Insoluble Particles in Hail and Rain

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ABSTRACT

Varying concentrations of different sized, solid, water-insoluble particles in rainwater and hailstones collected during thunderstorms indicate the existence of several scavenging mechanisms of micron-size particles. It is shown that Stefan flow is probably the predominant mechanism of in-cloud scavenging of particles 1.5–5 μ in diameter when solid (ice crystal) and liquid (supercooled cloud droplet) phases are present simultaneously. Particles larger than 5 μ in diameter were scavenged primarily by impaction. On many occasions, concentration of particles larger than 100 μ diameter in rainwater was found to be inversely proportional to rainfall intensity. Studies of spatial distribution of solid particles in hailstones should be supplemented by isotopic analysis.

1. Introduction

List (1965) has pointed out many times that "a hailstone can be regarded as a sonde, fallen through a thunderstorm cloud, its life history imprinted in its structure." Attempts have been made to decipher the mechanism of hailstone formation by studying entrapped water-insoluble particles (Rosinski, 1966). By applying similar studies to rain it should be possible to learn more about the mechanism of rain formation. Concentrations of different sizes of solid water-insoluble particles in hailstones and rainwater were studied as functions of the lifetime of individual storms. During the 1966 summer program, emphasis was placed on severe storms of local origin (afternoon or evening convective thunderstorms). The synoptic situations for those isolated storms are not known because mobile laboratories were equipped primarily to sample aerosol particles and precipitation.

2. Experimental method

About 70 samples of rain (not single drops) and hailstones from 12 thunderstorms near the Colorado-Nebraska border were collected systematically between 13 June and 24 July 1966. Additional samples were collected in the Colorado mountains and in Minnesota, mainly for comparison.

Rainwater was collected in glass bottles in an 11.5-cm diameter nylon raingage. An attempt was made to change the container when the rainfall intensity visually changed. The amount of water collected was determined by weight, and the intensity of the rain was computed in gm min^{-1} m^{-2}. The collecting apparatus was cleaned to remove almost all particles larger than 1.5 μ in diameter. The standard of cleanliness set up for particle-free containers was: <10 particles cm^{-3} for $1.5<d<5$ μ; <1 particle cm^{-3} for $5<d<13$ μ, and zero particles >13 μ diameter in 10 cm^3 of particle-free water. The particle-free water contained a drop of formaldehyde and a drop of wetting agent. The apparatus was handled in a clean chamber and sealed in plastic bags before the field tests. Blank bottles, similarly cleaned and handled, showed no entrainment of particles above the background noise of the counter. Concentration and size distribution of solid insoluble particles were determined using the Coulter Particle Counter, Model B (Coulter Electronics Inc., Chicago, Illinois). All particles of a diameter larger than 100 μ were separated by filtration and counted on membrane filters.

Hailstones were gathered from grass surfaces, transferred to plastic boxes, and transported to the laboratory in freezers or cold boxes. They were subsequently transferred into freezers for storage. The temperature of the field and laboratory storage compartments was kept below −15C. Since the outer surfaces of the hailstones were cleaned prior to analysis, plastic storage boxes were washed only once before use.

Concentrations of condensation nuclei, aerosol particles and ice nuclei were determined during and between storms. Concentrations of condensation nuclei were determined by means of the type CN small-particle detector (Gardner Associates, Inc., Schenectady, New York) at its lowest expansion of 5 cm Hg. Values thus obtained are in excess of the concentration of cloud condensation nuclei, but they correspond more closely to the concentration of cloud condensation nuclei than to values measured at higher expansion ratios. Concentrations of condensation nuclei measured at ground level during thunderstorms were extremely irregular. Changes in concentration of two orders of magnitude

TABLE 1. Particle concentration in rainwater.

Date and place	Time of sampling	Rainfall rate (gm min^{-1} m^{-2})	Number of particles in $\Delta d\mu$ size range per cm^3										
			1.5–3	3–5	5–7	7–9	9–11	11–13	13–30	30–40	40–70	70–100	over 100
22 June 1966	1635–1645	13.5	(5)1.3			(3)1.7			(3)1.6		23		10
Big Springs	1652–1700	40.7	(5)7.2	(5)1.6	(4)3.2	(3)9.5	(3)7.5	(3)5.1	(4)1.2	(3)3.1	(3)3.7	47	82
Junction,	1742–1752	68.6	(5)1.8	(5)1.1	(4)1.3	(3)1.9	(3)4.3	(3)1.7	(3)2.7	(2)1.4	(2)2.3	0	24
Nebr.	1802–1811	80.2	(5)1.9	(4)8.8	(3)6.9	(3)1.9	(3)1.4	(3)1.8	(2)8.3	(2)1.5	(1)9.5	0	7
30 June–	1647–1653	77.0	(5)6.9			(4)2.1			(4)2.4		(2)1.8		48
1 July 1966	1654–1656	389	(5)6.8			(4)1.4			(4)1.5		(2)1.3		20
Julesburg,	1706–1719	9.8	(5)6.6			(4)2.0			(4)2.6		0		(2)2.0
Colo.	1748–1802	19.6	(5)7.4			(4)1.6			(4)1.6		(4)1.6		(2)1.0
	1810–1820	269	(6)7.9			(3)7.7			(3)9.8		60		7
	1837–1842	170	(5)4.2			(4)1.4			(4)1.6		(2)1.3		15
	1905–1909	177	(5)4.6			(3)4.5			(4)1.2		77		25
	1918–1921	382	(4)8.3			(3)4.0			(3)4.6		1		12
	1925–1932	161	(5)7.2			(4)1.8			(3)8.8		34		20
	1945–1958	253	(5)5.2			(3)4.2			(3)8.2		66		10
	2015–2027	154	(4)6.3			(2)2.8			(3)1.6		12		14
	2028–2035	43.5	(5)1.2			(3)2.5			(3)2.2		0		6
	2045–2105	31.4	(6)1.2			(3)9.0			0		0		(2)6.9
	2140–2158	50.9	(4)7.8			(2)7.4			(3)1.6		20		10
	2202–2218	32.4	(4)4.8			(2)5.8			(2)7.3		0		14
	2218–2250	11.4	(4)5.5			(3)1.3			(3)1.3		1		11
	0053–0120	17.4	(5)1.1			(2)8.8			(3)1.4		0		20
	0120–0150	20.3	(4)9.0			10			(2)5.4		0		11
	0230–0240	44.5	(6)1.5			(3)6.9			(3)4.1		1		4
	0317–0340	8.8	(5)2.1			10			0		0		14
1 July 1966	1830–1840	20	(5)3.2			(3)2.1			(3)6.6		(2)6.3		19
Sydney,	1840–1850	514	(4)4.4			(2)5.2			(3)8.4		(2)4.1		2
Nebr.	1845–1915	189	(5)1.9			(2)9.5			(3)6.8		25		3
	1857–1903	614	(5)1.3			0			(3)2.3		2		3
	1915–1945	2.2	(5)8.9			(3)6.7			(3)4.0		(2)7.7		63
6 July 1966	1750–1752	909	(5)3.4	(4)7.9	(4)2.0	(3)7.6	(3)5.9	(3)1.7	(3)5.3	(2)5.3	(2)1.3	0	37
Peetz, Colo.	1754–1756	653	(5)2.2	(4)3.8	(3)6.9	(3)2.0	(3)2.3	(3)1.3	(3)2.2	(2)1.1	63	2	10
	1800–1801	3310	(5)1.2	(4)1.7	(3)3.4	(3)1.2	(3)1.6	(2)7.6	(2)4.3	(2)1.1	10	0	7
	1803–1805	552	(4)8.3	(4)1.3	(3)4.3	(3)2.7	(3)2.2	(3)1.1	(3)1.8	78	0	0	7
	1817–1828	133	(5)1.5	(4)7.1	(3)7.4	(3)1.9	(3)1.4	(2)7.3	(3)1.3	63	0	7	5
13 July 1966	1903–1910	78.1	(5)2.5	(4)4.3	(4)1.8	(3)8.9	(4)2.6	(4)2.1	(4)2.9	(3)4.1	(3)3.7	(2)1.1	54
Lorenzo,	1912–1917	112	(5)4.2	(4)6.3	(4)1.6	(3)7.9	(5)7.0	(3)3.7	(3)6.3	(2)5.7	(2)4.4	11	36
Nebr.	1929–1939	27.6	(5)5.0	(4)8.2	(5)2.4	(3)6.8	(3)8.6	(3)2.9	(3)2.2	(2)2.2	0	0	(2)1.5
	1935–1939	413	—	(3)6.6	(2)8.5	(3)2.8	(3)1.7	(2)7.9	(3)1.3	(2)4.5	13	0	9
21 July 1966	1519–1520	1051	(5)1.8	(4)4.6	(4)1.7	(3)7.1	(3)4.8	(3)2.8	(3)4.1	(2)3.1	(2)1.7	9	35
Purcell, Colo.	1526–1530	7.1	—	—	—	—	(4)2.6	(3)2.7	(3)3.9	(2)7.7	12	0	(2)2.4
	1615–1617	718	(4)5.2	(4)1.4	(2)4.8	(2)2.5	(3)1.6	(2)5.4	(2)9.3	(2)1.1	54	8	11
	1620–1622	137	(4)6.4	(4)1.6	(3)6.0	(3)1.5	(3)2.6	(2)7.9	(3)1.2	(2)1.5	75	0	22
	1831–1838	86.9	(5)2.7	(4)5.7	(4)1.7	(4)1.0	(3)6.6	(3)3.7	(3)7.2	(2)3.5	(2)3.6	23	60
	1844–1854	31.9	(4)8.9	(4)2.4	(3)9.2	(3)3.7	(2)2.4	(3)1.0	(3)1.7	(2)1.1	74	0	43
23 July 1966	1636–1641	191	(5)1.3	(4)4.8	(4)1.3	(4)1.2	(4)1.0	(4)6.4	(4)1.0	(2)6.3	(2)2.6	13	13
Kimball,	1700–1702	1245	(5)1.2	(4)2.7	(4)5.0	(4)2.6	(3)9.9	(3)5.3	(3)8.4	(2)6.7	(2)3.1	32	36
Nebr.	1712–1714	650	(5)1.0	(4)2.0	(3)7.2	(3)3.3	(3)3.3	(3)1.9	(3)2.8	(2)1.8	73	4	12
	1741–1742	1160	(4)6.8	(4)1.3	(3)2.4	(3)1.8	(3)1.1	(3)4.1	(3)1.3	(2)1.0	32	13	14
	1758–1759	3350	(5)2.3	(4)3.4	(3)6.4	(3)2.9	(3)3.3	(3)1.3	(3)1.8	(2)8.5	33	7	7
	1809–1812	692	(5)1.6	(4)1.2	(4)1.2	(3)5.4	(3)4.5	(3)2.7	(3)5.2	(2)2.5	(2)1.9	21	19
	1818–1820	819	(5)1.8	(4)3.1	(4)1.0	(3)3.5	(3)4.2	(3)2.1	(3)4.3	(2)1.7	(2)1.3	13	24
	1828–1830	1190	(5)1.7	(4)2.1	(3)3.1	(2)9.9	(3)1.3	(2)9.9	(2)3.2	5	5	0	65
4 August 1966	1610–1613	92.9	(5)2.7	(4)2.5	(3)9.0	(3)2.2	(3)3.2	(3)1.1	(3)3.3	(2)2.2	(2)1.5	0	(2)1.2
Bemidji,	1658–1700	173	(5)2.7	(3)4.1	(3)1.2	0	(2)7.3	(2)2.6	(2)2.1	0	29	0	23
Minn.	1711–1713	199	(5)1.6	(3)7.5	(3)5.0	0	(3)7.8	(3)3.1	(4)6.6	0	(2)3.1	0	16
	1715–1718	58.5	(5)3.9	(3)3.3	(3)2.2	(3)2.3	(3)2.2	0	(2)4.5	(2)4.5	(2)1.1	0	16
	1744–1750	11.1	(5)4.2	0	0	0	0	0	0	0	0	0	74
	1750–1812	10.6	(5)2.3	(3)5.8	(3)4.9	(3)2.9	(2)4.9	97	(2)5.3	97	97	0	19
	1800–1825	8.7	(5)1.7	(3)3.8	(3)1.9	(2)1.9	(2)4.3	(2)3.9	(2)8.2	48	48	0	31
	2230–2239	100	(4)4.0	(4)1.0	(3)2.6	(3)1.2	(2)7.9	(2)6.3	(2)9.3	20	20	0	9
	2250–2303	8.9	(5)1.2	(4)1.3	(3)7.3	0	(2)7.6	(2)8.2	(3)1.1	0	0	0	10
12 August 1966	0912–0932	30	(4)4.2	(3)4.1	10	10	(2)1.0	(2)2.3	(2)1.3	82	80	10	54
Long's Peak	0932–0936	216	(4)2.3	(3)2.8	10	10	(2)1.1	(2)9.9	(2)1.4	71	71	14	23
Colo.	0936–0940	120	(4)4.8	(3)4.9	(2)4.4	10	(2)4.9	(2)2.4	(2)4.2	(2)1.1	22	0	29
	1000–1020	40	(4)3.8	(3)3.5	(2)3.1	10	(2)2.7	(2)1.3	(2)2.9	97	56	10	45

The numbers in parentheses indicate the power of 10 by which tabulated values are to be multiplied.

TABLE 1.—(*Continued*).

Date and place	Time of sampling	Rainfall rate (gm min^{-1} m^{-2})	Number of particles in $\Delta d\mu$ size range per cm^3 1.5–3	3–5	5–7	7–9	9–11	11–13	13–30	30–40	40–70	70–100	over 100
31 August 1966	1522–1530	36	(5)1.4	(4)1.8	(3)3.9	(3)1.9	(2)9.7	(3)1.4	(3)1.7	(2)1.5	49	12	32
Caribou,	1530–1540	9.6	(4)7.6	(3)8.6	(3)8.6	(3)2.1	(2)6.5	(2)2.2	(2)7.0	(2)2.6	27	0	46
Colo.	1540–1550	96	(4)4.7	(4)1.0	(3)2.5	(3)2.3	(2)6.3	(2)2.4	(2)4.9	(2)2.6	11	0	23
	1550–1600	86	(4)2.7	(3)4.8	(2)7.5	(2)7.5	(2)3.5	(2)2.5	(2)7.8	(2)2.5	13	0	11
	1600–1607	34	(4)9.4	(4)2.0	(3)3.8	(2)9.4	(3)1.2	(2)2.8	(2)9.9	(2)2.3	59	12	21

in 10 min were not uncommon. The range of concentrations recorded was 10^3–10^6 cm^{-3}.

Aerosol particle concentrations were determined using all-glass impingers (Greenburg and Smith, 1922). To prevent contamination of samples by particles generated in the impingers by ground glass connections, the connections were coated with silicone oil. Since neither isokinetic sampling conditions nor proper alignment of the sampling system to wind direction were attempted, estimation of concentration of aerosol particles larger than approximately 10 μ in diameter would be meaningless. Only aerosol particles 1.5–5 μ in diameter, and insoluble in water, were considered with this technique. The concentration of particles larger than 5 μ in diameter was determined by counting, under a microscope, the particles accumulated on membrane filters mounted in open-face filter holders. The concentration of aerosol particles varied as follows during the thunderstorms from which rain and hail samples were obtained.

Particle diameter (μ)	Concentration range (cm^{-3})
1.5–5	0.05–200
5 –13	0.05–2
13 –45	0.01–0.5

Variations in time were as great as three orders of magnitude in 20 min.

Concentrations of ice nuclei were determined using the NCAR Ice Nucleus Counter (Langer *et al.*, 1967). These measurements were backed up by the membrane filter technique (Langer and Rosinski[1]). Ice nucleus concentration measured at −21C did not show any correlation with aerosol concentration measurements except during one storm, which will be discussed in Section 3. The ratio of ice nucleus concentration from the NCAR Ice Nucleus Counter to that from the Bigg-Warner expansion chamber was approximately 0.25. The recorded ground-level concentrations of ice nuclei did not differ during stormy and stormless days. They ranged from 10^{-4} to 10^{-1} cm^{-3}. The average concentration for 3 km altitude in Colorado, during the year of 1966, was approximately 10^{-3} cm^{-3}.

[1] Langer, G., and J. Rosinski, 1966: Detection of ice nuclei: a new continuous automatic technique and an improved membrane filter technique. Paper presented at Amer. Geophys. Union, Sixth Western National Meeting, 7–9 September, University of California, Los Angeles.

3. Particles in rainwater and hailstones

It was impossible in *this preliminary study* to make continuous measurements of the concentration of aerosol particles. Collection of particles was interrupted by malfunction of some of the instruments and by failure to collect from those storms which changed their direction of travel and were not intercepted by the mobile laboratory. Results have been classified into a number of groups in an attempt to present as continuous a picture as possible of the history of aerosol particles in storms.

The results of the rainwater analyses are given in Table 1. The size class interval used in this table is not uniform and is larger for diameters above 13 μ than for smaller diameters. When the concentration of particles larger than 13 μ in a size interval is below 5, the concentration is reported as zero. All particles above 100 μ diameter were separated from the whole sample and counted under a microscope; numbers in the "over 100 μ" column therefore represent absolute concentrations.

The results of the hailstone analyses are given in Table 2.

Since every storm is associated with its own characteristic aerosol concentration, storms should be analyzed individually. The mechanism of scavenging of aerosol particles by cloud particles and different forms of precipitation may be similar for similar storms generated in the same area. Concentrations of scavenged particles, on the other hand, may differ considerably from one storm to another. In the temperate zone, removal of aerosol particles under storm conditions is probably as follows:

a) removal of cloud condensation nuclei by formation of cloud droplets;

b) removal of ice nuclei by formation of ice crystals;

c) removal of aerosol particles by cloud droplets due to Brownian motion;

d) removal of aerosol particles by cloud droplets and ice crystals due to Stefan flow;

e) aerodynamic capture of aerosol particles by raindrops, ice crystals and hailstones;

f) capture of aerosol particles by different forms of precipitation due to electrostatic attraction; and

g) capture of aerosol particles in the presence of turbulent diffusion.

TABLE 2. Particle concentration in hailstones.

Date and place	Hailstone Type	Size, (gm)	Number of particles in $\Delta d\mu$ size range per cm³ 1.5–3	3–5	5–7	7–9	9–11	11–13	13–30	30–40	40–70	70–100	over 100
6 July 1966 Peetz, Colo.	milky	0.15	(6)1.6	(5)3.0	(4)4.3	(4)3.8	(3)9.8	10	0	0	0	0	24
	milky	0.21	(6)1.5	(5)2.7	(4)3.4	(4)2.1	(3)8.8	(2)1.1	(2)9.8	(2)3.2	0	0	27
	milky	0.50	(6)1.1	(5)1.6	(4)2.4	(3)4.3	(3)5.0	(2)1.4	(3)7.4	(2)8.5	0	0	21
	milky	0.70	(5)6.5	(5)1.2	(4)1.4	(3)3.1	(3)3.6	10	(3)2.0	(2)1.5	(2)2.1	0	44
7 July 1966 New Raymer, Colo.	milky	0.33	(6)1.3	(4)4.3	(4)2.5	—	(3)9.0	(3)6.2	(4)1.7	(2)3.1	(2)3.1	0	(2)1.9
	milky	1.88	(5)1.8	(4)4.9	(4)1.1	(3)1.2	(3)4.9	(3)1.3	(3)2.9	(2)1.8	0	0	38
	mixed	0.28	(5)4.6	(4)8.2	(4)1.6	(3)4.1	(3)3.3	(2)9.2	(3)3.7	(2)3.1	0	0	38
	mixed	0.40	(5)3.6	(3)5.9	(4)1.2	—	(4)1.0	(3)2.9	(3)4.4	0	0	0	82
	mixed	0.43	(5)6.2	(5)1.2	—	—	(3)7.4	0	(3)3.7	0	0	0	(2)1.2
	mixed	0.52	(5)1.4	(4)1.3	—	(3)8.7	(3)1.1	(2).65	(2)6.5	(2)2.2	0	0	(2)1.1
	mixed	0.94	(4)5.7	(4)2.1	(4)1.2	—	(3)3.3	(2)6.0	(3)2.7	0	0	0	(2)1.2
	mixed	1.10	(4)4.2	(4)1.1	(3)2.2	(3)2.2	(3)1.3	(2)8.7	(2)4.3	(2)3.3	0	0	47
	transparent	0.15	(4)1.5	(4)1.5	—	—	(3)2.2	(3)4.5	(3)7.5	(2)7.5	0	0	95
	transparent	0.89	(4)1.5	(4)2.0	(3)5.6	(3)5.6	(3)2.6	(2)5.6	(3)2.6	0	(2)1.4	0	72
	transparent	1.41	(4)1.6	(4)1.3	(3)6.3	(3)2.1	(3)1.2	(2)7.1	(2)7.2	(1)9.2	(2)1.0	0	48
9 August 1966 New Raymer, Colo.	transparent (outside layer)	3.77	(5)1.3	(4)1.9	(3)5.8	(3)1.7	(3)1.2	(2)7.3	(2)7.3	29	29	29	19
	milky (center of stone)	0.52	(5)2.7	(4)5.4	(4)1.2	(4)8.9	(3)1.4	(2)7.7	(2)1.9	(2)1.9	(2)1.9	(2)3.8	69
8 July 1966 Bemidji, Minn.	mixed	12.46	(4)3.5	(3)6.9	(3)2.3	(3)6.6	(3)1.0	(2)3.6	(2)8.0	33	0	0	2
1 September 1966 Nederland, Colo.	milky	0.21	(6)1.6	(5)1.9	(4)3.5	(4)1.8	(3)7.7	(3)5.6	(4)1.9	(3)1.0	(2)4.4	24	(2)1.2

The numbers in parentheses indicate the power of 10 by which tabulated values are to be multiplied.

Analysis of water-insoluble particles captured by precipitation was performed only for particles above 1.5 μ in diameter. Smaller particles, though not detected by this technique, were present. Particles soluble in water were of course destroyed. Some hygroscopic particles were decomposed by hydrolysis, forming hydrosol. Agglomerates of clay particles were probably destroyed also.

Each of the processes by which particles are removed from the aerosol population operated with different efficiency for each particle size range.

Particles 1.5–3 μ in diameter. Cloud condensation nuclei are usually smaller than 1 μ in diameter and are soluble in water. Giant condensation nuclei (larger than 1 μ) are retained in suspension only if they consist of insoluble particles. Condensation nuclei 1.5–3 μ in diameter contribute only slightly to the number of water-insoluble particles found in precipitation.

Removal of 1.5–3 μ aerosol particles (other than cloud condensation nuclei) by cloud droplets due to Brownian motion was considered by Greenfield (1957), who also included turbulent coagulation in his calculation. Greenfield, using assumptions of 0.4 gm m^{-3} for the liquid water content of a cloud and 30 cm^2 sec^{-1} for the turbulent velocity gradient normal to streamlines, calculated that cloud droplets of 20 μ diameter will remove 1% of the 1.5–3 μ aerosol particles during a 15-min interval of aerosol particle-cloud droplet coexistence. The ground level concentrations of aerosol particles recorded were up to approximately 10 cm^{-3} before and during storms. The concentration of aerosol particles in clouds is unknown. At 3 km above ground the concentration was usually 1/100 of that on the ground on moderately windy days over the Colorado-Nebraska border (based on three measurements). Assuming that aerosol particle concentration in clouds during storms is 1/100 of that at ground level and that 1% of the particles were removed due to Brownian motion (Greenfield's method), the concentration of solid particles in cloud droplets, and consequently in rainwater, will be 2.5×10^3 cm^{-3}. Removal of aerosol particles should be higher in hailstorms because of the larger liquid water content of clouds (higher concentration of cloud droplets and larger droplets) and stronger turbulence. The increase of in-cloud scavenging should be four orders of magnitude to explain concentrations of up to 10^7 particles cm^{-3} found in precipitation.

Evaporation of raindrops during fall was not measured and is not taken into account; it would clearly increase the concentration of collected particles in rainwater. High relative humidity and high rainfall intensity during thunderstorms indicate that evaporation was not excessive. Therefore, some other efficient mechanism of aerosol particle capture must be operating in the 1.5–3 μ range.

Stefan flow (Davies, 1967) should always be considered when aerosol particles are mixed with evaporating and condensing water droplets. In thunderstorm

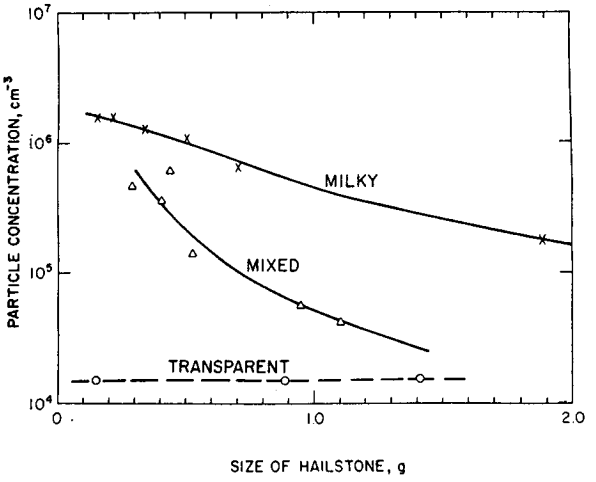

FIG. 1. Concentration of 1.5–3 μ particles in milky, mixed and clear hailstones.

clouds, Stefan flow should be effective because of the presence of ice crystals. The growth of the ice takes place in an environment of water vapor saturation with respect to supercooled cloud droplets, corresponding to ice supersaturation. Vittori and Prodi (1967) have shown that in a mixed cloud containing evaporating liquid droplets and growing ice crystals, aerosol particles are transported effectively in the direction of the growing ice phase. In their laboratory, capture of particles by a growing ice phase was found to be 16 times higher than capture on an equivalent water surface. If the same conditions operate in thunderstorm clouds, the highest concentration of 1.5–3 μ particles should be found in the centers of hailstones if the central ice crystal (grown by condensation of water vapor) originates the hailstone.

Concentration of particles of this size in milky, mixed, and transparent hailstones was plotted against the size of the hailstones (Fig. 1) collected during the 6–7 July 1966 storm activity. (Data from other storms cannot be plotted on this graph because every storm is associated with its own aerosol concentration.) Milky and mixed hailstones were distinguished by splitting the stones before analysis. Large (> 0.5 gm) milky hailstones contained irregular thin layers of clear ice, the amount of clear ice increasing with the size of the hailstones.

It can be seen that the concentration of 1.5–3 μ diameter particles is 100 times higher in small milky hailstones than in clear ice. The slope of the curves for milky and mixed hailstones represents the dilution of milky ice with clear ice. The low concentration of particles in clear hailstones was independent of hailstone size, indicating that the hailstones grew through accretion of cloud droplets. The high concentrations of particles in small milky hailstones indicates that the mechanism of graupel growth, in this particular case, should be by condensation of water vapor on an ice crystal. Consequently, the high concentration of particles found in some samples of raindrops indicates that the raindrops originated from melted graupel.

Particles 3–100 μ in diameter. Most natural ice nuclei appear to consist of particles of insoluble clay minerals. Clay particles which initiate ice crystals that later become raindrops will therefore be found in rainwater. Particle diameters range from 0.1 to 80 μ, depending on local conditions. Particles 0.1–10 μ in diameter (but mostly 0.5–1.0 μ) were found in snow crystals in the Greenland icecap by Kumai and Francis (1962). Soulage (1957), sampling ice nuclei during summer in Europe, found particles between 1 and 80 μ in size, with the majority in the 5–15 μ range. In the storms under investigation, the size of ice nuclei should be similar to that reported by Soulage. The size range 3–13 μ diameter should consist of ice nuclei and aerosol particles collected by aerodynamic capture.

Particles larger than 13 μ diameter were divided into 13–30 μ, 30–40 μ, 40–70 μ, and 70–100 μ diameter classes. Particles in the first two divisions usually showed a decrease of concentration in rainwater with progression of a storm, indicating washout by raindrops due to aerodynamic capture. Concentration of particles in the 40–70 μ diameter range, and especially in the 70-100 μ range, was very irregular, but no decrease in concentration with progression of the rainstorms was found.

Particles larger than 100 μ captured by rain. Aerodynamic capture by raindrops of 100-μ, or larger, particles is very efficient. Let us assume for the sake of this discussion that rain is relatively monodispersed for a short period of time. If rain is collected in such a way that the collecting device is changed with a visual change in rainfall, it is possible to collect a sample which approximates monodispersed rain. Actually, this means that a certain raindrop size can be assigned to the time interval which gives the largest contribution of captured aerosol particles. The concentration in rainwater of collected particles C can be expressed as

$$C=\frac{\text{number of particles captured by } n \text{ drops}}{\text{volume of } n \text{ drops}},$$

$$=\frac{\left(n\frac{\pi D^4}{4}haE\right)}{\left(n\frac{\pi D^3}{6}\right)}=1.5\frac{haE}{D}, \tag{1}$$

where

D = diameter of a collected raindrop (cm),
a = aerosol concentration (cm^{-3}),
h = length of cylinder swept by falling drop (cm),
n = number of raindrops falling (min^{-1} m^{-2}), and
E = efficiency of collection (fraction).

With the above definition of n, the denominator in Eq. (1) is equal to the intensity of rainfall, as deter-

mined experimentally. Experimental results are expressed as the concentration of scavenged aerosol particles relative to rainfall intensity. At a given rainfall intensity, the higher the aerosol concentration in air, or the longer the distance the raindrop falls, the more particles will be scavenged by the rain. The position of a curve on a graph is proportional to the product ahE (cm^{-2}). The concentration of particles in rainwater from Eq. (1) for $ah=1$, 5, 10, and 50 cm^{-2} and $E=1$ is given in Fig. 2 (solid lines). The corresponding raindrop diameters D (mm) for a different rate of rainfall n were added below. Experimental results for two mountain rainstorms (31 August, Caribou, Colo., and 12 August, Long's Peak, Colo.) were plotted for two different particle size classes. The slopes for particles larger than 100 μ in diameter are the same as those in Eq. (1). For particles 30–100 μ in diameter, however, the slopes are less steep, indicating probable loss of capture efficiency by smaller raindrops. There is no decrease of washout with time as these storms progressed, so there must have been a continuous influx of aerosol particles. Rain measurements from three other storms are plotted in Fig. 3. They show a straight line relationship for particle concentration relative to rain intensity. Slopes of the lines are different, though, for each storm.

The concentration of scavenged particles in collected rainwater should be higher than that in Eq. (1) because of evaporation of falling raindrops. A developed raindrop of some initial diameter falling through the air will partially evaporate before being collected, and so will actually sweep aerosol particles from the frustum of an inverted cone rather than from a cylinder. If half of a drop evaporates, the concentration in collected rainwater will be $C_e=1.9\ (haE/D)$ and the ratio C_e/C would

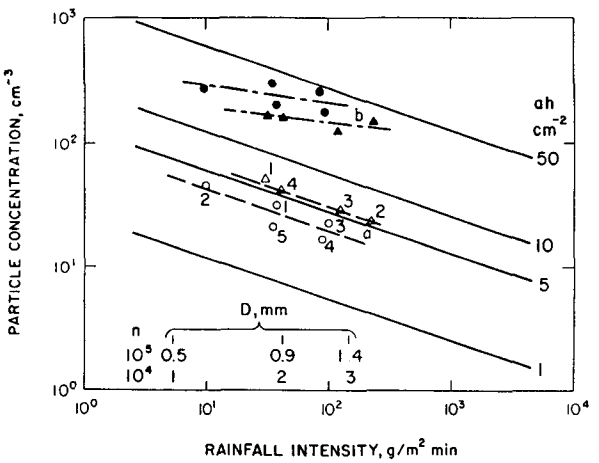

FIG. 2. Concentration of particles in rainwater from Eq. (1) and for two rainstorms plotted against rainfall intensity, the numbers representing successive samples. Data for 31 August 1966, Caribou, Colo., $d>100\,\mu$, open circles, $30<d<100\,\mu$, solid circles; for 12 August 1966, Long's Peak, Colo., $d>100\,\mu$, open triangles, $30<d<100\,\mu$, solid triangles. The solid straight line is a graph of Eq. (1).

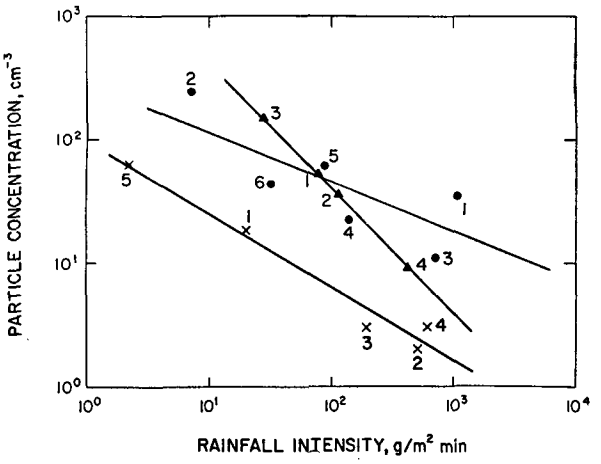

FIG. 3. Concentration of particles $d>100\,\mu$ in rainwater vs. rainfall intensity for three thunderstorms. Crosses, 1 July 1966; triangles, 12 July 1966; circles, 21 July 1966. Numbers represent successive samples.

be 1.3. This changes the position of the curve but not the slope.

During thunderstorms, it seems unlikely that aerosol concentration could remain constant; as soon as the ground is wetted by rain the supply of aerosol particles of local origin should be considerably reduced. Determinations of concentration of particles larger than 100 μ in diameter in rainwater collected during thunderstorms do not always verify this, however. Thunderstorms of 30 June–1 July, 1 July, 13 July, 18 July, 21 July, 23 July and 12 August did not show a decrease of concentration with progressing rainfall. Thunderstorms of 22 June, 6 July, 4 August and 31 August did show such a decrease as the storm progressed.

Many simultaneous measurements of concentrations of different aerosol particles were recorded during the 30 June–1 July storm. Rain started on 30 June at 1647 and continued until 0340 of 1 July. Concentration curves of condensation nuclei, ice nuclei and aerosols are given in Fig. 4, and of scavenged aerosol particles in Fig. 5. The ice nucleus concentration curve represents continuous sampling and the condensation nucleus curve was constructed from measurements taken every 15 min. Curves representing aerosol concentrations are smoother than the others because of longer sampling periods (10–20 min). Fig. 4 shows a definite correlation between condensation nuclei and ice nuclei, and a reflection of the same trends by aerosol particles. This is the first time that such correlation has been found. During this particular storm, ice and condensation nuclei must have originated from the same source, probably an aerosol cloud of soil particles lifted by the wind.

Particles larger than 100 μ in hailstones. Aerodynamic capture of particles differs for each form of precipitation. During thunderstorms, ice crystals are present in the clouds, but their lifetime is limited. They may collide with aerosol particles, but their efficiency of aerody-

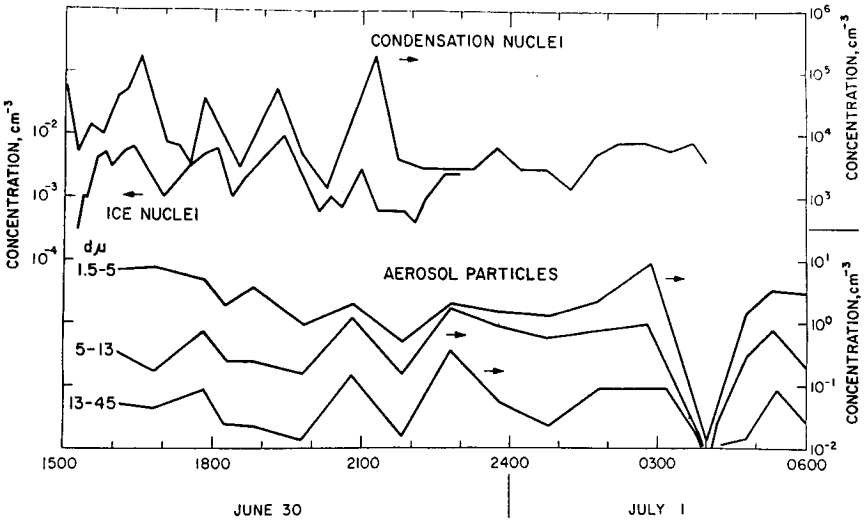

FIG. 4. Ice nucleus condensation nucleus, and aerosol particle concentration in the 30 June–1 July storm, Julesburg, Colo.

namic capture is low because of their small terminal velocity. Hailstones should collect a considerable fraction of the particles through which they fall. Because hailstones grow by accretion of cloud droplets and ice crystals, it is difficult to differentiate between particles captured as aerosol particles and those captured with cloud droplets. Particles larger than 100 μ in diameter (usually 35% grass fibers and 65% soil particles) are found in concentrations as high as 1000 cm^{-3} in the outer layers of hailstones. As has been shown previously (Rosinski, 1966), the concentration often decreases from the center of a hailstone through intermediate layers and then increases in the outer shell. Giant aerosol particles (larger than liquid cloud droplets) found in the center of hailstones may actually originate the hailstones as they fall through a cloud.

Hailstones were classified by appearance into milky (57%), mixed (26%) and transparent (17%), and by weight into small (0.1–0.4 gm), medium (0.4–1.0 gm), and large (1.0–2.0 gm). The distribution of 128 hailstones collected during the 21 July storm near New Raymer, Colo., is seen in Table 3.

Studies of spatial distribution of solid particles in hailstones should be supplemented by isotopic analysis, as described by Facy *et al.* (1963). One 8-cm hailstone from the storm of 5 July 1965 (Chadron, Nebr.), was

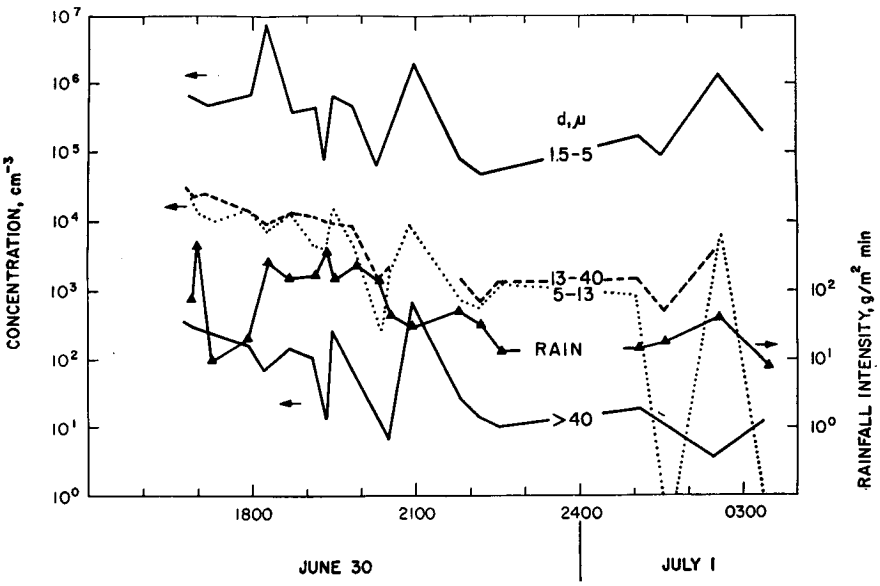

FIG. 5. Concentration of captured aerosol particles in rainwater, 30 June–1 July storm, Julesburg, Colo.

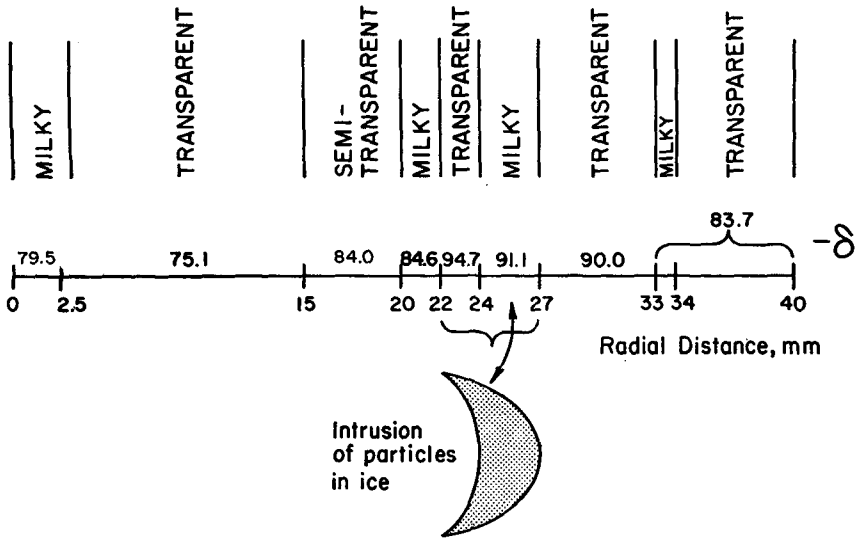

FIG. 6. Deuterium content in ice shells of a hailstone from storm of 5 July 1965.

sectioned, and ice from different layers was analyzed for deuterium content. The results are shown in Fig. 6. The deuterium content is expressed by

$$\delta = 1000\left(\frac{R_{\text{ice}}}{R_{\text{snow}}} - 1\right),$$

where $R = D/H$, and R_{snow} refers to the ratio for standard mean ocean water. The values of δ are negative. Lower values of δ (absolute values higher) in an ice layer indicate that the ice layer was formed from deuterium-depleted water. The transparent ice layer with $\delta = -94.7$ was formed in the most deuterium-depleted part of the cloud and, consequently, at the highest altitude relative to altitudes at which formation of the other layers of ice took place. It can be seen that the hailstone analyzed grew to $R = 15$ mm during a warmer period (release of heat during phase transformation and transfer into a warmer zone should be considered), grew up to $R = 24$ mm in a cooling period (updraft), and completed its growth during final descent. (78% of the total mass of this hailstone grew during terminal descent.) An interesting feature found in this hailstone is the presence, in a layer corresponding to the beginning of final descent, of a crescent-shaped intrusion made of captured aerosol particles up to 4 mm in diameter. The shape of this deposit indicated that the stone was definitely oriented during this portion of the descent.

TABLE 3. Per cent distribution of hailstones from hailstorm of 21 July 1966, New Raymer, Colo.

Type	Size Small	Medium	Large
Milky	55	27	18
Mixed	52	33	15
Transparent	50	45	5

4. Electrostatic forces and turbulent diffusion

Electrostatic forces and turbulent diffusion always exist in storm cells. Turbulent diffusion has been discussed and evaluated by Greenfield (1957). The role of electrostatic attraction in thunderstorms is extremely complicated, and the author will not attempt to speculate about its role, except to say that removal of very small particles may be influenced more by electrostatic forces than removal of large particles. Possibly, the high concentrations of 1.5–3 μ diameter particles is due to a combination of phoretic and electrostatic forces.

5. Conclusions

Despite the extreme complexity of storms studied, results of this field test program permit us to draw several broad conclusions. These will be listed according to particle size classes.

Particles 1.5–3 μ in diameter. In thunderstorms, these particles are scavenged by cloud ice crystals, primarily by motion in the diffusion field established when solid and liquid phases are present simultaneously. The high concentrations found periodically in rainwater indicate periodic formation of conditions favorable for in-cloud scavenging. From the viewpoint of in-cloud scavenging of particles there is a cellular character in a thunderstorm.

Particles 3–100 μ in diameter. Results were often so irregular for this size classification that general quantitative conclusions cannot be drawn at this time. Microscopic counts should be used in the future to insure accuracy of data.

Particles larger than 100 μ in diameter. The concentration of these particles in rainwater was found on many occasions to be inversely proportional to rainfall intensity. These particles, therefore, are removed primarily by impaction and interception (inertia forces).

Hailstones. Most of the hailstones collected had milky centers containing high concentrations of 1.5–3 μ diameter particles, which suggests that they were formed around ice nuclei grown from vapor. Isotopic analysis should be included in future studies to give additional information on the history of formation of hailstones.

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