Aerosol characteristics at a high-altitude location during ISRO–GBP Land Campaign-II

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We present detailed analysis of the extensively measured aerosol parameters at the Aryabhatta Research Institute of Observational Sciences (ARIES), Manora Peak Nainital, during ISRO–GBP Land Campaign-II. This land campaign was focused on the winter month of December 2004, using eight fixed stations distinctly located over the Gangetic belt in the North Indian corridor, where thick fog conditions generally prevail during winter. Among these stations, Manora Peak was selected due to its high-altitude location, at an altitude of ~2 km, having a pristine location in the Shivalik Ranges of Central Himalayas and allowing a free tropospheric site. In this perspective, observations of aerosol optical depths (AODs), black carbon (BC) mass concentration, total columnar ozone (TCO), mass loading of total suspended particulates (TSP), and number concentration of near-surface aerosols have been carried out from Manora Peak. These experiments barring AODs, were made for the first time at this location. The monthly mean AOD at 500 nm was found to be 0.056 (± 0.037). Temporal as well as the diurnal variation of BC mass concentration show almost similar variation as that of aerosol number (>0.3 μm) concentration, having relatively low values during night and early morning periods, and gradually increasing as the day advances reaching its maximum level around 1600 h local time. The monthly mean BC concentration was found to be 1.36 (± 0.99) μg m^{-3}. Mass loading of TSP was in the range 20–40 μg m^{-3}, with a mean value of 27.1 (± 8.3) μg m^{-3}. During the period under study, average BC mass fraction at Nainital was found to be ~6.3 ± 2%. The monthly mean TCO was found to be 268 ± 22 DU. The diurnal variation of BC mass concentration shows a typical behaviour compared with other low-altitude stations, where simultaneous measurements were made by other investigators during the campaign. This behaviour in the diurnal trend of aerosols is due to the topography and the boundary-layer dynamics over the high-altitude station of ARIES at Nainital.

Keywords: Aerosol optical depth, black carbon, columnar ozone, ISRO–GBP Land Campaign-II, size distribution.

IT has been noticed during the last few years that the entire northern part of India is covered with thick fog during winter. This scenario prevails more intensely over the Gangetic belt and persists tentatively during December and January every year. Satellite pictures also reveal thick layers of fog, especially in the northern corridor of India during these months. The occurrence of temperature inversion during the fog prevents pollutants to get mixed and be transported. The impact of this is discernible as poor visibility and high level of pollutants. During this season, dry, cold wind from the northwest picks up moisture from Punjab–Haryana region, making it more conducive for fog formation, as fog can occur if the wind is calm, and the air is sufficiently moist, cool enough and descending. In order to understand the origin of this unusual phenomenon, a comprehensive and coordinated campaign-based study was conducted as a part of Indian Space Research Organisation–Geosphere Biosphere Programme (ISRO–GBP) Land Campaign-II, during December 2004.

Aerosols have a direct radiative forcing in the atmosphere as they scatter and absorb solar and infrared radiation in the atmosphere[1] and indirectly affect the size distribution of cloud droplets[2]. Knowledge of the optical properties and temporal and spatial variability of aerosols is the basic requirement for understanding the direct effects of aerosol in the atmosphere[3]. In this perspective, the Indo-Gangetic belt provides special meteorological conditions during winter, when it is usually covered with a thick layer of aerosol and other pollutants. At higher altitudes above the mixing region and free troposphere, the retrieved aerosol characteristics may be an indicator of its background level. In order to evaluate the temporal and spatial aerosol characteristics, extensive and coordinated simultaneous measurements were made for building up a comprehensive picture of aerosol distribution over this region.

Measurements were carried out at eight fixed locations, extending from Hissar in Haryana to Kharagpur in West

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Bengal, covering the entire Indo-Gangetic belt. All these stations except Manora Peak, Nainital, fall in the low-altitude Gangetic plains, located in the downwind direction. Thus they provide an opportunity to study the horizontal transport of pollutants. Manora Peak (29.36°N; 79.45°E), is considered to be a free tropospheric site, as it is located at an altitude of ~2 km in the Shivalik ranges of Central Himalayas. Manora Peak is about 1.5 km away from Nainital town having a floating population of about one lakh, and further having insignificant anthropogenic activities during winter months. Hence it can be considered as an appropriate site for evaluating the characteristics of background aerosols. We present here detailed analysis of the simultaneous data obtained from Manora Peak station during December 2004 for aerosol characteristics, using ground-based passive instruments. The instruments operated during the campaign comprised of MWR (multi-wavelength radiometer), Microtops II, GRIMM aerosol spectrometer, aethalometer, a high volume air sampler (HVS) and an automatic weather station.

Experimental details and data

Aerosol optical depth measurements

Aerosol optical depth (AOD) was obtained using multi-wavelengh radiometer (MWR), and Microtops II sunphotometer. The MWR is based on the principles of filter wheel radiometer[4]. It takes continuous measurements of spectral extinction of ground-reaching, directly transmitted solar flux as a function of solar zenith angle. The raw data obtained from the MWR were analysed using the conventional Langley-plot technique to deduce AOD (t_p). More details on the instrumentation, data analysis and error budget are given elsewhere[5,6].

Microtops II (Solar Light Co, USA), sunphotometer and ozonometer are hand-held photometers that measure direct solar radiations and give instantaneous AODs, columnar water vapour content (W) and total columnar ozone (TCO). The performance and methodology of data acquisitions with Microtops II have variously been described[7–9].

Spectral extinction measurements were taken regularly with the MWR during periods of unobscured solar conditions, whereas with Microtops II measurements were taken even with partly clear sky conditions, as it acquires instantaneous AODs, TCO and W. During the study period, a total of 33 datasets were obtained from MWR measurement, while more than 650 datasets were obtained from the Microtops-II sunphotometer and ozonometer measurements. The ozonometer was also used to measure instantaneous values of total columnar ozone (TCO) and precipitable columnar water vapour.

Aerosol number concentration, BC mass concentration and TSP

Simultaneous measurements of the number concentration of near-surface aerosols and mass loading (M_T) of total suspended particulates (TSP) were conducted using a GRIMM aerosol spectrometer (Model 1.108 of Grimm Aerosol Technik, Germany) and a high volume air sampler (HVS: Model Envirotech – APM 230) respectively. The GRIMM is a portable, lightweight, battery-operated optical particle counter, which sorts out the particles into 15 different size bins in the diameter range between 0.3 and 20 μm. It uses light scattering technology for single particle counts in which a semiconductor laser serves as the light source. In the present study, the instrument was operated in the count mode to obtain the aerosol number concentration (N_t) and the measurement cycle (time base) was set to 5 min. Continuous measurements of aerosol number concentration were obtained throughout December 2004. Since the aerosol concentration at Nainital was low, the HVS was operated for two continuous days during daytime. For obtaining the night-time concentration, sampling was done from the evening of the first day till the morning of the second day. During the study period a total of 20 samples were collected, 16 were collected during daytime and four samples were collected during the night. From HVS measurements, the mass loading of the TSP was evaluated. For monitoring black carbon (BC) aerosols, a dual channel aethalometer (Model AE-21 of Magee Scientific, USA) was used. It uses a continuous filtration and optical transmission technique to measure the concentration of BC in near real time and aspirates ambient air using both its inlet tube and pump. The BC mass concentration is estimated by measuring the change in the transmittance of a quartz filter tape, onto which the particles impinge. More details on the instruments and measurement technique are available elsewhere[10–12]. The aethalometer was operated at a flow rate of 3.3 l/min and at a timebase of 5 min.

Prevailing meteorology

The prevailing meteorology at Manora Peak during December comprises a synoptic northwesterly circulation, dry ambient with low RH (~40 to 90%) and scanty rainfall. The prevailing wind during the campaign was ~2.0 m s^{-1} and the temperature varied in the range of 2 to 21°C (Figure 1). During most of the days of the present study, the morning sky was in general clear (cloud-free), followed by partly cloudy sky in the afternoon. The sunshine recorder data revealed that during the observation period, the sky was almost clear for 15 days (> 8 h), partly clear for 11 days (> 6 h) and for the remaining days it was partially cloudy or overcast. It is quite discernible from Figure 1 that the wind was > 5 m s^{-1} on 14 and 30 December and corresponding RH was as low as 40 and 15% respectively.

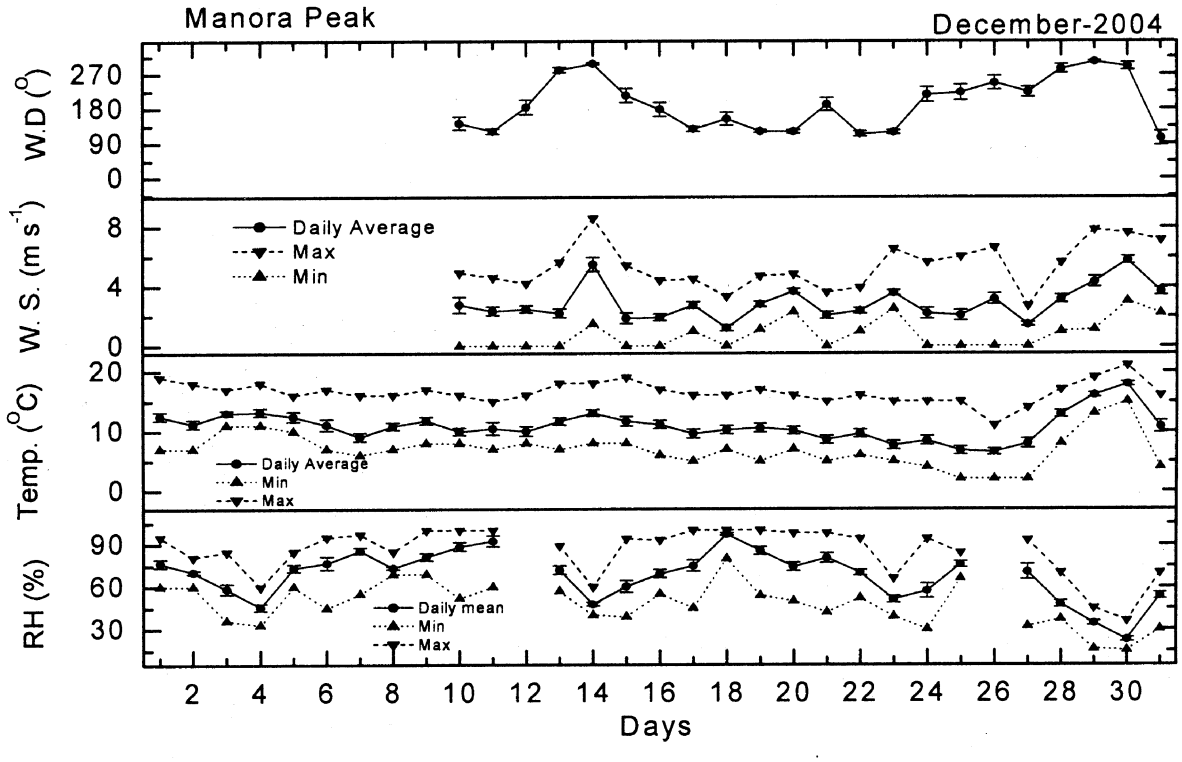

Figure 1. Monthly mean diurnal variations (daily average, maximum and minimum) of relative humidity (RH), temperature (Temp), wind speed (WS) and average wind direction (WD) during December 2004.

Results and discussion

Aerosol optical depth

Since AODs were retrieved from two different instruments (MWR and Microtops II), having wavelengths common to both for AOD measurements, it could be possible to intercompare the results obtained from both these instruments and thereby ascertain their mutual consistency, reliability and inter-usability. A comparison of the monthly mean spectral AODs derived from MWR and Microtops-II has been shown in Figure 2 *a*. Although mean AOD exhibits day-to-day variability as represented by the large standard deviations shown in Figure 2 *a*, both the instruments produced quite similar results that remained stable and were statistically consistent during the study period. This comparison validates the authenticity of AOD data retrieval from these two independent techniques. Although the monthly mean AOD at 500 nm was 0.056 (± 0.037), slightly higher values of AOD (> 0.1) were found on 1, 18 and 21 December 2004. Whereas, in contrast, the monthly mean value during this campaign was reported to be ~0.6 over Allahabad, an urban low-altitude site, located in the heart of the Indo-Gangetic basin[13]. Other locations in the Indo-Gangetic Plains have also reported similar variations in AOD from their respective stations, references to which have been reported in the preliminary results[13] on Land Campaign-II by various authors. It is quite discernible from Figure 2 *b* that AOD values at shorter wavelengths were higher than those measured at higher wavelengths for the entire month of December 2004. Larger values of AODs around shorter wavelength indicate abundance of smaller size aerosol particles. Spectral variation of t_p is important due to its significance in the relative evaluation of aerosol size characterization. It is well known that AODs contain information pertaining to their size distribution[14]. A simple way of expressing the wavelength dependence of t_p is through the Angstrom relation[15] expressed as

$$t_{pl} = b\, l^{-a},$$

where b is the turbidity coefficient and a the wavelength exponent. a is related to the size distribution and depends on the ratio of the concentration of large to small aerosols and b represents the amount of aerosol present in the vertical column[4]. The values of a and b are evaluated by linear least square fitting of $t_p - l$ estimates on a log–log scale. The estimated a value in the present study ranges from 0.4 to 1.2. Higher value of a implies relative dominance of smaller aerosol particles. This is attributed to the fact that sedimentation of coarse particles probably occurred before they could disperse high into the atmosphere and manage to cross the atmospheric boundary layer. This validates the pristine environment over the site, inherent characteristics of a free troposphere.

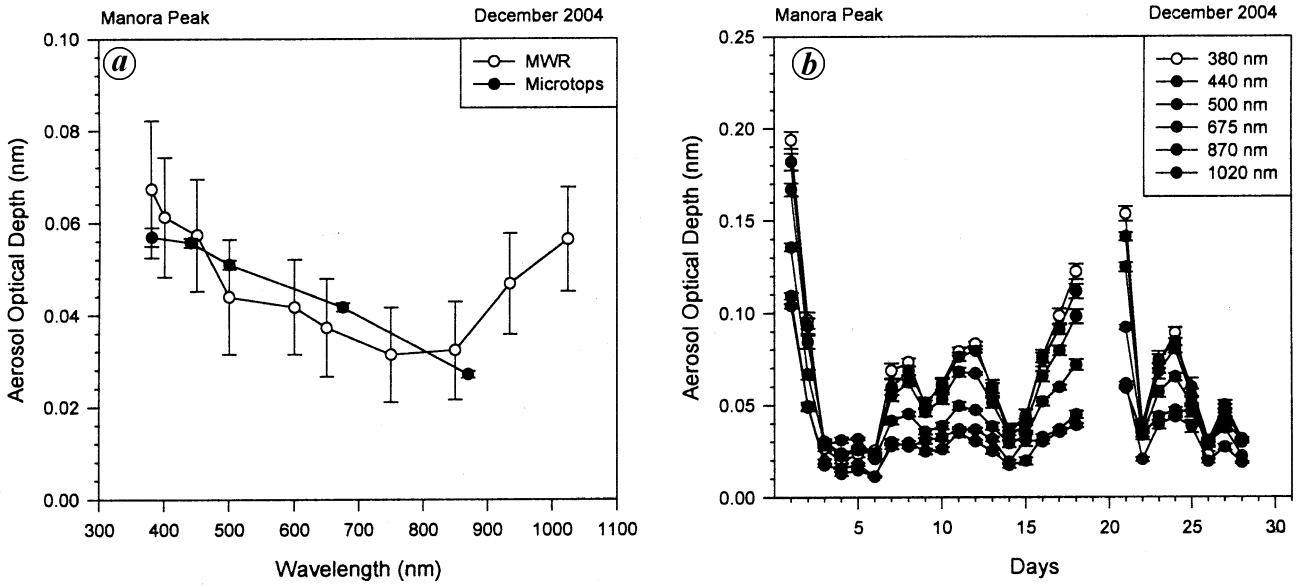

Figure 2. (***a***) Comparison between retrieved mean values of spectral AOD obtained using MWR and Microtops II and (***b***) spectral monthly mean AOD retrieved from Microtops II during December 2004.

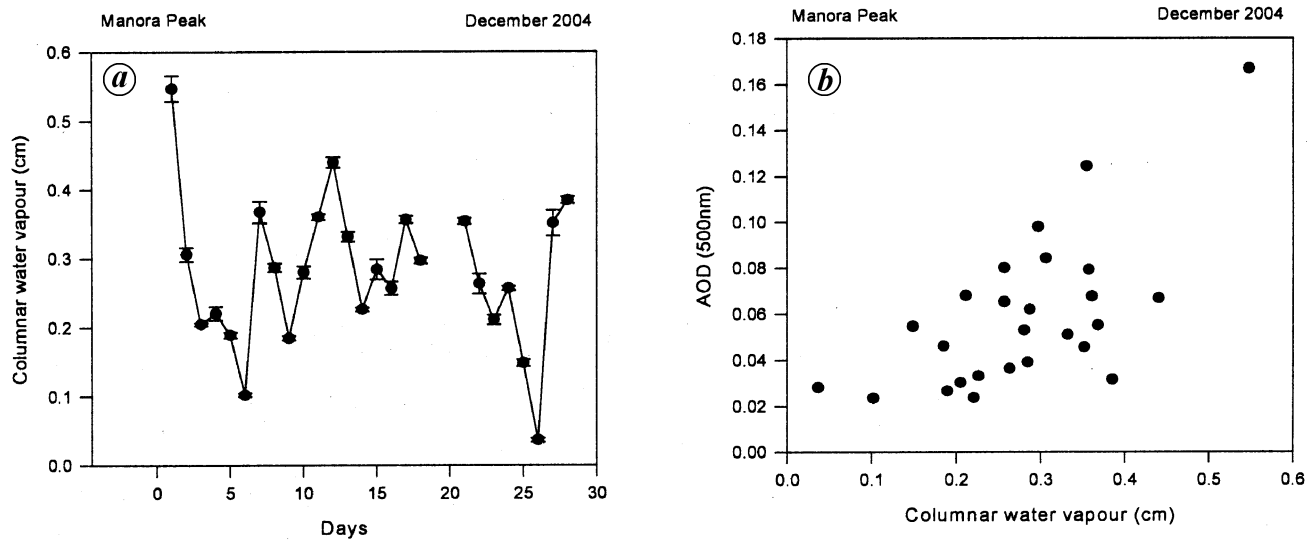

Figure 3. (***a***) Temporal variation of columnar water vapour (*W*) and (***b***) scatter plot of *W* against AOD at 500 nm measured by Microtops II during December 2004.

Columnar water vapour content

Figure 3 *a* shows the temporal variation of *W* derived from the Microtops II measurements. The temporal variations clearly show the day-to-day variability, with highest value (~ 0.55 cm) observed on 1 December 2004 and the lowest value (~0.04 cm) on 26 December 2004. The monthly mean value of *W* was 0.28 (± 0.11) cm, which reveals the prevalence of dry environment over Manora Peak, during the campaign period. The estimated water vapour contents are in good agreement to the reported values during the winter season over Manora Peak, based on three years' extensive measurements in the eighties[16]. Figure 3 *b* shows the scatter plot of *W* against AOD at 500 nm having no linear relation between the two, suggesting that an increase in RH and/or *W* causes a nonlinear increase in AOD at all wavelengths[17].

Total columnar ozone

The daily average columnar ozone measured using Microtops II ozonometer during the campaign period shows

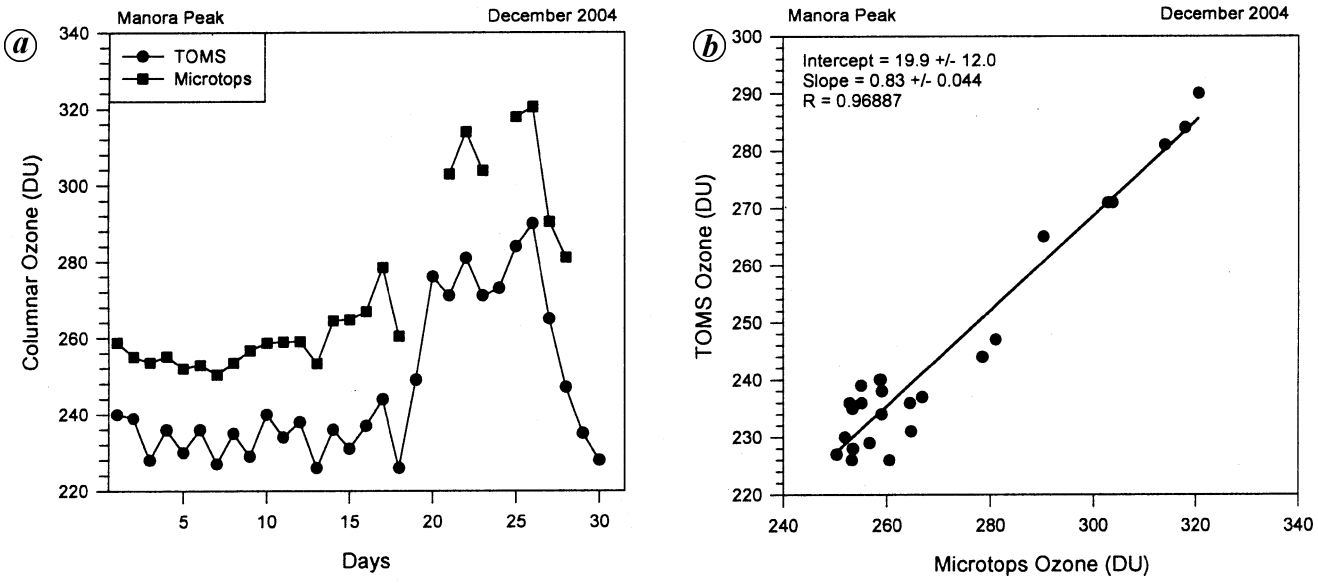

Figure 4. (***a***) Temporal variation of ozone obtained from Microtops II and TOMS and (***b***) Inter-comparison of ozone obtained from Microtops II and on-board TOMS data.

day-to-day variations. The temporal variation in daily mean of TCO is shown in Figure 4 *a*. It is noticeable that the daily mean values are in the range 260–300 Dobson Unit (DU). The monthly mean TCO was found to be 268 ± 22 DU. Temporal variations of total ozone measured from the Earth-Probe Total Ozone Mapping Spectrometer (TOMS) satellite have also been plotted in Figure 4 *a*. Although the daily mean values retrieved from both the ground-based Microtops II and satellite-based TOMS show almost similar trend in temporal variations (with correlation coefficient of 0.96), the TOMS measurements underestimate the total ozone values approximately by 20 DU (Figure 4 *b*). Temporal variations of TCO (Figure 4 *a*) show that for a majority of days, it was around ~260 DU during the initial phase of campaign. However, TCO increased significantly during 21–26 December 2004, when it reached up to as high as >300 DU. The prevailing columnar water vapour contents during this period were at the lowest level, as low as ~0.05 cm on 26 December (Figure 3 *a*).

Near-surface aerosol measurements

Aerosol number concentration

Measurements of the number concentration of near-surface aerosols were carried out using GRIMM aerosol spectrometer. Although the number concentration of near surface aerosol ($N_T > 0.3$ μm) shows day-to-day variability (Figure 5 *a*), it is clearly seen that the monthly mean diurnal variation of N_T has a well-defined diurnal pattern (Figure 5 *b*) with extremely low values occurring during night and early morning hours, while it gradually increases after sunrise to attain a peak in the afternoon around 1600 h IST (IST = UT + 5.5 h). The concentration decreases considerably thereafter at sunset and again reaches a low level by midnight. The afternoon peak can be attributed to the vertical transport of aerosols from the nearby polluted urban and valley regions, which were initially confined to lower heights in the night and early morning due to the low-level inversions, but are released to greater heights (>2 km), as the boundary layer evolves upward. It is an established fact that populated area produces more pollutants[18] confining them within the atmospheric boundary layer[19]. During night majority of aerosol concentration lies below the mountain peaks (in the valley as residual layer), due to the capping inversion process, that caps a convective boundary layer, consequently preventing the convective elements from rising higher into the atmosphere. As the day advances, the sun warms the surface, and convective plumes transport the emissions, including updraft of contained pollutants from the surface higher into the atmosphere[20], consequently reflected as an enhancement in aerosol number concentration during the afternoon.

To make a comprehensive analysis of the observed peak during the afternoon hours in the number concentration of composite aerosol particles, we have elaborately studied the temporal behaviour of particle size distribution from morning till evening as shown in Figure. 6. It reveals that the number concentration of finer particles grows faster than the coarse particles. This is quite evident in Figure 7, where number concentration of total particles (upper panel), sub-micron particles (middle panel), and super-micron particles (bottom panel) has been illustrated. The time series for total and sub-micron particles shows consistently higher values of number concentration from 16 to 24

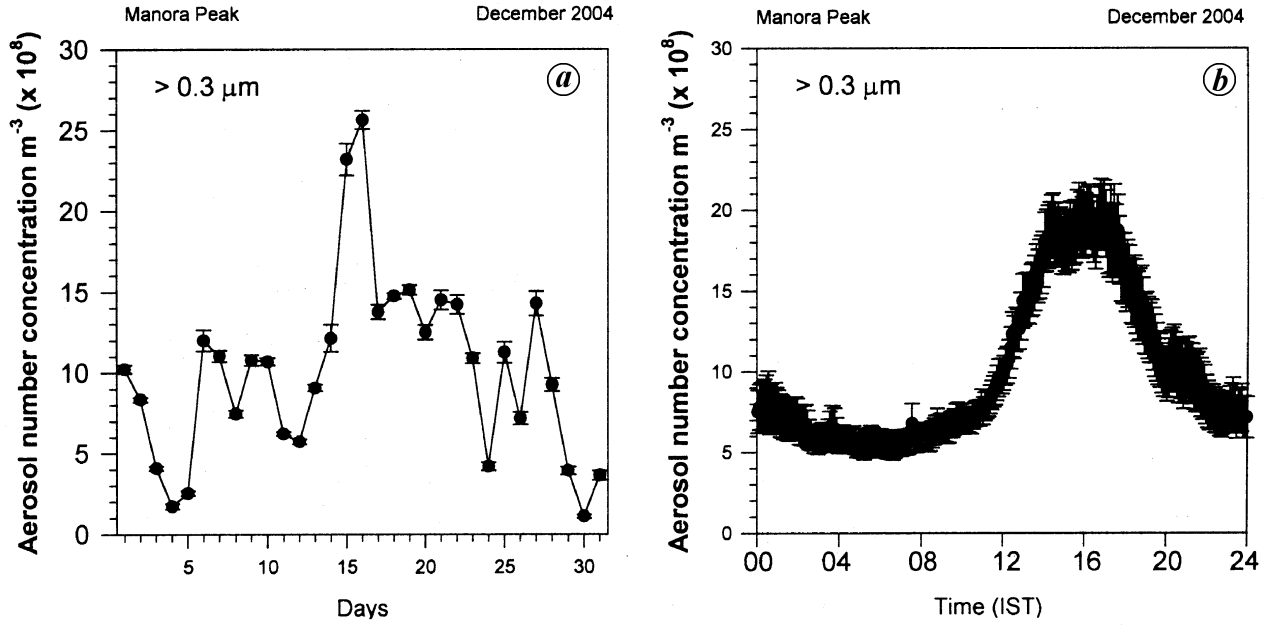

Figure 5. (***a***) Temporal variation of total number (>0.3 μm) concentration (N_T) of near-surface aerosols and (***b***) monthly-mean diurnal variation of N_T at Manora Peak.

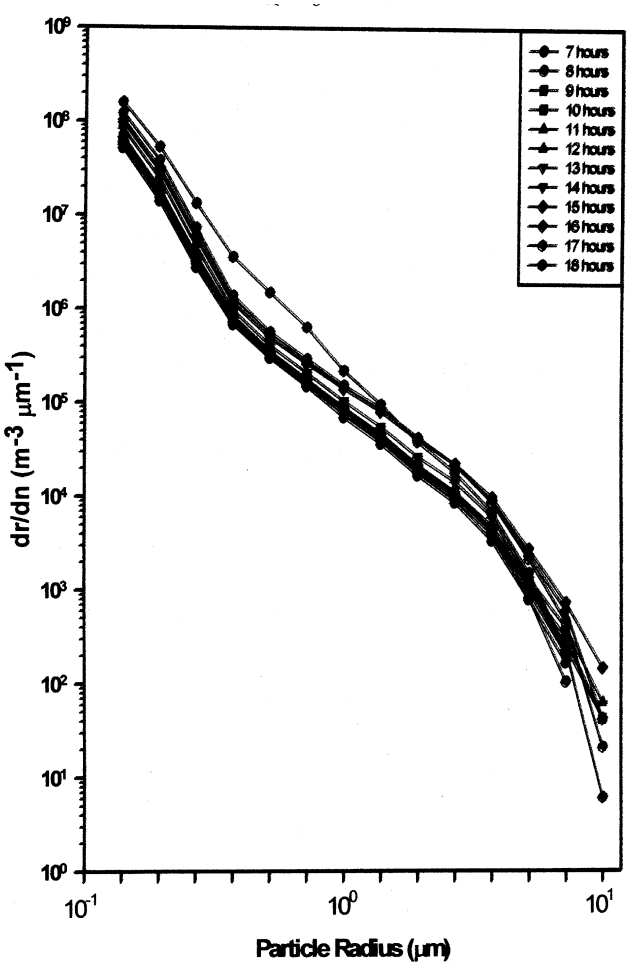

Figure 6. Hourly variation of particle size distribution from morning till evening.

December, whereas the super-micron particles show a peak during 12 to 15 December. Although the contribution of super-micron particles to the total particles comes out to be only about 3%, the afternoon concentrations are about 40% higher than the forenoon concentrations in all representative size ranges (Figure 7).

Black carbon mass concentration

Figure 8 *a* shows the temporal variations of BC mass concentration and Figure 8 *b* shows the monthly mean diurnal variation. The temporal and diurnal variations of BC mass concentration show almost similar trend as in the variations for N_T (Figure 5), thereby indicating similar processes (such as boundary layer dynamics) to be responsible in causing these variations. The monthly mean BC concentration was found to be 1.36 (± 0.99) μg m^{-3}, which is lower compared to other urban locations[11,21,22]. Simultaneous measurements of BC mass concentration were also performed by other investigators during this campaign, which were confined at a low altitude and in urban locations in the Gangetic basin[22,23]. An interesting feature is evident in their findings that the reported diurnal variation in the monthly mean of BC mass concentration shows that the higher value of BC concentration is dominated during night and morning hours, which progressively decreases as the day advances, attaining its minimum level around 1700 h IST; thereafter it again starts rising to its nighttime level. While for Manora Peak site the monthly mean variation of BC mass concentration shows lower values during night and morning hours and increases slowly thereafter, approaching its highest level around 1700 h

IST and then again decreases gradually reaching its minimum level around midnight.

The discrepancy in the diurnal variation of near-surface measurements of the BC mass concentration, between measurements from a high altitude and a low altitude station could be due to the geographical location of the observing site. To the northeast of the Manora Peak site, the hilly terrain of the Central Himalyan ranges (>2.0 km amsl) are prominently located, where anthropogenic activities are absent. On the other hand, the southwest is predominantly covered by the plains (<0.3 km amsl), merely at a distance of about <10.0 km, contributing to most of the BC emission from the heavy vehicular traffic and other anthropogenic sources of pollutants. Once the daytime temperature rises due to the convection processes, the BC aerosols as well as the trapped particles will be lifted into the atmosphere. This facilitates the transport of BC from the source regions to higher altitudes by convective mixing.

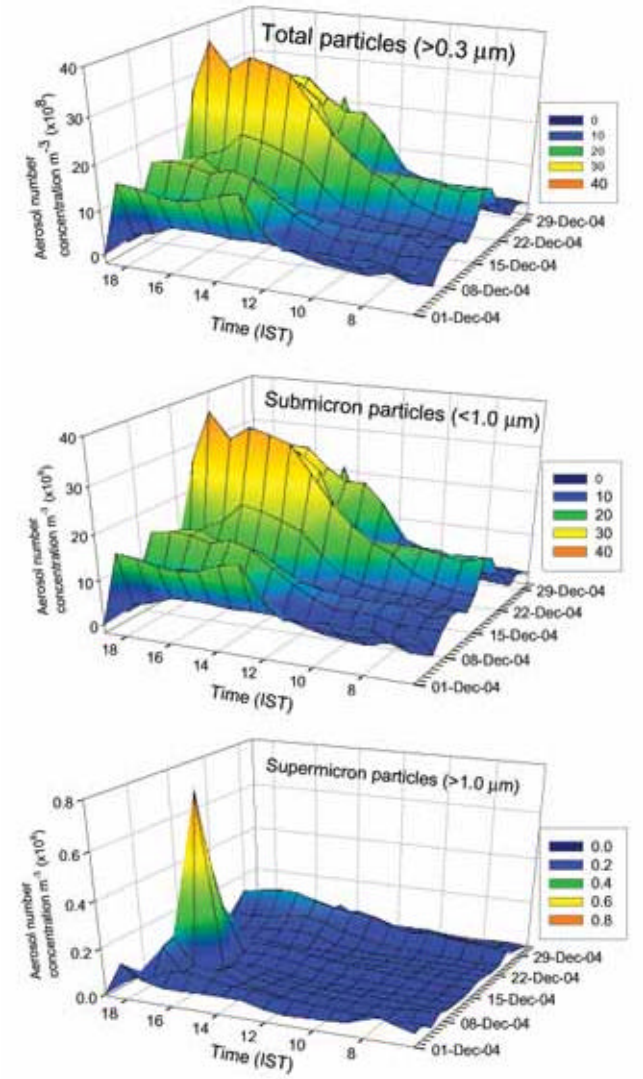

Figure 7. Diurnal variation of composite aerosol number concentrations of representative sizes during December 2004.

Owing to this, a reverse trend in the diurnal variaton in the observed BC concentration between a lower and higher altitude location attributes some sort of downdraft and updraft of BC aerosols, due to the atmospheric boundary layer dynamics during night and daytime respectivly. The mixing process of BC aerosols is also reflected as the observed daytime enhancement in the BC mass concentration over Manora Peak. In a broad sense, the diurnal depletion in BC concentration level at lower altitudes is reflected as the enhancement of its concentration at higher altitudes.

Total suspended particulates

The mass loading of TSP was in the range 15–40 μg m^{-3}, with a mean value of 27.1 (± 8.3) μg m^{-3}. Although BC contributes only a few per cent to the total aerosol mass, it produces significant radiative effects[24,25]. The apportionment of BC is thus important in modelling the aerosol radiative properties. To estimate the BC mass fraction, we have used the monthly mean aerosol mass loading (M_T) and BC mass concentration (M_B). The share of BC to total aerosol mass at Manora Peak was found to be ~6.3 ± 2%. It is lower than the values reported for the west coast location of Trivandrum, which showed 12% share of BC during December[11]. A comprehensive plot of BC, N_t and TSP has been shown in Figure 9. The temporal variation shows almost similar trend for all these parameters, which demonstrates that the share of sub-micron and super-micron aerosols to the total aerosol concentration is significant.

Conclusion

The monthly mean AOD at 500 nm was 0.056 (± 0.037), which is typical for a remote, high-altitude location. Moderately high values of AOD (>0.1) at 500 nm were found on 1, 18 and 21 December 2004 during the campaign. The estimated α value in the present study ranged from 0.4 to 1.2 during the campaign. The estimated AOD values at shorter wavelengths were higher than those measured at higher wavelengths for the entire month of December 2004.

The monthly mean value of W was 0.28 (± 0.11) cm, which reveals prevalence of dry environment over Manora Peak, during the campaign period. The monthly mean TCO was found to be 268 (± 22) DU. The daily mean values retrieved from the ground-based (Microtop II) and reported satellite-based (TOMS) values, show similar trend in ozone variation. However, TOMS measurements underestimate the total ozone values approximately by 20 DU.

The temporal and diurnal variations of BC mass concentration show almost similar trend as those of number concentration of aerosol particles (>0.3 μm). These monthly mean variations at Manora Peak show lower values during night and morning hours, which increase slowly

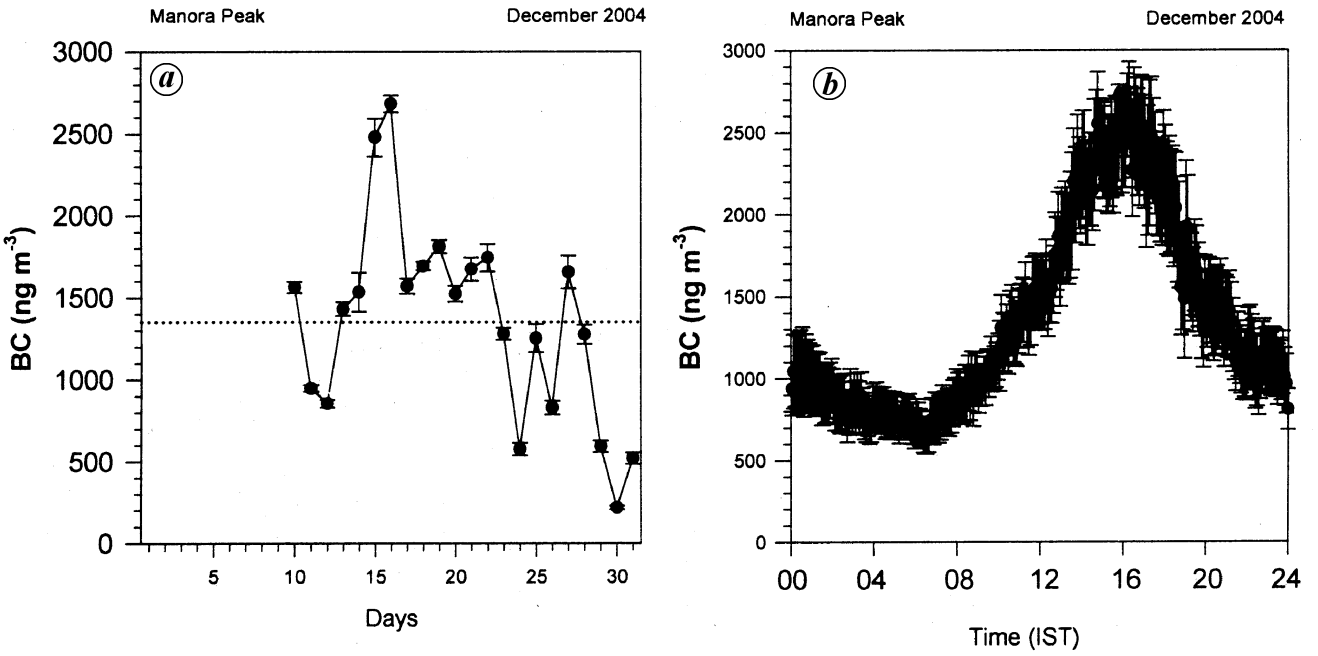

Figure 8. (***a***) Temporal variation of near surface mass concentration (M_B) of BC and (***b***) monthly mean diurnal variation of M_B at Manora Peak during December 2004.

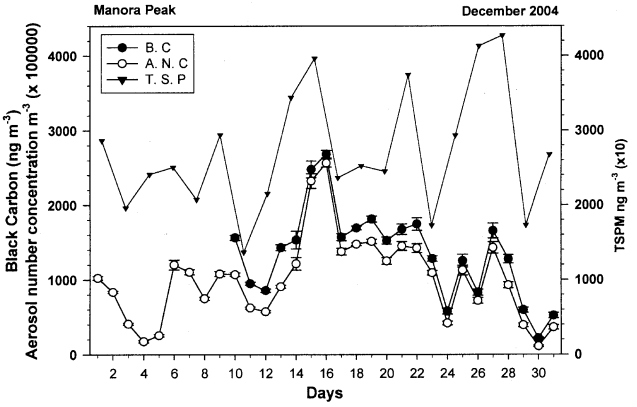

Figure 9. Temporal variation of black carbon (BC), aerosol number concentration (ANC) and total suspended particulate (TSP) during December 2004. Vertical bars represent standard error.

thereafter, approaching the highest level around 1600 h local time and then again decrease gradually, reaching night levels around midnight, thereby showing similar processes such as boundary layer dynamics to be responsible in causing these similarities. The monthly mean BC concentration at Manora Peak during December 2005 was found to be 1.36 (± 0.99) μg m^{-3}. The mass loading of TSP is in the range 15–40 μg m^{-3}, with a mean value of 27.1 (± 8.3) μg m^{-3} and the share of BC to total aerosol mass at Manora Peak was found to be ~6.3%.

The evolutionary trend in the size distribution of aerosol particle concentrations during December 2004 shows that the growth of super-micron particles is faster than particles of total and sub-micron size ranges as the day advances.

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