

Chemical composition of rain water and rain characteristics at Delhi

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ABSTRACT

Rain water samples collected with the progress of rain shower on a number of rain occasions in different seasons at Delhi have been analysed for chloride, sulphate, sodium, potassium and calcium. The trend of variations in the concentration of chloride ion in rain water with the progress of a rain shower would seem to suggest that the growth of rain drops in convective clouds at Delhi would involve a process which might be analogous to what was envisaged by Woodcock for Hawaiian rains. Further, it was noticed that the characteristics of precipitation at Delhi are dependent on the amounts of air-borne soluble particles present in the cloud forming air. Smaller cation concentration (< 1 ppm) and higher value of Cl/Na (> 1.8) distinguish what may be called "freezing" rain (rain from cold layer clouds) from that of the other type in which both freezing and non-freezing mechanisms operate (rain from cold convective clouds). Smaller ratio values of sulphate to chloride (< 6) and higher ratio values of chloride to sodium (> 1.8) characterize heavier rainfall. Probable explanations have been suggested for these observed features.

1. Introduction

Investigations in recent years have shown that development of rain in warm, cold convective and cold layer type clouds takes place by all-water (non-freezing), combined ice-water (combination of freezing and non-freezing) and all-ice (freezing) mechanisms respectively. Formation and growth of precipitable size cloud droplets leading to actual precipitation is facilitated by presence in the air, of giant size hygroscopic particles in the case of warm and cold convective clouds, and of ice-forming aerosols in the case of cold layer type clouds. As the nature and extent of the role played by the aerosols in rain formation is dependent not only on the size and concentration of these particles but also on their chemical composition, the chemical nature of rain water in precipitation caused by different mechanisms would be different. Measurements by Turner (1955) have indicated that it may be possible to distinguish between "freezing" and "warm" rain from a comparative study of raindrop salinity with size. However, as collection of rain water samples consisting of raindrops of specific size ranges would involve considerable

special efforts being made for such a study, it was thought that it would be of interest to see if measurements made on bulk rain water samples could help distinguish rain of one type from the other. Also, as the broad character of rain in the area under study may differ in different seasons, this being dependent largely on the dominant circulation pattern (according to Rossby & Egner (1955) the chemical climate varies in a striking manner with changes in circulation) it was intended to examine how the chemical composition of precipitation might differ on widely differing rain situation in different seasons. A study on chemical composition of rain water was undertaken at Delhi ($28^{\circ} 35' N$, $77^{\circ} 12' E$ and $714' A.S.L.$) with these objectives in view.

2. Collection of rain water

Samples of rain water were collected sequentially with the progress of a rain shower, using stainless steel funnels of 50 cm (sometimes 24 cm) diameter fitted on to 1000 cc capacity polythene bottles which had previously been cleaned and rinsed with double distilled

water. The number of samples collected on individual rain occasions varied from 1 to as many as 20 depending upon duration and intensity of rainfall. The duration of collection of a sample range from 2 to 10 minutes in heavy to moderate showers and from 20 to 40 minutes in light rainfall.

3. Measurements

The concentrations of the anions, Cl and SO₄, have been systematically measured in all the rain water samples collected since 1963. The cations measured are sodium, potassium and calcium and these measurements were commenced in 1965. The analytical techniques employed for measuring the anions are, precipitating chloride compounds as silver chloride and of sulphur compounds as barium sulphate, and measuring the resulting turbidity with Bausch and Lomb Colorimeter. Sodium, potassium and calcium have been measured by Kipp and Zonen Flame Photometer.

Information relating to the dominant type of precipitating cloud (warm/cold convective/cold stratiform) at the time of collection of rain samples was obtained from examination of pictures taken on a high power 3.2 cm radar in use at the site. Based on criteria of height of echo top vis-à-vis freezing level, and of association or otherwise of cold rain cells with melting band indicative of rain by ice process, the precipitating clouds have been classified under the three groups, warm, cold convective and cold stratiform. If the echo on radar was confined below freezing height, the precipitating cloud was classified as "warm". If the echo top was above freezing level but showed no melting band structure, it was classified as "cold convective". But, if the echo top was well above the freezing level, and also well developed melting band was seen, the cloud under question was classified as "cold stratiform".

4. Results and Discussion

The features studied are (i) trend of variations of chloride ion concentration in rain water with the progress of a rain shower, (ii) chloride, sulphate, sodium, potassium and calcium ion concentrations in different rain situations and (iii) ratio of (a) sulphate to chloride and (b)

chloride to sodium, in rain water collected on different rain occasions.

There is one aspect of important consequence which has not been considered in the present study and that is the possible effects of evaporation and of washout on the ionic concentrations. As the samples of rain water have been collected near ground level the effects of evaporation and of wash out which might vary with season and also with the type of precipitating cloud could, at times, be significant—see Gambell & Fisher (1964) for evaporation and Georgii & Weber (1964) for tropospheric wash out—and as reflected in the measurements by Oddie (1962), samples collected even at higher levels were not free from contamination. The interpretation of the data in terms of rain formation processes as attempted below may have to be treated, therefore, only as suggestive but not conclusive.

(a) Chloride ion concentration in rain water samples

The concentration of Cl in rain water varied from sample to sample by a factor of about 2 in the beginning of a shower, becoming progressively less at later stages. A close examination of data collected has shown that the variations in concentration conformed generally to the one or the other of the following four patterns:

1. Concentration decreasing progressively till the end of the rain shower,
2. Concentration decreasing progressively at first, but increasing towards the end stages,
3. Concentration increasing progressively till the end of the rain shower, and
4. Concentration fluctuating within narrow limits, tending to become nearly steady later.

Fig. 1 represents an example each of the four typical trends mentioned. Of the 57 rain instances in course of each of which 3 or more samples were collected, 18 showed variation of type 1, 20 of type 2, 10 of type 3 and 9 of type 4. The pattern of variation noted in the majority of rain instances, identified as rain from cold convective clouds, is one or the other of the first three types mentioned, while in the case of rain from cold layer clouds, the majority of occasions indicated variation of type 4. All the four occasions identified as rain from warm convective clouds showed variation of type 1.

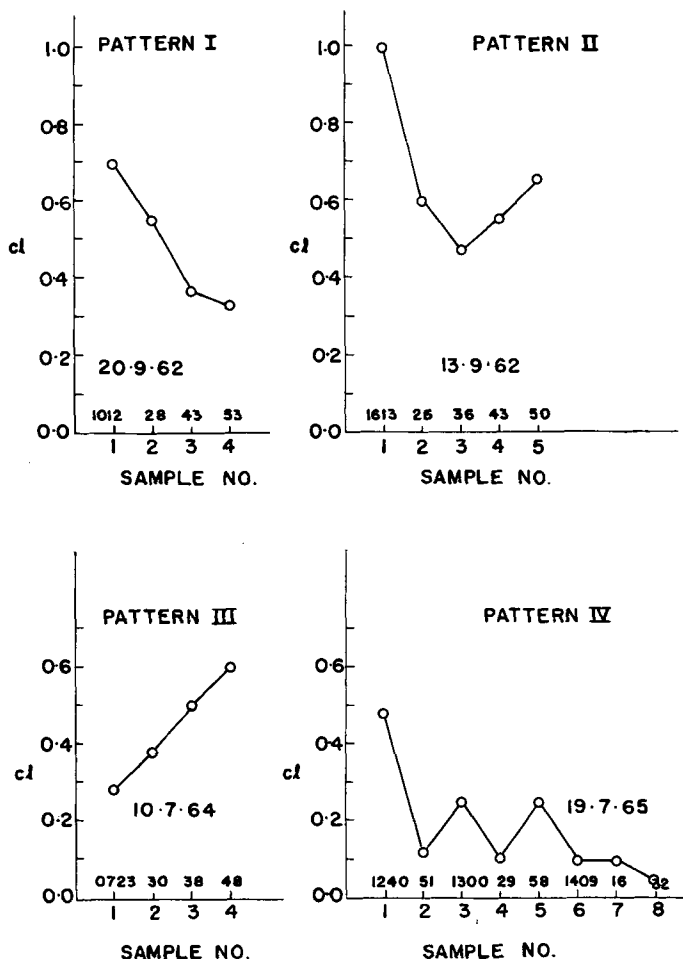


Fig. 1. Patterns of variation of chloride ion concentration (expressed in ppm) of rain water samples collected subsequently with progress of rain shower. Sample number and the mean time in I.S.T. of collection are given on the abscissa.

However, as rain from warm clouds occurs only with a random distribution in space and time, and as the net contribution by such rain to season's total rainfall at Delhi is extremely small, being only about 2% in the monsoon season (Ramana Murthy *et al.*, 1960), the mechanisms as may be considered to be effectively at work in the case of Delhi rains are either "all-ice" or a combination of "ice and water". A knowledge of the pattern of variation in the chloride ion concentration in rain water with the progress of rain shower would thus help distinguish rain situations by these two mechanisms.

Also, the observed trend of variation of

Chloride ion concentration as in patterns 1 to 3 which are nearly similar to salinity-dropsizes/salinity-time curves obtained by Turner (1955) in Hawaiian rains suggest that the giant hygroscopic aerosols which are found in sizeable concentrations on many occasions in the air at Delhi during the monsoon season (Ramana Murty *et al.*, 1962) could play an effective role in precipitation development from convective clouds in the region and that droplet growth on these particles could, perhaps, also involve a process which might be analogous to what was envisaged by Woodcock (1952) for Hawaiian rains, i.e. condensation followed by accretion of cloud droplets of uniform low salinity. The

Table 1. Average concentration of various ions in precipitation from different types of clouds during monsoon (1963-65)

Type of precipitating clouds	Concentration in ppm				
	Cl	SO ₄	Na	K	Ca
Warm convective	0.75	2.9			
Cold convective	0.35	3.3	0.63	0.36	1.76
Cold layer	0.27	1.5	0.11	0.07	0.13

general trend of dropsize distribution observed in rains support this contention (Ramana Murty & Roy, 1962). The pattern of the nearly steady chloride ion concentration (pattern 4) as observed in rain from cold layer type clouds lends support to the view that on such occasions the hydrometeors do not commonly originate around giant hygroscopic aerosols, but may have originally formed on ice-forming nuclei and then grown to raindrop size by coalescence with cloud drops whose mean salinity is near that of the resulting rain.

(b) Concentration of anions and cations in different rain situations

Rain from warm convective clouds showed high concentration of chloride (0.75 ppm) whereas that from cold layer clouds showed low amount (0.27 ppm). Sulphate also is found in low concentration in rain from cold layer clouds, (1.5 ppm) as against what it is in rain from cold convective clouds (3.3 ppm). The cations, sodium, potassium and calcium have been present in higher concentration (about five times or more) in cold convective rains than in the cold layer type ones. Considered season-wise, chloride and sulphate are found in high concentration in rains during summer (April-June) and in low concentration in rains during monsoon (July-September). The results of analysis which are given in Tables 1 and 2 are explained below.

High chloride in warm rain. The observed difference in Cl-concentration between warm convective clouds (0.75 ppm) and cold convective clouds (0.35 ppm) is found significant at 95 % level. It has been shown that the intensity of rainfall and the total amount of rain received from warm convective clouds are about an

order of magnitude lower than in the case of cold convective clouds (Ramana Murty *et al.*, 1960). As in general, in situations of low intensity rainfall the mean size of raindrops reaching ground is smaller, one would expect that, if each rain drop was considered as the product of growth on individual giant hygroscopic nucleus, warm convective rain should show higher content of chloride.

Low chloride in freezing rain. There is clear indication that the rain-forming process in such instances actually starts in the super-cooled regions (Ramana Murty & Srivastava, 1962). If, therefore, it is considered now that the solid hydrometeors in the super-cooled regions are formed on ice-forming aerosols which are largely chloride free and then grown by coalescence with cloud droplets of low uniform salinity, the resulting salinity of rain drops and therefore, of that of rain will be relatively small, as compared with rain from convective clouds.

Higher concentrations in showers than in rains. If rain situations classified under "warm" and "cold" convective types are considered as showers (duration of rainfall noticed in these cases is 10 to 30 minutes) and those classified under "cold layer" type as rains (duration of rainfall in this case often extends for hours) the results obtained are in sharp contrast with those of Macku *et al.* (1959) and Georgi & Weber (1960) who have found higher concentrations of Cl and SO₄ respectively in rains than in showers. Measurements by Mrose (1966) showed slight differences between showers and continuous rain, the value of SO₄ being higher in the case of the latter. There may be a correlation between the type of precipitation and the origin of airmasses, i.e., the chemistry of aerosols, but no precise information is available in this

Table 2. Average concentration of Cl and SO₄ in rain water during different seasons (1963-65)

Season	Concentration in ppm	
	Cl	SO ₄
Summer	1.90	8.8
Monsoon	0.43	2.9
Winter	0.67	6.2

Table 3. Concentration in ppm of Na, K and Ca in rain from cold convective (no bright band) and cold layer type (bright band) clouds

Rain from cold convective cloud					Rain from cold layer type cloud				
Date	Na	K	Ca	Total cation concentration	Date	Na	K	Ca	Total cation concentration
17.7.65	0.64	0.40	1.08	2.12	19.7.65	0.31	0.08	0.14	0.53
18.7.65	1.05	0.21	1.20	2.46	31.7.65	0.11	0.06	0.33	0.50
22.7.65	0.15	0.08	0.43	0.66	22.8.65	0.03	0.04	0.10	0.17
21.8.65	0.68	0.31	3.78	4.77	23.8.65	0.01	0.09	0.12	0.22
3.9.65	0.38	0.23	0.50	1.11	2.9.65	0.11	0.08	0.08	0.27
21.9.65	0.64	0.42	3.40	4.46	4.9.65	0.09	0.05	0.01	0.15
Average	0.59	0.28	1.73	2.60		0.11	0.07	0.13	0.31

regard. However, the following may be considered partly responsible for the observed finding. The atmosphere is relatively stable on days of rain from stratiform clouds, whereas latent instability is present on most days of convective rain (Ramana Murty *et al.*, 1960). The comparative stability and, as such, the less turbulent mixing obtaining on occasions of the former type does not allow free and rapid movement from below of aerosols upwards, whereas this is very much facilitated in situations characterizing development of convective type rain.

High concentration in summer rain. The period April to June is characterized by dry westerlies and the weather is invariably of intra-airmass character and predominantly of instability origin. Aerosols of local and land origin will continuously be lifted up and as a result a high level of aerosol concentration is maintained in the atmosphere. Also, occurrence of rain is infrequent. These features account for the highest concentration of Cl and SO₄ in precipitation when it occurs during this season.

Low concentration in monsoon rain. During the monsoon, the trough line demarcating the prevailing south-easterly bay monsoon air flowing up the Gangetic valley from the south-westerly Arabian Sea monsoon air dominating over the greater part of the Indian sub-continent runs very close to Delhi, and the relative position of this line with reference to station on individual days, determines the nature of the airmasses over the area and the weather which occurs there. Although some amount of aero-

sols would be picked up on the way while passing over relatively dry regions, the airmasses would get depleted, before reaching Delhi, of most of their original aerosol content along the path followed by them at every stage of rain development, which is most frequent during the season. Cl and SO₄ are, therefore, found on the average in very low concentration in rain water during this period.

Medium concentration in winter rains. During the winter period (November–March) the weather shows features somewhat similar to those of extra-tropical latitudes and is controlled chiefly by the passage of western disturbances. With the main western depressions following a track well to the north of Delhi, the area in question does not come always under direct influence of frontal weather systems, and rain occurrences are more often results of convergence within the same warm moist airmass drawn from more southerly latitudes under the influence of these depressions. Active weather development leading to rain over the area is more commonly associated with cold fronts, often occluded, rather than with warm fronts. Airmasses reaching the area during this period have a longer travel over land than in the monsoon. Also, occurrence of rain is infrequent, as compared with the monsoon season and a higher level of aerosol content is maintained in the atmosphere. These features account for the concentration of Cl and SO₄ in winter rains being higher than in monsoon rains.

Criteria for distinguishing different types of

Table 4. Mean concentration (in ppm) of SO_4 and Cl and mean value of the ratio SO_4/Cl in rain-water on rainy days categorised into "light" and "heavy"

Rainy day	SO_4	Cl	SO_4/Cl	Number of occasions
Light or passing shower (total rain recorded: < 20 mm)	6.2	0.70	8.9	13
Heavy or intermittent rain (total rain recorded: > 20 mm)	2.4	0.40	6.0	14
Light or passing shower (predominant size of rain drop: < 2 mm)	5.9	0.64	9.2	5
Heavy or intermittent rain (predominant size of rain drop: > 2 mm)	1.5	0.26	5.8	5

rain. Reference to Table 1 suggests that a knowledge of the content of the commonly occurring cations in rain water, namely, Na, K or Ca, may help indicate whether the rain in question is from cold convective cloud or from cold layer cloud. The day-to-day concentrations of these cations in rain under the two types of cloud situations are shown tabulated in Table 3. The values are substantially high in rain from convective clouds, the total cation content, on the average, being about an order of magnitude more. The values noticed for calcium differ to the maximum extent in the two different types of rain situations. As this element is by and large of long origin it occurs in higher concentration in convective rain situations in which rain develops by combined ice-water mechanism.

(c) *Ratio of sulphate to chloride*

The value of this ratio is found to be > 1 in all the rain water samples examined, being higher in the winter rains (average value is 9.2) and lower in the summer rains (average value is 4.6). Rain with higher sulphate content (4 to 6 ppm) has invariably shown higher SO_4/Cl also and such occasions have usually been those associated with lower rainfall activity. The latter feature has been critically examined by classifying rainy days into two categories, namely, light and heavy, based separately on the criteria (1) whether the total amount of rain received was less or more than 20 mm and (2) whether the predominant size of raindrop as indicated by raindrop imprints on dyed filter papers was less or more than 2 mm. The values of SO_4 and Cl for rain waters classified in this manner are given in Table 4. The observed differences in values between the two types of rain situations, which are found

significant at 95% level, point out higher value for SO_4/Cl in lighter rains.

However, considering that the value of the ratio SO_4/Cl might also depend on the wind direction (the direction of air currents affecting the Delhi region during monsoon is dependent upon the position of the monsoon trough) the feature has been further examined by classifying the rainy days in 1964 and 1965—these are years of very high and very low rainfall, the total amount of rain recorded in the two monsoon seasons respectively having been 1123.1 and 456.7 mm—according to the direction of wind at 1.5 and 2.0 km levels. The values of SO_4 and Cl for rain water classified in this manner, are given in Table 5. The observed differences which cannot be statistically proved suggest a high value of SO_4/Cl in light rains except in situations when winds are from the south east quadrant when no definite statement can be made.

The foregoing analysis of SO_4/Cl with respect to rainfall suggests that these two have certain interdependence, although it does not signify which is cause and which is effect. It may be that one of the parameters which determine the rain producing potential of the cloud is the chemical state of the cloud-forming air. The feature could, perhaps, be conceived as follows: As a considerable fraction of the atmospheric sulphate is found in aerosols of large size range (Junge, 1954), the presence of higher contents of sulphate could mean higher concentration of large size aerosol particles in the atmosphere. Clouds forming in such nucleus-rich air have higher droplet concentrations and, as demonstrated by Squires & Twomey (1958), such clouds are colloiddally more stable and their susceptibility to release rain is limited. However, as far as chloride aerosols are concerned,

Table 5. Values of the ratio SO_4/Cl and Cl/Na in rain water on different rain occasions considered wind wise

	N.W.		N.E.		S.E.		S.W.	
	1964	1965	1964	1965	1964	1965	1964	1965
Total number of days during July-Sept. when upper winds (1.5-2.0 km) were from the respective quadrants	41	54	9	11	31	23	11	9
Total number of rainy days	14	6	4	5	19	16	6	2
Total amount of rain received in mm on all rainy days	597.1	32.2	48.7	189.7	324.8	213.6	152.5	21.2
Average amount of rain received per rainy day (mm)	43	5	12	38	17	13	25	11
Average SO_4	3.0	4.4	2.0	1.2	3.8	2.7	3.5	3.3
Average Cl	0.63	0.51	0.25	0.26	0.37	0.33	0.48	0.33
Average Na		0.48		0.11		0.26		0.85
Average SO_4/Cl	4.8	8.6	8.0	4.6	10.3	8.2	7.3	10.0
Average Cl/Na		1.1		2.4		1.3		0.4

the effect is just the opposite, because a sizeable fraction of this component is composed by atmospheric aerosols in the giant size range and such giant size particles are considered to be the seeds for rain drop growth.

(d) *Ratio of chloride to sodium*

The value of this ratio in rain water varied with the character of rain in question. It was less than 1.8 (the mean value in sea water) on most of the rain occasions not associated with the development of bright band (rain from cold convective clouds) and more than 1.8 when bright band was present (rain from cold stratiform clouds). The mean values found respectively in the two types of rain situations are 0.67 and 2.36 (see Table 6). A knowledge of

Cl/Na in rain water could, therefore, help distinguish rain developed purely by ice mechanism.

Departures in the ratio of chloride to sodium from the sea water value have been generally attributed to possible modifications in the composition of sea salt particles during the process of separation from the sea surface or to addition of Na^+ or Cl^- from other sources (Rossby, 1955). While, on considerations such as these, it is possible to account satisfactorily for the low values of Cl^-/Na^+ (< 1.8), it is not easy to explain values more than 1.8 obtained systematically in certain type of rain situations as in the present study. Accumulation, with increasing distance from the coast, of Cl^- released from sea salt particles subsequent to their

Table 6. Value of Cl/Na in rain from cold convective and cold stratiform clouds

Rain from cold convective clouds				Rain from cold stratiform clouds			
Date	Concentration in ppm		Cl/Na	Date	Concentration in ppm		Cl/Na
	Cl	Na			Cl	Na	
17.7.65	0.26	0.64	0.41	19.7.65	0.18	0.31	0.58
18.7.65	0.33	1.05	0.31	31.7.65	0.22	0.11	2.00
22.7.65	0.23	0.15	1.53	22.8.65	0.31	0.03	10.33
21.8.65	0.50	0.68	0.74	23.8.65	0.28	0.01	28.00
3.9.65	0.30	0.38	0.79	2.9.65	0.26	0.11	2.36
21.9.65	0.68	0.64	1.06	4.9.65	0.33	0.09	3.67
Mean	0.38	0.59	0.64		0.26	0.11	2.36

formation might be expected to give rise to increase in the value of the Cl/Na ratio, but it is doubtful whether this process alone could be so effective as to make the ratio systematically higher than 1.8 in rain situations of a particular type. Also, if addition of Cl⁻ from other sources is to be considered responsible for the observed high value of Cl/Na, analysis of rain water should have indicated higher contents of the other commonly occurring cations like K and Ca with which that additional Cl⁻ could have been associated. But measurements have shown that in such rain situations, the concentrations of potassium and calcium have also been much less. In fact, as seen from Table 7, the decrease in the cation component has been more marked than that of the anion component in rain situations in which Cl/Na has been more than 1.8 (rain from cold layer clouds). These features indicate that the probable cause for the observed high value of Cl/Na lies primarily somewhere else. The authors are inclined to suggest in this connection that the possible absorption of trace gases by cloud drops in course of the water condensation process and during rest of the life cycle of the precipitating cloud could be responsible for the observed effect. Cloud and rain situations associated with bright band feature, as already stated, last for a much longer period than those not associated with it. Absorption by cloud drops of trace gases such as SO₂, NO₂ and Cl components (Cl₂ and HCl) could, therefore, be facilitated to a larger extent in the former type of rain situation than in the latter. Such phenomena can lead to formation of additional amounts of soluble anion component in rain water, without altering its cation content as under study. For example, as demonstrated by Cauer (1951), SO₄ always forms when water vapour condenses in the atmosphere and the process could be facilitated by traces of heavy metal ions such as Mn, Cu, Fe etc. (Junge & Ryan, 1958). Measurements by Gambell & Fisher (1964) support the theory that much of the SO₄ in rain water is formed from the oxidation of SO₂ in cloud droplets. While a possibility like this is strongly suggested it is not known how efficiently this mechanism could operate especially in the case

Table 7. Ratio of concentration of different ions in rain from cold convective clouds to that from cold layer clouds (Monsoon Season, 1965)

Rain from cloud	Cl	SO ₄	Na	K	Ca
"Cold convective"	0.38	2.9	0.59	0.28	1.73
"Cold layer"	0.26	1.3	0.11	0.07	0.13
Ratio	1.5	2.2	5.4	4.0	13.3

of the gaseous Cl component present in the atmosphere.

Further, as rain under prolonged periods of rain spells show higher Cl/Na (>1.8), it is to be noted that a knowledge of this ratio in rain water could help suggest the severity of the rain situation in question. Reference to the last column of Table 5 lends support to this view. The highest value (mean) of the ratio observed which is 2.4 was found on the occasion of maximum amount of rainfall, i.e. when 38 mm of rain was recorded per rainy day. It would be of interest to recall in this connection that Rossby (1955) also noticed highest amounts of chloride relative to sodium in precipitation when rainfall was profuse, up to 2 to 3 times the normal value.

Conclusion

The chemical content of rain water forms a useful parameter to help reveal the characteristics of rain in question. There may be what is called a "right type of chemical climate" for each region in the country which, when present, may help reduce possibility of clouds remaining colloiddally more stable in the area. Adequate knowledge in regard to this climate could only be gained from a detailed study of the chemical composition of rains in regions likely to be affected by known types of weather systems. All the same, it is to be noted in this connection that interpretation of chemical data on precipitation is a rather complex problem, and as Eriksson (1960) puts it "until more direct evidence is available, it is a matter of intuition and, perhaps, taste in which way they are interpreted".

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ХИМИЧЕСКИЙ СОСТАВ ДОЖДЕВОЙ ВОДЫ И ХАРАКТЕРИСТИКИ ДОЖДЯ В ДЕЛИ

Пробы дождевой воды, собранной в течение развития ливневого дождя для ряда случаев в различные сезоны в Дели, были проанализированы на хлориды, сульфаты, натрий, калий и кальций. Направление изменений в концентрации ионов хлоридов в дождевой воде, по-видимому, указывает, что рост дождевых капель в конвективных облаках включает процесс, который может быть аналогичным процессу, рассмотренному Вудкоком для дождей на Гавайях. Далее, было замечено, что характеристики осадков в Дели зависят от количества растворимых частиц, присутствующих в воздухе, формирующем облако. Малые концентрации катионов (<1 части на

10^6) и большие величины отношения Cl/Na ($>1,8$) отличают то, что может быть названо «охлаждающим» дождем (дождь из холодных слоистых облаков), от другого типа дождя, в котором действуют как охлаждающие, так и неохлаждающие механизмы (дождь из холодных конвективных облаков). Меньшие неохлаждающие механизмы (дождь из ховеличины отношения сульфатов к хлоридам (<6) и большие величины отношения хлоридов к натрию ($>1,8$) характеризуют более сильные дожди. Для объяснения наблюдаемых особенностей предложены возможные объяснения.