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Long-term record of aerosol optical properties and chemical composition from a high-altitude site (Manora Peak) in Central Himalaya

K. Ram[1], **M. M. Sarin**[1], **and P. Hegde**[2]

[1]Physical Research Laboratory, Ahmedabad – 380009, India
[2]Space Physics Laboratory, Trivandrum – 695022, India

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Correspondence to: M. M. Sarin (sarin@prl.res.in)

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Abstract

This MS reports on a long-term study of aerosol optical properties and chemical composition, conducted during February 2005–July 2008, from a high-altitude site (Manora Peak, ~2000 m a.s.l.) in the central Himalaya. The chemical analyses suggest that, on average, total carbonaceous aerosols (TCA) and water-soluble inorganic species (WSIS) contribute nearly 25% and 10% of the total suspended particulate (TSP) mass, respectively. Both, TSP and aerosol optical depth (AOD) exhibit significant increase during summer months, with simultaneous increase in the abundance of mineral dust under the prevailing south-westerly winds and long-range transport from desert regions (from middle-East and Thar Desert in western India). The temporal variability in the abundance pattern of carbonaceous species (EC, OC) is also significantly pronounced, with lower concentrations occurring during summertime (April–June) and monsoon (July–August) and relatively high during post-monsoon (September–November) and wintertime (December–March). The WSOC/OC ratios (range: 0.32 to 0.83) during summer and post-monsoon suggest significant contribution from secondary organic aerosols. The mass fraction of absorbing EC (elemental carbon) ranges from less than a percent (during summer and monsoon) to as high as 7.6% (during winter) and absorption coefficient (b_{abs}, at 678 nm) varied as 0.9–33.9 Mm^{-1} (1 Mm^{-1} = 10^{-6} m^{-1}). The linear regression analysis between b_{abs} and EC concentration (μgC m^{-3}) yields a slope of 12.2(±2.3) m^2 g^{-1}, referred as mass absorption efficiency (σ_{abs}) of EC. However, temporal data suggests lower σ_{abs} values during winter and higher in summer and post-monsoon. The change in the mixing state of aerosols and/or variability in the emission sources could be a plausible reason for the variability in σ_{abs} at this high-altitude site (Manora Peak).

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1 Introduction

The rapid urbanization and growth of industrial activities in south and south-east Asia have lead to a substantial increase in the atmospheric aerosol loading in this region (Adhikary et al., 2007; Jethva et al., 2005; Rengarajan et al., 2007). The high aerosol loading, persistent throughout the year, can induce significant dimming of the solar radiation at the surface and can counter the influence of warming caused by greenhouse gases (Carmichael et al., 2009). Black carbon (BC), a major absorbing species in the atmosphere, plays a significant role in Earth's radiation budget, regional climate and hydrological cycle (Jacobson, 2001; Lelieveld et al., 2001; Menon et al., 2002; Ramanathan and Carmichael, 2008). Recent studies have inferred that BC concentration over south-Asia contributes ~5–10% of the total aerosol optical depth (AOD) (Pant et al., 2006; Ramana et al., 2004) and has a significant influence on atmospheric heating with a warming potential of ~55% in comparison to that of CO_2 (Carmichael et al., 2009).

A detailed understanding and knowledge of aerosol chemical composition, size-distribution and optical properties (absorption coefficient, single scattering albedo and aerosol optical depth) is crucial for the estimation of aerosol radiative forcing on a regional scale. A recent study addresses the need for real-time data on natural and anthropogenic aerosols (sulphate, organics, sea salt and dust), trace gases (e.g. O_3, SO_2, NO_x and CO) and their vertical profiles in order to constrain the model derived parameters over Indian subcontinent (Adhikary et al., 2007). Based on aircraft measurements, conducted from an urban location in the Indo-Gangetic Plain, Tripathi et al. (2005) has suggested that BC concentration decreases rapidly as a function of increase in altitude (up to 600 m). Recent studies performed, based on real-time measurements and satellite retrievals, have reported relatively low single scattering albedo (SSA), suggesting absorbing nature of aerosols in the central Himalaya (Gautam et al., 2009; Hyvärinen et al., 2009; Ramana et al., 2004; Sagar et al., 2004). The aerosols at high-altitude site in the central Himalaya can originate from long-range transport of

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mineral dust (during April-June) and those associated with polluted air-masses from the Indo-Gangetic Plain (IGP) during wintertime (Carrico et al., 2003; Dumka et al., 2006; Ram et al., 2008).

We report here a comprehensive data set acquired through long-term measurements during February 2005 to July 2008 on organic and elemental carbon (OC and EC), water-soluble OC (WSOC), a suite of water-soluble inorganic species (K^+, Ca^{2+}, NO_3^- and SO_4^{2-}) and WSOC/OC ratios from a high-altitude site (Manora Peak, ~2000 m above mean see level, amsl) in the central Himalaya. In addition, measurement of optical properties includes aerosol optical depth (AOD), absorption coefficient (b_{abs}) and mass absorption efficiency (σ_{abs}) of EC. Our strategy of sampling from a high-altitude site is advantageous in order to evolve a better understanding of the evolution/transformation and change in the mixing state of aerosols through heterogeneous reactions occurring on mineral surfaces during their transport processes.

2 Methodology

2.1 Ambient aerosol sampling

Ambient aerosols were collected (from February 2005–July 2008) on to pre-heated (at 650 °C for ~6 h) tissuquartz filters (PALLFLEX™, 2500QAT-UP; size: 20.0×25.4 cm^2) using a high-volume sampler operated at a flow rate of 1.0±0.1 $m^3\,min^{-1}$ (Ram et al., 2008). The total suspended particulate (TSP) matter abundance was ascertained by weighing the filters (before and after the sampling) at constant temperature (22±2 °C) and relative humidity (35±5%). Overall, the sampling frequency was maintained at one sample every two weeks, except when the sampling frequency was increased to one sample per week (during December 2007–March 2008) and a total of 86 aerosol samples were collected during the sampling period spanned over 42 months. Of these, 38 samples were collected during wintertime (December–March), 20 samples during summer (April–June), 17 samples during monsoon (July–September) and 11 samples

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during post-monsoon months (October–November).

2.2 Site description and meteorological details

Manora Peak (29.4° N, 79.5° E) is located at an altitude of ~2000 m a.m.s.l. in the Shivalik range of mountains along the central Himalaya (Fig. 1). The sampling site is relatively free from anthropogenic activities within the immediate vicinity. The major source of carbonaceous aerosols includes biomass burning emission used for cooking and heating purposes and transport of polluted air-masses from the Indo-Gangetic Plain (IGP) during wintertime (December–February) (Dumka et al., 2006). During summer months (April–June), long-range transport of mineral aerosol (Fig. 1) dominate the atmospheric loading with a relative decrease in the abundances of carbonaceous species due to reduced biomass burning emission strength (Ram et al., 2008). The two types of aerosols and their emission strengths impart a strong seasonal and inter-annual variability in the chemical and optical properties of ambient aerosol at Manora Peak.

The meteorological parameters, namely rain fall, wind speed and direction and the emission strength of natural/anthropogenic species regulate the atmospheric aerosol loading, chemical composition and optical properties. The aerosol loading and the concentration of chemical species are inversely related to rain fall and thus, the amount and pattern of rain fall during a particular season of the year dictate the seasonal and intra-annual variability. The rain fall over Indian subcontinent is mainly (>80% of total rainfall) observed during the south-west monsoon (late June to September) (Fig. 2d) and leads to an efficient washout of atmospheric aerosols. The major wind regime (south-westerly) passes over the Deserts in middle-East and the Thar Desert (in western India). The higher wind speed can transport aerosols to north-eastern parts of India and over the Himalayan mountains (Hegde et al., 2007; Prasad and Singh, 2007; Ram et al., 2008). These transported aerosols are enriched in mineral dust and result in relatively high TSP abundances during summer months.

The boundary layer dynamics also play an important role in dictating the ambient concentrations of chemical species. Hegde et al. (2008) have reported that bound-

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ary layer height (BLH) is shallower during post-monsoon compared to that in pre-monsoon season. The BLH for the four seasons for the year 2006–2007 are 1300, 1450, 1000 and 600 m for winter, pre-monsoon, monsoon and post-monsoon, respectively (Hegde et al., 2008). Relatively lower temperature, poor thermal convection and shallower boundary layer height confine the aerosols and thus, lead to an increase in the abundances of carbonaceous species (OC and EC) during wintertime. The monthly mean values of the meteorological data (rain fall, temperature, wind-speed and wind-direction) during the sampling period are shown in Fig. 2.

2.3 Analytical methods

The concentration of carbonaceous species (EC, OC) in atmospheric aerosols are assessed on the EC-OC analyzer (Sunset Laboratory, Tigard, OR) using thermo-optical transmittance (TOT) protocol (Ram et al., 2008; Rengarajan et al., 2007). The carbonate carbon (CC) concentrations were obtained by manually integrating the EC-OC thermogram of aerosol samples between 210–220 and 280–290s (Ram et al., 2008). The CC peak in the EC-OC thermograms were confirmed by de-carbonizing aerosol samples with 6M HCl fumes in a desiccators (for ∼3 h), running the acidified aliquot and comparing the thermograph with an unacidified sample (Cachier et al., 1989). Simultaneously, blank filters were also run and all the reported OC concentrations are corrected for the OC blank (∼0.8 µgC m^{-3}) and carbonate carbon. The initial transmittance, ratio of the transmitted radiation (I) to the incident radiation (I_0), measured using a 678 nm laser source is used to define the split-point between OC and EC. The optical-attenuation [ATN; ATN=$-100\times\ln(I/I_0)$] measurements at 678 nm in the EC-OC analyzer have been used for the determination of absorption coefficient (b_{abs}) and mass absorption efficiency of EC (σ_{abs}). The detailed discussion, experimental substantiation and associated uncertainties in the determination of absorption properties of EC are described in our recent publication (Ram and Sarin, 2009).

The measurement of WSOC were performed using a total organic carbon (TOC) analyzer (Shimadzu, model TOC-5000A) as per the protocol described in Ram and

Sarin (2010). Briefly, one-fourth filter (~105 cm^2 area) was soaked in 50 ml de-ionized water (resistivity: 18.2 MΩ cm) and ultrasonicated for ~8 h. The resulting water-extract were filtered, transferred to pre-cleaned glass vials and analyzed for WSOC content. The analyses of water-soluble cations (K^+ and Ca^{2+}) and anions (SO_4^{2-} and NO_3^-) were preformed on the ion-chromatograph (Dionex) (Rengarajan et al., 2007). The clear solution of the water-extracts (the same used for the WSOC analysis) was analyzed for their water-soluble ionic constituents. Simultaneously processed blank filters were run to obtain blank concentrations of WSOC, cations and anions and were subtracted from their actual concentrations in aerosol samples. The details of the measurements of WSOC and water-soluble ionic species are described in our recent publications (Ram and Sarin, 2010; Rengarajan et al., 2007).

3 Results and discussion

3.1 Total suspended particulate (TSP) matter and chemical characteristics

The abundances of total suspended particulate (TSP) matter at Manora Peak show a large temporal variability and ranges from 13 to 272 μg m^{-3} during the sampling period (February 2005–July 2008) (Fig. 3a). A characteristics feature of TSP relates to the dominance of carbonaceous aerosols during wintertime (December–March) resulting from local biomass burning (used for cooking and heating purposes) and vehicular emissions whereas the contribution of mineral dust, originating from the long-range transport from the Thar Desert (in western India) and middle east, dominates during summer months (April–June). The highest TSP value of 272 μg m^{-3}, that occurred on 11 June 2006 (DOY: 162; Fig. 3a), was associated with dust storm (see Fig. 1) (Hegde et al., 2007). The abundances of carbonate carbon (CC) during dust-days showed an order of magnitude increase (compared to the average CC concentration) and co-varied with TSP mass (Fig. 3b). Furthermore, concentrations of water-soluble Ca^{2+}, a tracer of mineral dust, also exhibit an increasing trend (similar to TSP) (Fig. 3c). Mineral

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dust is primarily composed of calcium sulphate (or Gypsum), calcite ($CaCO_3$), $MgCO_3$, dolomite [$CaMg(CO_3)_2$] and alumino-slilicates. For example, Cong et al. (2008) have reported that aerosol samples, collected from Mt. Qomolangma (Everest) during May–June 2005, were mainly composed of alumino-silicates/silica (55%), calcium sulphate (16%), Ca/Mg carbonate (2%) and soot (8%). The reactive fractions of mineral dust such as $CaCO_3$, $MgCO_3$ and $CaMg(CO_3)_2$, can be replaced by the reactive gaseous acidic species such as H_2SO_4 and HNO_3 during the transport through the heterogeneous chemical processes occurring on mineral surfaces (Favez et al., 2008a). This enhances the solubility of mineral dust and thus, leads to an increase in the concentration of $Ca^{2+}$and Mg^{2+} which can change the mixing state (from external to more internal one). As a consequence, this can lead to an increase in the absorption characteristics and cloud condensation nucleation (CCN) properties of aerosols (Geng et al., 2009).

The chemical composition during dust storm events is dominated by mineral aerosols as evident from a relative increase in carbonate carbon (CC) and Ca^{2+} concentrations (Fig. 3b and c). The abundances of carbonaceous species show a decrease (Fig. 3d and e) and the fractional contribution of total carbonaceous aerosols (TCA, see Sect. 3.2) reduces during dust storm events. The concentration of K^+, used as an indicator of biomass burning, is also lower during storm events (Fig. 3g). The abundance of SO_4^{2-} does not show any increase during dust storm events suggesting lower contribution from anthropogenic emissions (Fig. 3i). However, NO_3^- concentrations show a significant increase during storm periods (Fig. 3j), suggesting possible formation of NO_3^- on mineral surfaces through heterogeneous uptake of reactive gaseous nitrogen species (Geng et al., 2009). The fractional contribution of water-soluble ionic species (WSIS; sum of the abundances of cations and anions) decreased to 7% on 11 June 2006. It is noteworthy that not only the contribution of WSIS decreased during dust events; the contribution of TCA was reduced to 6.6% on 11 June 2006. These results suggest that the contribution from mineral aerosols can be as high as ~85% during storm-days. On the contrary, aerosol chemical composition during normal days

shows higher abundances of carbonaceous species. On average, TCA contributes ~25% of the TSP during normal-days. However, the fractional contribution of TCA increases to as high as 65% of the TSP for some days during winter and post-monsoon seasons.

3.2 Temporal and spatial variability in mass concentrations of carbonaceous species

The concentrations of OC and EC at Manora Peak varied from 0.4 to 22.3 μgC m^{-3} (1.9 to 39.3% of the TSP) and 0.14 to 7.6 μgC m^{-3} (0.1 to 7.6% of the TSP), respectively (Fig. 3d and e). The abundances of OC and EC exhibit a large temporal and seasonal variability during the sampling period. The lower OC and EC values are typical of monsoon (due to efficient wash-out of aerosols) and summer months (due to lower biomass burning emission) whereas relatively higher values were observed for the samples collected during post-monsoon and winter months (Fig. 3d and e). The total carbonaceous aerosols (TCA) constitute ~30–35% (of the TSP) during winter and post-monsoon seasons and ~15–20% during summer and monsoon seasons. A value of 1.8 has been taken for converting the measured OC to organic matter (OM) (Cozic et al., 2008) and TCA is estimated as the sum of OM and EC (i.e. TCA=1.8×OC+EC). The conversion factor taken in this study is relatively higher than those used for urban locations (Rengarajan et al., 2007; Turpin and Lim, 2001) because aerosols at high-altitude sites are aged and contains oxygenated organic compounds (Zhang et al., 2007). This is further supported by relatively high WSOC/OC ratios at Manora Peak compared to those at urban locations of the IGP (Ram and Sarin, 2010). Carrico et al. (2003) have reported a similar seasonal variability for OC, EC abundances at high-altitude locations, Nagarkot and Langtang, with higher values occurring during October–January and lower during June–September. Adhikary et al. (2007) have found elevated values of OC and EC after the post-monsoonal months and dry months (up to May). The observed OC and EC mass concentrations, in present study, during April 2005 (9.6 and 1.5 μgC m^{-3}, respectively) are lower than the values of 51.0 and 4.0 μgC m^{-3}, respectively reported

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by Adhikary et al. (2007) for the same time period. In fact, OC concentrations never went beyond 25 $\mu gC\,m^{-3}$ and the highest OC (22.3 $\mu gC\,m^{-3}$) was observed on 26 October 2005 (26 October 2005; DOY 299; Fig. 3d). However, EC concentrations on some occasions can be as high as 7.6 $\mu gC\,m^{-3}$ (e.g. 4 March 2008; DOY 64; Fig. 3e).

The monthly average mass concentrations of OC, EC and WSOC for the sampling period are presented in Table 1. A clear seasonal and inter-annual variability could be noticed for the abundances of carbonaceous species with relatively high abundances during post-monsoon and winter months. The abundances of OC and EC are almost a factor of two lower in July 2006 to September 2007 compared to those in other sampling years. The lower abundances of carbonaceous species during these time period are attributed to the high rain fall during monsoon seasons. A similar variability in BC mass concentrations (measured using the Aethalometer) have been recently reported at a nearby high-altitude station, Mukteshwar, in northern India (Hyvärinen et al., 2009). The average BC mass concentration was 0.8 $\mu gC\,m^{-3}$ for the sampling period (September 2005–September 2007) with an average single scattering albedo (SSA) value of 0.81 at Mukteshwar (Hyvärinen et al., 2009).

The abundances of EC and OC at Manora Peak are more or less similar to the abundances at other high-altitude sites in the world. For example, Puxbaum et al. (2000) have reported that average EC and OC abundances at Nylsvley Natural reserve (altitude: 1100 m) were 0.85 and 14.1 $\mu gC\,m^{-3}$ during May 1997. Hitzenberger et al. (1999) have reported values of 3.8 and 16.2 $\mu gC\,m^{-3}$ for EC and OC at a European background site (Mt. Sonnblick, altitude: 3100 m Austria). Han et al. (2008) have found a similar EC and OC abundances (3.1 and 19.9 $\mu gC\,m^{-3}$ respectively) at a rural-high mountain site (Daihai) in northern China. However, the EC abundances at Manora Peak are relatively higher compared to the average BC abundances of 0.15 and 0.45 $\mu gC\,m^{-3}$ at Mt. Krvavec (Bizjak et al., 1999) and 0.22 $\mu gC\,m^{-3}$ at Mt. Mitchell (Bahrmann and Saxena, 1998). Cozic et al. (2008) have reported that maximum EC and OC concentrations at Jungfraujoch (altitude: 3580 m) were 0.5 and 2.2 $\mu gC\,m^{-3}$, respectively. In a recent study, Cao et al. (2009) have reported an average values of

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0.055 and 0.48 µgC m^{-3}, respectively for EC and OC at Muztagh Ata, a remote mountain in China for the sampling period December 2003–February 2005. However, the abundances of EC and OC at Manora Peak are an order of magnitude lower compared to those at sampling sites in the Indo-Gangetic Plain (Ram and Sarin, 2010).

3.3 OC/EC ratio

The OC/EC ratios in aerosol samples varied from 4.0 to 27.2 during the sampling period (Av: 7.7±3.4, 1 standard deviation). The relatively high OC/EC ratios at Manora Peak indicate the dominance of scattering OC over the absorbing EC. The elevated OC/EC ratios also suggest that either these species have been derived from a primary emission source which is enriched in OC or there is a significant contribution of secondary organic aerosol (SOA) at Manora Peak. The biomass burning emissions produces relatively high fraction of OC compared to EC and thus, results in an enriched OC/EC ratios (Andreae and Merlet, 2001). Manora Peak is located at an altitude of ~2000 m and represents a typical remote site; emissions from vehicular and industrial activities are lower compared to those at the sampling lactations in the Indian Plains. Furthermore, a good linear relationship between OC and EC (R^2=0.83, n=86, Fig. 4a) suggest that emission sources of carbonaceous species have remained the same during the sampling period. The changes in the emission strength and meteorological conditions have lead to observed variabilities in the abundance patterns of OC and EC at Manora Peak. The majority of carbonaceous aerosols in India originate from the biomass burning (used for cooking and residential heating purposes) and agriculture crop-wastes (Gustafsson et al., 2009; Venkataraman et al., 2005). The emission from the former source increases during the wintertime (December–February) when ambient temperatures are relatively lower. Furthermore, relatively low boundary layer height and poor thermal convection, during wintertime, confine the aerosols. The increase in ambient concentrations of measured carbonaceous species during the wintertime are, thus, attributed to the enhanced biomass burning emissions from residential heating/cooking

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and shallower boundary layer height.

As stated earlier, high OC/EC ratios at Manora Peak could also result from the enhanced SOA formation. However, this would reflect in the seasonal variability in OC/EC ratios as SOA formation exhibit a seasonal pattern (Castro et al., 1999; Chang and Lee, 2007). These studies have suggested relatively higher SOA formation during summer months (due to higher photochemical activity) compared to that in winter months. The seasonally averaged OC/EC ratios at Manora Peak are similar and statistically indistinguishable. However, the median values of OC/EC ratios for summer season (8.0) are relatively higher compared to those during winter (6.7) and post-monsoon seasons (7.2). The estimated secondary organic carbon (SOC), using the EC tracer method (Castro et al., 1999; Ram et al., 2008), exhibit a seasonal pattern and average SOC contribute to ~30, 48, 37 and 20% (of OC) during winter, summer, monsoon and post-monsoon seasons, respectively. Thus, the higher OC/EC ratios during summer could also be due to the enhanced SOA formation. However, this would lead to an increase in the WSOC/OC ratios during summer months because most of the SOA are soluble in water (Kondo et al., 2007; Weber et al., 2007). The results indicate that WSOC/OC ratios do not show any seasonal variability during the sampling period (also see the discussion in Sect. 3.4). Thus, it can be inferred that higher OC/EC ratios at Manora Peak are derived from the transport of biomass burning emissions in the IGP and western parts of India with a significant contribution from secondary organic aerosols.

3.4 Water-soluble organic carbon (WSOC) and WSOC/OC ratio

The WSOC mass concentration at Manora Peak varied from 0.9 to 15.4 $\mu gC\,m^{-3}$ during the study period (2005 to 2008). The measured WSOC concentration shows a good correlation with OC concentration with all the data falling on a line with a slope of 0.54 (R^2=0.79, n=69, p <0.05, Fig. 4b). Although, a significant linear relationship is observed between WSOC and OC for the entire sampling period with a slope of 0.55, some of the data points show different values of WSOC/OC ratios for the same time-period. For example, the lower WSOC/OC ratios (0.35) in aerosol samples collected

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during winter season probably indicate fresh emission and less chemical processing and transport from the Indo-Gangetic Plain (IGP). This observation is also corroborated by lower WSOC/OC ratios observed at urban locations (range: 0.32 to 0.40) in the IGP during wintertime (Ram and Sarin, 2010). Furthermore, a good correlation between WSOC and SO_4^{2-} during wintertime (R^2=0.57, n=29, figure not shown), compared to that during summer months (R^2=0.25, n=14), suggest their transport from the Valley. However, the average WSOC/OC ratio observed at Manora Peak (0.55±0.15; median 0.53) is lower than that reported for the Himalayan Nepal Climate Observatory-Pyramid (0.65±0.15) situated at an altitude of 5079 m a.s.l. (Decesari et al., 2009). Thus, an increasing trend in the WSOC/OC ratio is observed as we move from sampling sites in the IGP to high-altitude sites. It has been suggested that WSOC mass concentration and WSOC/OC ratios can be used as an indicator for the secondary organic aerosol (SOA) formation (Kondo et al., 2007; Weber et al., 2007). Therefore, the increasing trend in WSOC/OC ratios suggest the aging, chemically processed aerosols and/or possible contribution from the SOA (Zhang et al., 2007).

However, a recent study have reported that a significant fraction of freshly formed SOA in a semi-arid region can be water-insoluble in nature (Favez et al., 2008b). The water-insoluble OC (WIOC; defined as the difference between OC and WSOC) could also be derived from primary emissions such as biomass burning and vehicular exhausts (Favez et al., 2008a). The WIOC concentration shows a linear relationship with thermal EC concentrations (R^2=0.48, n=69, figure not shown) and the slope (2.6) similar to WIOC/EC ratios in the IGP for primary emission sources (Ram and Sarin, 2010). The median WIOC/EC ratios at Manora Peak, during the four seasons, are 3.0 (winter), 3.8 (summer), 3.9 (monsoon) and 2.6 (post-monsoon). The resemblance of the WIOC/EC ratios to those in the IGP further substantiates the advective transport of primary emitted aerosols from Indian Plain.

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4 Optical properties

4.1 Aerosol optical depth (AOD)

The dust storm does not only affect the chemical composition, they also change physical and optical properties (e.g. AOD and fine-mode aerosol fraction, FMAF) of aerosols (Jethva et al., 2005). The aerosol size-distribution during storm days shows an increase in coarse-mode particles (Hegde et al., 2007) and a decrease in the FMAF values (Jethva et al., 2005). During the dust-storm event in June 2006, Hegde et al. (2007) have reported that the number concentration of coarse-mode particles and aerosol optical depth (AOD) values increased by a factor of five compared to the respective monthly mean values. We have compiled the AOD values reported in the literature (from 2002 onward) and those measured during 2006–2008 at Manora Peak and AOD values are presented in Fig. 5. The monthly averaged AOD values and TSP mass concentrations, along with measured chemical parameters for the sampling period, are presented in Table 1. It is noticeable AOD values at Manora Peak show a significant increase (factor of two to six) during summer months (April–June) compared to those during winter months (Fig. 5; Table 1). Based on one-year measurements of AOD with a sunphotometer at the Himalayan Nepal Climate Observatory-Pyramid, Gobbi et al. (2010) have shown that AOD values show maximum during summer (May–September) and minimum in winter. The higher AOD values during summer months are attributed to the increase in abundance of mineral aerosol. The lower AOD values during monsoon months are attributed to an efficient wash-out by rain.

4.2 Absorption coefficient

The absorption coefficient (b_{abs}) and mass absorption efficiency of EC (σ_{abs}) were simultaneously assessed using the thermo-optical EC-OC analyzer and all the values presented in this study are given at 678 nm (Tables 1 and 2). The measured b_{abs} values ranged from 0.9 to 33.9 Mm^{-1} (1 Mm^{-1}=10^{-6} m^{-1}) and exhibit a large temporal

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and inter-annual variability during the sampling period (Figs. 6 and 7a). The higher b_{abs} values were observed for the aerosol samples collected during winter and post-monsoon months whereas lower values were associated with summer and monsoon months. The monthly average EC mass concentration, b_{abs} and σ_{abs} values are shown in Fig. 7b. Generally, b_{abs} values decreases April onward and the minimum were observed in July (monsoon month) and then it continues to increase until end of the March. The trend of b_{abs} values at Manora Peak closely follows EC concentration trend (Fig. 7b) suggesting EC as a major absorbing species. Recently, Hyvärinen et al. (2009) have reported that b_{abs} values measured using the Aethalometer varies from 4.5 to 23.2 Mm^{-1} during September 2005–September 2007 at Mukteshwar, a high-altitude site in northern India located nearby Manora Peak. The BC mass concentrations, based on Aethalometer based measurement, also exhibit a large spatio-temporal variability over Indian regions (Beegum et al., 2009). If BC mass concentrations are converted to b_{abs}, one can find that b_{abs} values at Manora Peak are an order of magnitude lower compared to those in the Indo-Gangetic Plain (Ram and Sarin, 2009). Furthermore, the annual average b_{abs} values at Manora Peak are factor of two higher than those at Mt Abu, another high-altitude site in western India (Ram and Sarin, 2009). A comparison of EC mass concentration, absorption coefficient (b_{abs}), mass absorption efficiency (σ_{abs}), AOD and single scattering albedo (SSA) in the Himalayas and other high-altitude sites are presented in Table 2.

4.3 Mass absorption efficiency of EC (σ_{abs})

The σ_{abs} values at Manora Peak ranges from 4.4 to 21.2 $m^2\,g^{-1}$ for the entire sampling period (February 2005–July 2008). Despite of a large temporal and sample-to-sample variability in σ_{abs} values, measured b_{abs} values and thermal EC (in unit of $\mu gC\,m^{-3}$) show a good correlation for the data with EC<2.0 $\mu gC\,m^{-3}$ and/or b_{abs} <25 Mm^{-1} (R^2=0.72, n=73, Fig. 6) and the slope of the best-fit line provides a value of 12.2 $m^2\,g^{-1}$. However, the linearity ceases for the data with EC >2.0 $\mu gC\,m^{-3}$ and/or

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b_{abs} >25 Mm^{-1}. Most of these data points represent collection of aerosol samples during wintertime (Fig. 6). The samples collected during wintertime have relatively lower σ_{abs} values compared to those for other seasons (Fig. 7 and Table 1). Recently, Cozic et al. (2008) have reported that absorption coefficient (at 630 nm) measured by the Multi-Angle Absorption photometer (MAAP) and thermal EC with a semi-continuous EC-OC analyzer were highly correlated (R^2=0.97). The σ_{abs} values showed a seasonal variability with an average values of 7.6±0.2 $m^2\,g^{-1}$ (winter) and 11.1±0.2 $m^2\,g^{-1}$ (summer).

The variability in σ_{abs} values, reported in the literature, has been explained in terms of the sources, chemical composition, measurement protocols, aerosol aging and the mixing state of aerosol (Liousse et al., 1993; Ram and Sarin, 2009; Sharma et al., 2002). For example, Liousse et al. (1993) have reported that σ_{abs} values varied from 2 to 25 $m^2\,g^{-1}$ for aerosol samples derived from different emission sources. Sharma et al. (2002) have explained the variability in σ_{abs} values (6.4 to 20.1 $m^2\,g^{-1}$) in terms of the sources and atmospheric processing of aerosols reaching at rural, urban and suburban sampling sites in Canada. Cozic et al. (2008) suggested that higher σ_{abs} values in summer probably resulted because of greater coating of BC due to the photochemical activity. The mixing state of aerosols (external or internal) could be a probable reason for observed seasonal variability in the mass absorption efficiency at Manora Peak. The aerosol particles collected at Manora Peak (during winter) are relatively drier, located near the source regions (i.e. freshly emitted) and may exist as an external mixture of aerosols and thus, probably have lower σ_{abs} values. On the other hand, aerosol particles collected during summer months are aged and chemically processed during the transport processes and may exist as an internally mixed particle, thus resulting in higher σ_{abs} values. However, the reasons for the variability in mass absorption efficiency of EC need to further investigated.

The σ_{abs} values, obtained in this study, are higher than those reported during the PRIDE-PRD 2004 experiment, e.g. 7.7 $m^2\,g^{-1}$ (Andreae et al., 2008); 7.2±1.0 and 9.3±1.4 $m^2\,g^{-1}$ for PM_1 and PM_{10} aerosols (Cheng et al., 2008). Bond and

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Bergstrom (2006) have suggested a value of 7.5±1.2 $m^2 g^{-1}$ for the σ_{abs} at 550 nm for the freshly emitted soot particles. However, if we assume an enhancement of 50% in the absorption for coated and aged aerosols (Bond et al., 2006); the observed σ_{abs} (12.2±2.3 $m^2 g^{-1}$) value is in the similar range as suggested in the literature. The average σ_{abs} value at Manora Peak is relatively higher than the commonly cited value of 10.0 $m^2 g^{-1}$ and that used in the Particle Soot Absorption Photometer (PSAP) for the determination of BC mass concentrations (Sharma et al., 2002). In a recent study, Miyazaki et al. (2008) have reported σ_{abs} as 9.8±0.1 $m^2 g^{-1}$ for a suburban site in Thailand based on a newly designed Continuous Soot Monitoring System (COSMOS) for the measurement of BC. Based on Aerosol Robotic Network (AERONET) retrievals, Schuster et al. (2005) have derived an average values of 10.5 and 10.0 $m^2 g^{-1}$ were obtained for Asian continental aerosols for the years 2000 and 2001. Dey and Tripathi (2006) have reported σ_{abs} as 7.9±1.8, 9.7±3.4 and 12.7±2.9 $m^2 g^{-1}$ at an urban location (Kanpur) in northern India, for the years 2001, 2002 and 2003 respectively. The average σ_{abs} value, obtained in this study, is similar to those reported for Asian aerosols derived based on the AERONET retrievals.

5 Conclusions and implications

Our long-term study carried out for over three years from a high-altitude site (Manora Peak) located in the foot-hills of the central Himalaya has provided important information on temporal variability in the abundance pattern of EC, OC, WSOC and secondary organic aerosol formation. In addition, optical properties and mass absorption efficiency of EC have been evaluated. The results documented in this study are useful for the assessment of single scattering albedo (SSA) and aerosol radiative forcing on a regional scale.

The important findings are:

Although, Manora Peak represents a high-altitude location; mass concentration of

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absorbing EC and absorption coefficient are significantly higher compared to other high-altitude sites of the world. The EC concentration and absorption coefficient varies by an order of magnitude (range: 0.1 to 7.6 $\mu gC\,m^{-3}$ and 0.9 to 33.9 Mm^{-1}, respectively) during February 2005–July 2008. The representative mass absorption efficiency of EC, at Manora Peak, is 12.2(±2.3) $m^2\,g^{-1}$. However, mass absorption efficiency of EC shows relatively lower values during winter season and higher during summer and post-monsoon seasons.

The monthly mean AOD varied from 0.03 to of 0.47 and exhibit maximum during summer months. However, the temporal variability in TSP mass is not significantly pronounced, except for the sampling year of 2006 marked by dust storm events. The relatively high TSP and AOD, during summer, are attributed to the transport of mineral dust originating from desert regions in middle-East and Thar Desert in western India.

Another conspicuous feature of carbonaceous aerosols is reflected in high WSOC/OC ratios (average: 0.55±0.15) at Manora Peak (compared to those in the Indo-Gangetic Plain) (Ram and Sarin, 2010) and is attributed to the enhanced secondary organic aerosol (SOA) formation and/or aging of aerosols during transport processes. Although, the contribution of EC was lower during summer and monsoon months; its contribution was significant during wintertime (as high as 7.6%) at Manora Peak.

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Table 1. The monthly average values (±1 standard deviation, s.d.) of optical and chemical parameters in ambient aerosols at Manora Peak during February 2005–July 2008.

Month	n	TSP	AOD[a]	b_{abs}	σ_{abs}	EC	OC	WSOC	WSOC/OC	NO_3^-	SO_4^{2-}	K^+	Ca^{2+}
		$\mu g\,m^{-3}$		Mm^{-1}	$m^2 g^{-1}$	$\mu gC\,m^{-3}$	$\mu gC\,m^{-3}$	$\mu gC\,m^{-3}$		$\mu g\,m^{-3}$	$\mu g\,m^{-3}$	$\mu g\,m^{-3}$	$\mu g\,m^{-3}$
Jan	6	52±22	0.06±0.04	13.9±8.2	11.3±0.8	1.3±0.8	9.3±4.4	5.2±3.3	0.57±0.24	0.4±0.4	2.9±2.6	0.3±0.3	0.8±0.5
Feb	12	76±49	0.11±0.07	18.9±7.9	10.5±2.2	1.9±0.9	10.6±5.5	6.1±3.2	0.56±0.15	0.3±0.3	6.1±4.4	0.6±0.3	1.4±0.8
Mar	14	86±32	0.18±0.15	12.7±4.5	12.6±4.7	1.5±0.5	8.8±3.1	3.8±1.0	0.46±0.11	0.6±0.3	3.6±1.3	0.5±0.3	1.9±0.5
Apr	6	106±59	0.25±0.09	13.9±5.7	11.4±3.2	1.4±0.8	9.1±5.8	5.3±2.7	0.46±0.06	1.0±0.8	3.5±1.6	0.9±0.2	2.0±1.1
May	6	87±68	0.31±0.12	11.5±6.6	14.0±3.5	0.8±0.5	4.7±2.0	2.7±1.3	0.51±0.12	1.2±1.0	4.9±3.0	0.2±0.1	2.0±1.6
Jun	8	86±111	0.33±0.03	9.3±4.6	14.5±3.6	0.6±0.2	4.8±2.7	2.4±1.1	0.58±0.09	1.5±1.6	3.3±1.1	0.2±0.2	1.2±1.1
Jul	7	55±68	0.16±0.11	4.1±2.3	12.2±3.5	0.3±0.2	2.9±1.8	1.5±0.5	0.44±0.10	0.2±0.2	2.5±1.5	0.1±0.05	0.7±0.8
Aug	5	49±21	0.16±0.04	6.7±2.1	13.7±2.6	0.5±0.2	4.3±2.1	1.9±1.0	0.44±0.05	0.4±0.5	3.7±1.7	0.2±0.1	1.4±0.9
Sep	5	52±18	0.13±0.06	12.9±3.9	15.3±2.2	0.9±0.4	6.9±2.6	4.8±2.1	0.68±0.11	0.1±0.1	5.0±1.1	0.5±0.3	1.2±0.7
Oct	6	78±47	0.10±0.03	20.0±7.8	14.2±3.4	1.5±0.8	12.2±6.3	8.4±4.8	0.67±0.13	0.9±1.2	8.5±4.5	0.9±0.6	2.2±1.7
Nov	5	47±9	0.10±0.04	14.8±4.2	12.3±3.7	1.3±0.6	9.1±4.3	5.2±2.2	0.61±0.19	0.4±0.2	4.8±3.1	0.4±0.2	1.0±0.1
Dec	6	48±20	0.08±0.04	19.7±3.8	12.6±2.6	1.7±0.7	12.3±6.3	6.7±2.8	0.57±0.11	0.5±0.6	4.3±2.8	0.5±0.2	0.9±0.4

[a] AOD: average values (@ 500 nm) for the respective months of the years 2002, 2006, 2007 and 2008; b_{abs} and σ_{abs} data are given at 678 nm wavelength.

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Table 2. The measured optical parameters in ambient aerosols at Manora Peak and an inter-comparison with those reported in literature for the Himalayan region and other high-altitude sites. Numbers in parenthesis represent median values.

Sampling sites	Elevation	Sampling period	b_{abs}	σ_{abs}	SSA	AOD	EC/or BC	Reference
	(km)		Mm^{-1}	m^2g^{-1}			$ng\,m^{-3}$	
Manora Peak	1.95	winter	17.0±7.0 (16.8)	10.9±3.5 (10.4)		0.11±0.09 (0.08)[a]	1850±1440 (1840)	Present study
Manora Peak	1.95	summer	11.3±5.8 (10.5)	12.1±2.5 (12.9)		0.29±0.09 (0.31)[a]	940±690 (815)	Present study
Manora Peak	1.95	monsoon	7.4±4.6 (7.5)	13.5±3.0 (14.0)		0.09±0.09 (0.09)[a]	540±330 (520)	Present study
Manora Peak	1.95	Post-monsoon	17.6±13.3 (15.5)	13.3±3.5 (14.8)		0.08±0.02 (0.10)[a]	1440±690 (1240)	Present study
Manora Peak	1.95	Feb 2005–Jul 2008	13.9±7.4 (13.8)	12.3±3.6 (12.2)		0.16±0.11 (0.14)[a]	1320±1160 (970)	Present study
Manora Peak	2.18	Sep 2005–Sep 2007	11		0.81 (0.75–0.85)		800	Hyverinen et al. (2009)
Mt. Abu	1.7	May 2005–Feb 2006	5.8±4.3	9.8±2.1			500	Ram and Sarin (2009)
Manora Peak	1.95	Feb–Apr 2006					400–2900	Dumka et al. (2008)
Manora Peak	1.95	Dec 2004			0.9 (0.87–0.94)	0.06±0.03	1360	Pant et al. (2006)
Manora Peak	1.95	Jan–Dec 2002				0.0–0.69		Sagar et al. (2004)
Kathmandu	1.35	winter 2003			0.7–0.9	0.34 (0.16–0.55)		Ramana et al. (2004)
Godavari	1.6	winter 2003			0.7–0.9			Ramana et al. (2004)
Nagarkot	1.975	winter 2003				0.06–0.29		Ramana et al. (2004)
NCO-P	5.079	winter 2006			0.6–0.9	0.04[b]		Gobi et al. (2010)
NCO-P	5.079	May–Sep 2006			0.6–0.9	0.14[b]		Gobi et al. (2010)
Jungfraujoch	3.58	Feb–Mar 2005	0–4.2	7.6±0.2		0.06±0.03	0.0–500	Cozic et al. (2008)
Jungfraujoch	3.58	Aug 2005	0–4.2	11.1±0.3				Cozic et al. (2008)

[a] Average of AOD data (@ 500 nm) for the years 2002 and 2006–2008. Seasons are defined as: winter (December–March), summer (April–June), monsoon (July–September), post-monsoon (October–November). [b] Median values (@ 500 nm).

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Fig. 1. The sampling site, Manora Peak (white circle) located at the foot-hills of central Himalaya. A major dust-storm that occurred on 12 June 2006 over the Indus Valley (along the border between Pakistan and India) is also shown in the picture (brown rectangle). This dust-storm had resulted in the high TSP abundances and AOD values at Manora Peak. The event was captured by the Moderate Resolution Imaging Spectroradiometer *(MODIS)* flying onboard NASA's *Aqua* satellite on 12 June 2006 (http://earthobservatory.nasa.gov) and was combined with the Google Earth.

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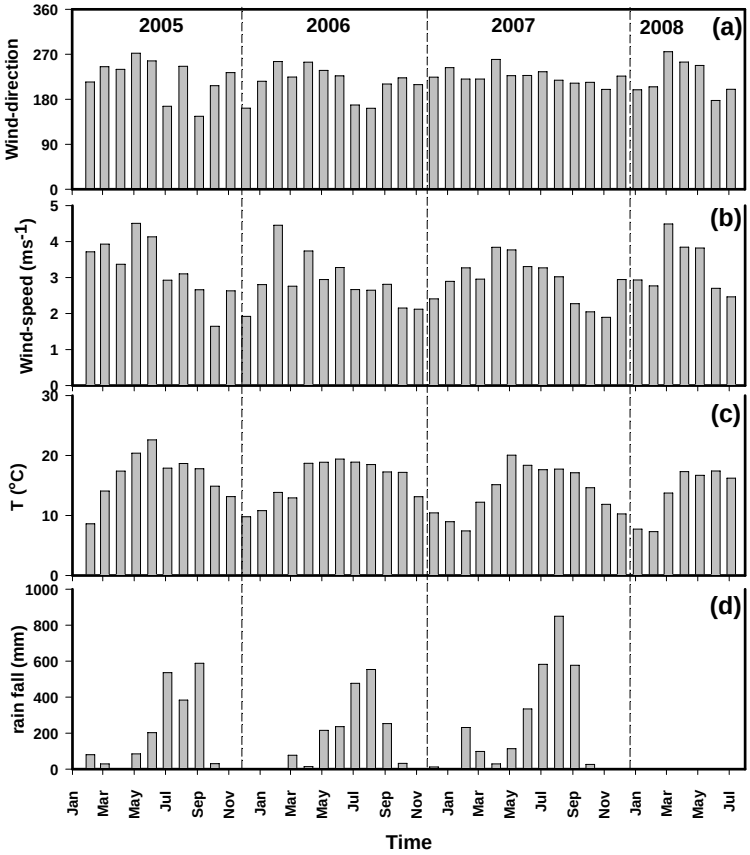

Fig. 2. The monthly average meteorological data for **(a)** wind-direction, **(b)** wind speed, **(c)** temperature and **(d)** rain fall during the sampling period (February 2005–July 2008). Rain-fall data for 2008 is not available.

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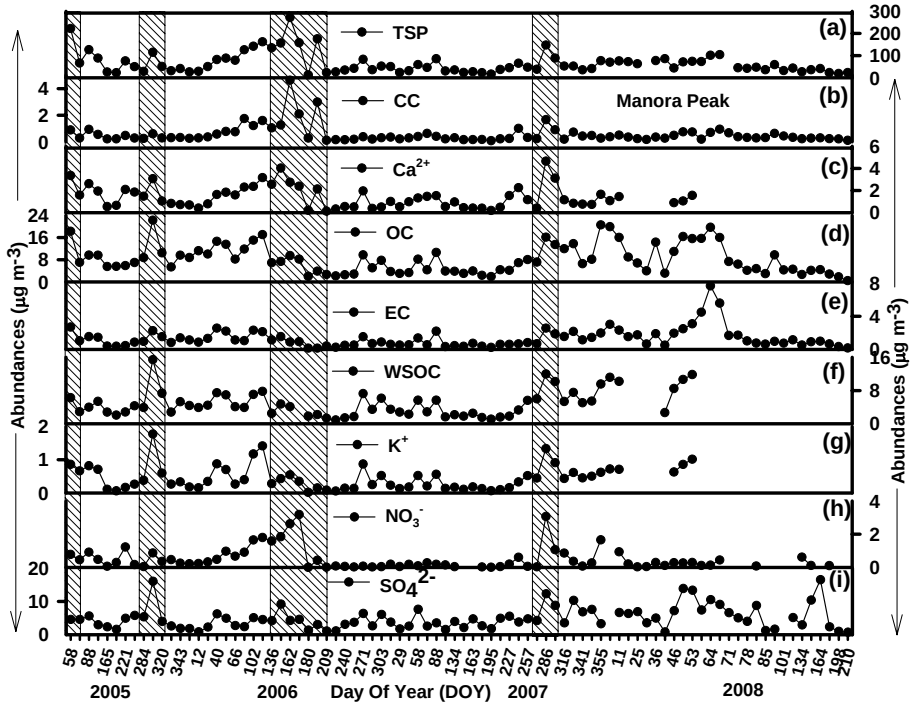

Fig. 3. Temporal variability in the abundances of **(a)** total suspended particulate (TSP) matter, **(b)** carbonate carbon (CC), **(c)** Ca^{2+}, **(d)** OC, **(e)** EC, **(f)** WSOC, **(g)** K^{+} **(h)** NO_3^- and **(i)** SO_4^{2-} during the sampling period. The shaded area for the sampling in 2006 represents dust-storm events; whereas other three shaded area show predominance of pollutants.

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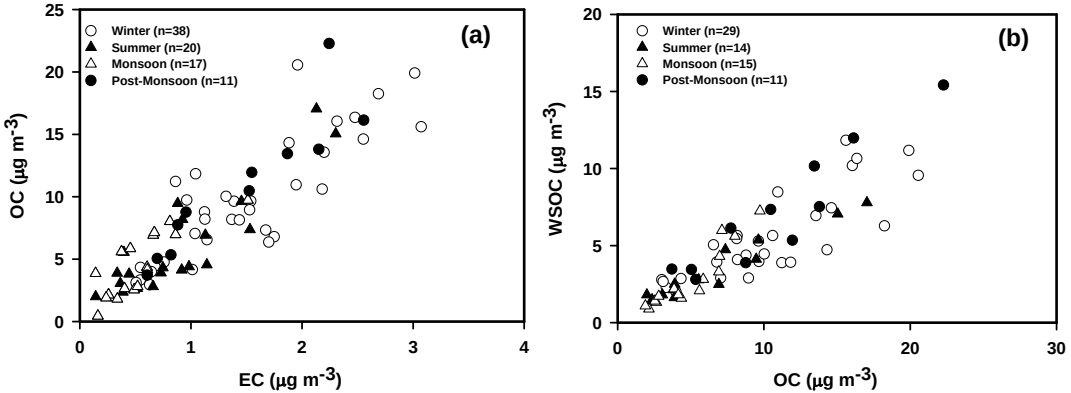

Fig. 4. Scatter plots between **(a)** OC and EC and **(b)** WSOC and OC.

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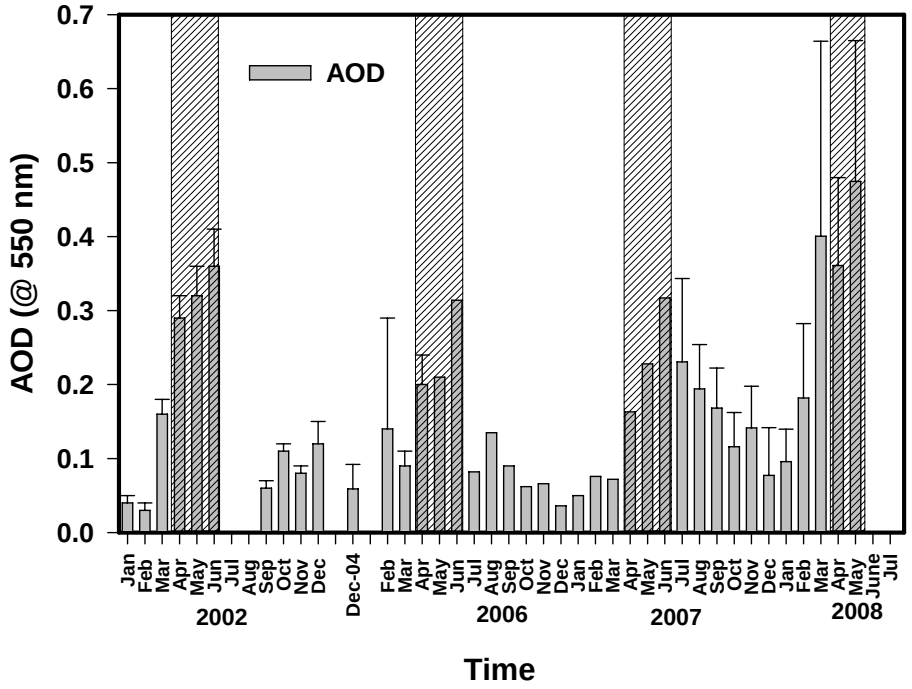

Fig. 5. The monthly mean aerosol optical depth (AOD @ 550 nm) at Manora Peak. The AOD data for the years 2002 and December 2004 are taken from Sagar et al. (2004) and Pant et al. (2006), respectively. The vertical strips indicate the high AOD representing summer season (April–June) of the respective years.

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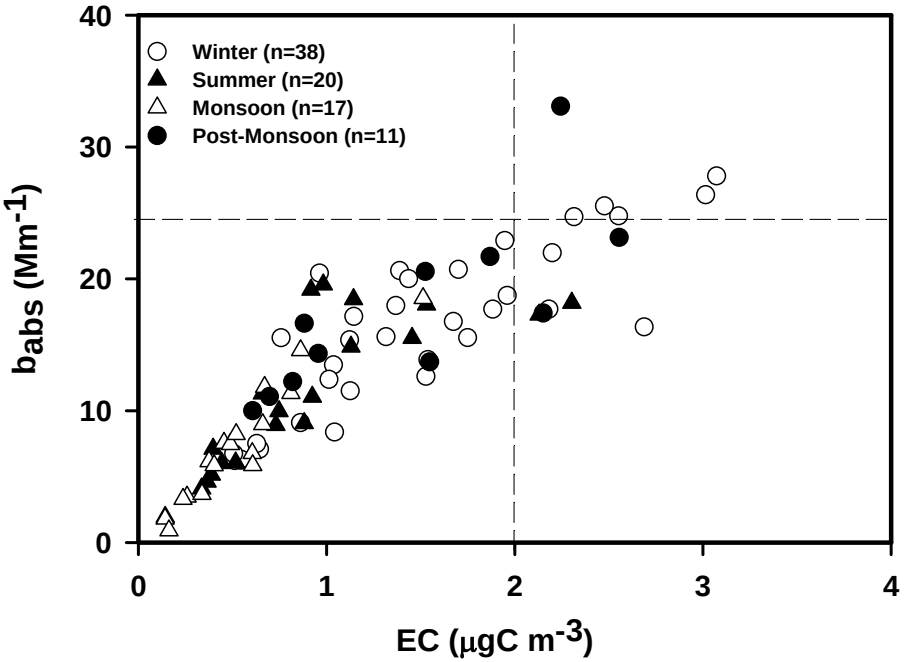

Fig. 6. The scatter plot between b_{abs} and EC concentration exhibit significant correlation up to 25 Mm^{-1} (shown by horizontal dashed line) and 2.0 $\mu gC\,m^{-3}$ (shown by vertical dashed line). The linearity ceases at higher EC concentrations and high b_{abs} values.

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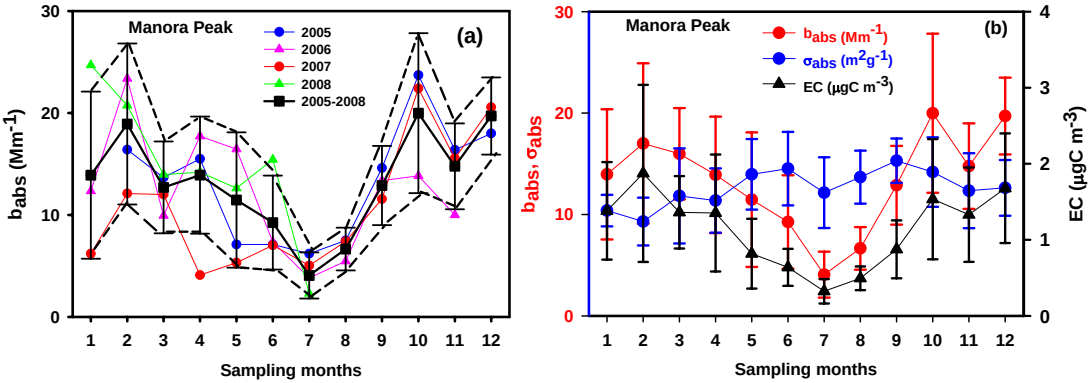

Fig. 7. **(a)** The temporal variability of average b_{abs} for the years 2005, 2006, 2007 and 2008 (shown in different colours). Data points shown by filled squares refer to monthly average values of b_{abs} for four years. **(b)** The monthly average values of b_{abs}, σ_{abs} and EC mass concentrations.

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