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Altitudinal variation of surface aerosol with change in site: A comparative study

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The study of size distribution of atmospheric aerosols is an important tool in understanding and managing aerosol effects on health, visibility and climate. The measurements of surface aerosols, both mass-size distribution and mass concentration, were carried out in the campaign mode at Leh (34°09′N, 77°34′E, 3441 m asl), Hanle (32°47′N, 78°58′E, 4517 m asl) and Delhi (28°39′N, 77°13′E, 220 m asl) during July 2003 to examine their variation with altitudinal and anthropogenic activity. A 15 channel portable particle size aerosol spectrometer GRIMM was used for carrying out the experiment. The aerosol spectrometer is equipped with class 3-B laser as the light source and uses a light scattering technology for single particle counts in the range > 0.3 μm to > 20.0 μm. During the period of observation at all three sites, the pattern of total mass concentration was found to be influenced by supermicron range mass concentration. The higher values of aerosol mass concentration observed at Delhi as compared with those observed at Leh and Hanle on all the channels show a large amount of anthropogenic activity taking place around Delhi *vis-à-vis* the other two sites. It is further observed that the mass-size distribution changes with the change in altitude and anthropogenic activity.

Keywords: Surface aerosol, Mass-size distribution, Submicron range, Supermicron range, Mass concentration
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1 Introduction

Study of the temporal and spatial changes of aerosol characteristics at different wavelength give insight into the dynamics of aerosol population[1], their production and removal processes[2], i.e. the origin of particles[3,4] and their lifetime[5]. Surface aerosols are those components of atmospheric aerosol system that reside within the boundary layer and exhibit a very high degree of spatial and temporal variability at short scales and highly susceptible to the boundary layer dynamics[1,6]. Size distribution of surface aerosols is highly dependant on their sources and sinks, meteorological conditions and the boundary layer circulations[7-10]. Nevertheless, surface aerosols contribute significantly to the total aerosol loading at any given location[11]. The measurement of the mass-size distribution is thus an important tool for characterizing atmospheric aerosols[12,13].

A comparative study of surface aerosols was carried out in a campaign mode at Delhi (28°39′N, 77°13′E, 220 m asl), an urban polluted city, and high altitude sites at Leh (34°09′N, 77°34′E, 3441 m asl) and Hanle (32°47′N, 78°58′E, 4517 m asl) in India. Hanle and Leh are located in the mountain range of the western Himalayas in the vast Nilamkhul plain of the Changthang, Ladakh region of the Jammu and Kashmir state in India. Hanle is a hilly desert with very little snow cover and vegetation and free from pollution and anthropogenic sources of aerosol. The site is so dry that the annual precipitation of rain and snow is less than 10 cm. Leh, the other high altitude site, is a small town with little population, where some anthropogenic activity was present during the observational period. On the other hand SPM or near surface aerosol concentration in Delhi's environment is not only contributed by vehicular and industrial activity but also significantly because of soil originated particles and suspended dust generated by winds and construction activity[14]. The model studies suggested that the Delhi aerosol is best represented by a combination of urban and desert aerosols. This is because, the coarse sand and dust particles are gradually added in the already polluted (of vehicular and industrial origin) atmosphere of Delhi[15].

The measurements were carried out during 4-7 July 2003 at Leh, 8-10 July 2003 at Hanle and 11-14 July 2003 at Delhi. For the comparative study of aerosol mass concentration and mass-size distribution, data of one complete day are used for each aforementioned site. On 6, 9 and 13 July 2003 the data were available

throughout the day i.e. for the entire 24 h without any gap. The aim of this experiment was to examine the variation of surface aerosols, both mass-size distribution and mass concentration, with altitude and anthropogenic activity.

2 Experimental setup

The surface aerosol measurements were carried out using a 15 channel portable particle size aerosol spectrometer (Model 1.108 of GRIMM). The aerosol spectrometer contains a class 3-B laser as the light source and uses a light scattering technology for single particle counts. It operates at a flow rate of 1.2 l/min and gives total mass concentration (M_t), over first channel, and mass concentration (m_{ci}), explained in the following text, in one of the two basic modes, particle counts in counts/liter or mass concentration in $\mu g/m^3$. The data used in this analysis, was collected in mass concentration mode. The details of channels, their lower cut-off point, i.e. particle diameter of each channel, and their geometric mean diameter are given in Table 1. The data used for analysis is divided into two parts, as (i) directly measured parameters and (ii) derived parameters.

2.1 Directly measured parameters

(i) *Total mass concentration* (M_t) — Total mass concentration (M_t) was measured in the size range > 0.3 µm to > 20.0 µm. The unit of measured quantity was $\mu g/m^3$. This complete range is divided into 15 channels. This division is unequal and is partitioned with respect of particle diameter (d_{pi}) as given in Table 1.

(ii) *Mass concentration* (m_{ci}) — Each channel gives the mass concentration (m_{ci}) of surface aerosols as the sum of mass concentration over each channel (m_i) up to the last channel, i.e.

Table1 — Channels and cut points of GRIMM aerosol monitor

Channels	1	2	3	4	5
Particle diameter (d_{pi}), µm	0.3	0.4	0.5	0.65	0.8
Geometric mean diameter (d_{gi}), µm	0.35	0.45	0.57	0.72	0.89
Channels	6	7	8	9	10
Particle diameter (d_{pi}), µm	1.0	1.6	2.0	3.0	4.0
Geometric mean diameter (d_{gi}), µm	1.26	1.78	2.44	3.46	4.47
Channels	11	12	13	14	15
Particle diameter (d_{pi}), µm	5.0	7.5	10	15	20
Geometric mean diameter (d_{gi}), µm	6.12	8.66	12.24	17.32	—

$$m_{ci} = \sum_{i=1}^{15} m_i \quad \ldots(1)$$

Therefore, $M_t = m_{ci}$, where $i = 1$. Values of M_t and m_{ci}, are provided by the instrument, need no analysis or pre-processing and can be directly used.

Statistical and mathematical operations were performed on the obtained dataset to study the aerosol characteristics and their response to different size range and diameter.

2.2 Derived parameters

(i) *Mass concentration over each channel* (m_i) — As the channels are of unequal width, the mass concentration over each channel (m_i) is the result of contribution of all the particles lying within the size range of two adjacent channels, i.e. difference between two adjacent channels. This can be calculated from directly measured mass concentration (m_{ci}) as follows

$$m_i = m_{ci} - m_{c(i+1)}, \text{ where } i = 1 \text{ to } 14 \quad \ldots(2)$$

and mass concentration over last channel is the values given by the instrument over the 15th channel. This channel has no upper cut-off and shows the concentration for all values above 20 µm.

(ii) *The mass size distribution*, dm/dr — The mass size distribution is given as follows:

$$dm/dr = m_i/\Delta r_i, \text{ where } \Delta r_i = \tfrac{1}{2}(d_{p(i+1)} - d_{pi}) \quad \ldots(3)$$

where d_{pi} is particle diameter on ith channel.

3 Results and discussion

3.1 Diurnal variation of total mass concentration, submicron range (> 0.3 µm to < 1.0 µm) and supermicron range (> 1.0 µm to < 20 µm) mass concentration

Diurnal variation of total mass concentration (M_t), submicron range (M_a) and supermicron range (M_c) mass concentration on 6, 9 and 13 July 2003 for Leh, Hanle and Delhi are shown in the Figs 1(a), (b) and (c), respectively.

Figure 1(a) shows steady decrease in the total mass concentration over Delhi, from 0000 hrs IST onwards till 0700 hrs IST and the same starts increasing around 0800 hrs IST. This increase is due to peak traffic hours and continues up to 1200 hrs IST. The observation suggests that the daytime (during 1200 to 1600 hrs IST) minimum values may be resulting from dilution of boundary layer during its vertical

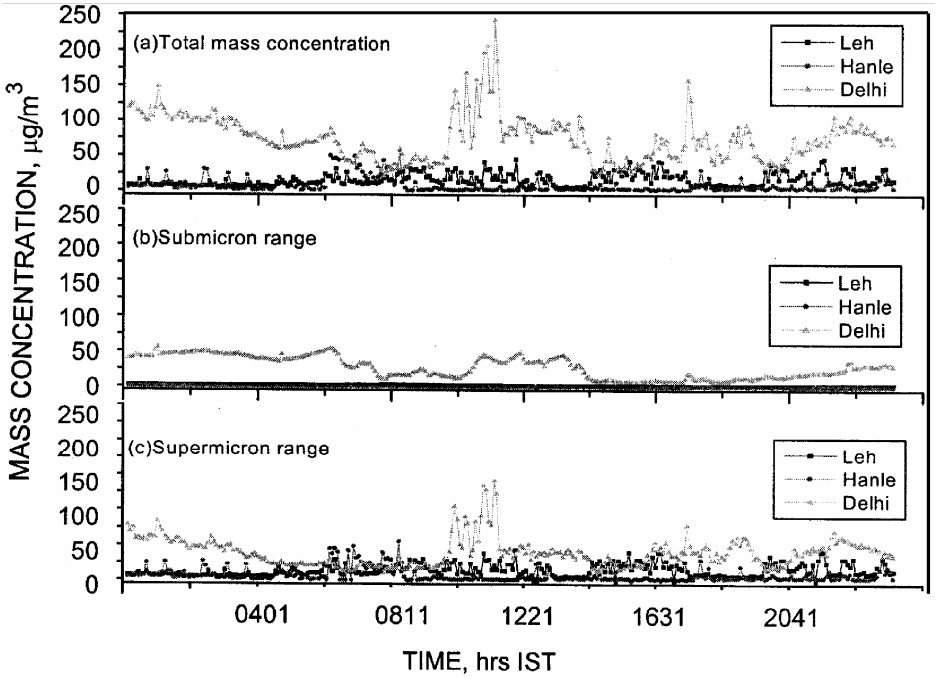

Fig. 1 — Comparison of diurnal variation of (a) total mass concentration, (b) submicron range and (c) supermicron range mass concentration at Leh, Hanle and Delhi on 6, 9 and 13 July 2003

expansion and due to light rainfall during the daytime in spite of additional injection of aerosols during the daytime. The second peak was observed during evening hours around 1700 hrs IST, which is again due to peak traffic time followed by strong inversion. At Leh the total mass concentration was found to be very low and comparatively less variations in the measured concentration was observed. At still higher altitudes, i.e. Hanle, the total mass concentration was found to be very low as well as very stable with minor fluctuations, which can be subjected to local meteorological conditions.

The mass concentration in the submicron range (M_a) and supermicron range (M_c) are estimated separately from the size segregated mass concentration (m_i) as follows:

$$M_a = \sum_{i=6}^{5} m_i \quad \ldots(4)$$

$$M_c = \sum_{i=6}^{14} m_i \quad \ldots(5)$$

for the entire day.

Figures 1(b) and (c) show variations of submicron and supermicron range mass concentrations, respectively. The submicron range mass concentration over Delhi, shown in Fig. 1(b), was found to be very stable, from 0000 hrs IST onwards till morning around 0600 hrs IST. After sunrise around 0600 hrs IST boundary layer height increased, diluting the submicron range mass concentration and the values went as low as 5-10 μg/m^3. It is also clear from Fig. 1(b) that the value of submicron range mass concentration over Delhi on 13 July 2003 never exceeded 50 μg/m^3 any time of the day. On the other hand the values of submicron range mass concentration over Leh and Hanle, the values were almost constant with no variation. On close observation of Fig. 1(b) it can be seen that the values of submicron range mass concentration at Leh were little higher than Hanle.

The supermicron range mass concentration over Delhi had a decreasing trend, from 0000 to 0600 hrs IST and the values came down from 75 to 15 μg/m^3, as depicted in Fig. 1(c). The same started building up and reached again a value of 75 μg/m^3 on an average around 1200 hrs IST. The values also crossed the 140 μg/m^3 mark for sometime, around 1100 hrs IST and got almost stabilized around 40 μg/m^3 for the rest of the day. The supermicron range mass concentration for Leh was almost stable around 10 μg/m^3 from 0000 to 0700 hrs IST and started building up reaching values around 20 μg/m^3 from 0800 hrs IST onwards and sometimes even reached 45 μg/m^3 as shown in Fig. 1(c). This again got stabilized with minor

variations to 10-12 μg/m^3 in the evening around 1800 hrs IST. On the other hand the supermicron range mass concentration over Hanle was almost stable through out the day with average values around 5 μg/m^3, as depicted in Fig. 1(c) and occasionally crossed the 45 μg/m^3 mark in the morning, which can be attributed to local meteorological conditions. From Figs 1(a), (b) and (c), it is clear that at all the three sites, the pattern of total mass concentration was influenced by supermicron range mass concentration than by submicron range mass concentration.

3.2 Location-wise distribution of mass concentration

To examine how the mass concentration changes in the various size intervals with respect to altitude, the entire size spectrum measured was divided into three intervals: Large (d_{pi} = 5-20 μg/m^3), Coarse (d_{pi} = 0.8-5.0 μg/m^3) and Fine (d_{pi} = 0.3-0.8 μg/m^3). Table 2 shows the location-wise percentage share of aerosol mass concentration in three different aforementioned intervals. From Table 2 it is clear that as the altitude goes on increasing and the sources of anthropogenic aerosols goes on decreasing, the mass concentration, in percentage of surface aerosols in fine mode goes on decreasing, where as in coarse and large mode increases. This is true with the percentage share, where as the total mass concentration goes on decreasing, as the altitude goes on increasing and the source of anthropogenic aerosol goes on decreasing.

3.3 Channel-wise share of mass concentration

The mass concentration over each channel is calculated using Eq. (2) for all the 15 channels, and averaged for the day. Figure 2 shows the averaged mass concentration over each channel with vertical bars showing standard deviation. From Fig. 2 it is clear that the mass concentration over Delhi is found to be higher on all the channels, compared to Leh and Hanle, with higher standard deviation. This shows the influence of anthropogenic activity on the amount of variation in the mass concentration of surface aerosols at a site in Delhi.

As shown in Fig. 3 it is clear that at all the three sites, irrespective of altitude, bimodal pattern of aerosol mass concentration is observed. One peak is observed at the first two channels and the value of mass concentration goes on decreasing along with decrease in standard deviation, as the particle diameter goes on increasing. The values of mass concentration on channels 4-7 are stable, with lower mass concentration and standard deviation. Again the mass concentration increases from channel 8 onwards and attain highest value on channel 11, with higher standard deviation. The mass concentration again goes on decreasing up to channel 14. Channel 15 is not taken into account for this, since it covers all the values of aerosols, having particle diameter above 20 μm and have no upper cut-off.

Table 2 — Location-wise share of mass concentration (in %)

	Fine	Coarse	Large
LEH	10.16	59.05	30.79
HANLEY	5.66	54.78	39.55
DELHI	40.00	26.59	33.41

3.4 Mass-size distribution

Measurements showed that the mass-size distribution changes with the change in altitude and anthropogenic activity. The mass-size distribution is calculated from hourly averaged mass concentration over each channel (m_i) and radius-width of that

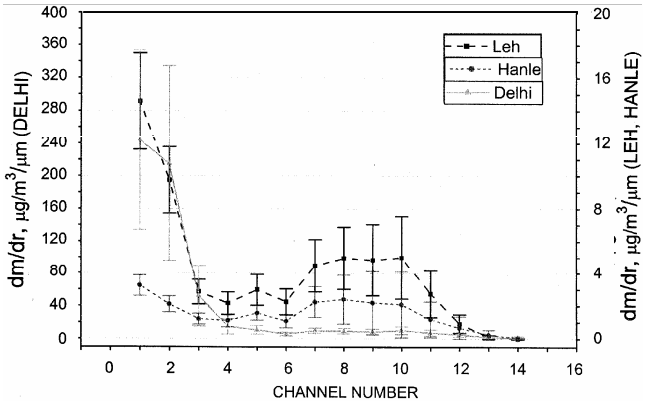

Fig. 2 — Typical features of the mass-size distribution at Leh, Hanle and Delhi on 6, 9 and 13 July 2003

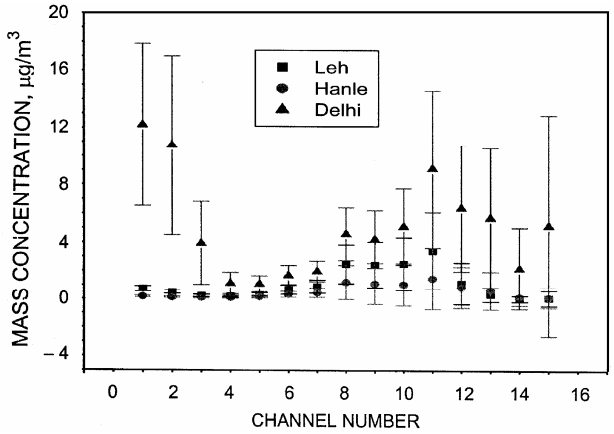

Fig. 3 — Averaged mass concentration over each channel with vertical bars showing standard deviation for Leh, Hanle and Delhi on 6, 9 and 13 July 2003, respectively

particular channel. This is done using Eq. (3) and then averaging the values for that channel.

The typical mass-size distribution is shown in Fig. 2, with the continuous line representing values for Delhi, dashed line for Leh and doted line for Hanle with vertical bars showing standard deviation. It can be seen that the mass-size distribution is found to be trimodal with domination at channel 1, then at channel 11 and channel 5 at all three sites. But at Delhi, the mass concentration at channel 1 is much higher than other two sites Leh and Hanle, and they are in the ratio of 80:5:1. Similarly at channel 5 and channel 11 the ratios are 10:2:1 and 4:2:1, respectively. This shows that there are no prominent sources of aerosols at high altitude site, due to which the mass-size distribution changes with respect to altitude.

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