Nucleation and the Wet Removal of Fallout

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ABSTRACT

The theory of condensation nucleation applied to radioactive fallout constituents shows that most tropospheric fallout particles are efficient nuclei in the production of warm rain. As a result it is found that the concentration of fallout in precipitation is a maximum soon after the onset of rain. This is supported by measurements made during many individual storms over an extensive period. The influence on fallout concentration of variations in rainfall rate through a storm is found to be complicated by the opposing effects of the liquid water content of the cloud and the generation of downdrafts within it.

1. Introduction

Of the various methods of removal of tropospheric fallout the most efficient are undoubtedly those associated with rainfall. Theoretical discussions of the mechanisms such as that by Greenfield (1957) have generally considered scavenging by collision between fallout particles and cloud or rain drops. Such models are lacking in that they fail to recognize the role of nucleation by the radioactive debris itself.

Although the mechanisms of ice nucleation are not completely determined, the theory of condensation nuclei in warm rain situations is well documented (Fletcher, 1962). In the latter case it is possible to discuss in precise terms the behavior of radioactive particles in supersaturated air.

2. Nucleation

Recent investigations into the chemical composition of atmospheric pollutants have shown that most particles of stratospheric origin exist either as sulfates or with a layer of sulfated material surrounding their core. The remaining particles are similarly composed of chlorides, nitrates and carbonates (Junge, 1954; Whitehead and Feth, 1964). Almost all such particles are soluble. Bearing in mind the known chemical behavior of the common fallout nuclides (strontium, cesium, cerium, zirconium, rhodium, etc.), we would expect a large proportion of fallout particles to be hygroscopic.

The fractional supersaturation, S, required for nucleation of a hygroscopic particle to occur and for the resulting droplet to continue to grow is a function of the particle mass m, its gram molecular weight M, the corresponding van't Hoff dissociation constant i, and the absolute temperature T (Fletcher, 1962). Thus,

$$S \cong 3.52\times10^{-8}(T^3 im/M)^{-\frac{1}{2}}.$$

For a particle of mean diameter d and density ρ, a direct substitution can be made for the particle mass so that, approximately,

$$S = 4.86\times10^{-8}(T^3 d^3 i\rho/M)^{-\frac{1}{2}}.$$

Fig. 1 illustrates the dependence of S on d for various values of the parameter $i\rho/M$ and for a temperature of 275K. Also shown in Fig. 1 is the fraction of the total tropospheric radioactivity F associated with particles having diameters less than d (Kalkstein *et al.*, 1959).

A representative fallout material may, for example, have a density of 3 gm cm^{-3}, a molecular weight of 200 and a dissociation constant of 2. Such a particle has a value of 0.03 cm^{-3} for $i\rho/M$. Reference to Fig. 1 shows that a supersaturation of 0.2 per cent is sufficient to ensure nucleation of all such particles of size greater than 0.1 micron. This size range corresponds to over 90 per cent of tropospheric fallout. Although the above values were chosen at random, it is felt unlikely that any fallout particle would differ by more than an order of magnitude from the result of 0.03 cm^{-3} for $i\rho/M$. The conclusion that most fallout particles are capable of acting as efficient condensation nuclei in conditions conducive to the production of warm rain is inescapable.

An indication of the ease with which the fallout particles under consideration can be removed by nucleation can be obtained from estimates of supersaturations achieved in clouds of various types. Twomey (1959) has calculated that supersaturations of between 0.12 and 0.17 per cent can be expected in a maritime cloud having an updraft of 10 cm sec^{-1}. For an updraft of 1 m sec^{-1}, Twomey obtains a range of 0.57 to 0.75 per cent. Clearly these values are sufficiently large to cause efficient nucleation.

To compare the nucleating efficiency of radioactive debris and naturally occurring particles it is interesting to consider the sodium chloride nuclei thought to be

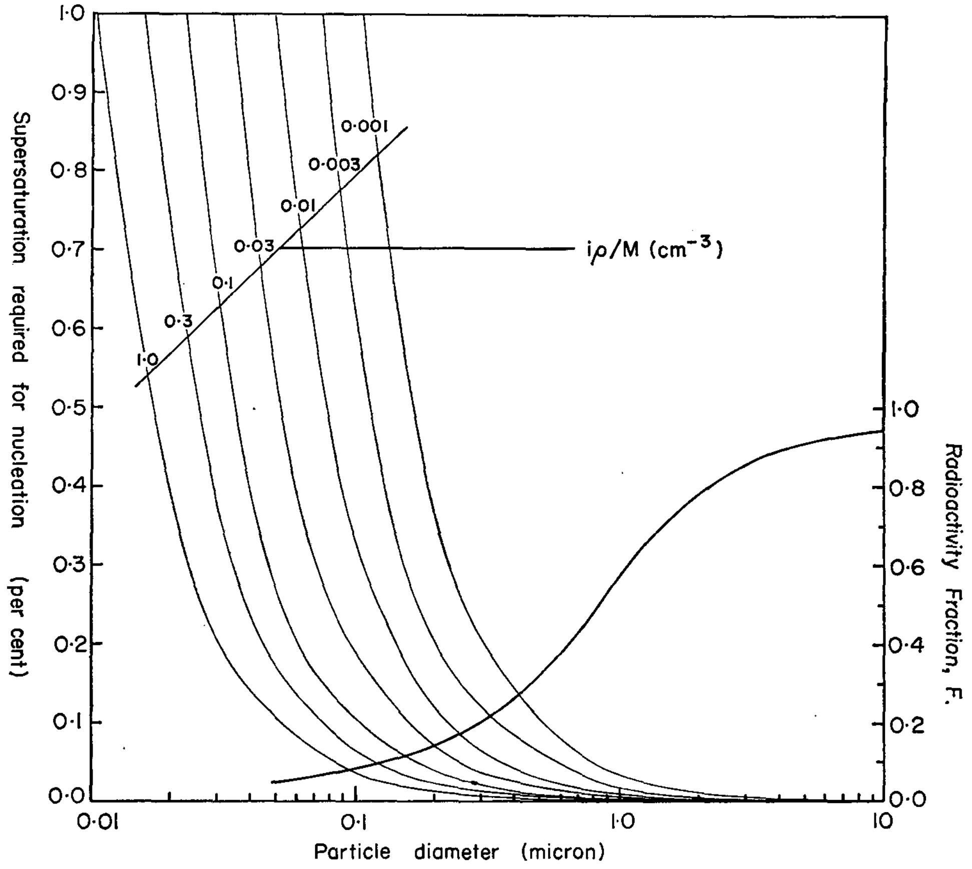

FIG. 1. Supersaturation required for effective nucleation by atmospheric particles and the size distribution of tropospheric radioactive fallout (Kalkstein *et al.*, 1959).

responsible for the large cloud droplets found early in the life of a convective storm. Measurements made by Wright (1940) of the sizes of sea-salt nuclei indicate that the mean mass of these particles is about 2×10^{-14} gm. The corresponding supersaturation required for effective nucleation is 0.029 per cent. From Fig. 1 an average fallout particle has a diameter of about 0.8 micron. Using the same values of i, ρ, M, and T employed earlier, the necessary supersaturation for nucleation of our mean fission product appears to be 0.015 per cent. It is apparent that fallout and naturally occurring sodium chloride particles are similar in their effectiveness.

The role of other aerosols in nucleation is not completely understood. It is known that a large proportion of continental nuclei are composed of ammonium salts while other nuclei contain compounds of calcium, magnesium, potassium and silicon (Whitehead and Feth, 1964). However, since sea salt forms the majority of maritime hygroscopic nuclei (Twomey, 1954), we must conclude that in coastal regions fallout particles are removed in the same way as the majority of the larger condensation nuclei.

3. The effect on rainout

Essentially, the process of nucleation will remove the more efficient particles first. Thus the larger nuclei will be consumed preferentially whether these are naturally occurring salts or radioactive compounds. It is interesting to consider the deposition of solids in rainfall in this light.

The structure of the simple convective cell has been thoroughly investigated in the Thunderstorm Project (Byers and Braham, 1948). Three stages of growth of a shower have been identified. In the cumulus stage the cell contains a central updraft being fed from below and partly from the sides. As the updraft increases, the cloud droplets grow through collision and coagulation until they reach sufficient size to be precipitated as rain. In this mature stage of its life the cell exhibits a dual character in that both an updraft and a downdraft exist in conjunction. The updraft is supported by flow from below the cloud base and through its sides. Later still, when the cloud is dissipating, downdrafts only exist. Except in this last case, we certainly have a flux of fresh nuclei into the cloud. A pure downdraft due to local circulations within the cloud, however, cannot cause such a flux.

Table 1. Distributions of radioactivity and precipitation in rainfall, Aspendale, Australia.

Date	Time	Rain	Beta activity***		Remarks
	(local)	(mm)	$(\mathrm{pC\ l^{-1}})$	$(\mathrm{mC\ km^{-2}})$	
24 Sept. 1962	0900–1200	1.22	118.6	0.145	Light showers 0.1 mm, 1105–1115
	1200–1300	2.75	46.2	0.127	First part of shower commenced 1235
	1300–1330	0.52	32.1	0.017	Second part of shower ended 1330
	1330–1530	0.13	38.2	0.005	Single shower, very light rain
	1530–1630	0.11	64.5	0.007	Light showers
	1630–0900	2.72	43.7	0.119	Continuing showers overnight
28 Sept. 1962	1405–1415	0.76	7.21	0.0055	Continuous period of heavy rain, commencing at 1040. Between 1040 and 1405, 3 mm fell in four showers. Rain ceased at 1615
	1415–1425	0.47	9.95	0.0047	
	1425–1435	0.39	13.00	0.0051	
	1435–1455	1.12	20.13	0.023	
	1455–1545	1.15	11.93	0.014	
	1545–1600	0.44	6.77	0.0030	
	1600–1615	0.64	4.21	0.0021	
4 Oct. 1962	0850–0915	0.80	94.9	0.076	First shower lasted from 0850 until 0925
	0915–0945	0.57	34.6	0.020	
	1015–1045	0.20	107.4	0.021	Sudden downpour 1035–1038 (0.20 mm)
	1045–1115	0.46	104.9	0.048	Gradually increasing intensity
	1115–1145	1.29	81.3	0.104	Steady continuous rain
	1145–1215	1.08	198.0	0.213	Rain ceased 1155. Recommenced 1205
	1215–1245**	0.71	—	—	Heavy rain stopped. Drizzle started 1230
	1315–1345*	0.35	142.3	0.050	Very light continuous rain
	1345–1445*	0.43	243.6	0.104	Continuous rainfall
	1445–1515*	2.22	69.2	0.154	Thunder commenced at 1515
	1515–1530*	2.75	19.5	0.054	Thunderstorm ended 1530
	1530–1600*	0.25	64.7	0.016	Light rain ended 1550
	1600–1045	2.00	36.1	0.072	Further rain overnight
17 Oct. 1962	1645–1648*	1.30	406.8	0.529	Sudden heavy downpour
	1648–1651*	1.12	73.8	0.083	
	1651–1654*	1.25	64.5	0.081	
23 Oct. 1962	0925–1005*	0.45	16.6	0.007	Drizzle commenced 0925
	1005–1045*	0.38	66.6	0.025	Continued drizzle
	1045–1055*	0.75	8.8	0.007	Heavy rain
	1055–1105*	0.33	10.7	0.004	Heavy rain
6 June 1963	1455–1500*	0.51	12.9	0.0066	Rain commenced 1455
	1500–1504*	0.82	3.6	0.0030	Steady rain continuing
	1504–1507*	0.56	3.4	0.0019	
	1507–1509*	0.68	5.0	0.0034	
	1509–1511*	0.57	4.1	0.0023	
	1511–1514*	0.76	2.7	0.0021	
	1514–1518*	0.92	3.7	0.0034	
	1518–1600	0.55	5.9	0.0032	Rain stopped 1520. Small shower at 1545
	1600–1605	0.41	3.7	0.0015	Steady rain recommenced 1600
	1605–1615	0.50	4.2	0.0021	Steady rain
	1615–1623	0.46	3.5	0.0016	Steady rain
10 Sept. 1963	1614–1620	0.33	52.2	0.017	Visible squall line. Rain started 1614
	1620–1629	0.26	31.2	0.0082	Steady rain, intensity decreasing
	1629–1640	0.11	45.9	0.0050	Rain ceased 1640
	1650–1701*	0.52	30.8	0.016	New storm commenced 1650
	1701–1706*	0.25	15.1	0.0038	Rainfall rate nearly constant
	1706–1710*	0.28	12.6	0.0035	Steady rain
	1710–1715*	0.25	10.4	0.0026	Steady rain until end at 1715
5 Dec. 1963	1432–1437	0.40	19.2	0.0077	Steady, heavy rain commenced 1432
	1437–1444	0.20	6.8	0.0014	Rain ceased 1440. A little drizzle
	1444–1449*	0.32	15.6	0.0050	At 1444 heavier rain started
	1449–1454*	0.30	14.4	0.0043	Continuous rain, intensity decreasing
	1454–1459*	0.15	7.2	0.0011	Rainfall continuing
	1459–1504*	0.15	7.2	0.0011	
	1504–1513*	0.22	6.0	0.0013	
	1513–1543*	0.28	3.3	0.0009	Rain stopped at 1533

* Samples used for analysis during continuous periods of rainfall.

** No measurement due to loss of sample.

*** Beta activity in picocuries per liter and millicuries per square kilometer.

TABLE 1. (Continued).

Date	Time (local)	Rain (mm)	Beta activity*** (pC l^{-1})	(mC km^{-2})	Remarks
17 June 1964	1356–1406*	0.49	19.9	0.010	Steady light rain, intensity decreasing
	1406–1416*	0.28	24.3	0.0068	Rainfall continuing
	1416–1436*	0.30	9.3	0.0028	
	1436–1501*	0.36	6.8	0.0025	End of rainfall at 1501
3 July 1964	0900–0925*	0.25	16.6	0.0042	Very light rain
	0925–0931*	0.65	8.3	0.0054	Heavier rain
	0931–0935*	0.28	1.5	0.0004	Rainfall rate decreasing
	0935–0945*	0.24	2.1	0.0005	Decreasing intensity
	0945–0950*	0.25	2.4	0.0006	Sudden increase in intensity
	0950–1000*	0.22	2.2	0.0005	Rain ceased 1000
	1000–1700	0.57	6.8	0.0039	Showers throughout day

A parcel of tropospheric air entering the vicinity of a cloud gradually becomes supersaturated. As it enters the cloud the largest and therefore the most efficient nuclei must be consumed first. As the air continues along its trajectory, progressively smaller particles will be removed. Subsequently most of the large particles in our parcel of air will be nucleated near the edge of the cloud. It follows that rain from near the extremities of the storm should be preferentially enriched with dissolved solids, including fallout, since this rain is derived from the vertical section receiving the greatest flux of particles.

Huff and Stout (1964) have commented on a second effect involving the correlation with rainfall rate. The fallout concentration in rain, A, will be an inverse function of the liquid water content, W, of the cloud, the important factor being the amount of radioactivity available per unit volume of water.

According to Best (1950), W is directly related to the precipitation rate, I, i.e.,

$$W = CI^{r},$$

where C and r are constants. The value of r was found to be about 0.85. All other factors being equal we would expect to find A inversely proportional to W. Subsequently the fallout and total solid concentrations should be nearly inversely proportional to the rainfall rate. Because the onset of rain is usually gradual, this effect

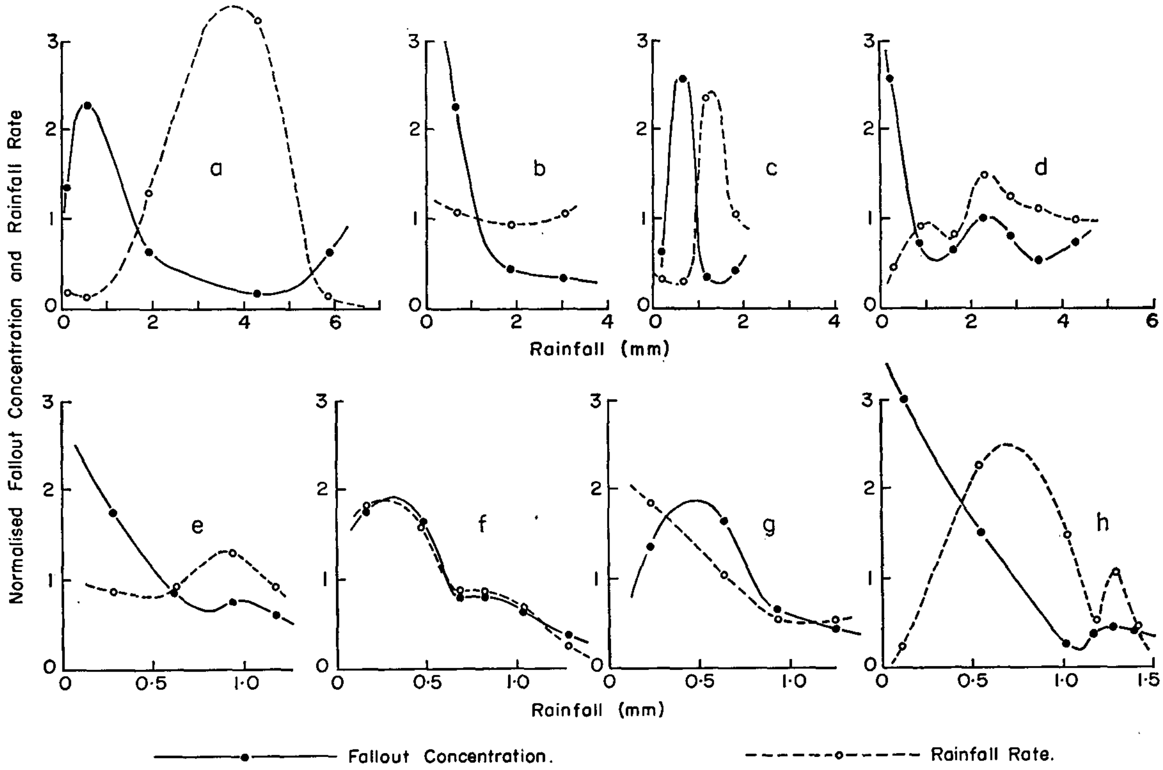

FIG. 2. Normalized rainfall concentration of radioactivity and precipitation intensity during continuous rain at Aspendale.

will enhance the early maximum due to preferential scavenging.

Such effects can be seen in each of the four types of beta concentration distributions given by Huff and Stout (1964). The series of measurements of the dissolved solid content of precipitation during a storm in January 1958, made by Whitehead and Feth (1964), shows a general maximum in concentration of solids early in the storm. Gambell and Fisher (1964) report maximum sulfate, nitrate and ammonium ion concentrations early in the passage of two thunderstorms.

The above considerations may have a more universal application. Although the effects of ice nucleation are not yet calculable, it seems likely that larger particles would be more efficient as freezing nuclei. Hocking (1959) concludes that the process of scavenging by cloud droplets becomes more efficient as the particle size increases, reaching a maximum when the cloud droplets and particles are the same size. Both these effects suggest that the preferential removal of most fallout near the edges of a storm may be quite general. This may account for the early maximum in the chemical composition of a snowstorm observed by Gambell and Fisher (1964). The inability of the present model to explain the thundershower of 17 August 1963 (reported by the same authors) when a maximum chemical concentration was observed at the middle of the storm may be due to the existing synoptic conditions being such that air entrained in the cloud may have previously been cleansed in other clouds in the squall line. In this case the entrained air would be deficient in large and giant nuclei and the greatest removal of particles must occur nearer the center of the storm.

4. Aspendale data

A study similar to that of Huff and Stout but not confined to convective storms has been conducted at Aspendale. Successive samples of rainfall collected during ten periods of rain were analysed for gross beta activity. A collecting area of 4 m^2 was used, so that samples of sufficient size were obtained from 0.1 mm of rain. The effects of dry fallout were eliminated either by careful washing or, later, by covering the collector when not in use. Samples were counted to an accuracy of better than 2 per cent. Results are listed in Table 1.

Of the ten periods investigated only eight include a continuous rainfall of sufficient duration for use in the present context. These eight showers are indicated in the table. The measured rainfall rates and fallout concentrations during these periods have been normalized by taking the ratio of each value to the mean through the storm. Fig. 2 illustrates the results.

In all cases in Fig. 2 the fallout concentration was a maximum early in the storm. While the corresponding precipitation rate was low in most cases, the storm began suddenly on three occasions (b,f,g). The early fallout peaks during these showers must be attributed to effects other than changes in the rainfall rate.

Secondary maxima in fallout concentration are to be seen on four occasions and these are all positively correlated with the rainfall rate. It seems reasonable to connect these maxima with the greatest downdraft through a mature raining cloud. The Wokingham hailstorm investigation (Browning and Ludlam, 1962) shows that such a downdraft is fed not only by convergence within the cloud but by an influx of air from the middle and upper troposphere. Thus there exists a downward flow through that part of the cloud below which we expect a peak in the precipitation intensity. Associated with this flow there must be a large particle flux from which fallout will be scavenged either by collision with falling raindrops or as a result of turbulent mixing within the cloud causing some of the entrained air to become sufficiently saturated to support nucleation.

Towards the ends of three storms the fallout concentration was seen to rise, indicating in at least one occasion (Fig. 2d) that preferential scavenging, as postulated above, may be occurring in this region of the storm.

5. Conclusions

That nucleation is of prime importance in the removal of tropospheric contaminants is supported by theoretical and experimental results. Observations of a large concentration of fallout at or near the commencement of a storm are attributable largely to this effect. Another contributing mechanism is the inverse relationship of the concentration on precipitation rate, which is often small at the start of a shower.

Rates of rainfall have a further influence in that the greatest flux of particles into a mature storm is associated with the greatest intensity of rain. Consequently the overall relationship between concentration and intensity will be difficult to resolve. However, it is possible that further careful work may enable atmospheric contaminants and radioactive fallout in particular to be used as tracers in the study of cloud structure in much the same way as they have been employed in general circulation studies.

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