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A study of the chemical components of aerosols and snow in the Kashmir region

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ABSTRACT

Atmospheric aerosols in surface air were sampled for a total period of 2 weeks, from February 25 to March 11, 1971, during the winter season, at Gulmarg and Srinagar. Snow samples were also collected on days of snow occurrence during the period. Aerosols were sampled using cascade impactor and millipore filter assembly. Samples collected through impactor were categorized into hygroscopic and non-hygroscopic nuclei while chloride, ice-forming and total nuclei were evaluated from millipore filters.

Analysis of the aerosol data showed higher counts for all the nuclei at Srinagar with respect to Gulmarg. Fractions of chloride, hygroscopic and ice-forming nuclei among the total nuclei were larger at Gulmarg. Aerosol counts during the days were higher as compared to the corresponding night values. All the chemical constituents, except Cl^-, gradually reduced from the initial to the end stage of the snowfall. Snow sample from Srinagar indicated higher cation concentrations. Variations of aerosols under different weather conditions have been discussed.

1. Introduction

Atmospheric aerosols occupy an important place in the realm of cloud physics on account of their crucial role in the formation and growth of precipitating clouds. Any study on aerosols and associated precipitation activity would, therefore, considerably contribute for a better understanding of physical and dynamical processes of precipitation development in clouds. Measurements on atmospheric aerosols at a few places in India reported earlier, suggested that the dominant source of ice-forming aerosols is land and that of hygroscopic aerosols is sea. These observations further indicated that the development of rain in coastal regions, at least in the tropics, is associated predominantly with a collision-coalescence process while in inland regions clouds do develop rain by ice-crystal mechanism as well (Ramana Murty et al., 1967; Kapoor et al., 1969, 1979). Simultaneous examination of ionic components in rain-water and aerosol state in different types of rain situations further suggested that the chemical state of air and precipitation activity are closely associated (Kapoor et al., 1972).

Reiter et al. (1976) observed that the level of aerosol concentration and the configuration of the aerosol chemical matrix are chiefly controlled by air mass character. Bigg (1977) observed distinct differences in the composition of atmospheric aerosols sampled during the day and during the night. Rhinehart (1973) noticed that a previous day's rain or morning fog usually accounted for an increase in the total number of particles captured and their sulphate content; higher relative humidity was an associated causative factor in increasing the particulate number as well as their sulphate content. Tsunogai et al. (1975) found that the chloride in the snow correlated fairly well with not only sodium of maritime origin but also with calcium, sulphate and radio nuclides. According to them, besides sea salt, snow probably contains aerosols transported far from the source area via the upper troposphere or the stratosphere. Kumai (1976) found that the central nucleus of snow crystals, sampled at the South Pole, were mainly clay minerals and sodium chloride particles. He further observed that all the nuclei found in the snow crystals were terrestrial substances from the oceans and the continents.

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The northernmost part of India often comes under the influence of western disturbances during winter (December to March) which give a considerable amount of rain and snow over the region. The tropopause over the region attains the extra-tropical characteristics and the height of the tropopause sinks down to about 11 km during this period. The authors got an opportunity for undertaking a study of the aerosol state of air for a total period of 2 weeks during one winter season, at stations Gulmarg (34°03′N, 74°24′E, 2655 m MSL) and Srinagar (34°05′N, 74°50′E, 1586 m MSL) situated in this region. They also collected snow samples on occasions of snowfall during that period. The chemical composition of snow water as well as the aerosol state of air, along with the prevailing meteorological conditions, are presented and discussed in this paper.

2. Measurement and analysis

The equipment used for sampling aerosols was the millipore filter assembly initially developed and applied by Lodge (1954). Millipore filters of diameter 47 mm, pore size 0.8 μm and type AA were used for this purpose. The sampling was done at two different rates, i.e. 16 litres and 0.2 litre min^{-1}. The high rate was used for sampling during the day over short periods, while the slow rate was used for sampling during the day and during the night, over long periods. The concentration of total nuclei (all atmospheric particles of radii 0.3 μm and above) was estimated by analysing a portion of the exposed millipore filter under a high power oil immersion microscope. The maximum error in the estimation of concentrations was estimated to be 10%. The chloride component of the atmospheric aerosols was evaluated from the exposed filters by analytical methods using a spot technique as described by Kapoor and Ramana Murty (1966). A uniform factor of 6, as determined in the laboratory, was used for obtaining the particle size from the corresponding spot size. Ice-forming nuclei concentration at −15 °C was determined as described by Bigg et al. (1963).

The hygroscopic and non-hygroscopic aerosol concentrations were measured using a cascade impactor (May, 1945). The aerosol samples were collected on clean glass slides with the impactor sampling at 17.5 litres min^{-1}. The sizes of the aerosol particles were evaluated by an optical microscope method. Each glass slide containing the aerosol deposit was kept in a perspex box maintained at 95% relative humidity. The hygroscopic particles within a few minutes transform into solution droplets in the shape of a plano-convex lens on the slide while non-hygroscopic particles appear opaque and are irregular in shape. The second slide of the impactor which collects the bulk of aerosols in the giant size range (radii one micron and above) was analysed at these stations. The details of the measurement procedure have been reported elsewhere (Kapoor et al., 1979).

The snow samples were collected through a polythene funnel in polythene bottles thoroughly rinsed with distilled water. The snow water was analysed for different chemical constituents employing techniques as described by Khemani and Ramana Murty (1968). The measurement site at Gulmarg was the High Altitude Research Laboratory. The site was almost free from contaminants of local origin. The period of measurements was from February 25 to March 2, 1971. The measurement site at Srinagar was the Meteorological Observatory. The Observatory is situated near the city and, as such, the area is not free from pollutants of local origin. Moreover, a large quantity of wood is burnt during the winter period by local residents to protect themselves from severe cold. The period of measurement was from March 3 to March 11, 1971.

The weather situations at Gulmarg and Srinagar were quite different during the period of measurement. Gulmarg had snowfall on 4 out of 6 days, whereas Srinagar had snow on only 1 day for a short period. The weather features, viz. wind, cloud amount, temperature, visibility, relative humidity and precipitation phenomena, based on surface observations at both the stations are given in Table 1.

3. Results

3.1. *Chemical components of snow*

Snow samples were collected from February 25 to March 2 at Gulmarg. Snowing continued with little interruption up to February 27. It started again on March 2. At Srinagar, snowfall occurred only in the morning of March 4 for a short duration. The concentrations in p.p.m. (by volume) of different constituents are tabulated in Table 2.

All the chemical constituents except Cl^- in-

Table 1. *Weather features at Gulmarg and Srinagar (surface observations)*

Date	Hours (IST)	Wind direction	Total cloud amount Oktas	Surface temperature (°C)	Visibility (km)	Relative humidity (%)	Precipitation phenomena, etc., at hours of observation
GULMARG							
Feb. 1971							
25	0830	W	8	−2	0.5–1.0	80	
	1730	E	—	−2	0.05–0.2	100	Heavy snow
26	0830	E	—	−2	4.0–10.0	100	Thunderstorm
	1730	E	—		0.05–0.2		Heavy snow
27	0830	W	—	−3	0.05–0.2	100	Heavy snow
	1730	W	—	−3	0.05–0.2	100	Dense fog
28*	0830	W	8	−5	2.0–4.0	100	
March 1971							
1	0830	W	6	−6	4.0–10.0		
	1730	W	5	−2	4.0–10.0	90	
2	0830	W	1	−6	4.0–10.0	85	
	1730	W	8	−4	0.5–1.0	80	Snow
SRINAGAR							
March 1971							
3	0830	SE	7	2	4.0–10.0	92	
	1730	SE	7	8	10.0–20.0	60	
4	0830	N	8	2	4.0–10.0	85	Rain with snow
	1730	SE	7	5	4.0–10.0	76	
5	0830	N	8	1	4.0–10.0	85	
	1730	NW	2	8	10.0–20.0		
6	0830	S	5	1	10.0–20.0	76	
	1730	NW	1	9	10.0–20.0	45	
7	0830	N	7	1	10.0–20.0	86	
	1730	SW	7	10	10.0–20.0	45	
8	0830	SE	7	3	10.0–20.0	85	
	1730	NW	8	10	4.0–10.0	55	
9*	0830	NW	7	5	10.0–20.0	88	
10	0830	NW	5	7	10.0–20.0	83	
	1730	NW	6	13	10.0–20.0	49	
11	0830	SE	4	5	10.0–20.0	88	
	1730	NW	6	15	10.0–20.0	40	

* No observations for 1730 hours. — Sky obscure.

dicated decrease in concentration from the beginning to end phase of snowfall. The chloride component remained nearly steady. The snow sample of Srinagar pointed out high Na^+, K^+ and Ca^{++} concentrations compared to all the snow samples collected at Gulmarg. Analysis in respect of other components for the Srinagar sample could not be possible due to the small quantity of snow. Relatively more SO_4^{--} was noticed amongst the anions in the Gulmarg snow. Ca^{++} was maximum amongst the cations. The mean values (in p.p.m.) of anions Cl^-, SO_4^{--} and NO_3^- were, respectively, 0.92, 3.01 and 1.32, and those of cations Na^+, K^+, Ca^{++} and NH_4^+ were, respectively, 0.40, 0.34, 0.79 and 0.42. Also, the mean ratio values NO_3^-/NH_4^+ and NH_4^+/SO_4^{--} were, respectively, 3.19 and 0.14 for the snow samples.

3.2. *Hygroscopic and non-hygroscopic aerosols*

The mean concentrations of hygroscopic and non-hygroscopic aerosols are shown in Table 3. The hygroscopic fraction in the total aerosol (sum of hygroscopic and non-hygroscopic aerosols as collected on the second slide of cascade impactor) is also given in the same table. The hygroscopic and non-hygroscopic aerosol concentrations at Srinagar were respectively 4 and 200 times the values measured at Gulmarg. The total aerosol content at Srinagar and the hygroscopic fraction at Gulmarg were very high.

Table 2. *Concentration in p.p.m. of chemical constituents in snow samples at Gulmarg and Srinagar*

Date 1971	Hours (IST)	Cl^-	SO_4^{--}	NO_3^-	Na^+	K^+	Ca^{++}	NH_4^+	Snowfall/ rainfall (mm)
GULMARG									
February									
25	1300–1400	0.90	4.10	1.60	—	—	—	—	
	1600–1915	0.85	3.65	1.82	0.82	0.87	1.17	0.59	28
26	0730–1000	0.80	4.60	1.42	0.73	0.49	1.23	0.58	
	1050–1400	1.28	2.10	1.33	0.36	0.28	0.58	0.46	169
	1610–1830	0.92	2.15	1.52	0.15	0.15	0.72	0.37	
27	0730–1000	0.92	1.90	0.90	0.25	0.13	0.67	0.19	
	1700–1920	0.76	2.05	1.05	0.12	0.13	0.38	0.34	28
March									
2	1200–1400	—	3.55	0.88	—	—	—	—	3
Mean		0.92	3.01	1.32	0.40	0.34	0.79	0.42	
SRINAGAR									
March									
4	1000–1110	—	—	—	1.39	1.33	1.88	—	2

— Not measurable due to small sample volume.

3.3. *Chloride, ice-forming and total nuclei*

The number and mass concentrations of chloride nuclei, and number of concentrations of ice-forming and total nuclei as evaluated from millipore samples at 16 litres min^{-1} are given in Table 4. For computing chloride mass, a density of 2.15×10^3 kg m^{-3} as applicable to sodium chloride was used.

The mean concentrations per litre of chloride aerosol were 20.0 at Gulmarg and 30.9 at Srinagar. The chloride mass varied from 0.24 to 1.24 μg m^{-3} at Gulmarg. At Srinagar, the corresponding variation was from 0.55 to 2.42 μg m^{-3}. The mean mass concentrations were 0.81 and 1.13 μg m^{-3} at Gulmarg and Srinagar, respectively. The number and mass concentrations of chloride aerosol measured at these stations were not much different from those reported in marine air at sea by Woodcock (1953). The order of values were in agreement with measurements made elsewhere (Gillette and Blifford, 1971; Isono et al., 1966). The ice-forming nuclei concentrations were 14.2 and 29.3 m^{-3} of air at Gulmarg and Srinagar, respectively. The order of these concentrations was similar to those obtained earlier during the winter season at a coastal station, Calcutta, compared to a station well inland, Delhi. The ice-forming nuclei concentrations in this case, were 10.3 and 39.5 m^{-3}, respectively, at Calcutta and Delhi (Kapoor et al., 1969). Also, the values were in agreement with

Table 3. *Mean concentration of hygroscopic and non-hygroscopic particles (number per litre) and percentage of hygroscopic in the total aerosol at Gulmarg and Srinagar. Approximate size range: 2–7 μm (dia.)*

Station	Hygroscopic aerosol	Non-hygroscopic aerosol	% hygroscopic in total aerosol
Gulmarg	0.17 (7)	0.80 (7)	17.5
Srinagar	0.69 (15)	165.77 (15)	0.4

Figures within parentheses indicate the number of observations.

Table 4. *Number and mass concentration of chloride aerosol, number concentration of ice-forming and total nuclei at Gulmarg and Srinagar (sampling rate: 16 litres min^{-1})*

Date 1971	Hours (IST)	Chloride nuclei (l^{-1})	Chloride mass ($\mu g\ m^{-3}$)	Ice-forming nuclei at −15 °C (m^{-3})	Total nuclei (l^{-1})
GULMARG					
February					
26	1447–1512	16.97	0.24	10.0	2160
	1753–1818	17.60	0.88	10.0	2031
27	1410–1435	18.22	0.53	5.0	2160
	1735–1800	19.48	1.13	25.0	2074
28	0720–0745	21.37	0.50	30.0	2117
	1240–1305	20.74	0.45	20.0	1555
	2035–2100	21.37	1.12	0.0	1728
March					
1	0903–0928	20.11	0.99	0.0	1814
	1340–1405	19.44	1.24	20.0	1382
	1805–1830	20.74	1.08	5.0	1599
2	0820–0845	21.37	0.80	5.0	1599
	1140–1205	22.63	0.73	40.0	1555
		20.00	0.81	14.2	1814
SRINAGAR					
March					
3	1445–1510	30.80	1.10	20.0	4536
	1745–1810	28.91	0.85	55.0	4738
4	1040–1105	35.19	1.51	25.0	16431
	1445–1510	27.66	1.56	40.0	1814
	1720–1745	26.40	0.98	30.0	2419
5	1100–1125	29.54	0.88	60.0	2722
	1602–1627	28.91	0.55	45.0	1663
6	0952–1017	28.29	1.94	30.0	2808
	1720–1745	35.20	0.79	10.0	3024
7	1055–1120	54.68	1.74	60.0	29484
	1949–2014	38.97	1.38	0.0	3578
8	0905–0930	37.08	1.41	5.0	4335
	1440–1505	32.06	0.83	15.0	4687
	1755–1825	20.72	0.67	8.3	3948
9	1038–1058	29.01	1.02	12.5	3831
	2045–2105	33.72	2.42	50.0	4032
10	1016–1041	23.88	0.73	35.0	2722
	1910–1935	23.17	0.56	15.0	8115
11	1030–1055	22.63	0.60	40.0	4234
Mean		30.89	1.13	29.3	5743

the most comprehensive measurements on a global scale as reported by Bigg and Stevenson (1970). The total nuclei concentrations were 1814 and 5743 litre^{-1} at the respective places (Gulmarg and Srinagar). The standard deviations of the values of total nuclei at Gulmarg and Srinagar were respectively 272 and 6419 litre^{-1}. Application of the Mann-Whitney test to the total nuclei data suggested that the aerosol state at Srinagar was significantly (significant at 0.003% level) different from that at Gulmarg. The higher concentration of particles at Srinagar may be attributed to urban activity. The percentage of Cl^- and that of ice-forming aerosols in the total nuclei were respectively 1.2, 0.8 × 10^{-3} at Gulmarg and 0.5, 0.5 × 10^{-3} at Srinagar.

3.4. *Variation of total nuclei during the day and night*

A small Teflon pump with a suction rate of 0.2 litre min^{-1} was used for sampling aerosols. Millipore filters were exposed for long periods during the day and also during the night. The concentrations measured are tabulated in Table 5. Average concentrations were 5520 and 4223 $litre^{-1}$ respectively for day and night samples at Gulmarg. Corresponding values at Srinagar were 9219 and 4793 $litre^{-1}$. Both places indicated lower values at night compared to day. The standard deviations of the values of total nuclei in respect to the day and night samples at Gulmarg were 1393 and 581 $litre^{-1}$ and at Srinagar were 3030 and 1047 $litre^{-1}$, respectively. Application of the Mann-Whitney test to the data collected during the day and during the night suggested that such variations were not statisticaliy significant at Gulmarg (significant at 20.0% level), whereas at Srinagar they were highly significant (significant at 0.1% level).

4. Discussion

The Gulmarg and Srinagar regions often remain under the grip of western disturbances during the winter period. These disturbances, in addition to bringing continental air masses along with it, also pick up maritime air masses from the seas of the sub-continent. The air mass prevailing over the region is therefore mainly continental as well as maritime in character.

4.1. *Variation in snow components*

The analysis of snow water collected at Gulmarg (see Table 2) indicated the same ratio values, NO_3^-/NH_4^+ and NH_4^+/SO_4^{--}, as reported in the case of cold convective rain situations in the Delhi region (Kapoor et al., 1972). Since the composition of rain/snow may be considered as an index of the chemical state of air in the region, it appears that these aerosol components may play a role in the formation of snow in the region under consideration.

A thunderstorm occurred at Gulmarg on February 26. SO_4^{--} and Ca^{++} concentrations were highest in snow on that day at 0800 IST, just before the thunderstorm occurred. After the thunderstorm, at 1100 IST, concentrations of all the ions, except Cl^-, reduced and this reduction was maximum for SO_4^{--} and Ca^{++}. SO_4^{--} was again found high on March 2 when snowing began after a

Table 5. *Number concentration of total nuclei at Gulmarg and Srinagar during day (D) and night (N) (sampling rate: 200 ml min^{-1})*

GULMARG		SRINAGAR	
Date 1971	Total nuclei (l^{-1})	Date 1971	Total nuclei (l^{-1})
February		March	
27 D	7488	3 D	15984
27 N	5040	3 N	4828
28 D	4464	4 N	2630
28 N	3744	5 D	9949
March		5 N	4404
1 D	4608	6 D	7489
1 N	3885	6 N	4590
		7 D	8960
		7 N	6536
		8 D	9180
		8 N	5670
		9 D	6480
		9 N	5057
		10 D	6488
		10 N	4628
Mean for days (D)	5520	Mean for days (D)	9219
Mean for night (N)	4223	Mean for night (N)	4793

gap of 2 days. This feature suggests that the presence of SO_4^{--} may probably be necessary for the formation of snow. This inference is corroborated from laboratory investigations which pointed out that supercooled water-drops freeze more readily when they contain sulphates than when they do not. Also, the drops containing higher concentrations of sulphates were found to freeze in warmer temperatures (Ramachandra Murty and Ramana Murty, 1972).

4.2. *Variation of aerosols*

Both Gulmarg and Srinagar indicated lower values of total nuclei at night compared to day. Higher values obtained during the day may be attributed to the lifting of soil dust from the surface by the convection process. This feature is similar to the one reported by Bodhaine and Pueschel (1972) who observed a distinct difference in the composition of the "day time" and "night time" aerosols.

At Gulmarg, chloride and ice-forming nuclei were highest on March 2 (1140–1205 IST) just before the reoccurrence of snowfall. Further, a relatively higher total nuclei during the day with respect to night were more prominent on February 27, a day of snowfall and dense fog (Table 5). It appears that the total nuclei may increase remarkably prior to the occurrence of snow/fog.

At Srinagar, the total nuclei, as observed from high rate sampler, were appreciably higher on the single occasion of snowfall, i.e. March 4 (Table 4). During the end phase of snow/rain, chloride nuclei reduced to some extent although mass concentration was the same, suggesting that relatively smaller particles were washed out in snow/rain. At 1745–1810 IST on March 3, ice-forming nuclei was quite high (55.0 m^{-3}) just before a thunderstorm occurred at 1825–1900 IST.

Aerosol measurements made at Gulmarg and Srinagar indicated higher total nuclei in situations associated with snow. Total nuclei as well as ice-forming nuclei concentrations enhanced with an increase in cloud cover and higher relative humidity, while chloride nuclei remained nearly steady.

4.3. *Average variation of the chemical composition of snow and aerosols*

Higher values of the cations Na^+, K^+ and Ca^{++} were observed in the snow sample of Srinagar compared to the values in the snow of Gulmarg (Table 2). The concentrations of hygroscopic and non-hygroscopic aerosols were more at Srinagar (Table 3). Also, chloride, ice-forming and total nuclei concentrations as estimated from millipores were higher at Srinagar (Table 4). These variations could be attributed to the following factors:

(a) Air in Gulmarg (altitude: 2655 m MSL) was less polluted compared to that at Srinagar (altitude: 1586 m MSL) due to greater height.
(b) There were more pollution sources in and around Srinagar but not so at Gulmarg.
(c) Washout effect was more at Gulmarg (due to heavy snowfall) than at Srinagar.
(d) Comparatively fewer particles were lifted up during the day from Gulmarg surface as it was much cooler than Srinagar surface (Table 1).

During the observational period of Gulmarg, surface wind direction both at Gulmarg and Srinagar was, in general, westerly. As Gulmarg lies southwest of Srinagar, the possibility of relatively more polluted air from Srinagar moving to Gulmarg was ruled out.

5. Conclusions

A study on atmospheric aerosols vis-a-vis chemical composition of snow was undertaken for a total period of 2 weeks during one winter season at Gulmarg and Srinagar. The study pointed out that the concentrations of chloride and ice-forming nuclei were greater at Srinagar, although their fractions in the total nuclei were higher at Gulmarg. It further showed that the total aerosol was greater at Srinagar compared to Gulmarg, although the percentage of hygroscopic aerosol was more by an order at the latter station. Total nuclei counts, at both places, were lower at night compared to day. Concentrations of chloride, ice-forming and total nuclei increased prior to the occurrence of snow/fog. Ice-forming nuclei were also higher prior to a thunderstorm.

In snow, all the chemical constituents, except chloride, decreased from the beginning to the end phase of snowfall. Snow at Srinagar showed higher cation concentration than snow at Gulmarg, a feature analogous to the one observed for aerosol state of air at the two places. The short observation period and the fact that the observations were not carried out simultaneously at both sites limit the value of the conclusion.

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ИЗУЧЕНИЕ ХИМИЧЕСКОГО СОСТАВА АЭРОЗОЛЕЙ И СНЕГА В РАЙОНЕ КАШМИРА

С 25 февраля по 11 марта 1971 года в течение полных двух недель в Гульмарге и Сринагаре собирался аэрозоль в воздухе у поверхности земли. В тот же период отбирался и снег в дни, когда он выпадал. Аэрозоль собирался с помощью каскадного импактора и системы фильтров с микропорами. Пробы, отобранные импактором, были разделены на гигроскопические и негигроскопические ядра, в то время как хлоридные, льдообразующие ядра и полное их число оценивалось по фильтрам с микропорами.

Анализ аэрозольных данных указывает на

более высокие отсчеты для полного числа ядер в Сринагаре, чем в Гульмарге. Фракции хлоридов, гигроскопических и льдообразующих ядер среди полного их числа были больше в Сринагаре. Отсчеты аэрозоля днем были больше, чем ночью. Все химические компоненты, за исключением хлора, постепенно уменьшались от начала до конца снегопада. Пробы снега в Сринагаре указывали на более высокие концентрации катионов. Обсуждаются вариации аэрозоля при различных погодных условиях.