Air Cleansing by Convective Rains[1]

A. NELSON DINGLE AND DONALD F. GATZ
The University of Michigan, Ann Arbor

ABSTRACT

The rain cleansing patterns observed in five rain situations, all characterized by convective structures, are presented. The data comprise radioactivity and pollen concentrations measured in sequential samples of rain water from each rain. The rain systems are categorized, three being classed as "well-organized," and the remaining two as "diffuse" convective systems. The temporal patterns of rain water contamination are relatively consistent in the well-organized showers, but much less so in the diffuse ones. Because the scavenging patterns of the pollens are quite parallel with those of the long-lived radioactive debris, it is inferred that both classes of contaminant enter the rain-producing parts of the showers by the same route, namely, in the low-level air that forms the convective updraft. It is therefore proposed that contamination that enters the troposphere from above (the stratospheric nuclear test debris reservoir) is not importantly scavenged until it passes through a low level tropospheric trajectory. The cleansing accomplished by the diffuse showers shows a tendency to be quite spotty, leaving pockets of contaminated air relatively unaffected between showers.

1. Introduction

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 *et al.*, 1962) as well as from sample to sample within a single rainfall. Variations within rains have been reported by others (Dingle, 1961; Salter *et al.*, 1962) and are also present in the five rains discussed in the present paper. Because the variability within a system is pertinent to the understanding of that between systems, the principal focus of the present paper concerns variations within convective rains.

2. Observational program

A basic premise of our research 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. The sampling pans. Rain was collected in two large pans, which together total 5.2 m² in area. The sampling pans were located on the flat roof of a temporary structure, shown in Fig. 1, in which sample-bottling operations were carried out. The pans were tilted slightly to provide drainage. The rainwater was conveyed from the pans to the bottling station directly below by flexible plastic tubing.

b. Sampling procedure. 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-gal samples were taken for radiochemical analysis and 1-qt samples were taken for pollen analysis. When radionuclides alone were to be determined, successive 1-gal samples were taken.

c. Complementary data. In addition to data obtained from the radiochemical analyses of rain samples, supporting data on the nature of the precipitation

FIG. 1. View showing location of rain sampling pans.

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system which produced the rain were taken at the collection station and acquired elsewhere. These additional data include conventional synoptic weather data and radar observations of movement of precipitation areas and height of echo tops.

Rainfall rate data were obtained by a tipping-bucket recording rain gauge. This instrument records an event mark on a strip chart recorder after every 0.01 inch of rain. Rate of rainfall is therefore inversely proportional to the time between successive marks.

Rainfall rate data from the tipping-bucket rain gauge were used to provide the detailed time profile 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.

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

3. Sample analysis

a. 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 and counted for radioactivity. The filters were dried, placed into identical planchets, and similarly counted. Early in the program, the samples were counted only for gross beta activity 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.

In each rain water sample, fission-product radionuclides were determined in the following three groups by gamma spectrometry:

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. The necessity for grouping the radionuclides as indicated arises from the analysis method.

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 photo-peak in the spectrum of an unknown may be assigned to a specific radionuclide. In practice, however, it is sometimes found that gamma rays from different nuclides are of nearly equal energy and cannot be separately resolved by the spectrometer. It is this complication which leads to the above groupings of radionuclides. In such cases only the total of the several radionuclides may be determined.

In the investigation of the mechanisms whereby fission products are brought to earth by rain, the changes of activity levels with time provide the best clues. Thus, one is more concerned with the precision of the analysis procedure than with its accuracy. The precision of sample preparation and counting is estimated to be approximately 15 to 20 per cent. This is

Table 1. Common names and approximate diameters of all of the pollens determined in the rain water samples. The diameters reported are the means of a small number of measurements made on the pollens found in the rain water.

Pollen	Common name	Approximate diameter (μ)
Ambrosia artemisiifolia	Ragweed	20
Carya	Hickory	47
Chenopodiaceae-Amaranthaceae	Goosefoots-Pigweeds	25
Gramineae	Grasses	29
Juglans	Walnut	42
Picea	Spruce	75
Tilia	Linden	42

Table 2. Summarization of characteristics that tend to distinguish "well-organized" from "diffuse" convective rain-producing systems.

Characteristic	Well-organized	Diffuse
Total rainfall	Over 15 mm	Less than 15 mm
Duration	Order 1 hr or less	Order 2 hr or more
Overall average intensity	15 mm hr^{-1} or higher	7.5 mm hr^{-1} or less
High intensity portions	Considerable above 25 mm hr^{-1}	Little or none above 25 mm hr^{-1}
Electric acitvity	Usual	Not usual
Cleansing effect	Prominent at beginning	Obscured

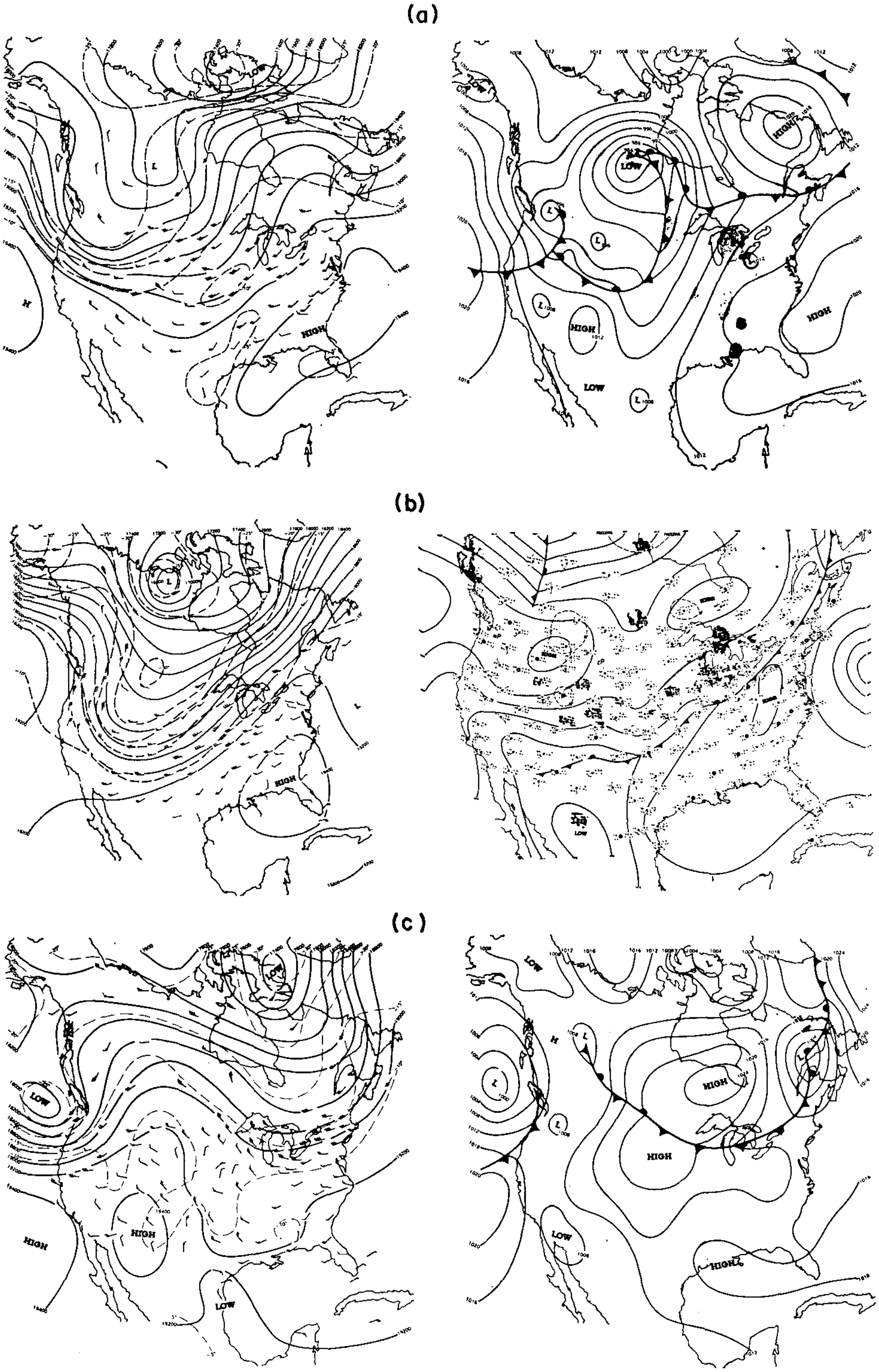

FIG. 2. Synoptic weather data for well-organized systems: (a) data for rain of 1 September 1961, 500-mb data at 1900, 1 September, surface data at 1300, 1 September; (b) data for rain of 23 September 1961, 500-mb data at 1900, 23 September, surface data at 0100, 24 September; (c) data for rain of 25 June 1962, 500-mb data at 1900, 25 June, surface data at 1300, 25 June. All times are EST.

entirely acceptable in view of the magnitude of observed sample-to-sample variations.

It is shown below that time variations of the several radionuclide groups are largely parallel in nature. In addition, random errors should be minimized by computing the total of the measured radionuclides. Therefore, unless specified to the contrary, only the variations of the total are considered in the discussion. For convenience, the "concentration of total measured radionuclides" is sometimes referred to as the "concentration."

b. Pollen analysis.[2] Insoluble material was separated from each sample by centrifugation. A known amount of a readily-identifiable tracer pollen, which would not normally be found in rain and a small amount of glycerine jelly were thoroughly mixed with the insoluble residue. Several microscope slides were prepared from each sample. Each slide was made by placing a single drop of the jellied mixture on a slide and covering it with a glass cover slip. The number of grains of each pollen species of interest and the tracer pollen were counted under a microscope for one or more slides from each sample. Under the assumption that equal fractions of the total number of tracer and "natural" pollens were present on the slides counted, it was possible to compute the total number of pollens of each species in the whole sample. These figures were converted to concentration units by dividing by the volume of the sample. A standard deviation of less than 10 per cent is estimated for most samples. Table 1 gives the common names and sizes of the pollens determined in the rain water samples.

4. Results

Among the rain-producing events that we wish to present here, a broad categorization into two types appears to be reasonable. We shall, therefore, consider three of these events as having involved relatively well-organized convective systems in contrast to the relatively poorly organized, or diffuse, convective activity of the other two. The descriptive characteristics and quantitative criteria which pertain to each category and indicate the distinctions between them are presented in detail below, and summarized in Table 2.

a. Well-organized systems. Three of the observed convective rain-producing events are considered to fall into this category. In broad terms these three events are distinguished by a rainfall yield of 15 mm or more within a period of the order of 1 hr or less. The events that fit these criteria are those of 1 and 23 September 1961, and 25 June 1962.

1. Synoptic conditions. It is appropriate to examine the weather maps to note superficial indications of similarity, and difference, among these three events. U. S. Weather Bureau Daily Map Series analyses for the nearest appropriate synoptic times, for the 500 mb and the surface levels are shown in Fig. 2.

Whereas the rain of 1 September was associated with a non-frontal convergence zone which had migrated northward from the Gulf of Mexico, both of the other rains were associated with cold frontal passages.

In the case of 25 June, the flux of Gulf air into the system over southern Michigan appears very poor, whereas that for 1 September is excellent throughout the lower half of the atmosphere, and that for 23 September is much less vigorous, depending mainly on a very weak divergent flow from the anticyclone just west of the Appalachian chain.

2. Rainfall intensity. The rainfall intensity graphs were formed by detailed reduction of the tipping-bucket rain gauge records. The results are shown in Figs. 3, 4, and 5. Here the 1 September rain is distinguished from the other two by the persistence of rainfall rates of 20

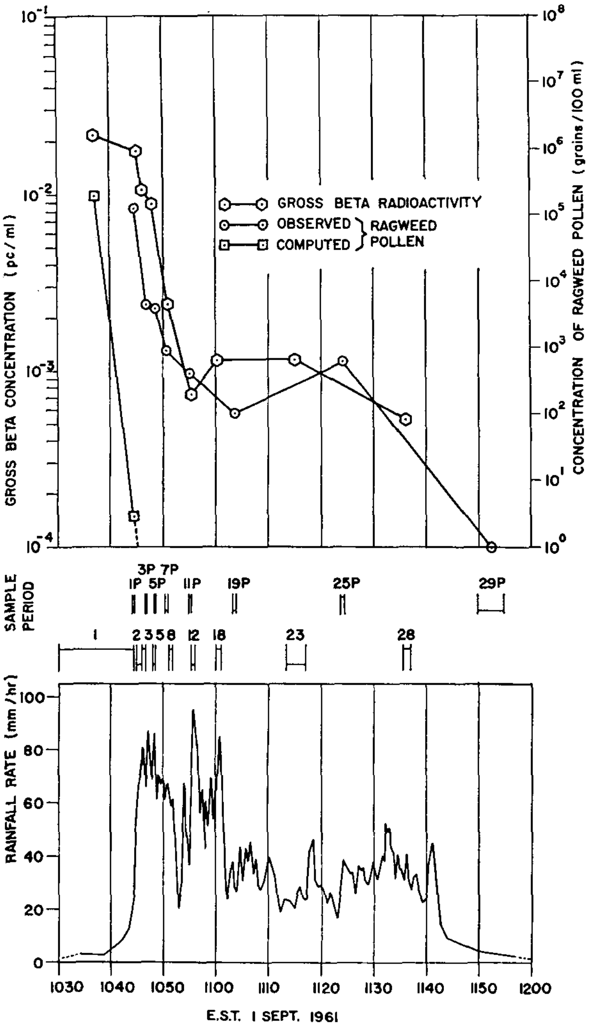

Fig. 3. Results of rain water analyses and rainfall rate data for 1 September 1961. The curve of computed ragweed pollen concentration is explained in the text.

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[2] An alternate procedure to the one described here was used for the rain of 1 September 1961, and has been described by Gatz and Dingle (1963).

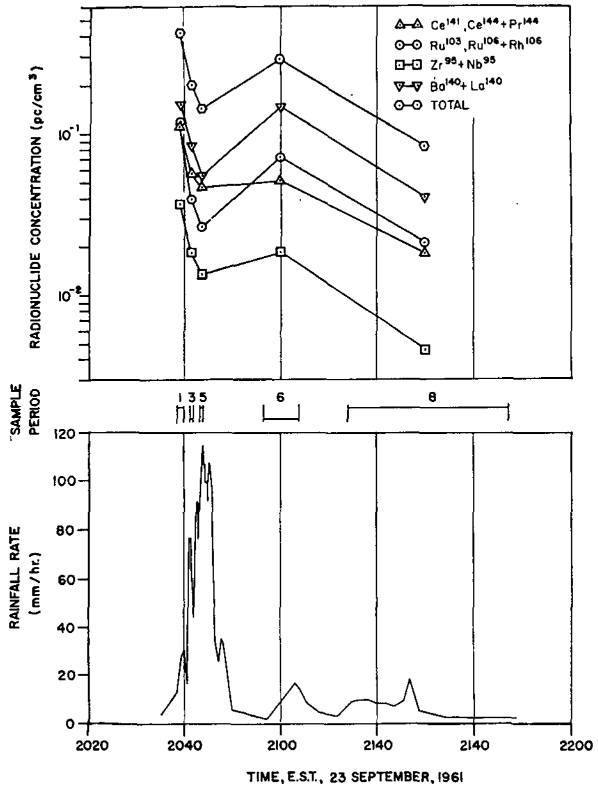

FIG. 4. Results of rain water analyses and rainfall rate data for 23 September 1961.

mm hr^{-1} and above. The two initial showers, which averaged about 70 and 60 mm hr^{-1}, respectively, lasted for about 10 min each. This feature is quite similar to the 12-min showers of 60 to 70 mm hr^{-1} average intensity which initiated both the 23 September and the 25 June rain. As a result of the sustained moderate intensity of the 1 September rainfall, it yielded a total of 47.2 mm whereas the yield of the 23 September rain was 17.8 mm and that of the 25 June rain was 16.3 mm. Maximum intensities observed were 95, 114, and 133 mm hr^{-1}, respectively, showing an inverse correlation of this characteristic with the total yield. Both cold frontal rains exhibited suppressed shower activity toward the end of the rainfall, that on 25 June reaching nearly 50 mm hr^{-1}, and that on 23 September nearly 20 mm hr^{-1}. All of the showers exhibited lightning and thunder, but data on the duration and intensity of electrical activity are lacking.

Interpretation of the rain intensity graphs and data on rates of motion of the respective weather systems suggests that the basic major shower units have a dimension of the order of 4 to 5 n mi along the direction of motion. Those of the 25 June system appear to be smaller. The fact that the initial shower in this case developed very rapidly just prior to the onset of rain at the station suggests that, at a more mature state of development, the basic elements would have joined and given an impression of structure more nearly comparable to the other systems.

Finer structure, of the order of 1 min or less in duration, or 0.5 n mi or less in extent, is indicative of the shaft-like character of heavy rain which is seen in radar RHI presentations. The relation of these two scales of rain structure to the cleansing action of the rain and to the entrainment of contaminated air is explored below.

3. Contaminant concentrations. Graphs of the concentrations of pollen and of radioactive materials are given in the upper parts of Figs. 3, 4 and 5. The periods during which the samples of rain water were collected are shown in the middle of the figures. The samples whose number designations are modified by "P" were analyzed for their pollen content only. Since the natural

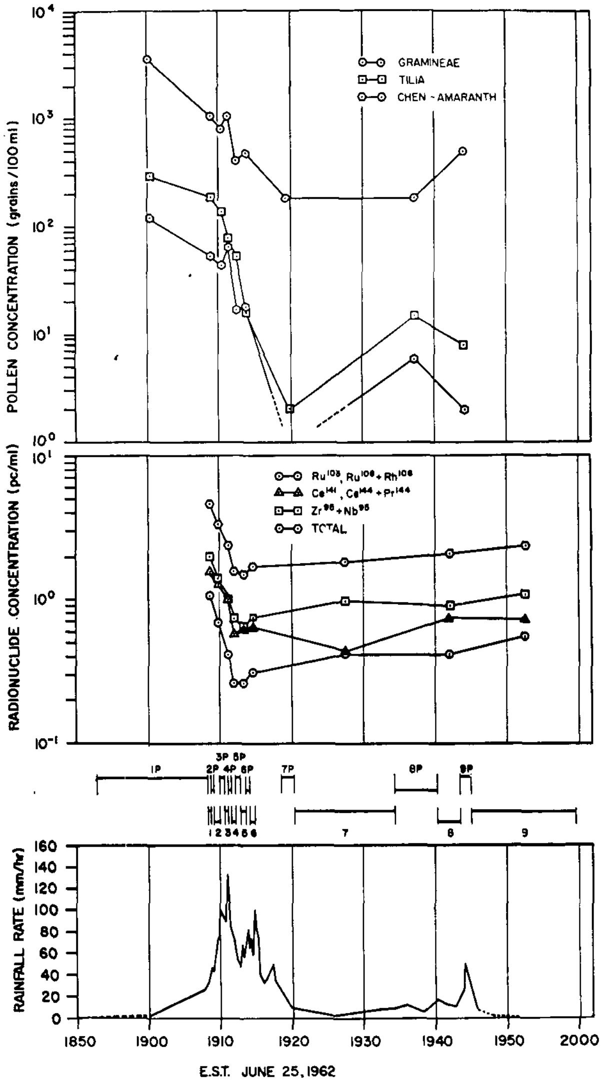

FIG. 5. Results of rain water analyses and rainfall rate data for 25 June 1962. The common names and diameters of the several pollens are given in Table 1.

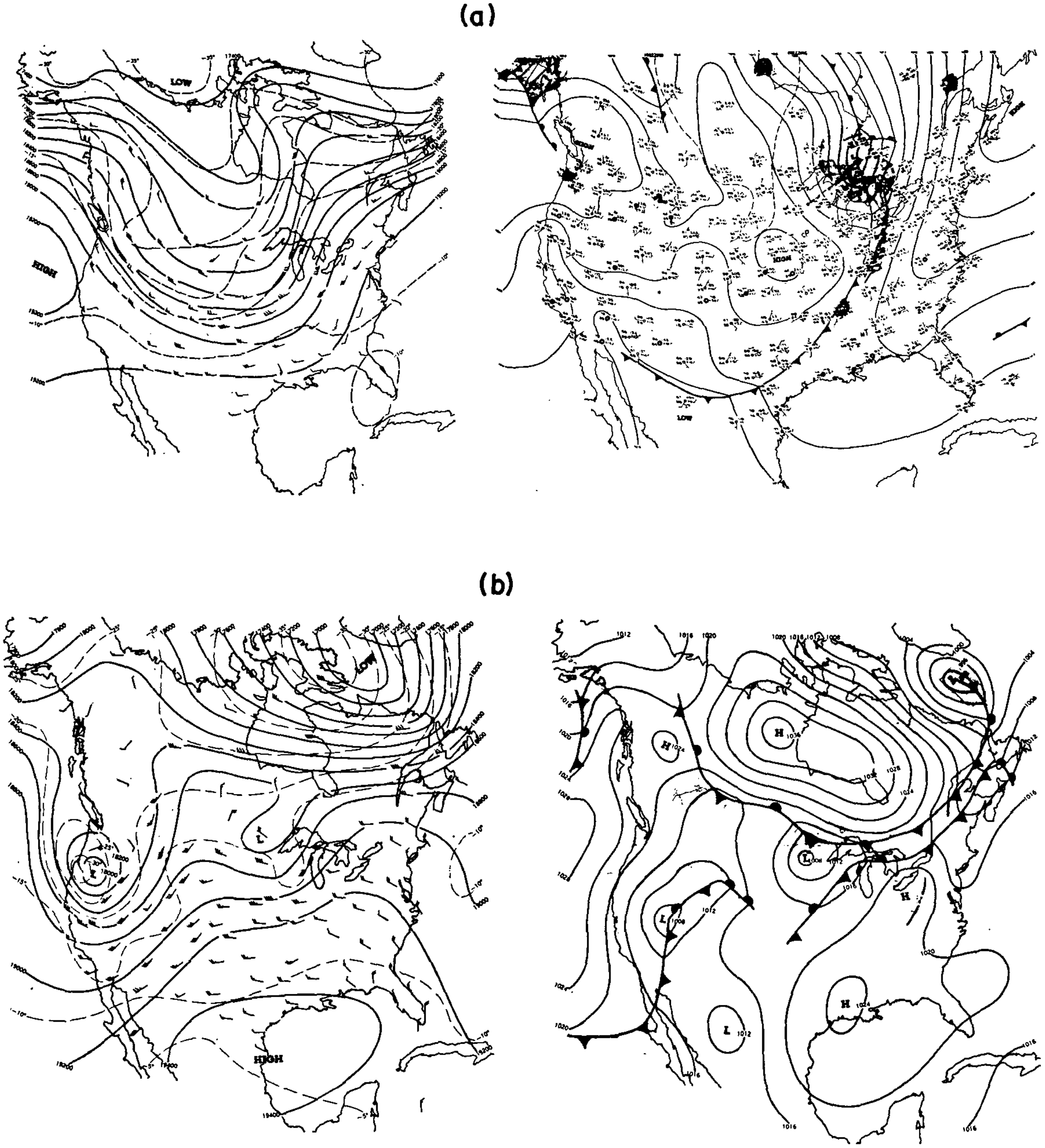

FIG. 6. Synoptic weather data for diffuse systems: a) data for rain of 30 September 1961, 500-mb data at 1900, 30 September, surface data at 0100, 1 October; b) data for rain of 19 May 1962, 500-mb data at 1900, 19 May, surface data at 1300, 19 May. All times are EST.

pollination seasons ended before 23 September, no pollen analysis was made for this rain.

In each of the three cases, and for both classes of contamination, an early sharp decrease of concentration is observed in the initial heavy shower. Subsequent increases and decreases also demand interpretation. Unfortunately, the complete continuum of samples was not analyzed for any of these three rains. The results are sufficiently interesting that this error will not be repeated.

b. Diffuse systems. Two rain-producing events among those reported here are considered to fit the category of diffuse convective activity as described above. These are the events of 30 September 1961 and 19 May 1962.

1. Synoptic conditions. The nearest appropriate U. S. Weather Bureau maps for the 500-mb and surface levels, are shown in Fig. 6. The rain collections on 30 September were obtained from two prefrontal squall lines, the first of which preceded the cold front by about

90 n mi. No thunder was observed, and the structure in general was a loose composite of relatively weak convective cells.

The rain of 19 May took place in apparently convergent flow in warm air without benefit of a direct association with identifiable frontal or squall passages. Radar observations showed no well-developed cells of strong convection, all echo tops occurring below about 36,000 ft.

2. Rainfall intensity. The rainfall intensity graphs for these two events are shown in Figs. 7 and 8. Transit of the station by rain-producing cells is clearly indicated. The precise position of the station with respect to the central or most intense rain shaft is not known, but a near-diametric transit is probably indicated by the maximum intensity spikes of 38 mm hr^{-1} on 30 September and 14 mm hr^{-1} on 19 May. The low intensity portions are apparently composed of stable rain and spray between active cells. The total rainfall amounts from these systems were 9.4 mm and 6.9 mm, respectively.

3. Contaminant concentrations. In both cases, the changes of the contamination levels of the rain samples are considerably different from those observed in the well-organized systems. Again, pollens were not present in sufficient number for a meaningful pollen analysis of the 30 September rain samples. In the 19 May rain, the pollens appear to have been progressively washed out except for the upward tendencies of samples 4P and 6P. The radioactive materials, on the other hand, show peculiar tendencies in both cases. It should be noted that there is considerable likelihood that tropospheric (intermediate fallout) radioactive debris was present in both cases. Necessarily, a low frequency of sampling is associated with low intensity rainfall. In both of these situations, however, samples were taken continuously throughout the rains.

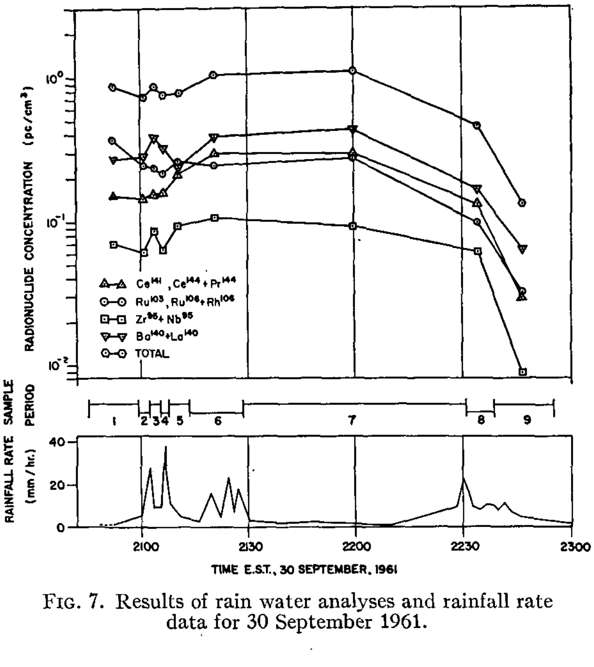

FIG. 7. Results of rain water analyses and rainfall rate data for 30 September 1961.

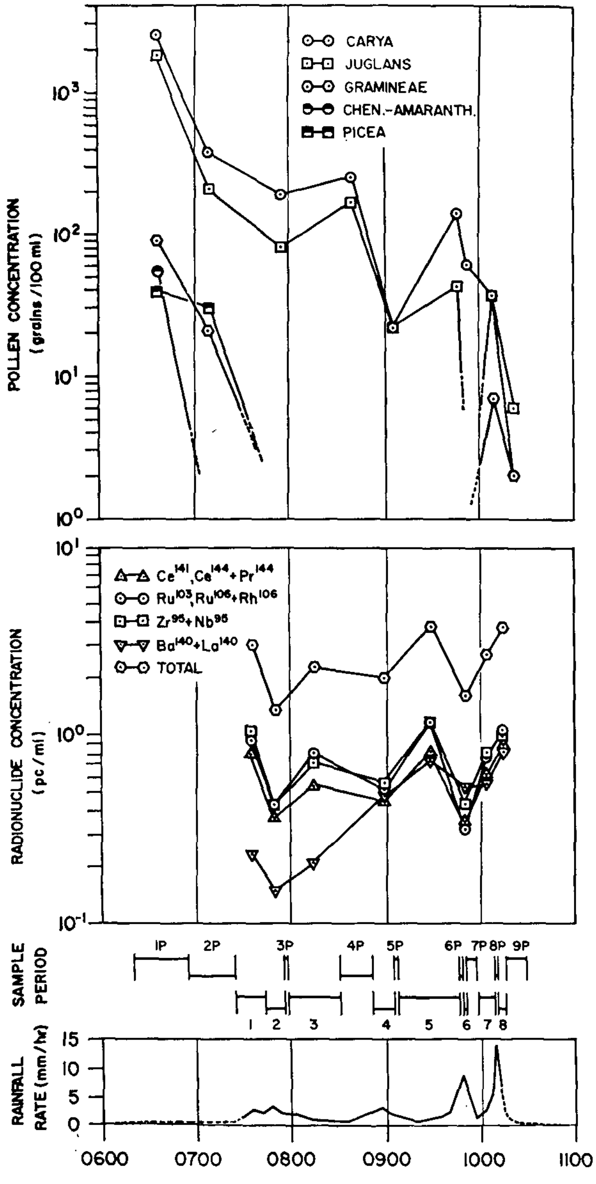

FIG. 8. Results of rain water analyses and rainfall rate data for 19 May 1962. The common names and diameters of the several pollens are given in Table 1.

5. Discussion of the results

A consistent pattern was observed in the time variation of the concentration of contaminants in rain samples during the early portions of the well-organized storms. Concentrations of plant pollens were determined in two of these storms and long-lived radioactivity was determined in all three. In the cases for which data are available on both kinds of contamination, concentrations of radioactivity and pollens exhibit similar patterns of temporal change, both showing a rapid decrease in the heavier rain portions with partial recovery between separated showers.

The similar trends of the two kinds of contamination suggest that the processes of removal of both were quite similar. Yet the pollens are characterized by sizes ranging over various species from 20 to 75 μ diameter, and by densities near 1.3 gm cm^{-3}, whereas the radioactive particles are necessarily much smaller when they enter the troposphere (Junge, 1963).

In a previous study (Gatz and Dingle, 1963) of ragweed pollen concentrations in the rain of 1 September 1961, computations of washout were made assuming a "stagnant" atmosphere beneath the rain-generating level. The results of this computation are plotted in Fig. 3. Comparison of the curves of computed and observed pollen concentrations indicates that a more or less continuous flux of contamination into the storm was necessary in order for pollen to be found in rain which fell more than a few minutes after the beginning of rainfall. This requirement is fulfilled as long as contaminated low-level air can be entrained into the storm. Most such entrainment must be associated with the convective updraft which also supplies the water vapor.

Circulation models for persistent convective storms (e.g., Newton, 1950) usually show a strong updraft to be located at the leading edge of the storm. This feature is consistent with the observation of high concentrations of contaminants in the first rain collected from such storms, even though present information does not permit complete specification of the trajectories followed by the raindrops arriving at a point on the ground at any specific time.

In the diffuse rains of 30 September and 19 May, peculiar trends of contaminant concentrations were noted. The absence of a consistent temporal pattern of concentration changes in these cases might be expected from a consideration of the likely circulations of weak convective systems. As Weickmann (1963) has pointed out, heavy storms with high rainfall rates are associated with circulations of a high degree of organization and persistent updrafts. Weak storms lack the organized circulation and their updrafts are more transient. It is consistent with these observations that an organized pattern of concentration changes, such as that observed during the early portions of the heavy rains, should be associated with an organized and persistent updraft, whereas the absence of an organized pattern should be associated with weak and disorganized convection.

In view of the high concentration of contaminants found in the first rain to arrive at the station, it is clear that this rain must be associated with a large value of the product: (volume of the air column swept out) × (mass, or activity, of contaminants in that volume). This criterion may be met by various combinations of drop trajectory, distribution of contaminants, and storm circulation pattern. The present data do not permit a definite specification of these parameters.

6. A mechanism for rainout of stratospheric contaminants

Two generally agreed-upon observations in regard to radioactive fallout point to the broader implications of the present study. The first is that the middle-latitude maximum of soil contamination by long-lived radioactive nuclides is the result of delayed fallout from the stratospheric reservoir that was created by atmospheric tests of nuclear devices at both high and low latitudes. The second is that the large majority of this contaminant has been brought to earth by rain.

The size distribution of the particles that carry radioactivity into rainstorms has not been well-established. In the case of the long-lived radioactive species that we are concerned with, the material is known to be stored in the stratosphere, necessarily as a finely divided aerosol, and to enter the troposphere from the top.

Certain entrainment mechanisms along with Brownian diffusion, etc., offer the possibility that these small particles may become associated with and collected by cloud particles at the tops and around the entraining edges of rainclouds. These processes are so different from those whereby the pollens are known to be scavenged that one should expect them to follow a totally different pattern in contaminating the falling rain. But this is not observed. Attention to the dynamics that prevail in the production of rain appears also to be important.

The rather strong evidence of observation and of atmospheric dynamics is that ordinarily the tops of rainstorms are divergent, and that therefore it would be very difficult for small particles to move directly from the air around the tops of rainclouds into the precipitating regions of these clouds. Rather, there is the strong suggestion that the cloudy regions accessible to the stratospheric aerosol are most likely to be those that diverge from the storm system aloft, and in which the cloud water re-evaporates as the elements move downstream and fall slowly under gravity. This process certainly serves to upgrade the size distribution of the original aerosol by associating it with tropospheric particles.

The observed similarities between the scavenging patterns for long-lived radioactive debris and airborne pollens suggest that the processes which ultimately bring these contaminants into contact with rain must be quite similar. This is true even though the known source regions for the two classes of contaminant are the top and the bottom, respectively, of the troposphere.

These facts strongly indicate that stratospheric contamination is brought to earth by rain mainly after an intermediate process of mixing with tropospheric air in a trajectory that penetrates to the lower troposphere (below about 700 mb). Such a sequence of events is very similar to that suggested by Staley (1960). In the course of this mixing process an association of the stratospheric aerosol with the tropospheric aerosol and with water vapor prepares it and locates its favorably for involvement in rain-generating processes. Efficient scavenging by the rain may then proceed primarily by 1) nucleation on the aerosol particles and 2) impact-collection of the aerosol particles by falling raindrops.

Examination of several cases in which large quantities of iodine-131 were detected in milk within several days after heavy rain from severe convective storms over the respective milksheds led Machta (1963) and List *et al.* (1964) to infer ". . . a cause and effect relationship between the penetration of thunderstorms into high concentrations of nuclear debris and the subsequent amount of iodine-131 in milk." Although it is not stated explicitly, the inference is apparent that the debris entered the cloud at high levels. No mechanism for the debris to become attached to particles that rain out and thus reach the ground was offered. Dingle (1965) has suggested that the cloud-top vortex development observed in some of these well-organized storms may provide a means of drawing heavily contaminated air from the lower stratosphere directly to the lower troposphere. Here the divergence from the vortex that is required at the lower boundary should serve to place the highly concentrated debris into the storm updrafts in a favorable position for rain scavenging by nucleation and impact collection processes.

7. Conclusions

The observation of parallel temporal changes in concentrations of radioactivity and plant pollens in rain samples from three well-organized convective rain systems is interpreted as evidence that both classes of contaminants were removed from the same air. Because plant pollens are released into the atmosphere at the earth's surface, it is inferred that both radioactivity and pollens were removed from an updraft of warm moist air which entered each system at a low level.

Inasmuch as the patterns of convection in the two observed cases of diffuse convective rains are such as to leave pockets of contaminated low-level air between the relatively weak and isolated precipitation cells, the irregular trends of contaminant concentrations observed in these cases are entirely consistent with the above hypothesis. We have collected more comprehensive data on the scavenging patterns and delineations of severe storms in central Oklahoma. Our analysis of these is under way, and will be offered for publication within a few months.

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