Trace substances in rain water: concentration variations during convective rains, and their interpretation[1,2]

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ABSTRACT

The temporal changes of contaminant concentrations in rain water samples collected sequentially at a fixed point are dependent upon both the rain scavenging and drop growth processes and the advective effects of the motion field upon the scalar field of concentration variations in the rain cloud. The observations that rainfall intensity and contaminant concentrations are usually negatively correlated, but occasionally for short periods are positively correlated, are explained. Periods of positive correlation appear mainly because of a predominating advective effect, whereas, all drop growth and scavenging processes operate to produce the negative correlation.

In the massive storms characterized by Browning (1964) as "SR-storms" and by Newton (1966) as "persistent", an extraordinarily well-organized downdraft is inferred. The strong evaporative effect of this downdraft upon rain in contact with it produces an atypical variation of concentrations in relation to rainfall rates. An example of this is presented.

I. Introduction

The large and sudden rainfall rate variations of convective rains are well known. Less well known are the oftentimes larger and more sudden variations of rain water impurity concentrations which also occur in these storms. Interesting relationships between rainfall rate and impurity concentration have been observed and speculated upon. This paper deals with these relationships.

Table 1 summarizes some of the literature on concentration–rainfall rate relationships for particulate impurities.

Previous experimenters looked at different substances and used different averaging times for concentrations and rainfall rates. Because of these differences it is important to examine experimental methods closely. In particular, one would ask whether sampling practices were adequate to distinguish the *inverse* (low concentration in periods of high rainfall rate) from the *direct* relationships. We have explored this subject, and concluded that, except when rainfall rates were averaged over long (15-min) periods, the data have sufficient resolution to distinguish the two types of relationships.

The investigations listed in Table 1 were concerned only with trace substances present as particles in the atmosphere. Oxygen and hydrogen isotopes (Bleeker et al., 1966; Ehhalt et al., 1963) are not included because they are present in the atmosphere mainly as water vapor. They appear to be useful in assessing the role of condensation as an attachment mechanism. This point is considered in more detail later. Radon daughter radioactivity (Jacobi, 1962) has also been excluded.

A. *Suggested mechanisms*

Direct relationships have been observed in connection with artificial radioactivity concentrations. A number of papers (Kruger & Hosler, 1963; Huff & Stout, 1964; Bleeker et al., 1966) have suggested that direct relationships occur when bomb debris enters at the top of a convective cloud and becomes attached to the heavy rain which forms at high altitudes. Dingle

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Table 1. *Summary of observed relationships between trace substance concentrations and rainfall rate in individual convective rains*

Trace substance	Reference	No. of cases	Predominant relationship reported	Rainfall per sample (mm)	Method of rainfall rate measurement
Pollen	Dingle & Gatz (1966*a*)	3	Inverse	0.73–1.5	*t–b* gauge[a]
	Gatz (1966)	2	Inverse	0.25–3.0	*t–b* gauge
Artificial beta radioactivity	Bleichrodt et al. (1959)	11	Inverse	0.20	Sample collection
	Huff & Stout (1964)	87	Inverse	1.0–1.5	Weighing gauge
	Huff (1965)	190	Inverse	1.0–1.5	Weighing gauge
	Dingle & Gatz (1966*a*)	5	Inverse	0.73–1.5	*t–b* gauge
	Dingle & Gatz (1966*b*)	3	Inverse	1.25	*t–b* gauge
	Gatz (1966)	2	Inverse	0.25–3.0	*t–b* gauge
	Hall et al. (1964)	11	Inverse	1.3	Weighing gauge
	Hall (1965*a*)	29	Inverse	1.3	Weighing gauge
	Hall (1965*b*)	46	Inverse	1.3	Weighing gauge
Inorganic ions	Georgii (1965)	Not reported	Inverse (single showers) Direct (series of showers)	Continuous sampling	Not reported

[a] Tipping-bucket rain gauge.

(1965) points out that the physics of this process is difficult to explain unless the deeply penetrating storms are characterized by strong vortex action capable of pulling the high level debris down and injecting it at low levels. Hicks (1966) proposed that direct relationships could be caused from a mid-level entrainment of airborne debris into a severe storm followed by collection of the debris by heavy rain falling in the downdraft.

Mechanisms have also been suggested to explain inverse relationships, and particularly the high concentrations often found in the first rain collected. Huff & Stout (1964) and others have suggested that evaporation of raindrops falling through relatively dry air at the leading edge of a storm could cause the high initial concentrations. They have also suggested that dust from the ground or lower atmosphere would be collected and deposited in the first rain to be collected. Bleeker et al. (1966) suggested that a combination of diffusive capture of aerosols, evaporation of the first rain, and dilution of the trace substance in the heavy rain produces inverse relationships. Hicks (1966) suggested that a presumed preferential nucleation of the radioactive particles followed by fallout of the raindrops thus formed early in the shower causes inverse relationships.

This review of proposed mechanisms indicates that most writers have taken the view that different mechanisms produce the two different relationships. However, there is no general agreement on the cause of either the direct or inverse relationship, therefore, we should like to emphasize the possibility that both relationships can result from the same physical cause.

B. *Particle attachment processes*

In considering the removal of particles from the air by rain, it is necessary to evaluate the processes of particle attachment to cloud and precipitation elements. These processes, in turn, are very sensitive to particle size, and they must be specified in any technical discussion of the subject. Our particular effort has concentrated on two materials of different sizes: Long-lived artificial radioactivity and plant pollens.

The pollen types that we have identified and counted in collected rain samples range in diameter from about 15 μ to about 70 μ. The pollen species found, their shapes and sizes are listed in Table 2, and some typical pollen grains are pictured in Fig. 1.

The particles bearing radioactivity in the lower troposphere are much smaller, being found in the diameter range below about 3 μ. Shleien, Galvin & Friend (1965) collected radio-

Table 2. *Pollen types determined in water samples*

Type	Taxon	Shape	Diamter (μ) Measured[a]	Literature[b]
Willow (*Salix*)	Genus	Spheroidal or oblate	18 × 16	17–18
Oak (*Quercus*)	Genus	Spheroidal (flattened and angular when moist)	25 (3)	Range: 30 × 25 to 30 × 26 (expanded)
Black walnut (*Juglans nigra*)	Species	Oblately flattened	34[c]	31–34
Hickor (*Carya*)	Genus	Spheroidal or oblate	44[c]	40–52
Pine (*Pinus*)	Genus	Ellipsoidal body, two spheroidal air bladders	67 × 44 (1)	46–65[c]
Chenopods/Amaranths	Families	Spheroidal	25 (1)	19–33 (moist and expanded)
Grass (*Gramineae*)	Family	Spheroidal, ovoidal, or ellipsoidal	25–59[c]	22–100
Composites (*Compositae*)	Family	Spheroidal	20	19–40
Sorrel (*Rumex*)	Genus	Spheroidal or ellipsoidal	19	18–32

[a] Average of measurements (parentheses indicate number measured if < 10) of non-acetolysed pollen in glycerine jelly. [b] Wodehouse (1935). [c] Largest diameter.

active aerosols on three filters in series and concluded that particles having diameters less than 1.75 μ contained at least 88% of the total fission-product gamma activity. Their samples were collected in the spring of 1964. They also concluded that "long-lived fission-product activity in air at ground level is for the most part associated with airborne materials having the same diameter as particles prevalent in the surrounding air". Lockhart et al. (1965), using a series of four filters, came to the same conclusions based on samples collected during 1963 and 1964. They found fission-products in tropospheric air to be associated with particles having an average diameter between 0.3 and 1.1 μ (assuming spherical particles of 1.8 g/cm^3 density). Shalmon (1964) determined the radioactivity associated with different sizes of ground-level airborne dust collected from April to September, 1963, and found that 90% of gross beta radioactivity was attached to particles larger than about 0.7 μ diameter. Taken together, these three papers indicate that about 80% of low-level airborne radioactivity was associated with particles between 0.5 and 1.75 μ diameter. These samples were all collected several months after all atmospheric testing had stopped. Therefore, the results may be said to apply to the rain samples collected in our program.

The size ranges of plant pollen grains and gross beta radioactivity are summarized in Fig. 2, which also includes the size ranges in which the important attachment mechanisms are effective. The mechanisms listed and their ranges of effectiveness have been described by Junge (1963). It is clear from Fig. 2 that impaction and condensation are important processes in the attachment of both pollens and artificial radioactivity.

Recent evidence (Dingle, 1966; Hicks, 1966; Storebø, 1966) points strongly to droplet nucleation in rising air as the primary mechanism for attachment of radioactive bomb debris to precipitation elements. For effective removal by this process, stratospheric debris particles need a period of residence near the earth's surface so that they may become attached to the tropospheric aerosol particles in the condensation nuclei spectrum at the same time as they become associated with water vapor (Dingle & Gatz, 1966*a*).

The evidence for nucleation of condensation on pollen grains is less certain. Because pollens

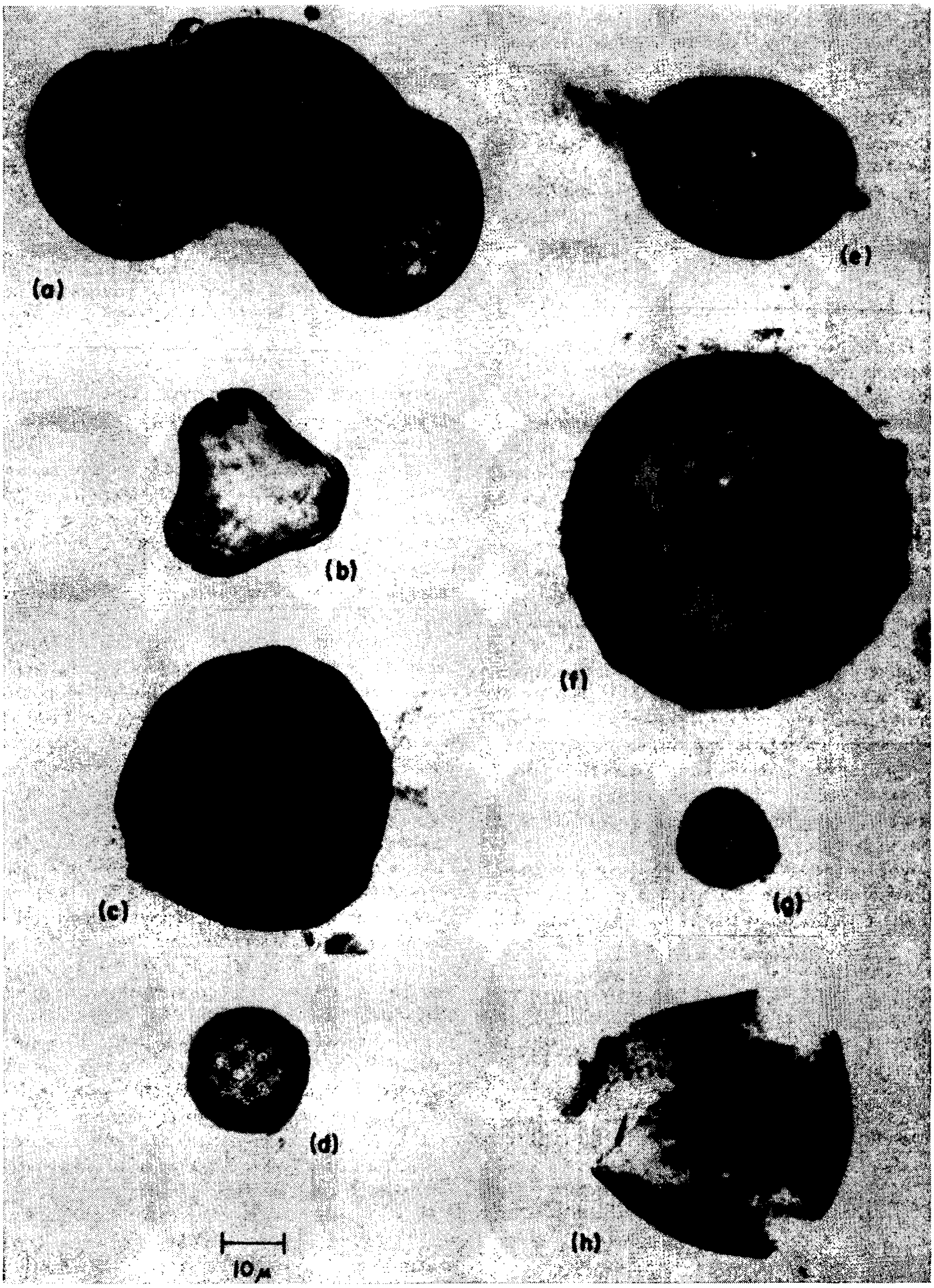

Fig. 1. Some pollen species found in rain samples: (*a*) pine, (*b*) eucalyptus (exotic species added for standardization), (*c*) hickory, (*d*) chenopod/amaranth, (*e*) black walnut, (*f*) grass, (*g*) willow, (*h*) oak.

are produced near the earth's surface, they undoubtedly enter a storm with the warm moist air, but their attachment mechanism is not clear. Their large size is definitely favorable for nucleation (Dingle, 1966) but the surface properties required for this role have not been clearly shown (McDonald, 1964).

In any case pollen grains are large enough to be collected very efficiently also by the impaction of falling raindrops.

II. Experimental

Each year since 1964 we have collected rain samples and measured rainfall rates at Chickasha, Oklahoma, in the National Severe Storms Laboratory (NSSL) cooperative spring program. Samples ranging in volume from 2 to 8 liters were collected sequentially using three collecting funnels, each 2.5 m² in area (Gatz, 1966). Samples were changed manually to permit maximum flexibility in changing samples in response to rapidly changing rain situations. Both plant pollens and artificial beta radioactivity concentrations were determined for each sample. The standard error, σ, of the radioactivity measurements, including counting and handling errors, is about 5%. The uncertainty associated with the pollen concentrations is greater. Therefore, 3 σ limits are shown for the pollen concentrations in Figs. 4, 5, and 7.

Rainfall rates were computed from a calibrated tipping bucket rain gauge for every 0.25 mm of rain.

During the 1964–1967 spring seasons fourteen individual rains were sampled in detail. Ten of these were convective rains. They ranged in

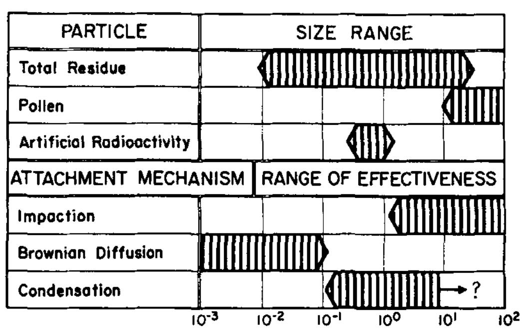

Fig. 2. Diagram showing approximate size ranges and attachment mechanisms for pollens and artificial radioactivity.

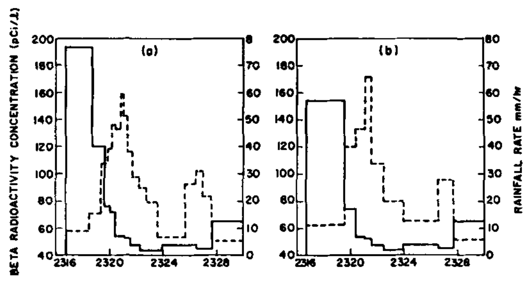

Fig. 3. Rainfall rate in mm/hr (dashed line) and β-radioactivity in pCi/l concentration profiles for rain of 11 May 1966 at Chickasha, Oklahoma. Abscissa is Central Standard Time.

intensity from light to severe. The data presented below have been selected from these convective rains.

Fig. 3 shows a typical time profile of tracer concentration and the typical relationship to the rainfall rate profile in a single shower. The concentration is high at the start of the rain, decreases rapidly as rainfall rate increases, and reaches a minimum sometime after the heaviest rain.

Fig. 4 shows concentration variations which occurred as two separate convective rain cells crossed the station. The rain did not stop between the two showers, which were imbedded in a severe squall line. These showers both produced heavy rain and the NSSL radar showed them clearly within the squall line but they lasted only a few minutes.

Again concentrations were high at the beginning of the rain. This is true of both the radioactivity and pollen. In fact, concentrations of the two impurities rose and dropped together all during this rain. Concentrations dropped slowly during the first hour of light rain. Then, as the first shower began, the concentrations dropped sharply and reached the lowest values of the shower just after the heaviest rain. They increased again, reaching a peak as the second shower started, and fell rapidly once more as the rain became heavier. The lowest concentrations of the second shower occurred in the heaviest rain. From that point, concentrations increased slowly as the rain became lighter.

It is tempting to say that these data show a simple inverse proportionality between concentrations and rainfall rates. It is not that for two reasons: (1) the concentration peak early in the second shower occurred several minutes

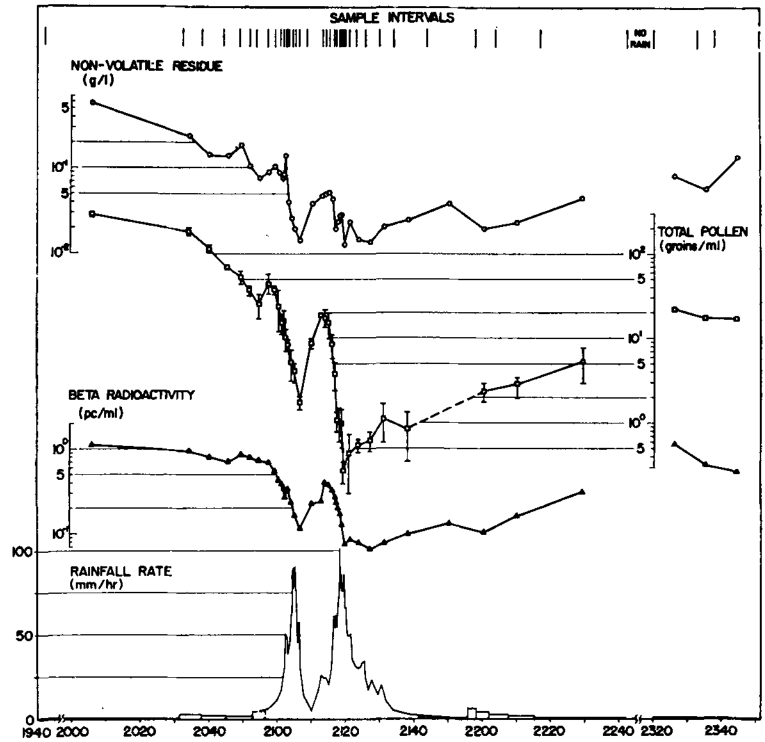

Fig. 4. Rainfall rate, pollen and β-radioactivity concentration profiles for rain of 9 May 1964 at Chickasha, Oklahoma.

(and 2 samples) after the time of lightest rain as the rain intensity was increasing, and (2) the extremely rapid decrease of concentration approaching the climax of the second shower was not matched by any comparable increase following the shower climax, even though the rates of rainfall increase and decrease were similar. That is to say, before the heaviest rain, the concentrations dropped very rapidly; after the heaviest rain, the concentrations remained nearly constant, and following that, in the very light decadent rain stage, they rose slowly.

Fig. 5 shows the concentration variations during three successive showers. The radioactivity concentration was high at the start of the first shower and low during the first rainfall peak. It rose temporarily before the second shower began, then dropped to a low point at the height of that shower. It then held steady for about three minutes as the rainfall rate decreased sharply, and then fell steadily through the onset and past the climax of the third shower. In this case the usual single-cell concentration pattern appears to have been repeated with each new shower, but the effects of cell overlap have obscured the single-cell profiles somewhat.

The pollen concentrations, having a larger error, are less clear; yet one sees the rise and fall of concentration with the passage of each shower.

Another case of closely-following and overlapping showers appears in Fig. 6. The two rainfall rate peaks were five minutes apart. The concentration profile was typical in the first shower: high at the beginning with a sharp drop to a minimum in the heaviest rain. The second shower shows the atypical direct relationship. The highest concentration occurred in the heaviest rain. Notice that in both showers the shapes of the concentration profiles are the same as seen in single showers.

Of the rains mentioned so far, some produced heavy rain and strong winds, but none of the individual storm cells could be followed on radar photographs for more than a few minutes.

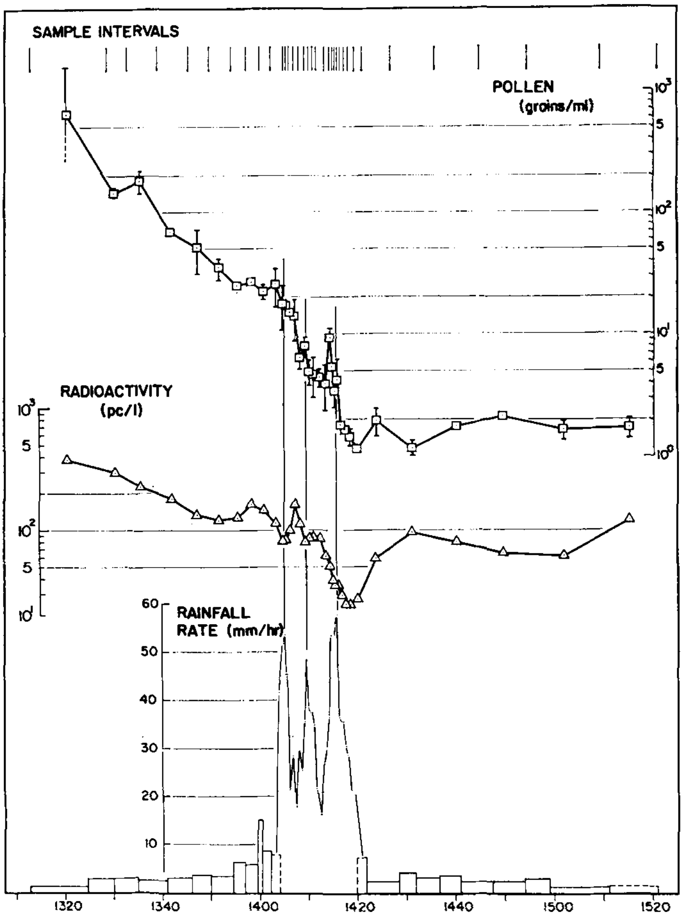

Fig. 5. Rainfall rate, pollen and β-radioactivity concentration profiles for rain of 9 May 1965 at Chickasha, Oklahoma.

On 10 May 1964, we sampled a storm that had attained the degree of organization and development described as "persistent" (Browning, 1964). Radar data show that the echo was identifiable for several hours, and that the strongest part of the storm echo passed directly over our station. It was associated with a squall line which produced considerable hail and spawned a series of tornadoes.

Concentrations and rainfall rates are shown in Fig. 7. During the heaviest rain we see a slight decrease in both concentrations, but the concentration decrease during the heavy rain is much less than occurred in any of the other convective rains sampled. The highest radioactivity concentration exceeded the lowest by a factor of only 3. In the other storms, this ratio is

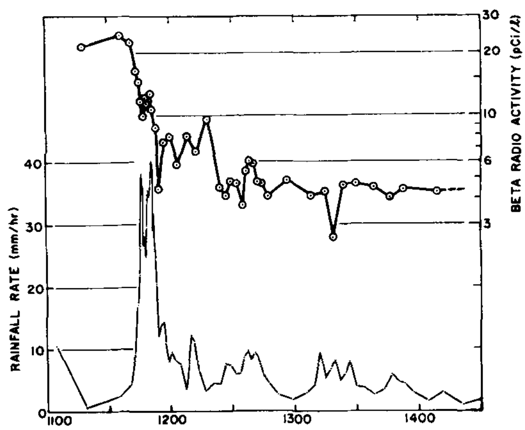

Fig. 6. Rainfall rate and β-radioactivity concentration profiles for rain of 22 April 1966 at Chickasha, Oklahoma.

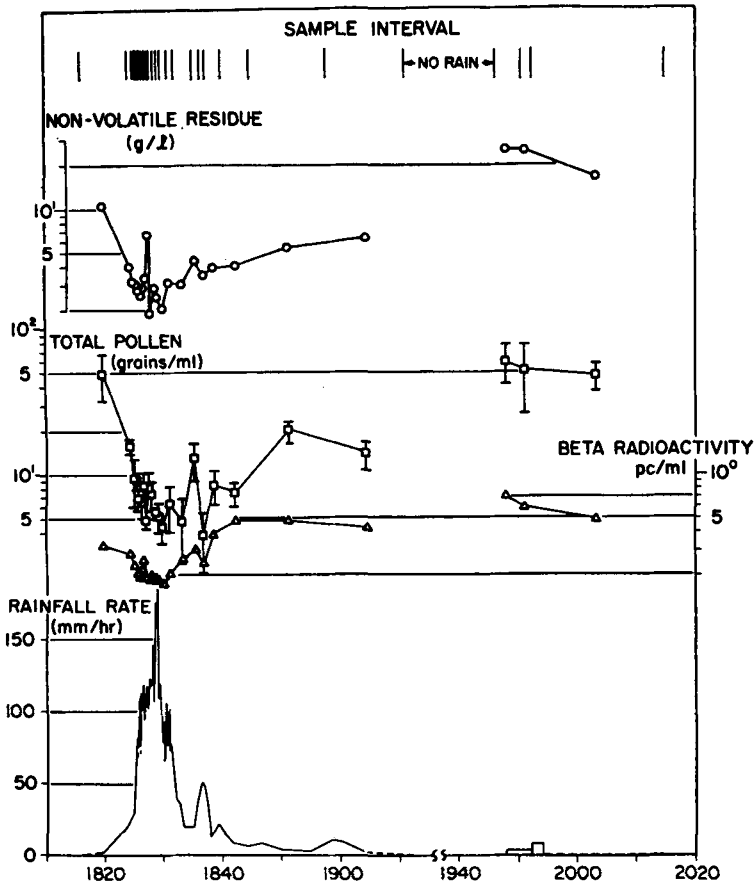

Fig. 7. Rainfall rate, pollen and β-radioactivity concentration profiles for rain of 10 May 1964 at Chickasha, Oklahoma.

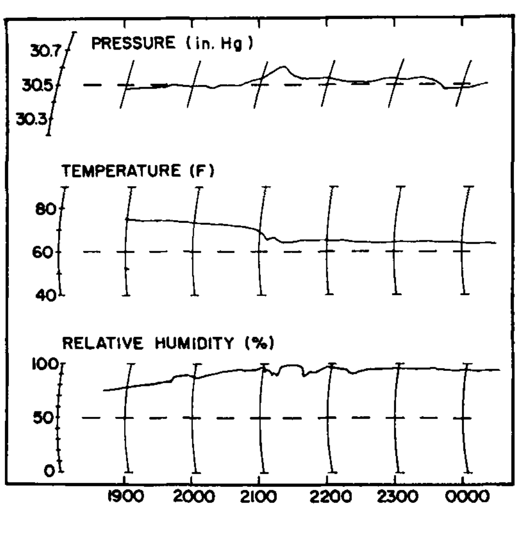

Fig. 8. Pressure, temperature and relative humidity traces for NSSL meso-network station no. 11 (approximately 500 m north of the University of Michigan rain sampling station), for 9 May 1964.

at least 10, often 20. At the same time, the rainfall maximum achieved in this storm was 179 mm/hr, whereas the other storms produced maxima of 70, 100, 57, and 40 mm/hr, respectively.

III. Discussion

Our examples of 1-, 2-, and 3-celled convective rains, and some of those in the literature (e.g., Georgii, 1965) show clearly that each convective cell produces a certain fluctuation in impurity concentrations. We and others, have found that the relationship between the concentration variation and the rainfall rate variation can usually be described roughly as inverse, but that direct relationships also occur at times.

The typical "inverse" relationship begins with a high concentration during the first light rain of a shower. Then the concentration drops sharply as the rainfall rate suddenly increases. At, or slightly after the time of the heaviest rain, the concentration descends to its lowest point. Then, as the rainfall rate drops off sharply, the concentration usually rises, although not at a rate proportional to that of the rainfall rate decrease.

A. *The evaporation mechanism*

A frequently suggested explanation for the high concentrations which are found in the first light rain is that the tracer has been highly concentrated by the partial evaporation from drops falling through relatively dry air. This explanation suggests the need to examine how much the evaporation of the first drops to reach a fixed station can enhance ground-level concentrations over those in the cloud. In the case of the second shower of 9 May 1964 (Fig. 4), because of the preceding rainfall, we note that the evaporation of the light rain must have been minimal, or perhaps totally absent. This inference is supported by the fact that rain fell at our station continuously from 1940 to about 2240—a 3-hr period. The two heavy showers occurred 10 min apart, at about the midpoint of this period.

The pressure, temperature, and relative humidity variations during this period at the NSSL meso-network station 500 m from our field station, are shown in Fig. 8. Although the humidity rose from about 80% at onset to about 97% at 2100 CST, just prior to the first shower maximum, this rise is well-correlated with the decrease of temperature during the same period. From 2100 to 2115, however, both temperature and humidity fell irregularly, the humidity reading a minimum of about 90%. During this period, the first shower climax passed, the onset of the second shower developed, and the maxima of radioactivity and pollen concentrations is found to correspond closely in time to the 90% relative humidity minimum. The next five minute period shows (1) a rise of humidity to 100%, (2) attainment of the second rainfall climax, and passage through it, (3) attainment of the second concentration minimum, and (4) constant temperature. If one assumes that the relative humidity of the entire sub-cloud layer was at least 90%, evaporation of the light rain at the beginning of the second shower is far too small to account for the concentration increase (Hardy, 1963). Aside from this argument, any evaporative contribution appears to have been nullified, for as the second shower began, both the rainfall rate and the concentration increased for a time before the concentration reached its peak. The concentration then began to decrease with further intensification of the rainfall.

One expects that the concentrating effect of raindrop evaporation should diminish as rainfall rate increases because of the smaller surface-to volume ratio of the larger drops of heavier rainfall. Therefore the observed concentration peak is clearly contrary to what is expected from the evaporation mechanism, and must have some other cause. Thus, even without clear evaporative effects, concentration peaks can and do occur early in convective showers.

B. *Mechanism for direct and inverse relationships*

Most writers on the subject (loc. cit.) appear to have assumed that the direct and indirect relationships are produced by different mechanisms. It appears to us that the mechanisms ordinarily at work in the atmosphere are capable of producing both effects, and the following points are offered in support of this view. First, the profile *shape* of the direct relationship in Fig. 6, and of those in the literature (Bleeker et al., 1966) is about the same as that found in cases of inverse relationship. Secondly, the direct relationship has been observed within 5 min of the inverse one at the same place (Fig. 6); and within 500 m distance at the same time (Hall, 1965). Huff (1965) also observed direct and inverse relationships occurring close together in time and space. One should not expect to find shifts from one relationship to the other on such short time and distance scales if greatly different physical mechanisms were responsible for them.

On examination, the time interval between concentration and rainfall rate peaks in our multiple shower cases tends to vary as a continuous, not a discontinuous parameter. This also suggests that it is more likely that the same basic mechanism produces both effects. That mechanism is complex, and the interactions of different components sometimes cause one relationship and sometimes another. Radically different input mechanisms are unlikely, however.

C. *A physical model*

The samples of rain under discussion were collected at a fixed station at ground level as each storm traversed the station. These measurements therefore may be interpreted as estimates of the local time deviative of contaminant concentration in the sequences of rain samples. Analytically

$$\frac{\partial C}{\partial t} = \frac{dC}{dt} - \mathbf{V} \cdot \nabla C \tag{1}$$

For our purpose, the motion is that of the liquid particles relative to the earth's surface, and C is defined as the ratio of the mass of any specified contaminant to that of the pure water of an individual raindrop or cloud droplet.

The term dC/dt includes all the effects that contribute to changes of concentration in the individual drop. These are evaporation, condensation, and the collection of contaminant. It may be presumed that collected contaminant is not lost from the drop except by drop breakup which cannot directly affect the concentration.

The advective term may be considered in terms of the horizontal and vertical components of motion:

$$\mathbf{V} \cdot \nabla C = \mathbf{V}_H \cdot \nabla_H C + w\frac{\partial C}{\partial z} \tag{2}$$

Here the horizontal component of motion, $\mathbf{V}_H$, is attributed to wind-induced transport of the drops, and this motion operating upon horizontal gradients of concentration must affect the local changes of concentration, $\partial C/\partial t$, observed at a fixed station. Equation (1) shows then that local increases of C $[\partial C/\partial t) > 0]$ may be contributed by horizontal motions away from the highly contaminated updraft core.

The vertical component of motion, w, may be written

$$w = w' - v_T \tag{3}$$

where w' is the vertical component of air speed, taken positive upward, and v_T is the terminal fall speed of the water drop. In falling rain, C must increase with height to contribute positively to $\partial C/\partial t$. A similar point has recently been made by Storebø (1968).

Others have studied the structure and circulations of the storms we are dealing with (Browning, 1964; Bates, 1961), and have indicated strongly that the updrafts feeding these storms ascend on a slope, not vertically. These models lend strong support to the concept that concentration gradients of considerable magnitude should be found within these convective storms. Considering, therefore, the above analytical discussion in conjunction with sound physical models of the subject storms it is evident that the advective effects, of both horizontal and vertical drop motions and concentration gradients, are of major concern and may frequently exceed in magnitude the individual changes (condensation, evaporation, collection) in convective situations.

Specifically, an adequate physical model of convective storm scavenging must explain the early concentration peak, the rapid lowering of concentration as rainfall rate (R) rises rapidly, the much slower increase of concentration following the shower climaxes, and the peculiar variations we have cited here in which on occasion rainfall rates and concentrations are observed to rise and/or fall together.

The mechanisms of condensation and evaporation, which contribute to dC/dt, generally operate to produce an inverse relationship between C and R. That is to say, as water is acquired by a droplet through condensation, making for an increase of R, C is reduced by dilution, and as evaporative effects occur, thus reducing R, C is enhanced. The collection of contaminant by rain—and cloud—droplets operates monotonically to increase C. But no mechanism is present in the individual change term to make C and R vary together in the same direction.

Advection, on the other hand, operates upon the horizontal and vertical gradients of C that occur in the cloud and precipitation fields. In convective storms we must anticipate that these are substantial and highly variable. Measurements are, however, difficult to obtain because of the violent motion fields encountered. Komabayasi & Isono (1967) report measurements of the vertical gradient of rainwater conductivity in Hawaiian tradewind orographic showers. A maximum of a tenfold change of conductivity in 200 m was found. If such a

Table 3. *Heights of origin of downdraft Using θ_w*

Observed at Chickasha				1800 CST sounding FSI		1800 CST sounding OKC		1700 CST sounding FSI	
Time (CST)	Temp. (C)	R.H. (%)	θ_w (C)	Pressure (mb)	θ_w (C)	Pressure (mb)	θ_w (C)	Pressure (mb)	θ_w (C)
(a) *May 9, 1964*									
2100	21.1	96	21.7	970	21.8	969	20.8		
2105	20.0	94	20.4	889	20.4	850	17.9		
2110	18.9	91	19.0	850	20.4	824	16.0		
2115	18.9	92	19.1	700	15.8	700	13.9		
2120	18.3	99	19.4	633	16.5	696	13.1		
2125	18.3	100	19.4	592	14.2	628	15.6		
2130	18.3	100	19.4	517	15.3	538	15.2		
				500	15.8	500	15.3		
(b) *May 10, 1964*									
1800	22.8	86	22.4					962	23.4
1805	21.2	76	19.7					900	21.9
1810	18.9	78	17.8					869	21.6
1815	18.3	82	17.9					850	21.8
1820	17.8	84	17.3					834	21.0
1825	17.2	92	17.9					750	19.7
1830	15.0	99	16.0					700	16.9
								650	16.6
								600	17.5
								550	16.4
								500	17.1
								430	19.0

gradient were maintained during descent of the rain, a tenfold conductivity change should be observed at the surface in a period of about 30 sec, assuming a mean fall speed of 6 m/sec for the rain. Whereas this is a more extreme rate of change than is commonly observed at the ground, the effects of turbulence and of drop sorting must operate to smooth sharp gradients. In the strong convective situations from which our data have been assembled, it is easily conceivable that more extreme in-cloud gradients are produced.

D. *Concentration profiles in a persistent severe storm*

The persistent severe storm of 10 May 1964 (Fig. 7) produced a scavenging pattern sharply different from that of the lesser storms. In particular, the radioactivity and pollen concentrations in the heaviest rain were only slightly less than those observed at the beginning and end of the rain. This suggests that some particular physical mechanism peculiar to such a persistent severe storm was present and produced relatively higher concentrations in the peak showers than were observed in the non-persistent storms.

The fact that both pollen and radioactivity concentrations behaved the same way insures that a high-level input of contaminants, as suggested by Kruger & Hosler (1963), and others (loc. cit.), cannot explain the observations. However, an enhancement of concentration must follow from partial evaporation of the raindrops during their fall, both in and below the cloud. Fujita (1959) has shown that the cold air outflow from thunderstorms results from cooling of the air by raindrop evaporation in the downdraft.

The different character of the concentration profiles in a persistent severe storm relative to non-persistent ones reflects a greater evaporation effect in the deeper downdraft of the persistent storm. Browning (1964) and others have suggested that persistent severe storms are better organized than less severe ones and draw in air from mid-tropospheric levels to compose the downdraft which reaches the surface as

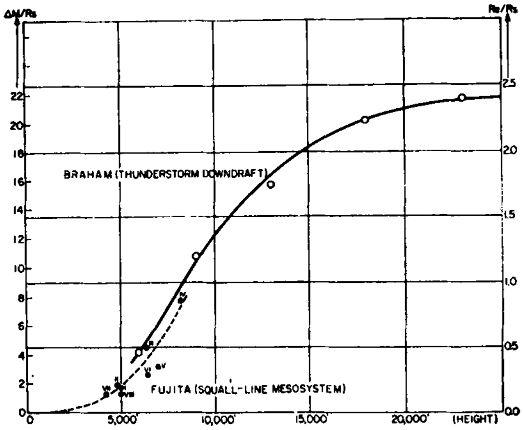

Fig. 9. Evaporative loss of water to downdraft air as a function of height of origin of downdraft: R_e is the amount of rain evaporated, R_s is the amount of rain that reaches the ground (after Fujita, 1959).

cold outflow air. The method of Newton (1950) was used to determine the approximate height of origin of the downdraft for the 9 May and 10 May 1964 storms. The method assumes that the potential wet-bulb temperature is conserved in the descending air, and hence its level of origin can be identified by means of soundings taken prior to the rainstorm. Table 3 shows θ_w values within the downdraft at our station and the profiles of θ_w at Fort Sill (FSI) and Oklahoma City (OKC) on 9 and 10 May 1964. The tables show that on 10 May the downdraft air originated between 700 and 500 mb, whereas the origin of the downdraft air on 9 May was below 700 mb.

The effect of evaporation on contaminant concentrations may be seen from Fig. 9, after Fujita (1959). The righthand scale is the ratio of the evaporated rain, Re, to the amount of rain reaching the surface, Rs, and the curve shows how this ratio changes as a function of the distance fallen. For a fall of 10,000 ft., the ratio Re/Rs is 1.3. Assuming that no drop disappeared entirely in the process, this implies a concentration enhancement factor of 2.3. If, on the other hand, complete evaporation of the smaller drops is allowed, their burden of contaminant should be lost from the incident rain water unless this liberated contaminant is rescavenged by the drops that retain their identity.

It will be noted that, despite the great evaporative effect required to conserve θ_w, the rainfall rate in this storm exceeded 100 mm/hr for more than 6 min and achieved a maximum of 179 mm/hr. Engelmann (1968) points out that the rain "washout coefficient", λ, may be expressed as

$$\lambda = \int_0^\infty FEA\,dD$$

where D is drop diameter, A is drop cross-sectional area, E is the collection efficiency, and F is the flux density of the raindrops. In the present instance, the integral of F over the diameter spectrum is the rainfall intensity, and as pointed out above, it is exceptionally high in the storm in question. The value of E is also of importance, and Mason (1957) has tabulated values greater than 1.0 for collector drops up to 2.0 mm diameter on droplets of 15 to 20 μ diameter. Thus in a downdraft such as that envisioned here, as small droplets approach 20 μ diameter in evaporating, the chance that they may be collected by a larger falling raindrop is enhanced. It is therefore reasonable to assume that the scavenging action of the intense rain in the downdraft region is so great as to prevent effective loss of contaminant by the complete evaporation of the smaller droplets.

In the 10 May case, the level of origin is estimated at 12 000 ft., for which a concentration enhancement of 2.6 times is given by Fujita's curve, whereas the level of origin on 9 May was about 7 000 ft., and the enhancement factor about 1.6.

The concentration profiles for the 9 and 10 May storms agree qualitatively with these indications. We therefore conclude that the absence of a large concentration decrease in the heavy rain on 10 May is partly explained by evaporative enhancement of the concentration in the downdraft.

IV. Conclusions

Trace analysis of sequential rain samples show that marked concentration variations in convective rains are clearly produced by individual shower cells. In most cases, concentration time profiles are related to rainfall rate time profiles in a particular way. Their relationship might loosely be termed "inverse", but close examination reveals that this is not strictly true, because opposing maxima and minima usually are not coincident.

The concentration time profile is quite distinctive. Most commonly the concentration peak occurs early in the shower, but the time intervals between the maxima of rainfall rate and concentration appear to vary continuously when a number of showers are studied. This suggests that the "direct" and "inverse" relationships are not necessarily produced by different kinds of processes.

Concentration variations in rain observed at a fixed point may be viewed as the sum of (1) individual changes within moving rain parcels, and (2) horizontal and vertical advection of concentration gradients in the three-dimensional rain field.

The data support the idea that the early precipitation of the largest and most favored nuclei produce high concentrations early in showers and low concentrations in the heaviest rain.

To the extent that direct relationships between concentrations and rainfall rates occur in sequential rain samples, a predominance of the advective term over the individual change term of Equation (1) is implied.

The one persistent severe storm sampled showed a smaller concentration depression during the heavy rain than that characteristic of non-persistent storms. A major contributor to this difference is inferred to be the much deeper downdraft of the persistent storm which leads to greater water loss by evaporation and higher-than-usual concentrations in the heaviest part of the rain. The limit on the rainfall efficiency of persistent storms, imposed by this mechanism, is also noted.

Because of the complexity of convective cloud and rain systems, the complete description of their circulations and evaluation of their processes has long been an elusive objective of meteorological research. In introducing trace analysis of rain samples as another means of obtaining quantitative measures of the products of these storms, we believe that a new set of constraints is added to our total information on individual storms, and that the interpretation of this information is thereby strengthened. The technique is still quite new, and much effort is required to exploit it adequately, but these more or less preliminary results appear to us, at least, to show that it holds promise of substantial contribution to understanding of the structure, dynamics and physico-chemical processes of convective storms.

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ТРАССЕВЫ В ДОЖДЕВОЙ ВОДЕ: ВАРИАЦИИ КОНЦЕНТРАЦИЙ ВО ВРЕМЯ КОНВЕКТИВНЫХ ДОЖДЕЙ И ИХ ИНТЕРПРЕТАЦИЯ

Временные изменения концентраций загрязнений в пробах дождевой воды, собранных последовательно в фиксированной точке, зависят как от процессов роста капель и дождевого вымывания, так и от влияния адвекции полем ветра на скалярное поле вариаций концентрации в дождевых облаках. Объясняются результаты наблюдений, что интенсивность дождя и концентрации загрязнений обычно отрицательно скоррелированы, однако, иногда в течение коротких промежутков времени скоррелированы положительно. Периоды положительной корреляции появляются, главным образом, благодаря доминирующему эффекту адвекции, в то время как рост капель и вымывание приводят к отрицательной корреляции. В массивных штормах, характеризованных Браунингом (1964) как «SR-штормы» и Ньютоном (1964) как «устойчивые», выявлено чрезвычайно хорошо выраженное опускание. Сильное влияние испарения на дожди в этих опусканиях вызывает нетипичные вариации концентраций по отношению к количеству выпадающей воды в дождях. Представлен пример такого рода.