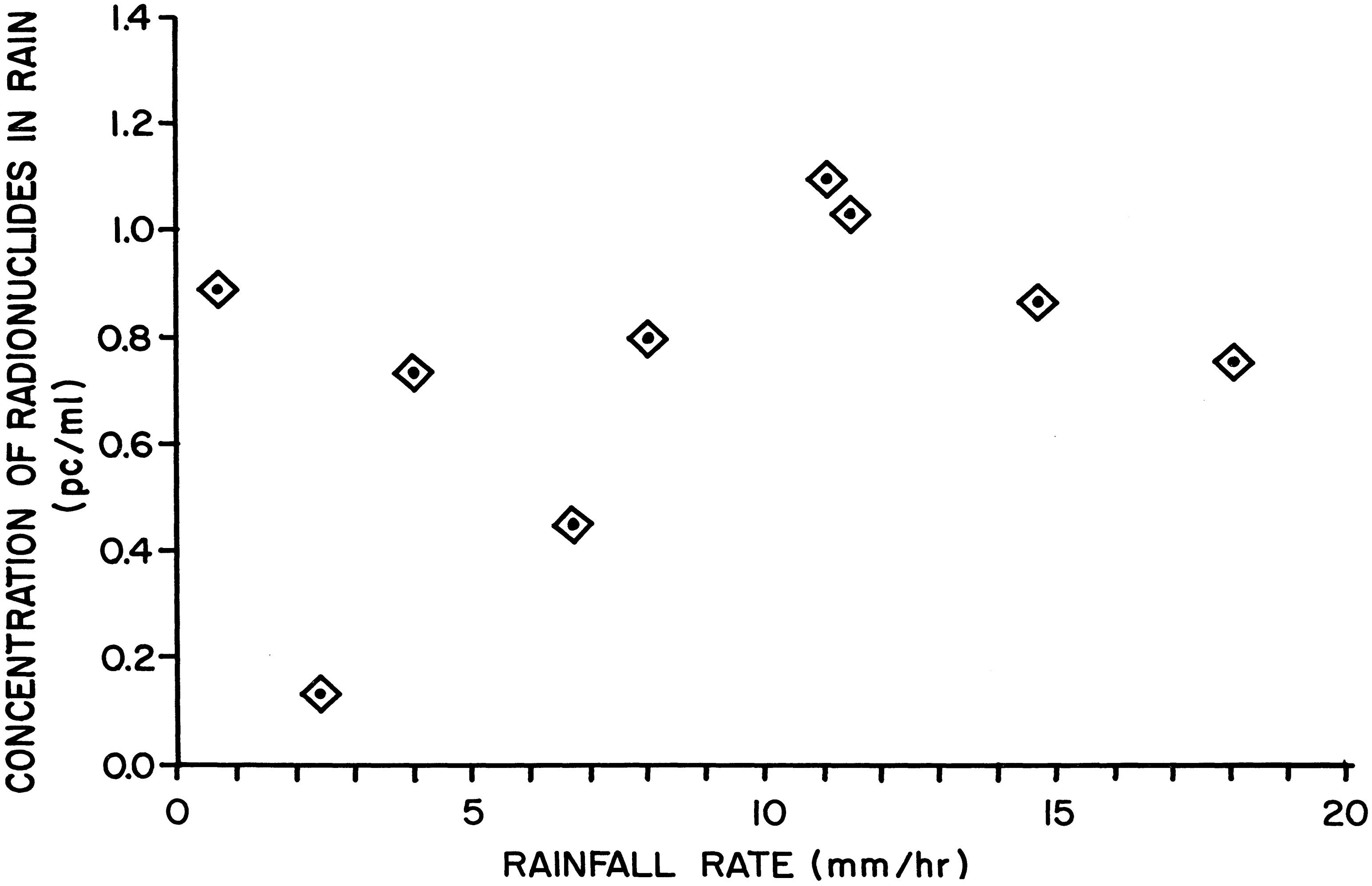

Figure 12. Graph of radionuclide concentration against rainfall rate for rain September 30, 1961.

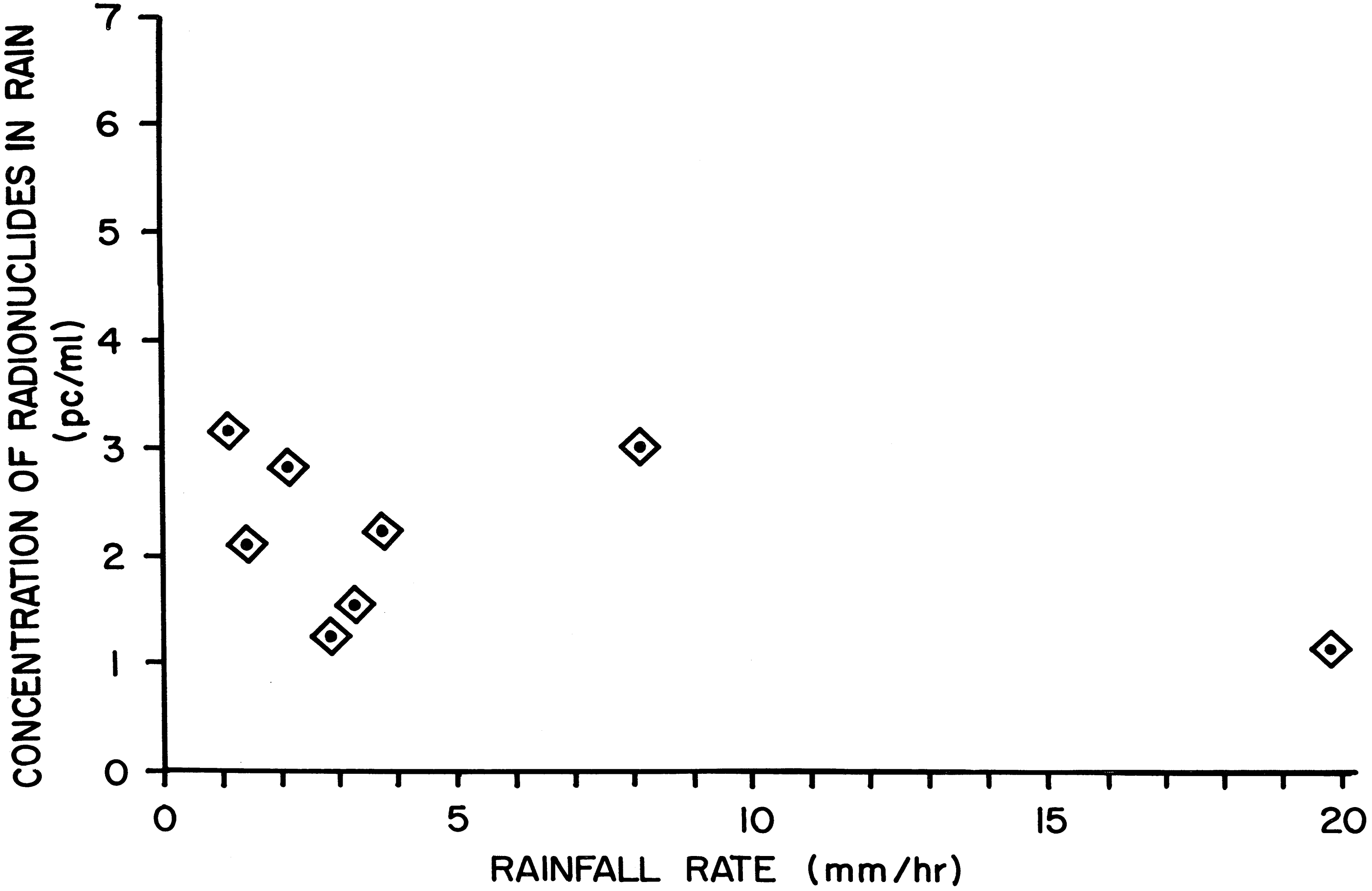

Figure 13. Graph of radionuclide concentration against rainfall rate for rain of May 19, 1962.

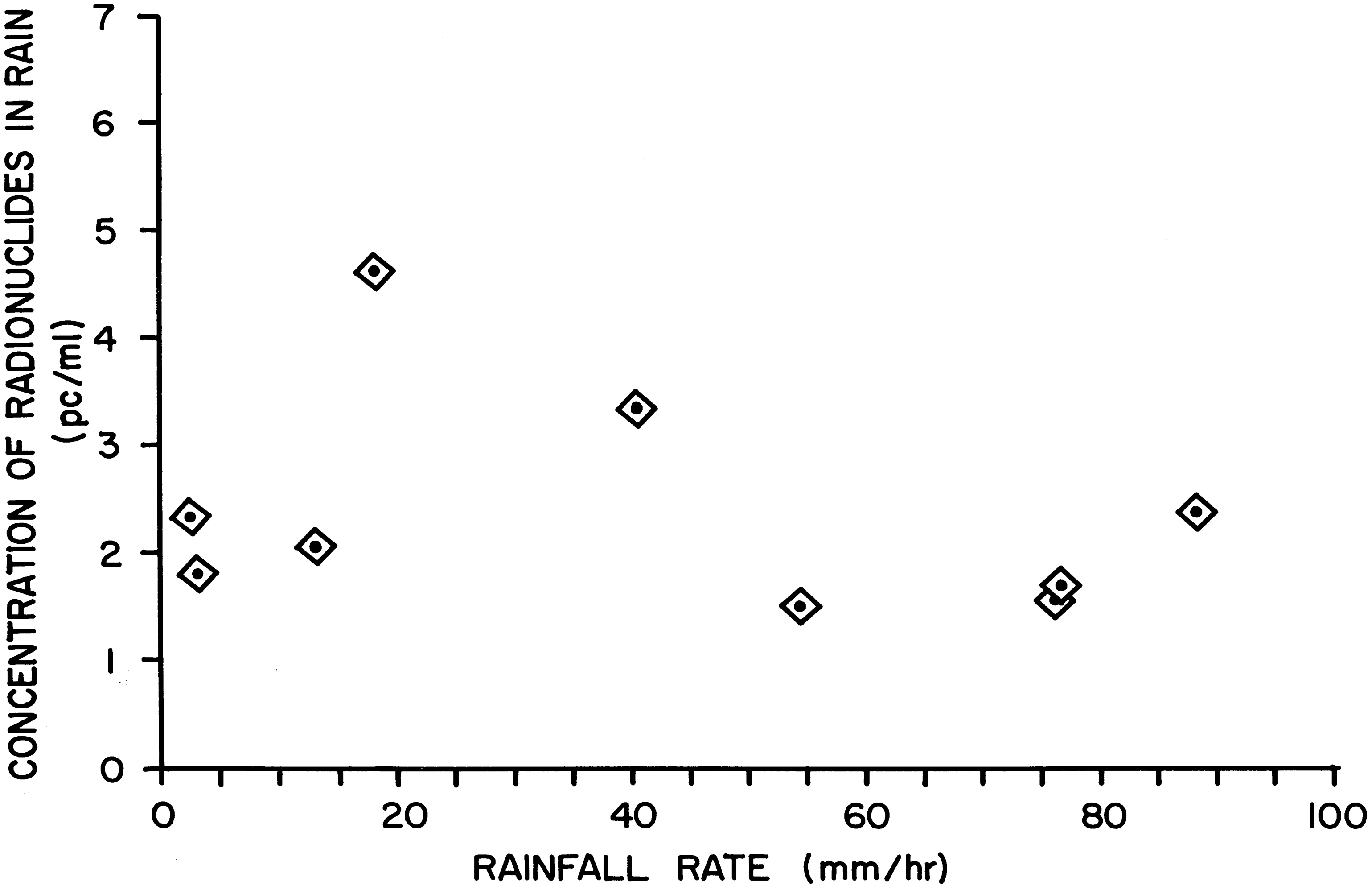

Figure 14. Graph of radionuclide concentration against rainfall rate for rain of June 25, 1962.

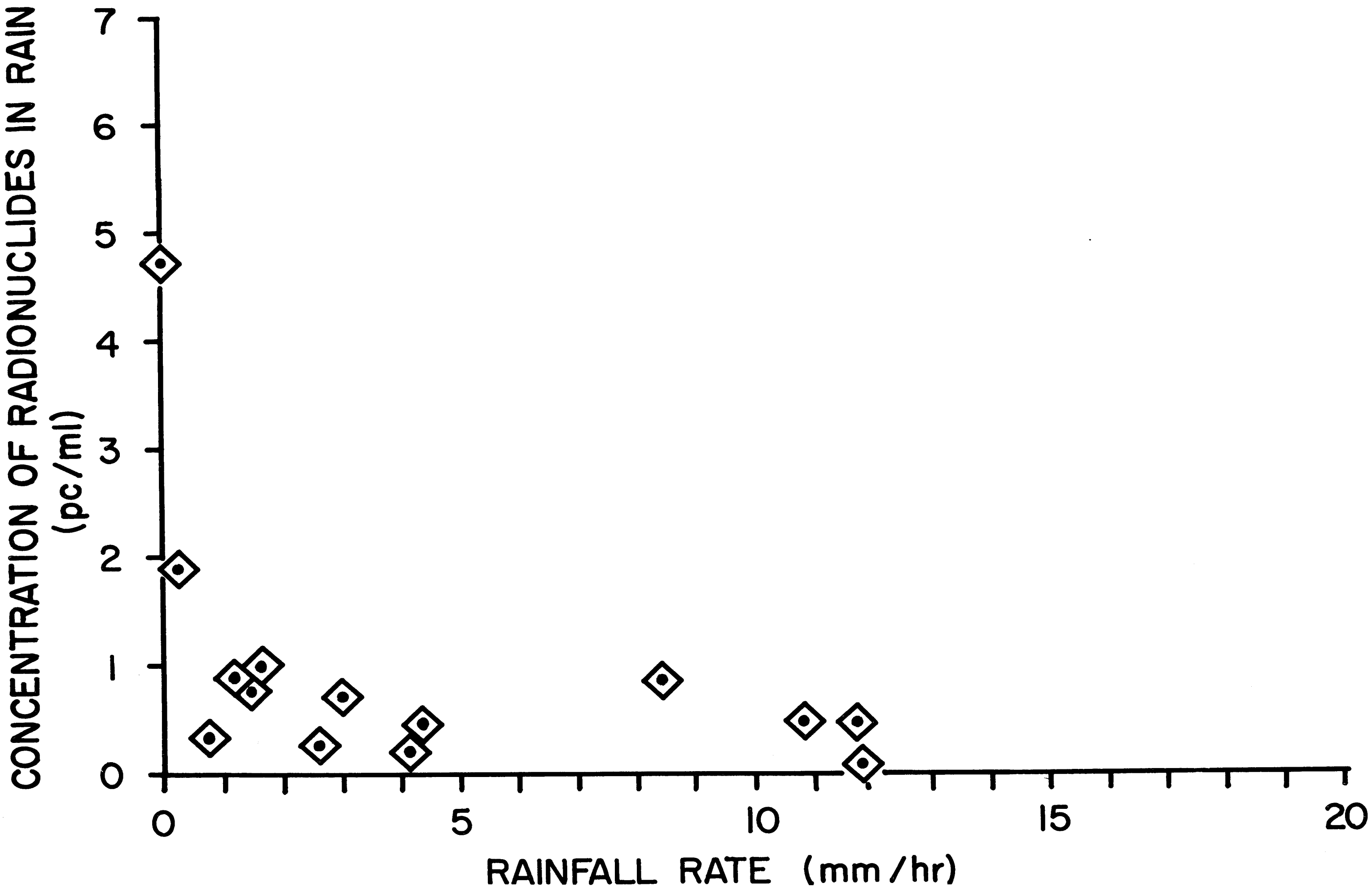

Figure 15. Graph of radionuclide concentration against rainfall rate for rain of July 2-3, 1962.

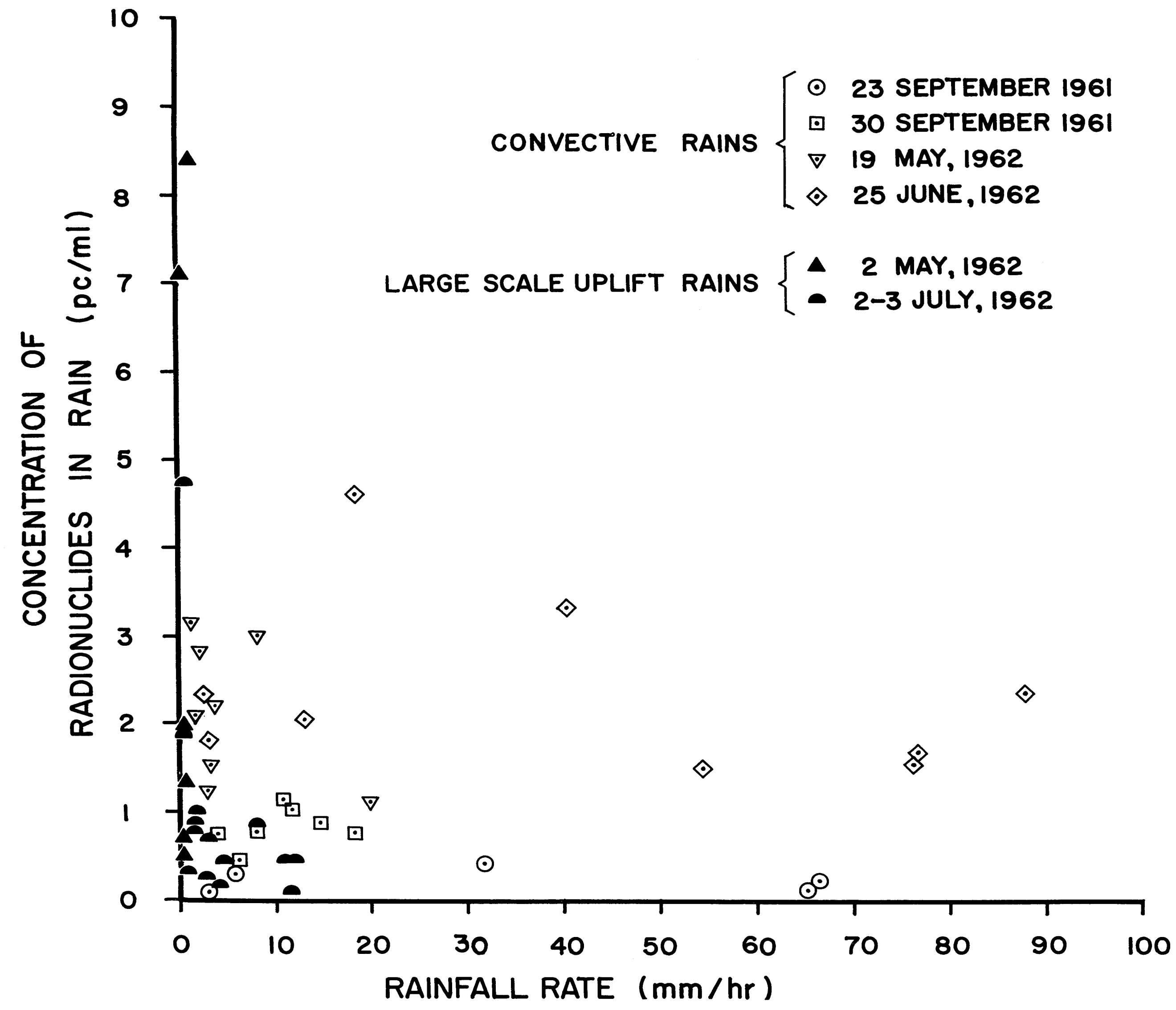

Figure 16. Graph of radionuclide concentration against rainfall rate for six rain systems.

trations are associated with the lowest rainfall rates. Furthermore, although the lowest concentrations are not associated with the highest rainfall rates, the concentrations observed at the highest rainfall rates are certainly much lower than the highest observed concentrations.

By dividing these six rains into convective-type and large-scale uplift-type events, it is readily seen that there is a rather strong inverse relationship for the large-scale uplift rains (May 2, 1962 and June 2-3, 1962), whereas there is no similar relationship apparent for the convective rains. To eliminate effects due to time variations in the concentration of radioactive particulate matter in the atmosphere, the individual rainfalls have been plotted in Figures 11-15.

Because of some uncertainties in the rainfall rates for the rain of May 2, 1962, this rain is not included among these graphs. In Figure 7, where the data for this rain are presented, it may be seen that the rainfall rate remained quite constant at about 0.4-0.5 mm/hr. As has been mentioned earlier, because of instrumental limitations, the variation between rainfall rate as computed from rain gage measurements and as computed from sample data becomes large at very low rates. In this case, the variations were of the order of 100% of the mean value so that it would not be meaningful to make an individual plot of this rain. On the other hand, the uncertainty is not significant when compared to the scale of Figure 16; therefore the May 2 data have been included in this figure.

As noted in the case of the composite plot (Figure 16), individual plots of convective rains show that radionuclide concentration is independent of rainfall rate. The single remaining example of an individual large-scale uplift rain (July 2-3, 1962) shows in general an inverse correlation.

It may be useful to recall some of the observations reported previously. Of the three large-scale uplift rains reported by Salter, <u>et</u> <u>al</u>., one showed rainfall rate and radionuclide concentration to have a strong inverse correlation, whereas the other two failed to show this relationship. Bleichrodt, <u>et</u> <u>al</u>., (1959) showed nominal inverse correlations for 11 rains of unspecified type, but having rainfall rates of less than 20 mm/hr. Shirvaikar, <u>et</u> <u>al</u>., (1960) found inverse correlations in only a few cases of monsoon rains at Bombay. These rains were of widely varying intensity, but there was no tendency for the inverse correlations to occur in the lighter rains.

In attempting to interpret these observations, it must be remembered that the rainfall rate is only one of the parameters that can be used to indicate the influence of the evaporation process upon radionuclide concentrations. It is, however, the evaporation process itself that should be evaluated. Apparently, other available indicators need to be considered in combination with the rainfall rate when evaluating a particular rain event.

B. THE RELATIONSHIP BETWEEN CEILING HEIGHT AND CONCENTRATION

Salter, Kruger, and Hosler (1961, 1962) used the ceiling height as an index of the humidity profile and depth of the lower layer of air. They conclude that under conditions of a fairly constant precipitation generating level* and precipitation mechanism in large-scale uplift rains,..."the ceiling height serves as a useful index of the evaporating power of the atmosphere below the cloud level." Thus with low ceilings, less evaporation should occur than with high ceilings, from which it follows that strontium-90 concentrations should vary in phase with the ceiling height.

In the absence of a rationale for choosing the ceiling height as an index of the evaporating power of the lower layer of the atmosphere, (Salter, Kruger and Hosler, 1961), it seemed reasonable to investigate another such index, namely, the height of the base of the lowest cloud layer. Except when the lowest layer is scattered, these two indices are identical. For those occasions when they are different, some further investigation might be fruitful. Although the ceiling height index was intended for use in large scale uplift rain systems, not convective ones, the remnants of convective storms are frequently stable in nature and productive of small-drop, steady light rain. For this reason, an effort was made to see how well the ceiling height index would work in partially convective rains.

During the intense shower on September 23, 1961, concentrations and ceiling heights decreased simultaneously. After the heavy shower, both parameters increased. These observations are in agreement with the ceiling-height hypothesis. However, in Sample 8, the concentrations decreased significantly, whereas the ceiling height remained constant or increased. The increase in ceiling height near the end of the period of sample collection was accompanied by a similar but smaller increase in the height of the lowest cloud base. This variation is in opposition to that predicted and, importantly, comes at a time in the storm when the ceiling height hypothesis should most be expected to apply. It was not expected that the ceiling height indicator would be significant during thundershowers because the requirements of constancy of generating level and precipitation mechanism might not be met. On the other hand, during the period of light rain and drizzle which follow thunderstorms, it would be expected that these conditions might be met.

The rain of September 30, 1961, was characterized until 2200 by rather

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*Defined as the highest level at which cloud elements could be detected, either visually or by 3-cm radar (Hosler, 1963).

constant concentrations and constant ceiling heights, although a lower scattered cloud layer was associated with the passage of the first squall line. These observations are in agreement with the ceiling-height hypothsis. However, during the remainder of the rain, concentrations decreased under conditions of constant ceiling height. A decrease of the height of the lowest cloud base was observed with the onset of the second squall line; however, a concentration decrease was observed between Samples 8 and 9 while the lowest bases were constant. In summary, neither index gives a satisfactory correlation with concentration changes during this rain.

The rain of May 2, 1962, appears to be a case of a large-scale uplift rain in which the relationship between radionuclide concentration and ceiling height is exactly opposite to that suggested previously (Kruger and Hosler, 1962, 1963). Further examination reveals, however, that one of the limiting conditions—that of a constant generating level—was not met. Indeed, the generating level (echo top) decreased after 1600, from 17,000 ft to 8,000 ft. It is considered quite unlikely that the ice mechanism prevailed throughout the sampling period because the cloud tops failed to reach temperatures as low as -15°C at any time, and toward the end of the rain, the cloud top temperature was warmer than -10°C. Soundings at Flint show considerable cold advection aloft during the sampling period. Since the cooling aloft was accompanied by a lowering of the cloud tops despite the destabilizing effect, it is clear that the air advected aloft was very dry. The possibility that this air came from the lower stratosphere should not be overlooked, but further study is required to establish its most likely source.

Comparison of concentrations with ceiling heights for the convective rain of May 19, 1962 (Figure 8) shows fair agreement with the hypothesis during the first half of the rain and rather good agreement during the last half. Similarly, rather good agreement was observed in the convective rain of June 25, 1962.

The rain of July 2-3, 1962, presents an excellent opportunity to test the ceiling-height hypothesis. As described above, the ice mechanism prevailed throughout the rain. In addition, the generating level remained nearly constant for much of the rain. The period between 2000 and 0000 was characterized by variable echo tops, yet these variations are no greater than those observed by Kruger, Miller, and Hosler (1962) in the rain of January 6-7, 1962, at State College, Pennsylvania.

The three main concentration minima have already been pointed out. In the first of these, the concentration decrease between Samples 1 and 2 was accompanied by a lowering of the ceiling from 10,000 to 8,000 ft. The ceiling height was at 7000 ft for a short time during the collection of Sample 3, but rose to 8000 ft and remained at that level for the rest of the period. A large increase in concentration was observed in Sample 3.

Thus, the initial decrease of concentration was associated with a decrease of the ceiling height, but the substantial concentration increase that followed occurred with no effective increase in ceiling height. Investigation of the height of the lowest cloud base as an index reveals that this height remained constant during the period of decreasing concentrations, but increased during the period when concentration rose. As the concentration decreased toward the second major concentration minimum, there was a substantial lowering of the ceiling; however, the ceiling height decreased further or remained constant during the period when the concentration underwent a substantial increase. Both the decreasing and increasing periods adjacent to the third concentration minimum occurred under conditions of constant ceiling height. The concentration increase observed in Sample 18 was accompanied by a rising ceiling.

Thus, of seven major changes in concentration during this rain, only three cases conformed to the ceiling-height hypothesis. These results show that the relationship between the ceiling height and radionuclide concentrations does not account well for the observed changes in this uplift-type rain system. It is interesting to note that ceiling-height changes were a better indication of concentration changes in the convective rains described above.

Critique. A basic flaw of the "ceiling-height" hypothesis lies in its extreme oversimplification of a complex situation. Evaporation is only one of a number of mechanisms capable of producing concentration changes from sample to sample in rain: ceiling-height is only one index of evaporative power or potential. Other very basic considerations are those relating to the initial distribution of the radioactive contaminants in the atmosphere and the means by which they become attached to cloud and rain drops.

Indeed, since the ceiling-height hypothesis is based upon an assumption of constant concentration of radionuclides in time and space in cloud water, it avoids the basic question of what the scavenging mechanisms are. It is unfortunate that so little is known regarding the temporal and spatial variations of radionuclides in cloud water. However, one clue is provided by the work of Aldaz and Howell (1960) who found considerable variation of radiostrontium in rime collected on Mt. Washington.

It is reasonable to assume that evaporation of raindrops between the cloud and the ground must affect the radionuclide concentration of rain collected at the ground. However, the question remains whether such processes can account for a major part of the variation observed. The present observations suggest that evaporation is indeed not the predominant process contributing to the observed concentration variations. Kruger, Miller, and Hosler (1962) have related concentration changes to ceiling height changes through the empirical equation

$$\overline{\Delta C} = 1.25\ \overline{\Delta H} \pm 0.65$$

where $\overline{\Delta C}$ and $\overline{\Delta H}$ are the fractional changes of concentration and ceiling height, respectively. Since $\overline{\Delta H}$ very seldom has an absolute value greater than 1.0, this equation implies a minimum error of about 50% in $\overline{\Delta C}$. Kruger, et al., (1962) suggest infrequency of measurement as the cause for the error. It seems more reasonable to attribute the error to the fact that the evaporation process is only part of the reason for concentration changes.

It is further suggested that the relative importance of the evaporation mechanism may be determined by means of available computational procedures to be discussed later.

C. THE RELATIONSHIP BETWEEN TROPOPAUSE PROXIMITY AND CONCENTRATION

It has been suggested (Kruger and Hosler, 1962, 1963) that concentrations of strontium-90 in rain from convective showers will vary according to the proximity of the cloud tops to regions of high concentrations of strontium-90 in air. The specific source of high radioactivity implied is the stratosphere or the so-called tropopause break. During the passage of a convective storm it is assumed that the height of the tropopause above a fixed ground station will not change significantly. Therefore concentration changes within individual rains may be related to the vertical development of the rain cloud above the station. Radar data for the present rains are not sufficiently detailed to permit experimental evaluation of this hypothesis. Nevertheless, we may examine the concentration data for an indication of the kinds of changes that would be necessary, under this hypothesis, to produce the observed concentration changes. The concentration decrease between Samples 1 and 5 on September 23, 1961, took place in roughly 5 min. The hypothesis would require a corresponding rapid decrease in the height of the cloud top above the station. Such rapid decreases in the height of the cloud tops themselves are unlikely, but rapid decreases in the height of the top above the station are possible as convective towers move across the station. Figure 5 shows that the sharp decrease in concentration occurred as the rainfall rate increased. The highest rainfall rates are identified with the central or core regions of the respective convective cells, and these in turn would seem to be associated with the region of maximum vertical development. Thus it appears that the changes in cloud top during this short period could only have been upward, directly opposite to the change required by the hypothesis. The same conclusions would be drawn from further examination of the rains of September 1, 1961, and June 25, 1962. Thus, while proximity of the cloud tops to the tropopause could account for some variation in radionuclide content between rains, it apparently is not the reason for the very rapid variations observed within rains.

D. CONCENTRATIONS IN THE FIRST RAIN FROM CONVECTIVE RAINS

It has been suggested (Kruger and Hosler, 1962) that the first rain collected under convective shower conditions will originate in the lower parts of the cloud and utilize condensation nuclei from the lower parts of the atmosphere where concentrations of radionuclides in air are expected to be low. It follows that concentrations of radionuclides in the first part of the rain should be low. The results of our analyses of the five convective rains are contrary to this suggestion. In three of the five cases the first sample collected contained the maximum concentration. In the remaining two cases, the concentration in the first sample was not much less than the maximum observed during the rain. The pattern of concentration changes in rain from convective showers is discussed further in a later section in which a model for rain-cleansing in convective rains is suggested.

E. THE INFLUENCE OF THE PROXIMITY OF THE STORM CLOUD TO THE JET STREAM AND STRATOSPHERE

It has been shown that the vertical distance between the cloud top and the tropopause cannot account for the sharp concentration changes observed during single rains. We must still investigate the hypothesis that this distance, as well as the horizontal distance to the tropopause break, will influence concentrations between rains. It must be noted that the data of Kruger and Hosler (1962, 1963) were taken at the end of the 1958-1961 nuclear tests moratorium when the stratosphere was indeed the only source of nuclear debris. Huff (1963), in a partial test of the Kruger and Hosler hypothesis, found poor associations between gross beta concentrations in convective rains and the distance from the tropopause to the top of the storm cloud radar echo. However, Huff's data were collected during the spring and summer of 1962, i.e., during a period of active nuclear testing in the atmosphere. In such periods, the troposphere, as well as the stratosphere, must be considered a source of nuclear debris, and it is to be expected that the hypothesis of Kruger and Hosler would not hold.

Our own data, also collected during or soon after periods of atmospheric testing, have been partially examined for relationships between artificial radionuclide concentrations in rain and the parameters proposed by Kruger and Hosler. In four convective and two large scale uplift rains, there was no evidence that either the maximum or average concentrations of bomb radioactivity in individual rains was dependent upon horizontal distance to the jet stream. Although there were some uncertainties associated with several of the tropopause height analyses obtained from the National Meteorological Center, examination showed no evidence for an association between the vertical distance from the tropopause to the storm echo tops and the concentration of nuclear debris in rain. This is in agreement with the findings of Huff. Hence, although no verification of the Kruger and Hosler hypothesis for moratorium conditions is currently possible, their hypothesis is clearly not valid during and shortly after test series, when there is a strong tropospheric source of fallout radioactivity.

F. GEORGII'S RELATIONSHIP

It has been noted many times in the past that concentrations of both natural and artificial trace substances in rain water decrease as the amount of rain increases. Georgii (1960, 1961) made a rather extensive study of this relation for individual rainfalls, and found for an "unpolluted" atmosphere the relationship

$$K = \text{const}\ h^{-0.28}$$

where K is the concentration of certain chemical trace constituents and h is the amount of rainfall.

Georgii claims good agreement between this relationship and the results of other investigators (Hinzpeter, et al., 1959; Peirson, et al., 1960; and Martell, 1959) for fallout radioactivity in rain. It should be noted that in Georgii's analysis, the results of individual observations were averaged in several intervals of rainfall amount before the curve was drawn. Even with this smoothing procedure, some scatter of the points is observed. In effect, then, this relationship is only valid in a general way for a large number of observations.

Although the number of observations reported here is small compared to the number used in Georgii's study, it is useful to compare our observations to this previous work. As explained in the section on sampling procedure, alternate samples were taken for radiochemical and pollen analyses. Therefore the mean concentration derived from weighting the activities of the individual samples according to sample size is only an approximation to the true mean of the whole rain. Table II shows the fraction of the total rainfall in the collector which was analyzed radiochemically for the several rains. The calculations are based on the sum of the measured volumes of the individual samples, the surface area of the collectors, and the amount of rainfall measured by the rain gages. Table II also gives the data on radionuclide concentration and total rainfall which are plotted in

Figure 11. Note that if we consider only the 1962 rains, so as to eliminate the effects of differences in atmospheric activity levels, a straight line may be drawn through three of the remaining four points. The stray point corresponds to the rain of July 2-3, 1962, for which only 29% of the rain was analyzed. The straight line through the remaining three points is given by the expression

$$K = \text{const } h^{-0.26}.$$

TABLE II

RELATIONSHIP BETWEEN AVERAGE CONCENTRATION AND TOTAL RAINFALL

Date	% of Rain Analyzed	Amount of Rainfall (mm)	Average Concentration of Total Measured γ (pc/ml)
23 Sept 61	< 50	17.78	.200
30 Sept 61	> 65	9.65	.744
2 May 62	83	2.79	3.53
19 May 62	82	6.86	2.61
25 June 62	34	16.26	2.14
2-3 July 62	29	31.24	0.674

The exponent is in very good agreement with the value of -0.28 given by Georgii. Such a good agreement is felt to be largely fortuitous, however, because of the fact mentioned earlier that even Georgii's smoothed data show some scatter about the straight line.

It is possible to test Georgii's empirical relationship further through the use of data given by Walton, Fisher, and Krey (1962). These data are cerium-144, concentrations, in individual rainfalls at Westwood, N. J., from July, 1960, through June, 1961, during the nuclear test moratorium. The raw data were averaged in several categories of rainfall amount. Results are given in Table III and Figure 17.

As in the case of Georgii's chemical constituents, there is much scatter among the points, and it is possible to draw a straight line conforming to Georgii's formulation which fits the data as well as any other straight line:

$$K = 12.4\ h^{-0.28}$$

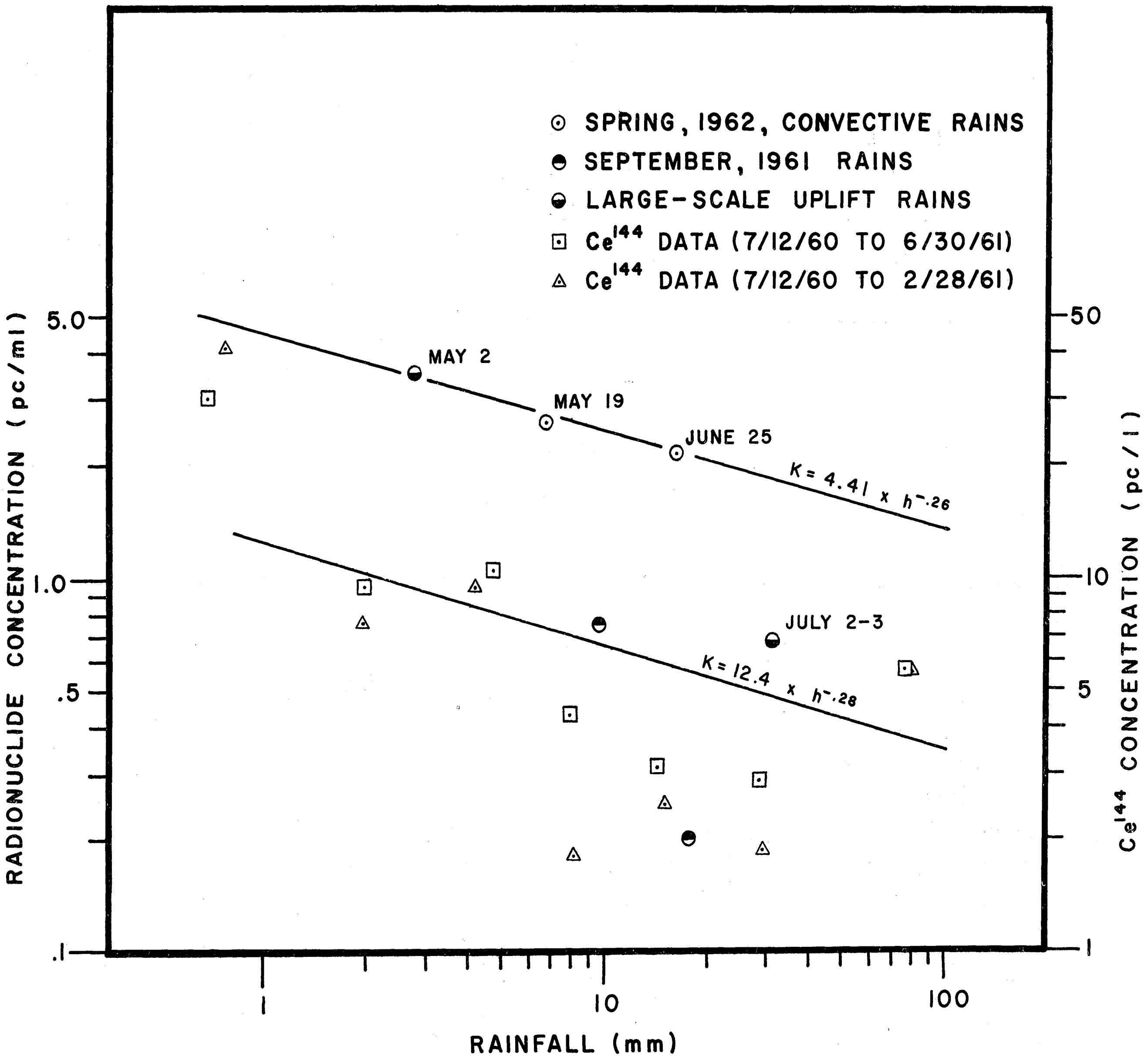


Figure 17. Comparison of data on radionuclide concentrations in rain with a relationship given by Georgii (1960).

TABLE III

RELATIONSHIP OF AVERAGE CONCENTRATION AND TOTAL RAINFALL FOR Ce^{144} DATA OF WILSON, FISHER, AND KREY (1962)

Amount of Rainfall (mm)	Mean Concentration of Ce^{144} (pc/ml)	No. of Cases	Mean Rainfall Amount (mm)
$\leq$ 1.0	30.46	4	0.70
1.1- 3.0	9.45	13	2.00
3.1- 7.0	10.41	12	4.74
7.1-11.0	4.35	8	7.97
11.1-20.0	3.11	10	14.26
20.1-50.0	2.88	17	29.00
$\geq$50.1	5.69	5	76.38

A difficulty is apparent in that the scatter of points is quite serious. Of course data on nuclear debris concentrations in rain over periods of a year or more suffer from the effects of seasonal variation of the concentrations in air, but this is presumably also the case for the fallout data mentioned by Georgii (1960), as well as for chemical trace substances. Therefore it is likely that the observed scatter in the case of the Ce^{144} data can be reduced only by increasing the number of samples. Since the Ce^{144} samples were collected over a period of one year, it appears that the power-law expression proposed by Georgii gives a reasonable relation for long-term averages, such as would be important in considering the long-term cumulative deposition of fission product radionuclides as a function of the total amount of rainfall.

The triangle points show means from the Walton, et al., (1962) data without the spring peak months. It appears obvious that Georgii's formulation fails effectively to express the relations of these observations to one another.

G. SUMMARY

Explanation of the present data with respect to two indices of the evaporation process showed that neither variations in rainfall rate nor ceiling height was adequate to explain corresponding variations of radionuclide concentrations in rain. Concentrations were found to be independent of rainfall rate in convective rains, but exhibited in general an inverse correlation in large-scale uplift rains, especially for the smallest rainfall rates. Rather serious deficiencies in the ceiling height relationship were found in the case of an extensive, well-documented rainfall during which the postulated limiting conditions were satisfied. Use of the height of the lowest cloud base yielded no significant improvement over the ceiling height. It is concluded that evaporation plays a limited role in the ultimate determination of radionuclide concentrations in rain.

The suggested relationship between concentration in rain and the proximity of the cloud top to regions of high radioactivity was found inadequate to explain observed rapid changes of concentration during individual rains. Similarly, the peak and average concentrations in individual rains appeared to be independent of the proximity of the tropopause and the jet stream during the period studied. It is expected that this relationship would be better fulfilled under moratorium conditions.

The present data show that the first rain collected from a convective rain is likely to contain a high concentration of radionuclides, contrary to an earlier suggestion.

VI. A MODEL FOR RAIN CLEANSING BY CONVECTIVE STORMS

Radiochemical analyses of samples of rain-water collected at fixed ground stations from rain systems of various types have shown substantial time variations of the concentration of radioactive substances in the rain. For the type of rain systems involving large scale uplift of air, it has been suggested (Salter, Kruger, and Hosler, 1962) that variations of the Sr^{90} concentration in rain are related to evaporation of the raindrops between cloud and ground.

The notion that the evaporation process is the only one which determines concentrations of radionuclides in rain at the ground is questioned in this report. In any case, no plausible theory has been put forth to explain rapid variations of concentration in rain from convective storms. The following discussion considers what <u>kinds</u> of parameters should be important in determining these variations.

Comprehensive studies of thunderstorms have shown that such rain systems are made up of one or more individual convective units called cells, and that any consideration of the storm as a whole must involve consideration of the characteristics of the individual cells. In the context of rain cleansing of atmospheric particulate matter by convective rains, it is therefore appropriate to consider the effects of individual cells.

In general, one can think of two categories of parameters which should govern the changes with time of radioactivity concentrations in rainwater from convective rains collected at a fixed point on the earth.

The first category includes those parameters which vary as the life cycle of individual cells. One of these—the vertical development of the cloud tops—has been suggested (Kruger and Hosler, 1962, 1963) as governing the Sr^{90} concentrations in rain at the ground. Other parameters which are partially or wholly in this category are rainfall rate, raindrop size spectrum, and the extent and velocity of the updraft. This category is hereafter called the "life-cycle" category.

The second category includes parameters which vary according to horizontal position of the collection station with respect to the center and boundaries of an individual cell. One such parameter is the relative amount of entrainment of outside air into the several parts of a rain-containing downdraft. Near the leading edge of the downdraft, where, according to the models of Browning and Ludlam (1962) and Newton (1950), the downdraft is adjacent to an updraft of unwashed environmental air, one would expect entrainment of the contaminated air into the downdraft, where the contamination would be picked up by the rain. One would further expect either that the amount of mixing of external air with downdraft air would decrease toward the center of the downdraft, or that contamination would be removed from the external air before it reached the center portions of the downdraft. In either case, the rain near the forward edges of the downdraft should contain a higher concentration of contaminants than that farther inside the same downdraft. Thus, as the cell proceeds across the station, the concentrations in the rain should decrease toward the rear of the cell. Other parameters which are partially dependent on spatial position are rainfall rate and drop-size distribution. This category is hereafter called the "cell-traverse" category.

From an examination of the characteristic time-scales of parameters in the life-cycle and cell-traverse categories, it is possible to indicate which of the two categories is dominant in controlling concentration variations. Life-cycle processes have a period near that of the lifetime of the mature and dissipating stages of a thunderstorm cell. This is about 1 hr (Byers and Braham, 1949). Cell-traverse processes have periods of the order of the time required for an individual cell to cross the station. A five mile diameter circular cell moving diametrically across the station at 20 mph will traverse the station in 15 min.

Since the life-cycle period is much longer than the time required for the traverse of an individual cell, concentration changes resulting from life-cycle parameters should be relatively small during the passage of an individual cell. On the other hand, concentration changes as a result of cell-traverse processes should be greatest during the traverse of an individual cell.

The character of the concentration changes during convective rains is quite variable. One of the prominent features observed, however, is a rather sharp decrease of concentration with time. Under the assumption that concentration changes are due predominantly to life-cycle processes, one expects that such rapid changes occur as the station leaves the influence of one cell and comes under the influence of another cell <u>at a different stage of its life-cycle</u>. Conversely, if such changes are due predominantly to cell-traverse processes, they must occur in the time during which an individual cell traverses the station.

In a rough way, it is possible to trace the passage of individual cells by observation of the rainfall intensity. Individual peaks in rainfall intensity may be assumed to correspond to passage of individual cells.

Thus, we should examine time variations of concentration with respect to time variation of intensity. If sharp changes in concentration occur <u>between</u> intensity peaks, the process responsible for such changes must be one which varies according to the life-cycle of individual cells. If on the other hand, such changes occur during intensity peaks, the responsible processes must vary according to the position of the station with respect to individual cells.

Ideally, one should have concentration of radioactive substances in rain recorded continuously with time. Due to sample counting requirements, however, this is not practical; samples of a finite size are required. The isotopes that we have chosen to determine and the gamma-spectrum analysis technique permit convenient analysis with samples of about four liters. With the degree of resolution of concentration changes possible with samples of this size, it is easily possible to supply the detailed information necessary to test the above hypothesis in heavy rains. This is more difficult to do in lighter rains, but usually enough samples are available to provide some information.

Several of the data sets described earlier in this report offer sufficiently detailed information on concentration changes to be examined in the light of this discussion. On September 1, 1961, a sharp decrease in gross beta concentration occurred during the passage of one or two intense cells. The concentration reached an apparent minimum value during the passage of the second cell. Only small concentration changes were observed during the remainder of the storm. On September 23, 1961, a sharp decrease in total measured gamma activity occurred during the passage of a single intense cell. A single sample collected during passage of a weaker cell later in the rain showed an increase in activity. Therefore, this rain showed strong evidence of substantial concentration changes occurring during passage of individual cells and weaker evidence, because only one sample was collected from the second peak, that changes also occur between cells.

The first squall line on September 30, 1961, is an example of light rain resulting from the passage of a series of rather weak small cells. Very little change in concentration was noted during passage of the first line, indicating at least that average concentrations during the several periods were about constant. Samples 8 and 9 were collected from a second weak squall line. The samples were not clearly representative of rain from individual cells, however, and no explanation of the observed decrease in concentration is attempted.

The rain collected on May 19, 1962, was again from a series of rather weak convective cells. Again, however, it is difficult to be certain that individual samples represent individual cells. This complicates the interpretation of the concentration changes, but there appears to be some degree of cell-to-cell variability. On June 25, 1962, rain was collected from a heavy multi-celled storm which began moments after the passage of a cold front. Radionuclide concentrations fell sharply during the passage of the first of two or three individual cells present in the main shower. Samples 5 and 6, collected from the second cell, showed no change from the minimum concentration reached in the first cell, indicating that there was no change between cells from the minimum reached in the first cell.

Several conclusions may be drawn from these data.

1. During heavy rain from single- or multi-celled thunderstorms, concentrations decrease sharply during passage of the first one or two cells, remaining constant for the duration of heavy rain from closely-packed cells.

2. When intense cells or groups of cells are separated in time and distance there is some evidence that increased concentrations may occur in the first rain from each shower. It is expected that these increases would be followed in each case by sharp decreases.

3. Concentration changes in rain from weak cells are difficult to interpret because of poor resolution of concentration changes within individual cells. There is some evidence, however, for some cell-to-cell variability in mean concentrations.

There is, therefore, strong evidence that time variations of radionuclide concentrations in rain from convective systems are controlled by processes which vary according to position within individual cells and cell complexes. Furthermore there is some evidence that patterns of sharp concentration changes are repeatable if cell complexes are some distance apart. These observations support the entrainment mechanism suggested earlier. When cell complexes are far enough apart to allow the establishment of a leading updraft, the characteristic pattern of concentration changes tends to be repeated. When cells are adjacent, the leading updraft is adjacent only to the leading cell, so that concentration changes occur only in the first one or two cells, and the concentration reaches and maintains a minimum during passage of the trailing cells.

VII. RECOMMENDATIONS FOR FUTURE RESEARCH

The present results indicate that further research would be profitable in several areas. Additional work is needed on the rain of May 19, 1962, to determine the source of the U.S. debris and the mechanism of its inclusion into the rain system. Similarly, more work is needed on the rain of May 2, 1962, to ascertain the role of the cold air advection aloft and the falling cloud tops on the changes in radionuclide concentrations.

The whole problem of the influence of the evaporation process on the concentration of radionuclides in rain at the ground needs further attention. Fortunately, much of the necessary computational and programming work has already been done at this laboratory by Hardy (1962) in a study of variations of raindrop size distributions with height. Following the procedure described by Hardy, it is possible to deduce the distribution of raindrop sizes at the melting level from known or assumed data on the drop-size distribution at the ground, the liquid water content of the cloud, the vertical profiles of temperature and relative humidity and the height of the cloud base and melting level. From drop-size distributions at the melting level and at the ground, the amount of evaporation may be computed. This procedure permits the evaluation of the ceiling height and the rainfall intensity as indices of concentration changes caused by the evaporation process. The ceiling height index may be evaluated by holding the rainfall rate constant while varying the cloud base. Similarly, the rainfall rate index may be evaluated by holding the cloud base constant while varying the rainfall rate. The drop size distribution is related to the rainfall rate through the relationship of Marshall and Palmer (1948)

$$N_D = N_o e^{-\Lambda D}$$

where D is the diameter, $N_D dD$ is the number of drops with diameters between D and D + dD in a unit volume of space, N_o is the value of N_D for D = 0 and found to be 8000 m^{-3} mm^{-1} for any intensity of rainfall, and

$$\Lambda = 41 R^{-0.21} \text{ cm}^{-1}$$

where R is the rainfall rate in mm hr^{-1}.

Additional data are required for confirmation of the proposed cleansing model for convective storms. To further substantiate the apparent results, it is necessary to examine in detail the cellular structure of convective rain systems by means of PPI, RHI, and vertically pointing radar displays and comprehensive mesoanalysis of a number of selected storms. Doppler radar would be a great aid in establishing circulation patterns within the storms. In addition, efforts should be made to improve the sampling procedure so as to provide for more effective regulation of the sampling intervals. Instead of merely filling a succession of sample bottles, the individual samples should be changed in accordance with changing storm conditions. This might be done on the basis of radar information or visual observations of rainfall intensity.

Close examination of the data from simultaneous determinations of fission product radionuclides and pollens in rain as given in Figures 4 through 10 suggest a new experimental technique for the study of the structure and circulation of precipitation systems.

The basic assumption is that airborne pollens, having their source near the ground, are tracers for low-level air; fission product concentrations on the other hand, usually increase with height in the troposphere. Furthermore, thin lamina of debris freshly injected from the stratosphere have been found in the troposphere. If these lamina are readily distinguishable from well-mixed radioactive debris (perhaps because of recent origin or a specific radioactive tracer) the radioactivity they contain can be used as a specific tracer for air at a certain level. Thus, determination of two or more tracers specific for various atmospheric levels in a series of sequential samples from individual rains may be an aid in deducing circulation in the rain system. In addition, such determinations may aid in the quantitative determination of entrainment of outside air into the storm system.

One may see by comparing the time variations of pollens and radioactivity in the rains of May 19 and June 25, 1962, that there are significant differences between the concentration curves of these two tracers. This suggests that there are corresponding differences in the relative amounts of air entrained from upper and lower levels.

It is suggested that development of the basic technique described here would lead to further knowledge in the field of storm circulation and entrainment.

VIII. RAINFALL CLIMATOLOGY OF THE WILLOW RUN METEOROLOGICAL FIELD STATION

The following tables (Tables IV-XV) and bar graphs (Figures 18-29), were constructed on the basis of rainfall data accumulated by the U.S. Weather Bureau from 1948 through 1961 at Willow Run Airport (YIP).

The principal interest of the project appears best served by the study of precipitation that falls as rain because our measurement techniques are best developed for rain as contrasted to snow. Summarizing graphs for all but the winter months (December-March) are given in Figure 30. Because our interest lies in obtaining enough rain to provide a good sequence of samples (from the large pans) or a good set of drop-size sorted samples (from the raindrop sorter), we regard it important to observe the time distribution of the relatively heavy rains throughout the year.

Considering rains of 1.0 in. and more (Table XVI), the months of April and October appear to provide the highest frequency (11 in 14 years) and July is a close competitor (10 in 14 years). The lowest frequency is found in June (7 in 14 years). Looking further, however, rainfalls of 0.5 to 1.0 in. appear to occur most frequently in June (19 in 14 years); and considering rains of at least 0.5 in., approximately three occur every 2 years in each month from April through September. Although it is not possible to choose a "best" month for our purposes from this evidence, it is clear that good data may be obtained in our area provided that an adequate sampling and observation program is fielded for about three years.

Considering the hourly rainfall amounts for the same part of the year (Table XVII), 1.0 to 1.99 in. in 1 hr tends to occur most frequently in July, August and September, whereas 0.5 to 0.99 in. in 1 hr was observed most often in June, July and August during the years 1948-1961. The rains of November through May appear to be relatively gentle, none having exceeded 0.99 in. in an hour during these years.

The time occurrence of the high rainfall rates is best observed by reference to Figures 22, 23, and 24. Five of the occurrences of 1.0 in. or more in an hour were observed between 1030 and 1530, two were in the early morning and two in the evening after 2130. Other physical factors that influence these occurrences appear to be at least as important as the diurnal effect. The timing of frontal passages is probably the most important consideration, and, in a measure, this means that the important systems are relatively predictable.

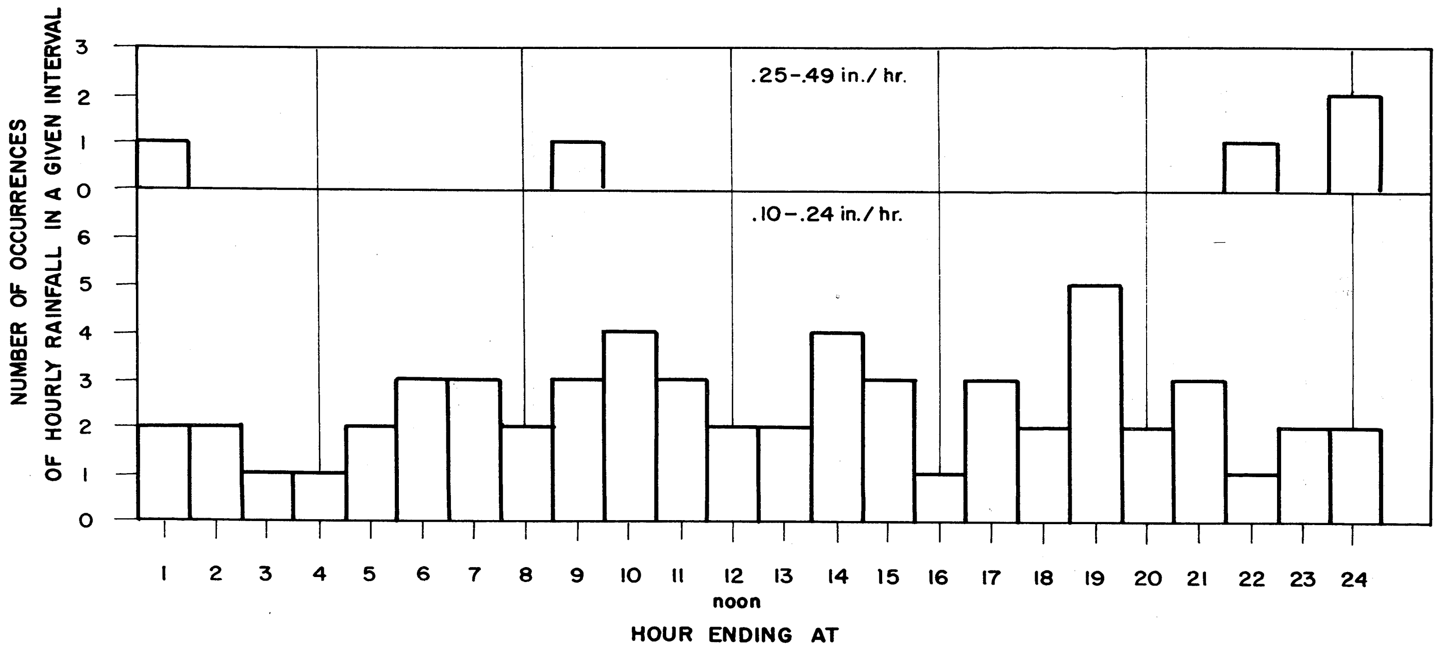
TABLE IV

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

January

Mean Hourly Rate, in./hr	Total System Rainfall,* in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01	7					7
.01-.019	13	10	3			26
.02-.029	3	8	3			14
.03-.039		3	4			7
.04-.049		2	1			3
.05-.099			3	3		6
Σ	23	23	14	3		63

*The amount of rain accumulated during periods of consecutive whole hours when at least a trace of rain was recorded. A "rain system" was considered to be terminated by a period of one hour with no rain.



January

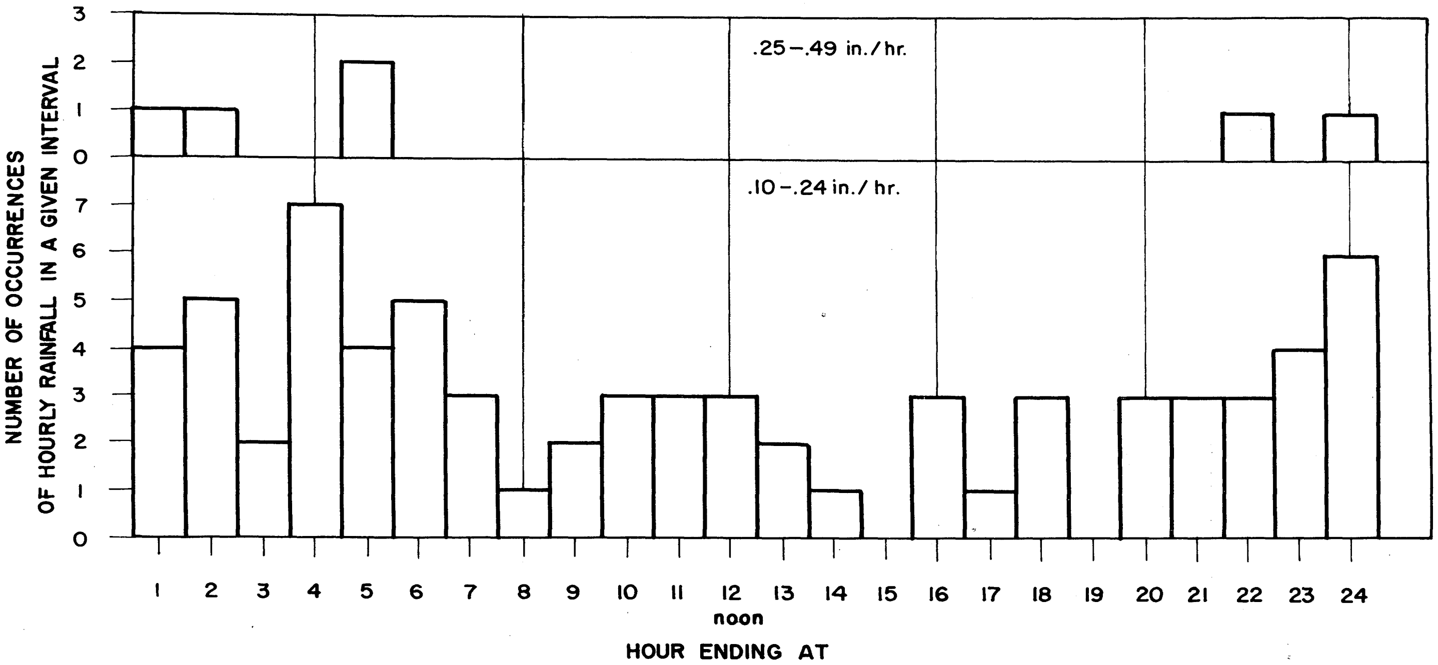
Figure 18. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE V

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

February

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01	9	1				10
.01-.019	12	3	2			17
.02-.029	8	7	3			18
.03-.039		4	2			6
.04-.049	1	3	3		1	8
.05-.099	3	5	4	1	1	14
.10-.199		1				1
Σ	33	24	14	1	2	74



February

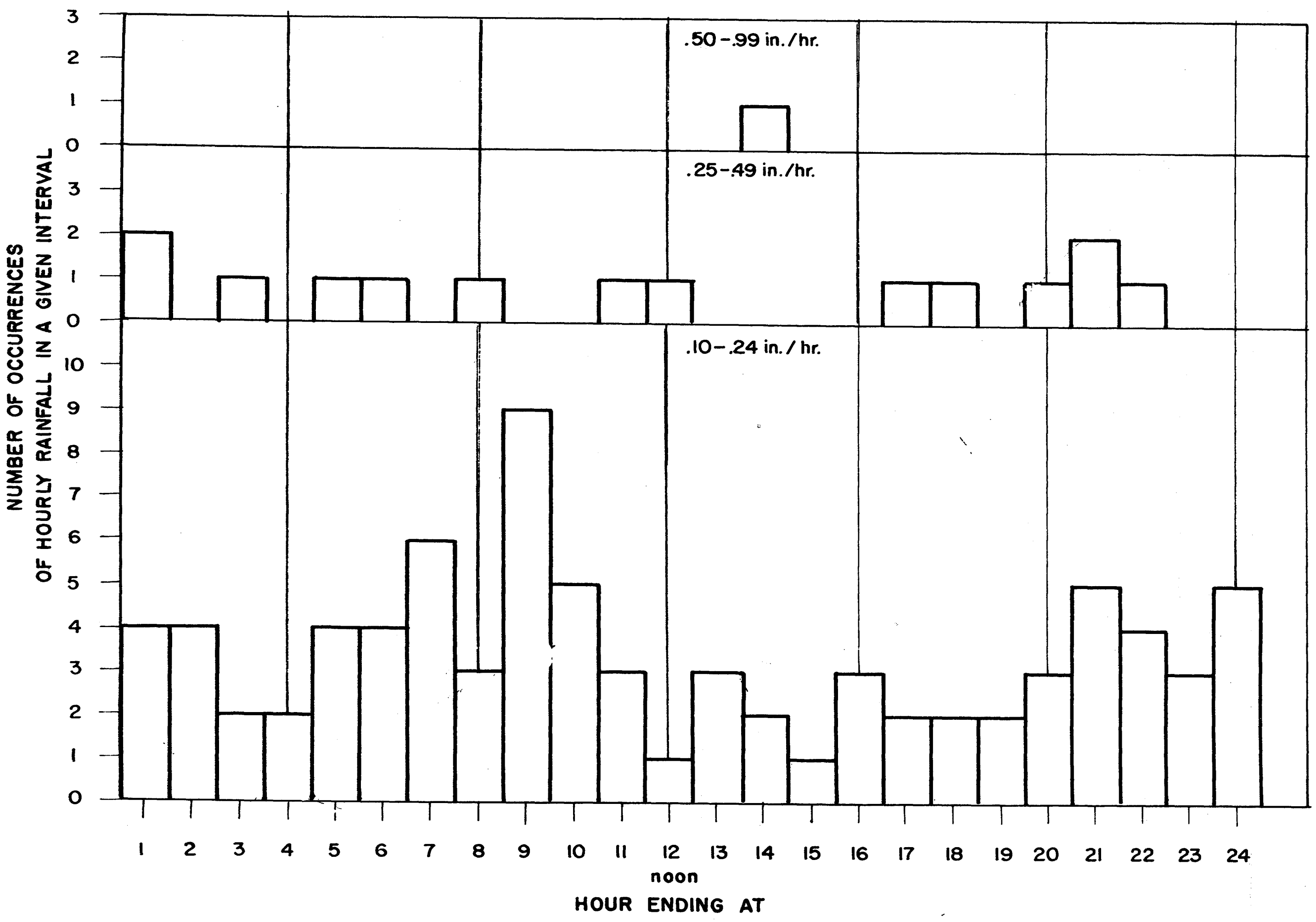
Figure 19. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE VI

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

March

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01	8	2				9
.01-.019	13	4	1			21
.02-.029	5	4	3	2		16
.03-.039	6	5	3			13
.04-.049	2	1	3			6
.05-.099	2	7	2	2		16
.10-.199	1	1		2	1	5
Σ	37	24	12	6	1	86



March

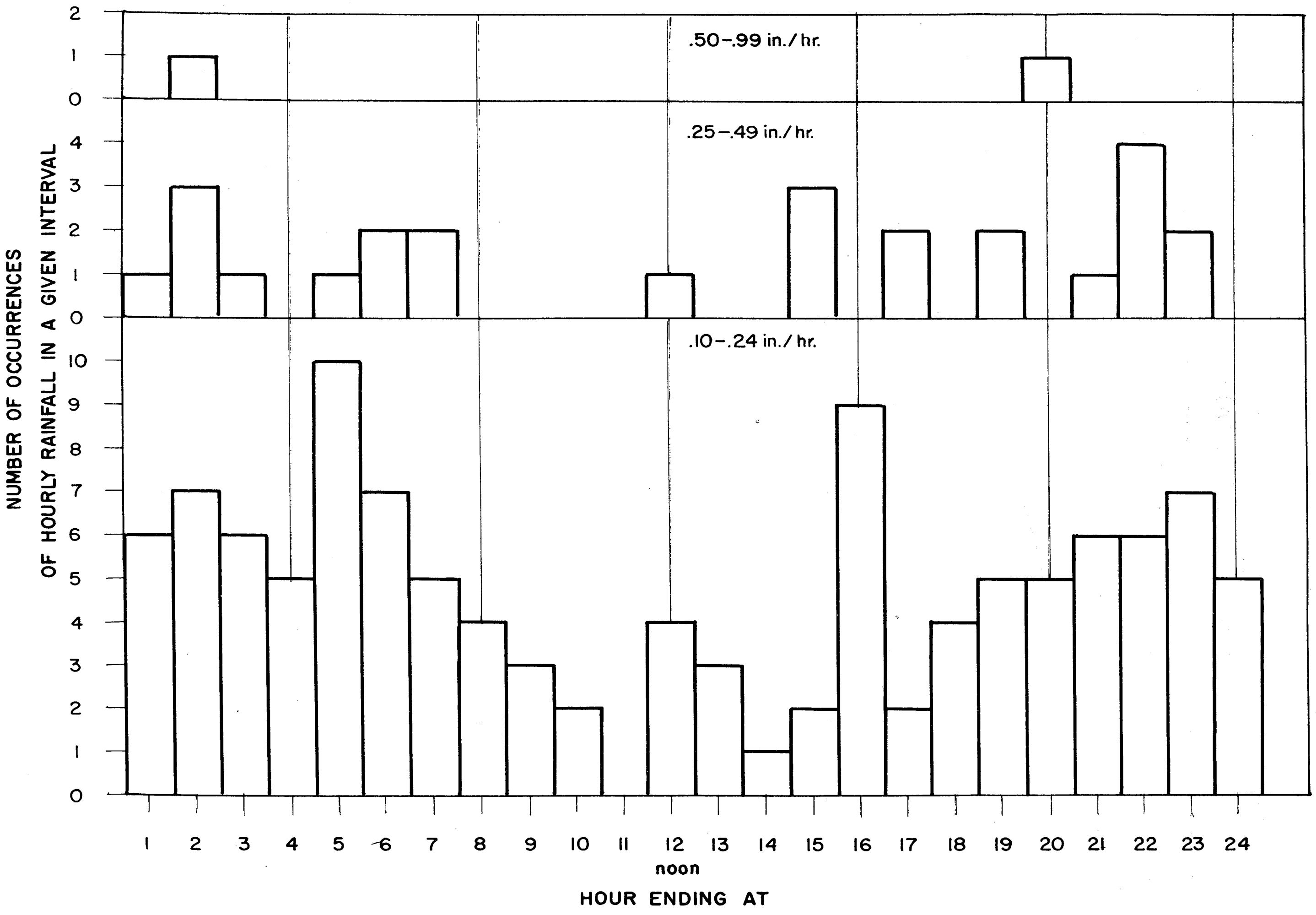
Figure 20. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE VII

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

April

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01	1	1				2
.01-.019	18	4	1			23
.02-.029	12	4		1		17
.03-.039	5	8	1	1		15
.04-.049	3	6	2			11
.05-.099	6	10	7	4		27
.10-.199	2	7	1	5		15
.20-.299	2	1				3
Σ	49	41	12	11		113



April

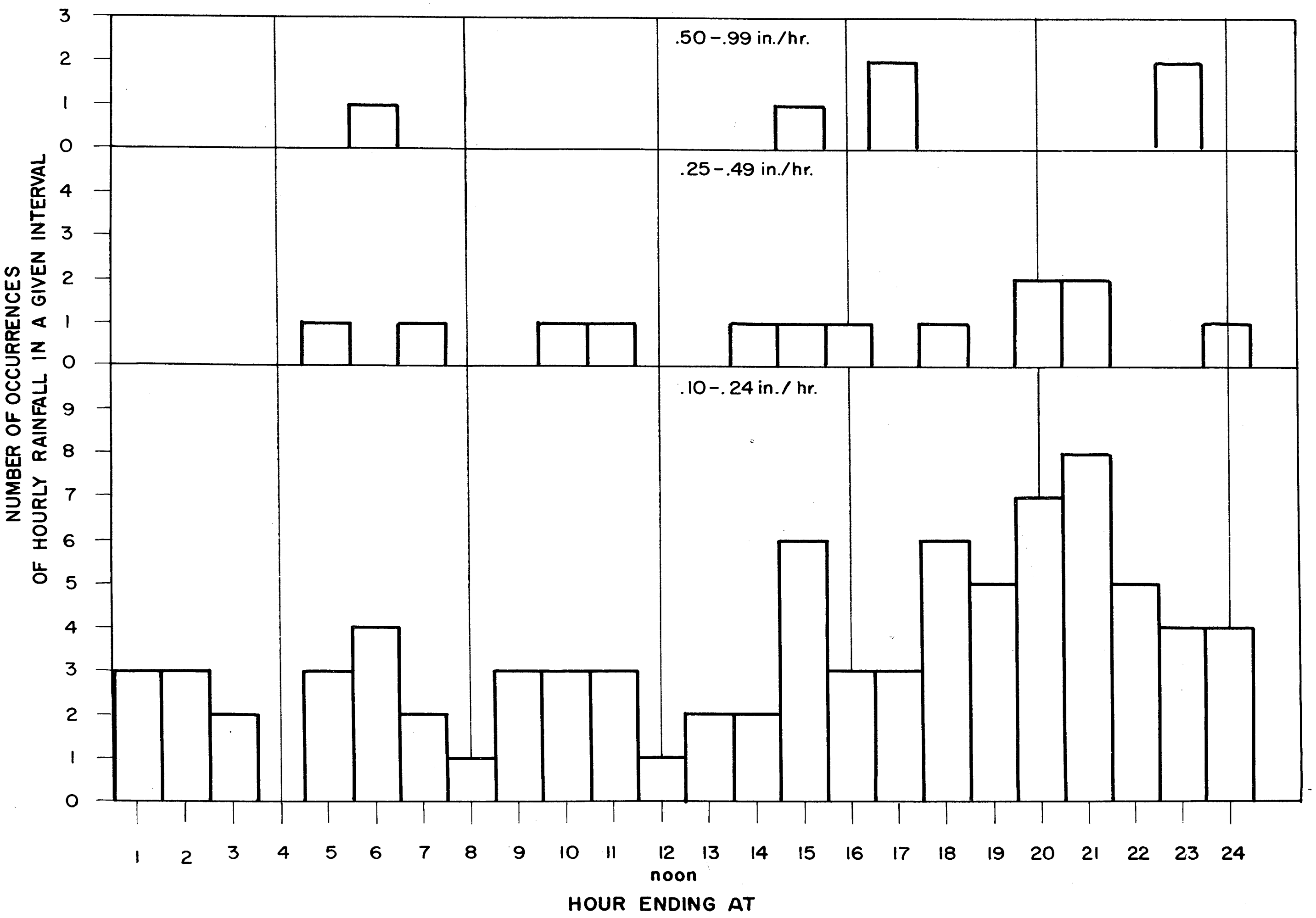
Figure 21. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE VIII

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

May

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01						
.01-.019	6	3	1			10
.02-.029	7	3				10
.03-.039	10	3		2		15
.04-.049	4	4	2	1		11
.05-.099	7	7	1	3		18
.10-.199	5	2	5	1		13
.20-.299			1	1		2
.30-.399			1			1
.40-.499			1			1
Σ	39	22	12	8		81



May

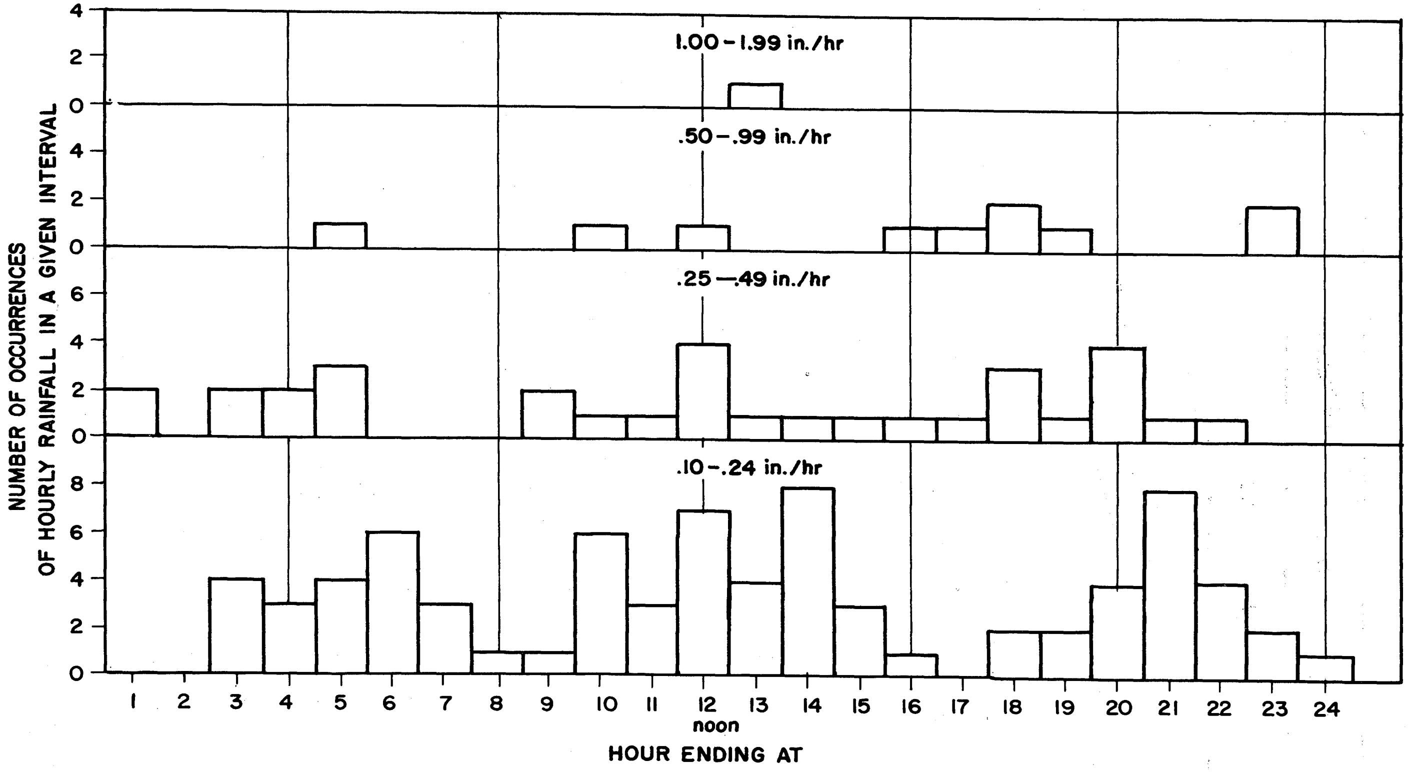
Figure 22. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE IX

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

June

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01						
.01-.019	8	3		1		12
.02-.029	4	4				8
.03-.039	6	4				10
.04-.049	2	5				7
.05-.099	17	11	8	4		40
.10-.199	1	8	8	1		18
.20-.299		2	1			3
.30-.399				1		1
.40-.499		1				1
Σ	38	38	17	7		100



June

Figure 23. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE X

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

July

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01						
.01-.019	3					3
.02-.029	6	2				8
.03-.039	7	1				8
.04-.049	3	1	2			6
.05-.099	12	14	1			27
.10-.199	5	6	6	5		22
.20-.299			2	2	1	5
.30-.399			1	2		3
.40-.499						
.50-.599						
.60-.699						
.70-.799			1			1
Σ	36	24	13	9	1	83

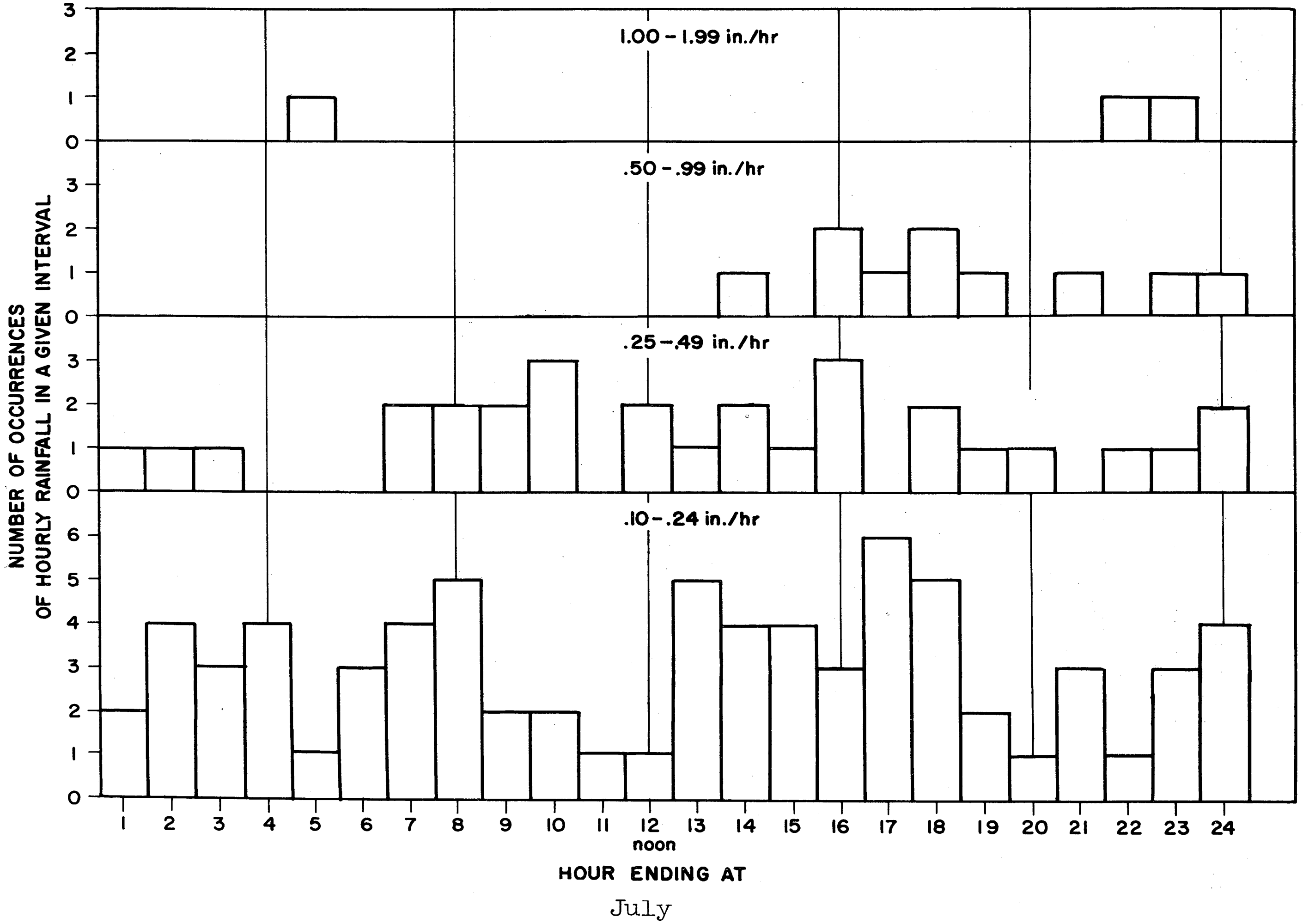


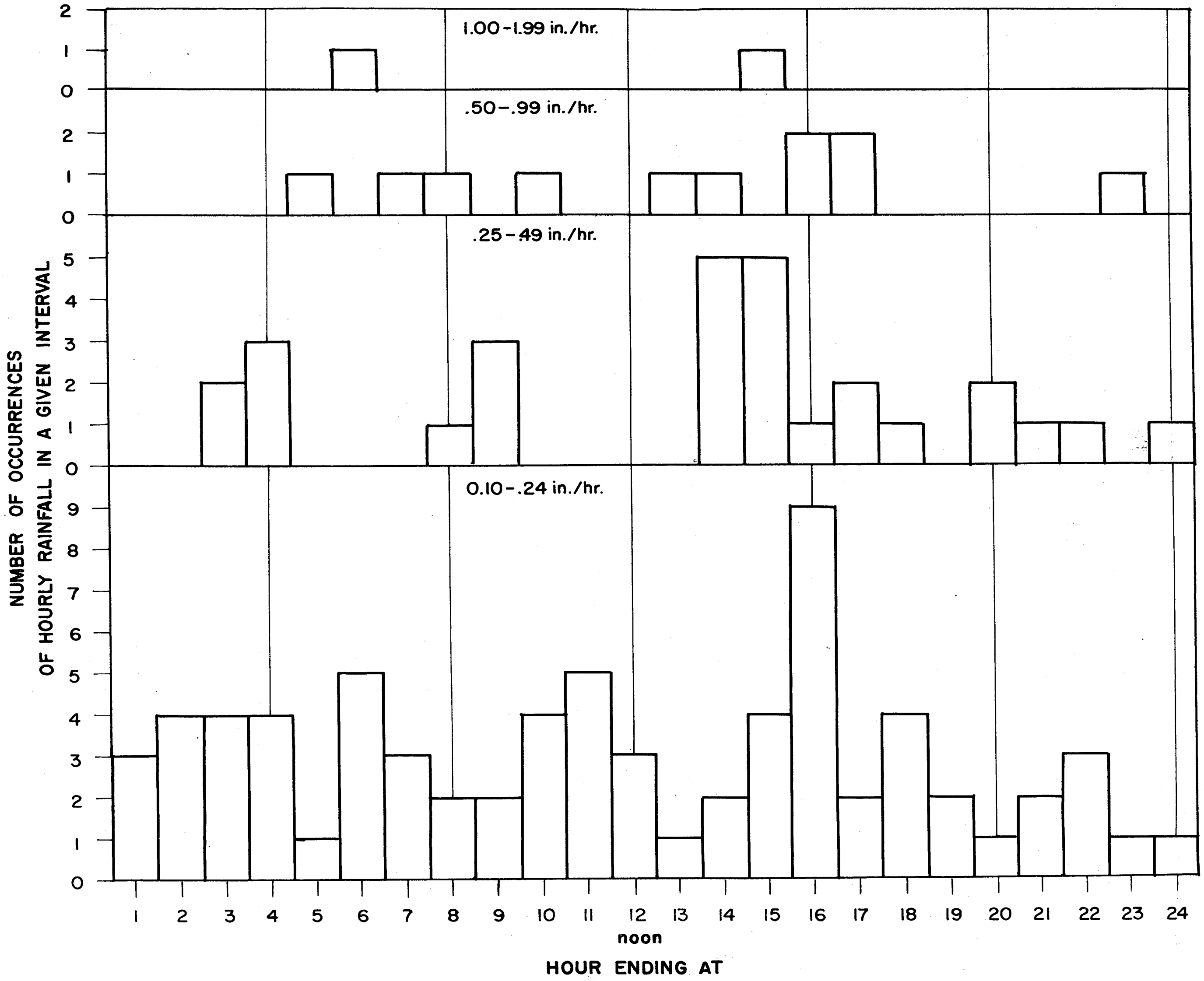
Figure 24. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE XI

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

August

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01						
.01-.019	4	1				5
.02-.029	4	3				7
.03-.039	9	1				10
.04-.049	4	5				9
.05-.099	13	9	3			25
.10-.199	3	8	9	2		22
.20-.299		1		3		4
.30-.399			1		1	2
.40-.499			1			1
.50-.599			1			1
.60-.699				1		1
Σ	37	28	15	6	1	87



August

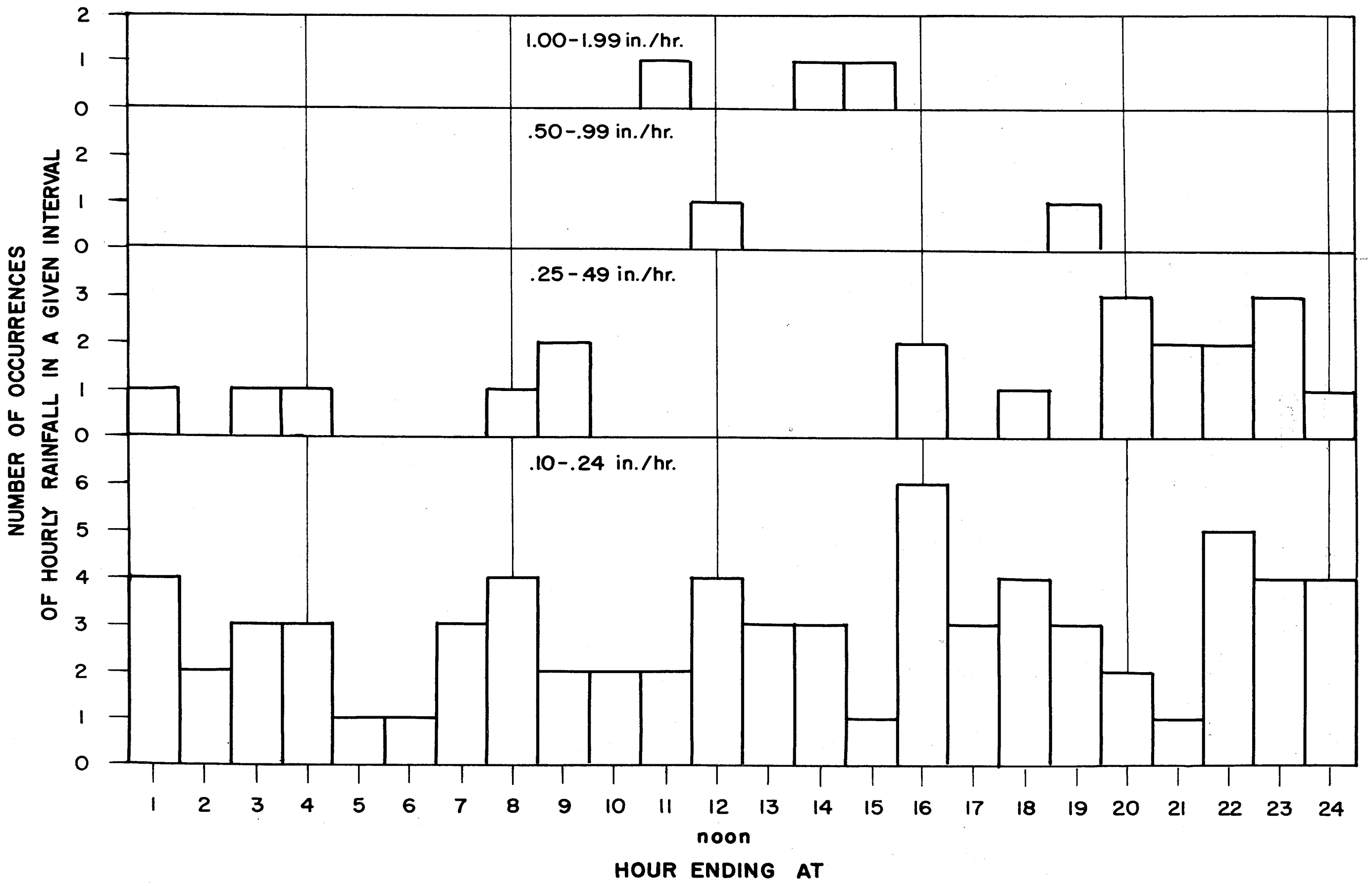
Figure 25. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE XII

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

September

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01						
.01-.019	2	2				4
.02-.029	8	2				10
.03-.039	4	5	2			11
.04-.049	3	2	1			6
.05-.099	6	6	3	3		18
.10-.199	1	5	5			11
.20-.299	1		1	2	1	5
.30-.399				1		1
.40-.499				1		1
Σ	25	22	12	7	1	67



September

Figure 26. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE XIII

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

October

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01	4					4
.01-.019	11	2				13
.02-.029	8	1				9
.03-.039	3	2			1	6
.04-.049	4	4	1	2		11
.05-.099	2	7	5	4	2	20
.10-.199	1	2	1	2	1	7
.20-.299	1				1	2
Σ	34	18	7	8	5	72

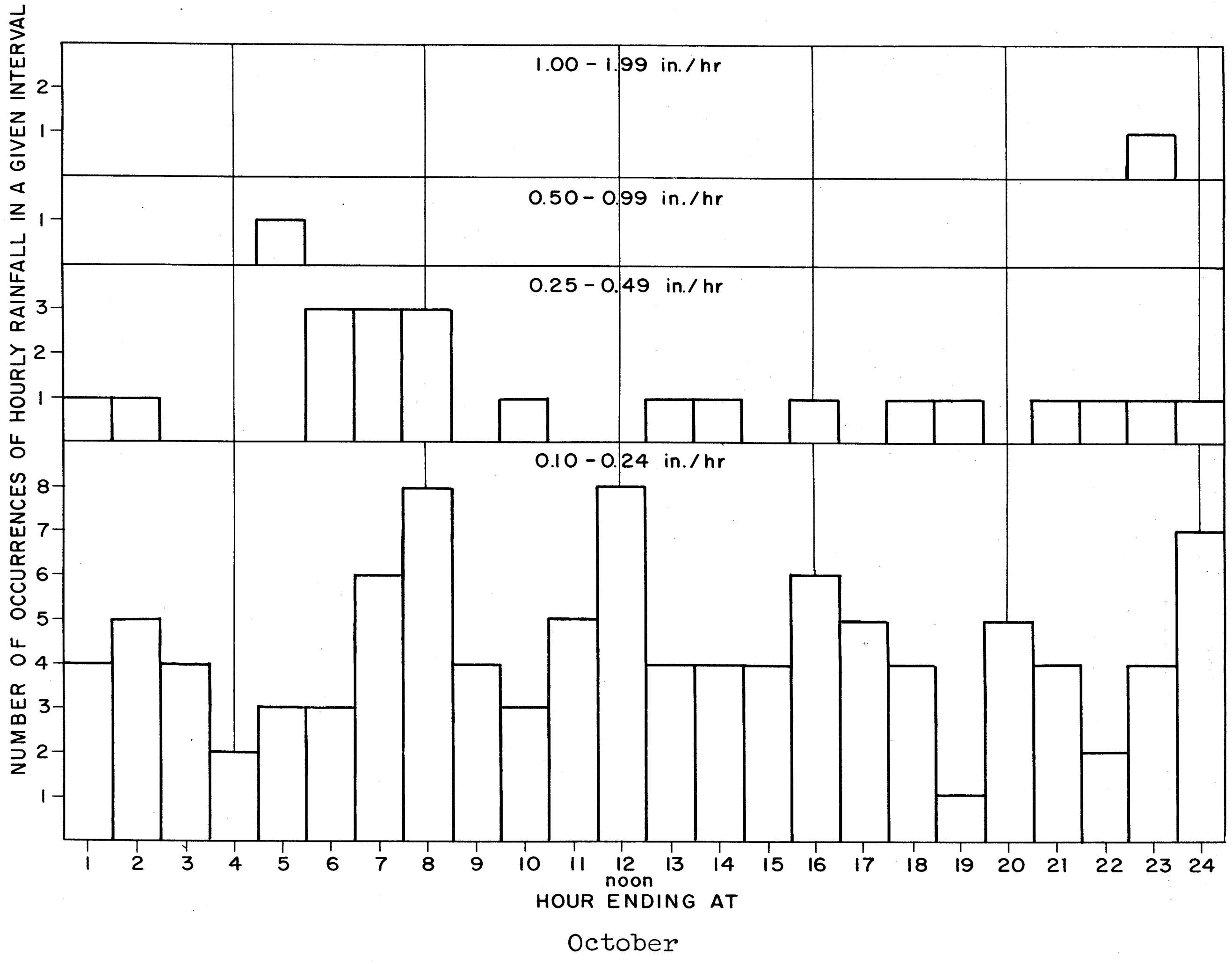


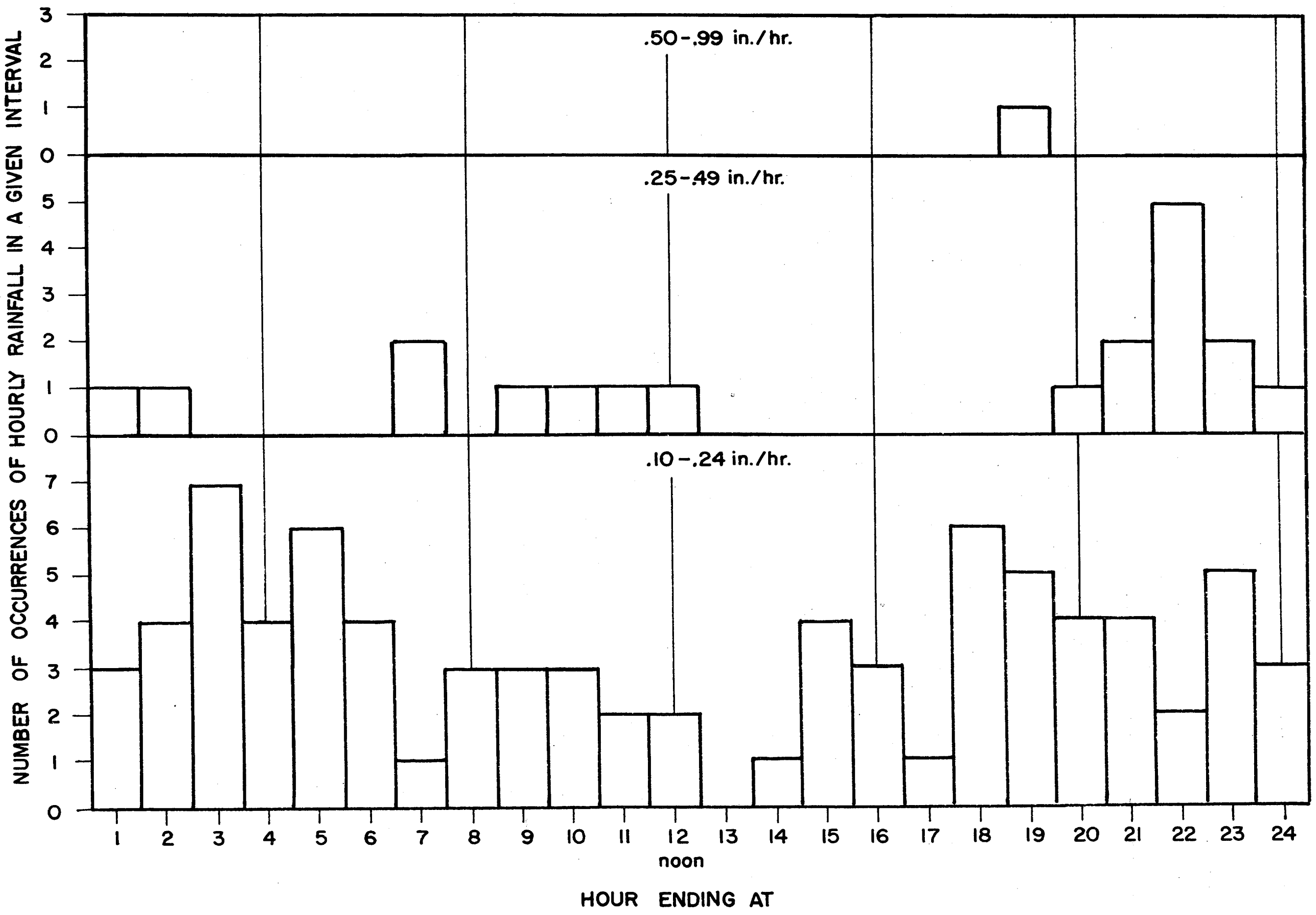
Figure 27. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE XIV

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

November

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01	3	2				5
.01-.019	17	3	1			21
.02-.029	11	6	1	2		20
.03-.039	1	4	1	1	1	8
.04-.049	2	2	2	1		7
.05-.099	3	3	5	3		14
.10-.199		1		1		2
.20-.299				1		1
Σ	37	21	10	9	1	78



November

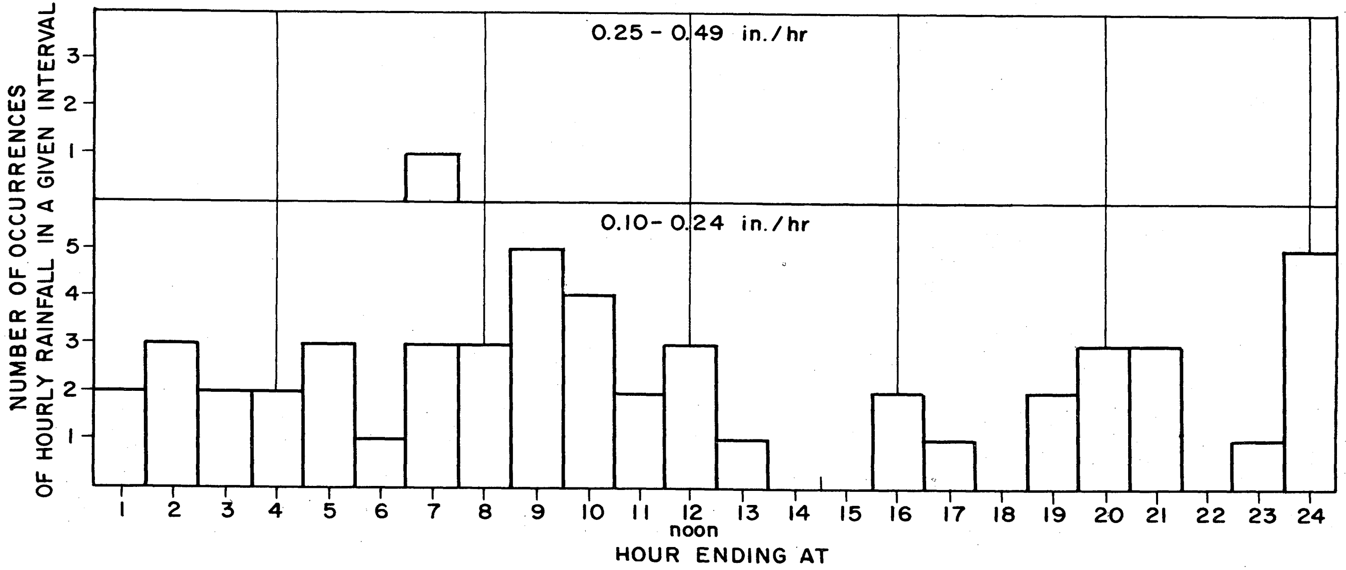
Figure 28. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

TABLE XV

CONTINGENCY TABLE—NUMBER OF OCCURRENCES OF SPECIFIED MEAN HOURLY RAINFALL PER SHOWER OF SPECIFIED TOTAL RAINFALL FOR YEARS 1948-1961

December

Mean Hourly Rate, in./hr	Total System Rainfall, in.					
	.10-.24	.25-.49	.50-.99	1.00-1.99	2.00+	Σ
<.01	9	1	1			11
.01-.019	11	4				15
.02-.029	5	10	3	1		19
.03-.039		3	6	2		11
.04-.049		1				1
.05-.099	1	2	2	2		7
Σ	26	21	12	5		64



December

Figure 29. Bar graphs showing frequency of occurrence of several classes of hourly rainfall totals for the years 1948-1961.

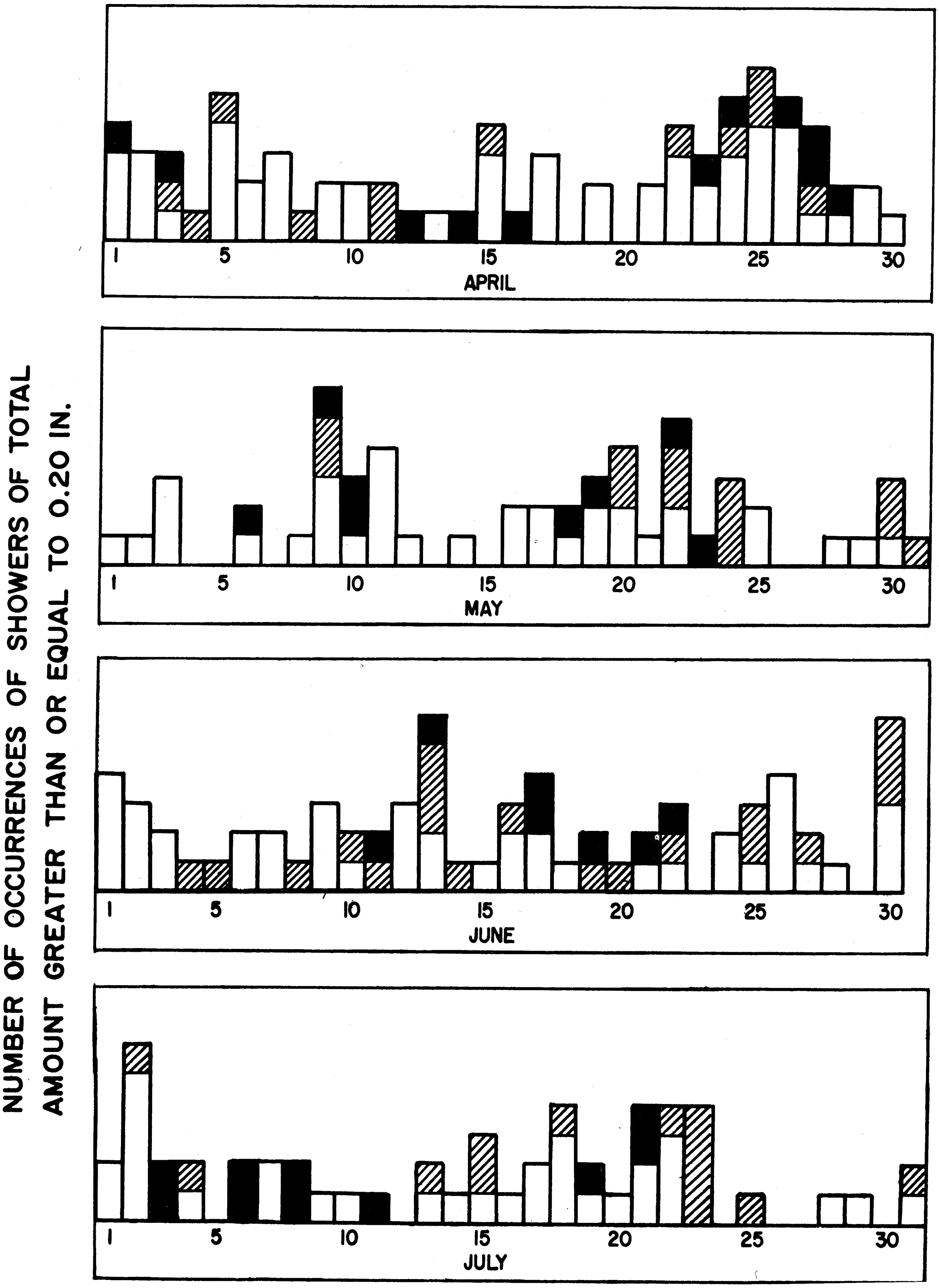


Figure 30. Number of rains of total amount at least 0.30 in. by categories (0.20 to 0.50; 0.50 to 1.00; and 1.00 and up) for the months April-November, 1948-1961.

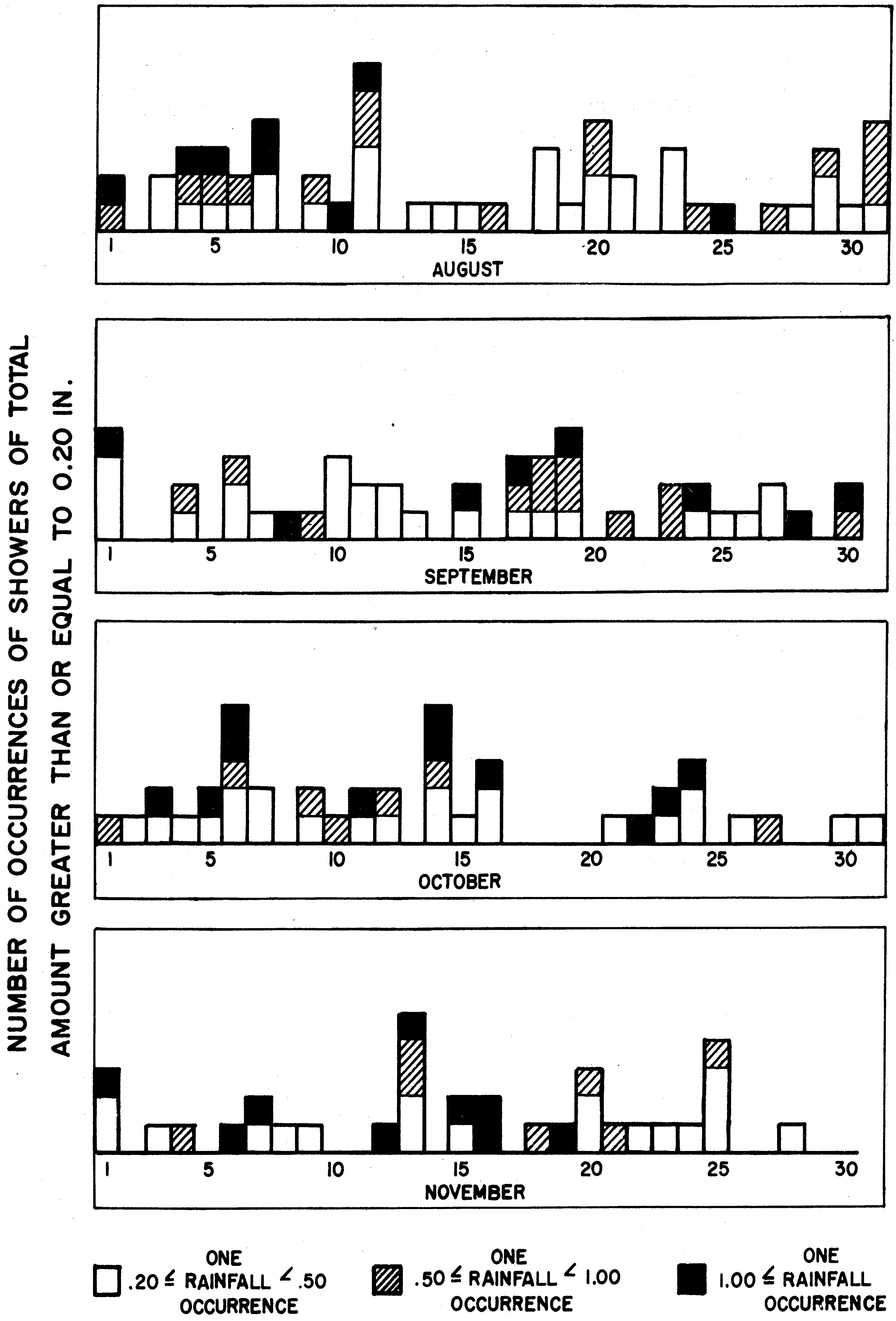


Figure 30 (Concluded)

TABLE XVI

NUMBER OF OCCURRENCES OF SPECIFIED TOTAL RAINFALL BY MONTHS, 1948-1961

Month	1.00 in.	.50-1.00 in.	.50 in. & Up
April	11	12	23
May	8	12	20
June	7	19	26
July	10	13	23
August	8	16	24
September	8	12	20
October	11	7	18
November	9	7	16

TABLE XVII

NUMBER OF OCCURRENCES OF SPECIFIED HOURLY RAINFALL AMOUNTS BY MONTHS, 1948-1961

Month	.50-.99 in.	1.00-1.99 in.
April	2	0
May	6	0
June	10	1
July	10	3
August	11	2
September	2	3
October	1	1
November	1	0
December	0	0
January	0	0
February	0	0
March	0	0

IX. THE RAINDROP SORTER

The first two reports of this series were primarily concerned with the preliminary study, design and construction of a device intended to sort raindrops into size-discriminated samples. The device chosen was modeled after the "mass-spectrometer" of Bowen and Davidson (1951). In this particular case, our initial analog studies indicated desirable modification of the simple horizontal wind tunnel design to provide for (a) better separation of the largest drop sizes and (b) better catch of the smallest drop sizes. The use of a 45° upward-tilted wind tunnel permits higher air speeds to accomplish separation at the same time that it provides a means of catching the small drops on the elevated tunnel floor within a reasonable downstream distance.

A basic assumption of the design is that the raindrops must enter the separator on a very nearly vertical trajectory at their terminal fall speeds. Obviously it is necessary to assume a trajectory and speed for design purposes. Any other assumption than this one introduces serious complications into the design and requires an expensively engineered system. In any case, it turns out that rain does not generally, nor usually, fall vertically in nature. It acquires horizontal momentum from air currents that it falls through, and at ground level these are frequently highly changeable. Consideration of a stilling basin, at the bottom of which the falling rain would have lost its horizontal momentum led to the conclusion that only a very large and deep depression could serve this purpose. It appeared not to be feasible to fulfill any reasonable design criteria for the required stilling basin. At the same time, it appeared that such a basin would itself introduce changes in the wind field, and distortions in the rain catch. In short, it is our considered opinion that the engineered solutions to the problems associated with variable low level winds, either in terms of machinery to orient the sorter with respect to the raindrop trajectories, or in terms of the stilling basin, are too costly to be clearly justifiable, and are further disqualified because of additional deleterious effects and errors.

One clear possibility to procure useful information with the raindrop sorter remains; this is offered by those situations in which the surface wind remains relatively steady for a considerable time. By operating the raindrop sorter throughout many rainy situations, in parallel with the other sampling devices, and by paying careful attention to the behaviour of the wind, changing sorter samples at principal changes of wind speed or direction, we believe that over an extended period we can obtain meaningful data.

A. RESULTS TO DATE

During the sampling periods of fall, 1961, and spring, 1962, the raindrop sorter was used to obtain samples of rain. The floor of the drop separator was divided into five catch-basins, each of which collects a portion of the raindrop-size spectrum under a given wind condition. The dimensions of the catch basins, and the drop-size interval collected by each under the no-wind condition are given in Table XVIII.

TABLE XVIII

SPECIFICATION OF THE DROP-SEPARATOR SIZE CATEGORIES
FOR CONDITION OF NO-WIND

Drop Size Category	Distance Interval from Entrance Slit	Drop Diameter Interval (mm)
1	40.64-65.25	0.65-0.79
2	15.24-40.64	0.79-1.3
3	7.94-15.24	1.3 -2.0
4	3.33- 7.94	2.0 -3.0
5	0.00- 3.33	> 3.0

These specifications apply for wind tunnel speed of 6.0 m sec^{-1}. Our calibration studies showed that something less than the computed separation for a 4 m sec^{-1} speed (Dingle, 1961) was attained in practice because of the influx of air at the entrance slit. This produces a fall speed in excess of the terminal speed, which reduces the drop separation, hence a higher sorter speed was used to obtain the separation indicated in Table XVIII.

Data collected to the present time are listed in Table XIX. The sample code numbers give the following information:

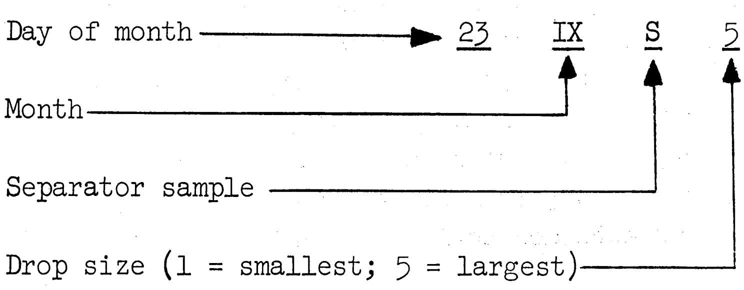


TABLE XIX

RAINDROP SORTER SAMPLES, 1961 AUTUMN AND 1962 SPRING, AND SAMPLE ANALYSES

Sample Code Number	Sample Volume (ml)		Total Solids	Activity (c/m/ml)
	Observed	Computed		
14 IX S 1 (2.5-)	75	16.5	.0003	0.0264 ± .00376
23 IX S 1+2	360		.0314	0.105 ± .00250
S 2+3	15		.0039	0.140 ± .0186
S 5	110		.0131	0.241 ± .00217
	485	178		
1 V S 2	62.4		.0116	0.954 ± .040
S 3	45.1		.0040	0.570 ± .026
S 4	44.2		.0058	0.405 ± .018
	151.7	*		
23 V S 1	69.4		.0148	1.92 ± .074
S 5	44.2		.0127	1.70 ± .063
	113.6	88.9		
4 VI S 1	33.7		.0099	1.90 ± .075
S 2	3.1		.0019	3.17 ± .153
S 3	2.2		.0028	4.67 ± .182
S 4	3.2		.0014	3.56 ± .155
S 5	4.1		.0033	1.93 ± .104
	46.3**	91.5		
11-12 VI S 1	69.8		.0199	1.88 ± .073
S 2	17.3		.0080	1.90 ± .075
S 3,4,5	43.8		.0068	1.10 ± .053
	130.9	100.0		

*Unknown, but certainly much less.

**This rain is probably all right. Samples were collected the following day, and some evaporation may have occurred.

An elementary check on the amount of water collected was made by summing the respective sample volumes and comparing these to the volume that should have been caught from the given rain falling through our entrance slit. In every case except 4 VI, the sorter collected much more rain than its share.

A convergence of water at the intake slit is expected by virtue of the flow of air through the intake into the working section. Preliminary analysis of the air intake to the drop sorting tunnel indicates that the air flow through the rain intake slit is about 0.1 that through the bell mouth, and that it attains a speed of about 8.7 m sec. It is not clear precisely what the quantitative effect of this flow may be upon the liquid water intake. This depends heavily upon the drop-size distribution of the rain, and must therefore be quite variable both within and between rains. A major source of error in the present case is probably associated with the entrainment of splash droplets produced by the impaction of rain upon nearby surfaces. This obviously has the greatest effect upon the smallest drop-size fraction, as observed in Table XIX.

B. POTENTIAL OF THE METHOD

Although various steps can be taken to reduce the variability of the data (e.g., splash suppression by means of fine screen over surfaces near the intake slit, careful recording of low level winds and separation of samples on the basis of wind-change criteria), it is clear that the sorter samples will always be rather "noisy." At this point, however, it cannot be said that the method has been given a sufficient trial to permit its complete evaluation.

We believe that such an evaluation should be based upon an accumulation of samples collected under all rain conditions and selected by reference to wind-behavior criteria derived from records of wind speed and direction near the intake slit. To collect a suitable set of samples, the raindrop sorter should be operated in all rainstorms in conjunction with other sampling devices, and with meteorological instrumentation designed to show the conditions under which the samples are collected.

Elsewhere in the present report, and in a published paper (Gatz and Dingle, 1963), we have pointed to the need for sub-meso-analysis of rain systems to determine details of the circulations into and through the parts of the rain systems that generate the rain collected by our samplers. A sound field program of this sort will be of great value in studying the rain scavenging function as well as the detailed structure of rain-producing weather systems. The information obtained will obviously be useful for specific analysis of the observed systems, but will also serve to provide the guidelines for model computations leading to generalization of the results. The raindrop-sorter should be used in conjunction with such a program.

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