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Characterization of aerosol black carbon over a tropical semi-arid region of Anantapur, India

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ABSTRACT

Black carbon (BC) aerosol mass concentrations measured using an aethalometer at Anantapur, a semi-arid tropical station in the southern part of peninsular India, from August 2006 to July 2007 are analyzed. Seasonal and diurnal variations of BC in relation to changes in the regional meteorological conditions have been studied along with the mass fraction of BC to the total aerosol mass concentration (M_t) and fine particle mass (FPM) concentration in different months. The data collected during the study period shows that the annual average BC mass concentration at Anantapur is 1.97 ± 0.12 μg m^{-3}. Seasonal variations of BC aerosol mass concentration showed high during the dry (winter and summer) seasons and low during the post-monsoon followed by the monsoon seasons. Diurnal variations of BC aerosols attain a gradual build up in BC concentration from morning and a sharp peak occurs between 07:00 and 09:00 h almost an hour after local sunrise and a broad nocturnal peak from 19:00 to 21:00 h with a minimum in noon hours. The ratio of BC to the fine particle mass concentration was high during the dry season and low during the monsoon season. The regression analysis between BC mass concentration and wind speed indicates that, with increase in wind speeds the BC mass concentrations would decrease and vice-versa. Aerosol BC mass concentration shows a significant positive correlation with total mass concentration (M_t) and aerosol optical depth (AOD, τ_p) at 500 nm.

1. Introduction

Black carbon (BC), the optically absorbing part of carbonaceous aerosols, is the major anthropogenic component of atmospheric aerosol system, which has significantly different optical and radiative properties, as compared to the other normal constituents (Badarinath et al., 2009). BC acts as an indicator of airmass affected by anthropogenic pollution (Penner, 1995). Current estimates of total global emission are approximately 8 Tg C yr^{-1} (IPCC, 2007). The chief sources of BC are the burning of biomass and fossil fuels, the automobile and aircraft emissions and forest fires. Being chemically inert, and in fine size range, the only removal mechanism of BC from the atmosphere is wet deposition, and as such, BC particles have long lifetime in the atmosphere (≳1 week in the lower troposphere; Babu and Moorthy, 2001), making them amenable for long-range transport. This long life, coupled with the strong absorption of light over a wide spectral range, makes BC an important contributor to radiative forcing of the atmosphere and 'green-house warming' (Jacobson, 2001; Chung and Seinfeld, 2005). This absorption can greatly offset the 'white-house cooling' due to sulphate aerosols (Schuster et al., 2005), and can even surpass the green-house warming effects of some of the atmospheric trace gas species (Jacobson, 2001). Recent studies suggest that BC can alter the cloud lifetime (Ackerman et al., 2000), precipitation patterns (Menon et al., 2002),

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reflectivity and melting of snow and ice (Hansen and Nazarenko, 2003).

The characterization of BC is attracting considerable interest in recent years due to its environmental and climate significance, as well as anthropogenic nature of its origin (Hansen et al., 2000). The atmospheric BC directly accounts for the reduction of the incoming short-wave solar radiation, leading to heating of the atmosphere. BC causes serious health problems too; as it can get easily deposited into the respiratory system through inhalation because of its fine sub-micron size. Recently, BC has been used as an indicator of exposure to diesel soot (e.g., Fruin et al., 2004), which has been classified as a toxic air contaminant and a suspected carcinogen. Considering the key role they play in atmospheric radiative transfer as well as chemical properties, studies on BC aerosols have become an important topic in recent years. The importance of anthropogenic emissions from South Asian countries in affecting the atmosphere over the north Indian Ocean has been significantly highlighted especially after the Indian Ocean Experiment (INDOEX) studies (Lelieveld et al., 2001; Ramanathan et al., 2001). However, there are very few data available on BC aerosols from the Indian region though some studies (Babu et al., 2002; Novakov et al., 2003; Latha and Badarinath, 2005; Moorthy et al., 2004; Tripathi et al., 2005; Husain et al., 2007; Safai et al., 2007; Nair et al., 2007; Sreekanth et al., 2007; Badarinath et al., 2009; Beegum et al., 2009) have reported on the characteristics of BC over the Indian region.

In this paper, the author reports the results of BC aerosol mass concentration obtained near the surface level during the months from August 2006 to July 2007 for the first time at Anantapur. A diurnal and seasonal variation of BC aerosol, simultaneous with total aerosol mass concentration derived using a 10-channel Quartz Crystal Microbalance (QCM) impactor is discussed along with the variations associated in local meteorological parameters and column aerosol optical depth. The mass fraction of BC, its annual variation, and their possible implications are discussed. The detailed summary of the data base (total number of days and number of measurements) with instruments is shown in Table 1.

2. Study area and synoptic meteorology

The actual measurements are carried out at the Department of Physics, Sri Krishnadevaraya University (SKU, 14.62°N, 77.65°E, 331 m asl), situated at the southern edge of Anantapur town. A site map is provided in Fig. 1, where locations of relevance are marked. The location of Anantapur (ATP) on peninsular Indian subcontinent is shown in the inset. Anantapur represents a very dry continental region of Andhra Pradesh, India. It is geographically situated on the boundary of a semi-arid and rain shadow region. The climate here is primarily hot and dry in the summer (March to May), hot and humid during the monsoon and post monsoon (June to November) and dry in winter season (December to February). This region receives very little rainfall, and the average annual rainfall is of the order of 450 mm during the whole year (about 300 mm in southwest monsoon and 150 mm in the northeast monsoon periods). Within a 50 km radius, this region is surrounded by a number of cement plants, lime kilns, slab polishing and brick making units. These industries, the national highways (NH 7 and NH 205) and the town area are situated in the north to southwest side of the sampling site (Fig. 1) (Kumar et al., 2010).

Table 1
Summary of the database.

Instruments operated	Parameters measured	No. of days	Total no. of observations
Aethalometer	Black carbon mass concentration	311	89,879
QCM	Size segregated and total mass concentration	40	348
MWR	Aerosol optical depths	161	189

QCM: Quartz Crystal Microbalance.
MWR: Multiwavelength Solar Radiometer.

The surface flow patterns obtained from NCEP/NCAR reanalysis (http://www.cdc.noaa.gov) have been used to ascertain the synoptic meteorological conditions and source variability to the observation site. Fig. 2 shows the synoptic pattern of monthly mean surface wind speeds during the months of August 2006 (Fig. 2a) and January 2007 (Fig. 2b) respectively over the Indian subcontinent. During August which represents the southwest monsoon season of June–September, the winds are stronger, moist, and are from the marine and western regions surrounding India. During October, wind patterns start shifting in direction from southwest to northeast. By November, when the northeast monsoon sets in, the winds are entirely from north/northeast. During northeast monsoon season (November–March), the winds are calm, north/northeasterly and are from the polluted northern regions surrounding India.

3. Instrument, attenuation cross section, and uncertainties

Black carbon aerosol mass concentration measurements began in Anantapur from August 2006 using a two channel aethalometer (AE-21 of Magee Scientific, USA). AE-21 measures BC mass concentrations at two wavelengths 370 and 880 nm. The measurements are made from an altitude of about 12 m above the ground using its inlet tube and pump. The BC mass concentrations are estimated using the optical method of measurement of the attenuation of a beam of light transmitted through the sample collected on a filter, which is proportional to the amount of BC mass loading in the filter deposit (Hansen et al., 1982, 1984). Aethalometer was operated with a flow rate of $3\,l\,min^{-1}$ at a time resolution of 5 min, round the clock. The BC measured at 880 nm wavelength is considered to represent a true measure of BC in the atmosphere as at this wavelength BC is the principal absorber of light while the other aerosol components have negligible absorption at this wavelength (Bodhaine, 1995). The measured location is a suburban and semi-arid station and is dominated by local sources, mainly anthropogenic. As at 880 nm the major absorbing species is BC, in this work, BC mass concentrations measured at 880 nm from August 2006 to July 2007 in SKU campus analyzed and reported. More details on the instrument and the principle of operation are given elsewhere (Hansen et al., 1984; Wiengartner et al., 2003).

This is a filter based technique that measures the light attenuation due to particles deposited onto a filter. The yielded attenuation absorption coefficient is then converted into BC mass concentration. The conversion of attenuation

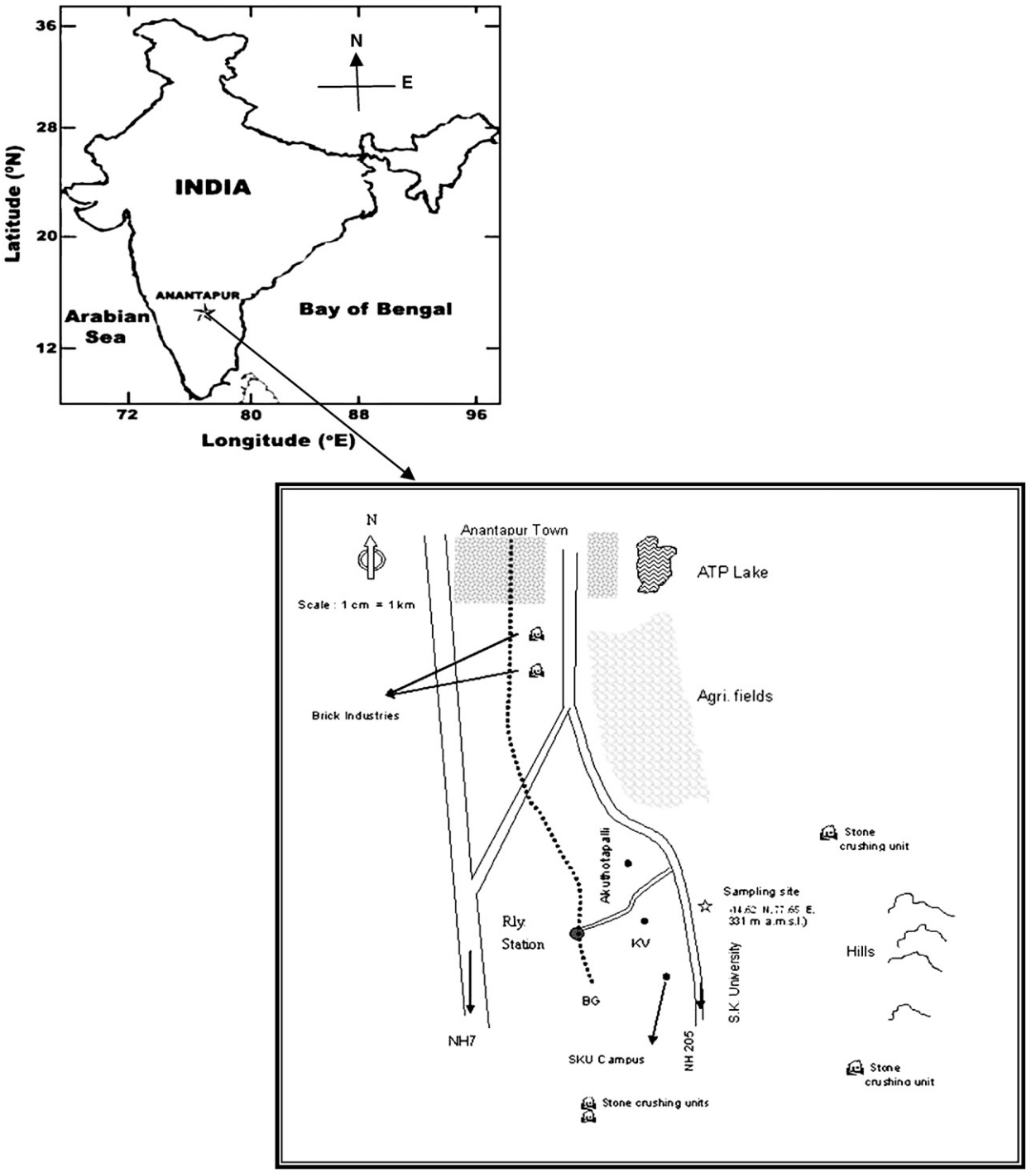

Fig. 1. Schematic site map of the sampling site Sri Krishnadevaraya University (SKU) (☆) showing the details mentioned in the text. Also shown is the position of the station (Anantapur) over the Indian peninsula, in the inset. Sampling site is the place where the observations are made.

absorption coefficient into BC mass concentration is done using appropriate absorption efficiency values. The absorption efficiency varies as a function of wavelength. The light transmission is detected using a set of two photo diodes, one through the sample sport and the other through a blank or an unsampled portion of the filter which is called the reference spot. The change in attenuation is obtained as a function of time and is related to the BC mass concentration in $\mu g\ m^{-3}$ as

$$\text{Black carbon mass} = \frac{-A100ln\left(\frac{I_2}{I_1}\right)}{KQ(t_2-t_1)} \quad (1)$$

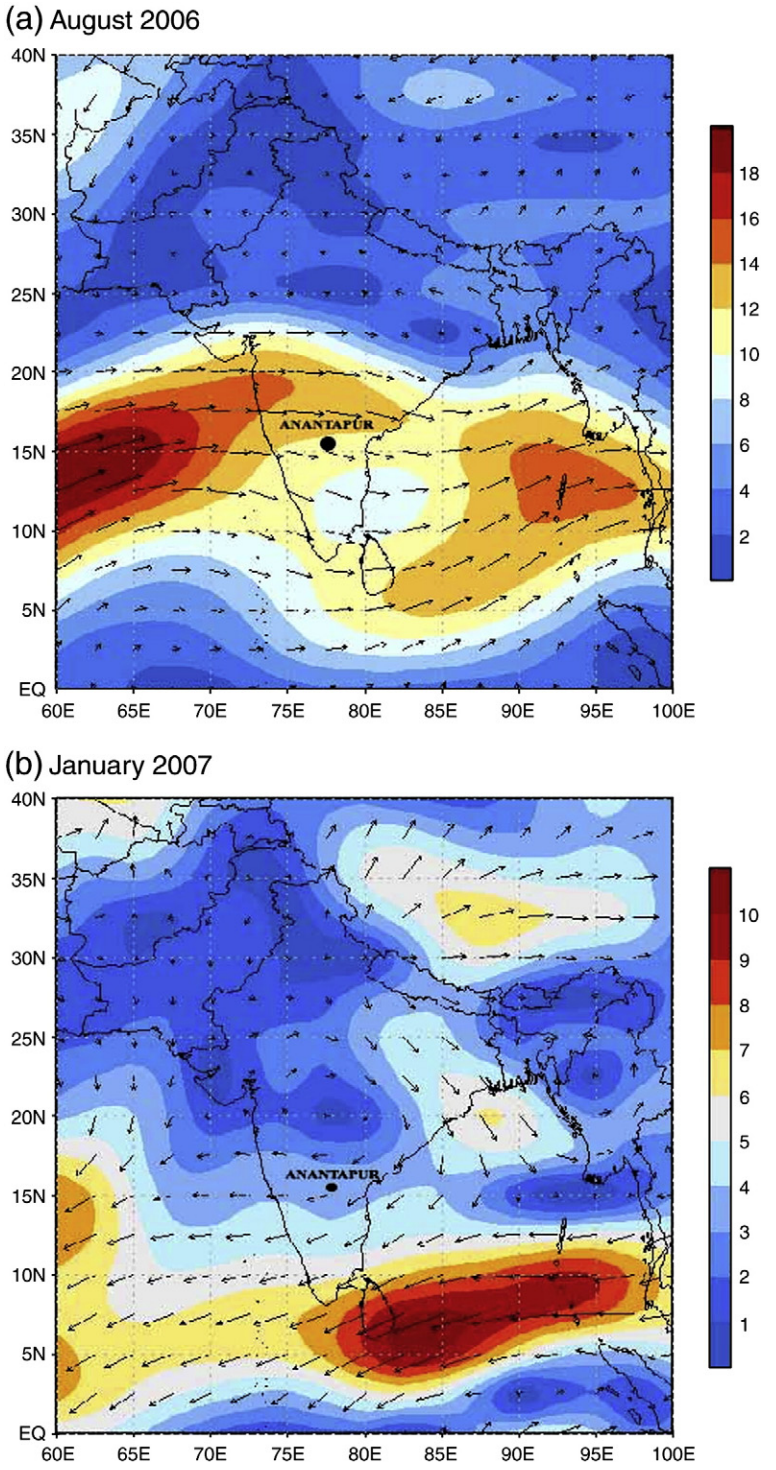

Fig. 2. Synoptic pattern of monthly mean surface winds over Anantapur for two typical months (a) August 2006 (southwest summer monsoon) and (b) January 2007 (northeast winter monsoon).

where I_1 and I_2 are the ratios of the intensities of sample beam to the reference beam at times t_1 and t_2 (seconds) respectively, Q in $m^3\ s^{-1}$ is the sample flow rate through the filter, A is the area of the exposed spot on the filter and is in units of m^2 and K ($m^2\ g^{-1}$) is referred to as the absorption efficiency.

Two values of spectral absorption efficiencies for aethalometer based BC measurements are provided by the manufacturers of aethalometers, namely, Magee Scientific. The "Classic BC calibration" by Magee Scientific at 880 nm is given as 16.6 $m^2 g^{-1}$, while the calibration factor derived using

thermal/oxidation reflectance (TOR) technique was found to be 12.6 $m^2 g^{-1}$. These absorption efficiencies were derived by operating aethalometers and quartz fiber filter samples side by side in Harvard School of Public Health for several years. It was found that the EC (elemental carbon) measured by TOR thermal analyses tracked well the aethalometer BC data but the numbers were higher. When particles are collected on a filter paper, EC is the material that remains after heating the filter to 700 °C, while BC is the part of the particulate matter that absorbs light. It was reasoned that the higher EC concentration indicates either BC is a subset of EC or alternatively the TOR method produces higher EC. It should be mentioned that most of the measurements treat EC as equal to BC. In addition to the above calibration, aethalometer measured BC mass concentrations have been compared with several other instruments such as Particle Soot Absorption Photometer (PSAP), laser integrating plate method, thermal EC analyzer etc., and the comparisons were found to be good (http://www.mageesci.com/book/aethalometer-book-2005.07.03.pdf).

Several investigations in different environments have been conducted using aethalometer and other analytical techniques to calibrate and obtain site specific absorption efficiencies for aethalometer, including Martins et al. (1998), Allen et al. (1999), Ballach et al. (2001), Sharma et al. (2002), and Clarke et al. (2004), to name a few. It has been shown that the absorption efficiency which relates particle light absorption to the BC measured with an aethalometer changes with age, type, and composition of the aerosol, which varies with time and space (Liousse et al., 1993; Lavanchy et al., 1999). Sharma et al. (2002) found that the absorption efficiency showed variations, from remote continental to rural, urban, suburban locations and also from summer to winter. Aethalometer and PSAP were found to agree well with each other (Sharma et al., 2002). The range of median site specific absorption efficiencies observed varied from a low of 6.4 $m^2 g^{-1}$ to 20.1 $m^2 g^{-1}$ for the BC mass concentration measurements made at different locations in Canada. Sharma et al. (2002) suggested that the variability is related to the distribution of sources and processes contributing to the carbonaceous aerosols at different sites. Absorption efficiencies were found to be in the range of 9.9 $m^2 g^{-1}$ to 19.5 $m^2 g^{-1}$ for the measurements made over Munich and Berlin (Petzold et al., 1997). The measurements in Munich and Berlin were conducted in residential areas without high traffic impact and in the immediate vicinity of main streets.

Martins et al. (1998) determined black carbon mass absorption efficiency of smoke particles for various types of biomass fires during the Smoke, Clouds, and Radiation-Brazil (SCAR-B) experiment from thermal evolution measurements for black carbon and optical absorption methods. The efficiency values ranged between 5.2 and 19.3 $m^2 g^{-1}$ with an average value of 12.1 ± 4.0 $m^2 g^{-1}$. It was noted that unrealistically high values of black carbon efficiencies were linked to high concentrations of K, which in turn influenced the volatilization of BC at lower temperatures than usual, leading to possible artifacts in the determination of BC by thermal technique. Although the aethalometer uses a standard absorption efficiency, the aethalometer measured BC mass concentrations are found to satisfactorily describe the concentration levels and trends in the urban atmosphere (Hansen and Novakov, 1990; Liousse and Cachier, 1992). As the BC mass concentration is inversely proportional to the absorption efficiency, a higher value of absorption efficiency would yield low BC mass concentrations.

It is to be noted that hematite (iron oxide, Fe_2O_3) is the other strong absorber found commonly in atmospheric aerosol (Fialho et al., 2005). It has been pointed out that about 200 times as much hematite (mass) as BC is needed for equivalent absorption (Bodhaine, 1995). It has been pointed out (Bodhaine, 1995) that the optics of aethalometer allows detection of changes in light intensity of 1 in 10^4 which corresponds to a noise level of about 1.5×10^{-8} m^{-1} in absorption coefficient. This translates into 1.5 ng m^{-3} (0.0015 μg m^{-3}) BC for a 1-hour collection period. It is to be noted that the lowest mean BC mass reported in this study is 0.86 μg m^{-3} and the highest is 3.47 μg m^{-3}. These two values are more than an order of magnitude higher than the noise level mass concentration of BC. In this study, an absorption efficiency value of 16.6 $m^2 g^{-1}$ corresponding to 880 nm is used to determine the BC mass concentrations over a suburban location. As the value of absorption efficiency used is either comparable or higher than the absorption efficiencies determined in different environments, the concentrations reported in this study represent the minimum values for BC concentrations (Ramachandran and Rajesh, 2007). The other sources of uncertainty in BC mass concentrations using an aethalometer arise from instrumental artifacts such as flow rate, filter spot area and detector response. The flow rate can be checked by the flow meter and value at which the center of the float lies. This has been checked on a daily basis during the measurement period and the flow rate was found to be quite stable. Taking into account all these effects, the overall uncertainty in the reported BC mass concentrations is estimated to be about a maximum of 10%.

There are three main uncertainties in BC measurements made with the aethalometer. The first uncertainty pertains to the assumption that the attenuation coefficient, σ in Eq. (1), is constant as the filter darkens during the collection of light absorbing particles. A second uncertainty is related to the influence of particle light scattering. By virtue of the scattering of light by the filter fibers in which the particles become collected, the light transmission measurement is sensitive primarily to the fraction of particle light extinction that is absorption rather than scattering. A third uncertainty is related to whether or not the attenuation coefficient, s, varies in response to changing aerosol properties.

Near-surface, size-segregated, total aerosol mass concentrations (μg m^{-3}) were measured using the Quartz Crystal Microbalance (QCM) cascade impactor (model PC-2, California Measurements Inc., USA), which provides real time measurements in 10 size bins, with 50% cut off diameters namely, >25 μm, 12.5, 6.4, 3.2, 1.6, 0.8, 0.4, 0.2, 0.1 and 0.05 μm for stages 1 to 10, respectively. Its pump aspirates the ambient air at a flow rate of 0.24 l min^{-1}. The QCM provides mass concentration of the particles collected in each stage (m_{ci}) as a function of particle diameter assuming a value of 2000 kg m^{-3} for ρ. Accordingly, it yields mass concentration in ten size bins; the 50% cut-off diameter and the geometric mean diameter of these bins are given in Table 2. Stage 1 collects all particles with diameter >25 μm, and hence no mean diameter is assigned to that stage (Pillai and Moorthy, 2001; Kumar et al., 2009b).

Table 2
Stage and cut points of QCM impactor.

Stage no.	Lower cut off diameter (μm)		
	Particle diameter (d_{pi})	Geometric mean diameter (d_{gi})	Aerodynamic diameter (d_{ai})
1	25.0	–	35.37
2	12.5	17.58	17.68
3	6.4	8.94	9.05
4	3.2	4.53	4.53
5	1.6	2.26	2.26
6	0.8	1.13	1.13
7	0.4	0.57	0.57
8	0.2	0.28	0.28
9	0.1	0.14	0.14
10	0.15	0.17	0.07

The instrument is operated from the top of the building of height ~12 m from the ground level and during periods when RH<80%. Below 80% RH, the instrument is not very susceptible to the moisture (Reddy et al., 2007). The dual unsealed crystal approach used in the model PC-2 reduces the thermal and humidity effects to a minimum. The optimum sampling time is taken as the time which is sufficient to impart a frequency change of 20 to 30 Hz with a tolerance of −5 to +10 Hz (Madhavan et al., 2008). So the typical sampling duration was kept as 5 to 6 min. On average, an aerosol sample was collected every 8th day over SKU (mostly on every Wednesday of the week) with nine to ten samples are taken on each day. A total of 270 samples for 27 days collected during August 2006–July 2007 at SKU formed the database for this investigation. Due to frequent occurrence of rains and RH>80% prevailing over the study region, there is lack of data from June to August 2007.

The aerosol optical depth (AOD) measurements were made using a Multiwavelength solar radiometer (MWR) designed and developed at Space Physics Laboratory following the principle of filter wheel radiometers. The MWR measures the field limited direct solar flux at 10 different channels centered at 380, 400, 450, 500, 600, 650, 750, 850, 935 and 1025 nm with a full width half maximum bandwidth at the range of 6–10 nm at different wavelengths as a function of solar zenith angle following the Langley plot technique as described in several earlier papers (Moorthy et al., 2003; Saha et al., 2005; Kumar et al., 2009a). Meteorological parameters like air temperature, relative humidity, wind speed, wind direction and rainfall have been measured using Automatic Weather Station (AWS) developed by Astra Microwave Products Ltd., India. The wind speed and temperature sensors operate with an accuracy of +2% of full scale and ±0.1 °C, while the accuracy of RH sensor is +3% of full scale reading.

4. Results and discussion

4.1. Monthly and seasonal variations in BC and FPM aerosol mass concentrations

The annual average BC concentration at Anantapur during August 2006–July 2007 was $1.97 \pm 0.12\ \mu g\ m^{-3}$ which is in good agreement with those reported by Safai et al. (2007) for high altitude rural stations, Sinhagad (located in the Sanhyadri mountain ranges of Western Ghat region) and Nainital (located in the foothills of Western Himalayan ranges) (Table 3). Whereas, this value is much lower as compared to urban and industrial locations like Delhi, Mumbai and Kanpur (Table 3). Fig. 3a shows monthly mean BC aerosol mass concentration during August 2006–July 2007 over the study area. Seasonal variations suggest large concentrations of BC ($\sim 3.5 \pm 0.59\ \mu g\ m^{-3}$) during the winter (November, December, January, and February), followed by those in the summer (March, April and May) and the post-monsoon (September and October) and low BC concentrations ($\lesssim 1 \pm 0.35\ \mu g\ m^{-3}$) during the monsoon months (June, July, and August). In fact, average BC concentrations in winter ($3.31 \pm 0.61\ \mu g\ m^{-3}$) and summer ($2.17 \pm 0.51\ \mu g\ m^{-3}$) were double than that in the post-monsoon ($1.55 \pm 0.35\ \mu g\ m^{-3}$) and monsoon months ($1.04 \pm 0.47\ \mu g\ m^{-3}$). The average BC concentrations were

Table 3
Average BC mass concentrations at different land locations in India.

Location	Environment	Study period	BC concentration ($\mu g\ m^{-3}$)
Trivandrum [a]	Urban Coastal	August 2000 to October 2001	0.3–5
Bangalore [a]	Urban	November 2001	4.2
Hyderabad [b]	Urban	January to July 2003	0.5–68 (dry season) 0.5–45 (wet season)
Mumbai [c]	Urban Industrial	January to March 1999	12.5
Delhi [d]	Urban Industrial	May 2001 to April 2002	17.9 (6.7–27.9)
Agra [e]	Urban	December 2004	20.6 (7.1–48.3)
Kanpur [f]	Urban	December 2004	6–20
Pune [g]	Urban	January to December 2005	4.1
Nainital [h]	Rural (high altitude)	December 2004	1.36
Sinhagad [e]	Rural (high altitude)	November 2004 to April 2005	1.5
Anantapur [i]	Rural (semi-arid)	August 2006 to July 2007	1.97

[a] Babu et al. (2002).
[b] Latha and Badrinath (2003).
[c] Venkatraman et al. (2005).
[d] Rai et al. (2002).
[e] Unpublished data.
[f] Tripathi et al. (2005).
[g] Safai et al. (2007).
[h] Pant et al. (2006).
[i] Present study.

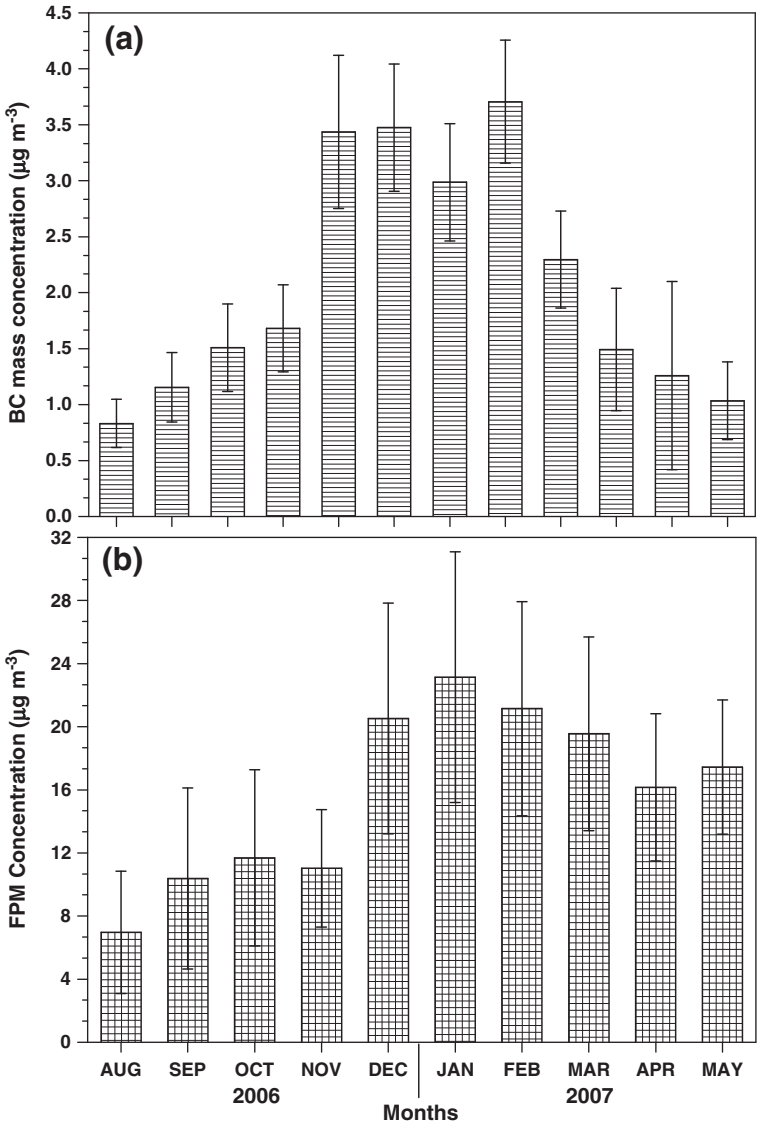

Fig. 3. Monthly mean mass concentrations of (a) BC and (b) fine particle mass (FPM) at Anantapur during the study period with $\pm 1\sigma$ deviation for the mean.

about 60% higher in winter than in summer (about 40%). As the measurement location is not highly industrialized and the seasonal and diurnal variations in BC mass concentrations can mainly be attributed to the synoptic meteorology, biomass burning and transport of airmass from the surrounding regions.

During dry months (December–May) large BC concentrations are associated with large diurnal oscillations in ambient temperature and scanty rainfall. Low BC concentrations occur during the monsoon months (June–November) when the station experiences high rainfall, low temperature and the indicative of the effect of washout of aerosols. This is mainly due to high convective activity and also responsible for the dispersal of aerosols (especially those in fine size). High BC values during the summer months of March and April have been attributed to the transport of airmass from continental regions in addition to an increase in fossil fuel consumption (diesel and petrol) for road transportation (Reddy and Venkataraman, 2002). The monthly mean fine particle mass (FPM) concentration is shown in Fig. 3b with $\pm 1\sigma$ deviation of the mean. It may be noted that the BC mass concentration was consistently 10% of the total fine mode particle mass as measured from a collocated near surface size segregated aerosol mass size distribution using QCM, which indicates that the BC aerosol is well mixed in the ambient fine mode aerosol.

4.2. Season-wise diurnal variations in BC and FPM mass concentration

The season-wise temporal variation of near surface BC mass concentration (top panel) and FPM mass concentration (bottom panel) are shown in Fig. 4, which indicates a significant diurnal variation during the winter month with a morning peak around 07:00–09:00 h with BC (FPM) mass concentration of 8.08 ± 0.92 μg m^{-3} (29.92 ± 2.13), a gradual decrease with minimum BC (FPM) mass concentration of

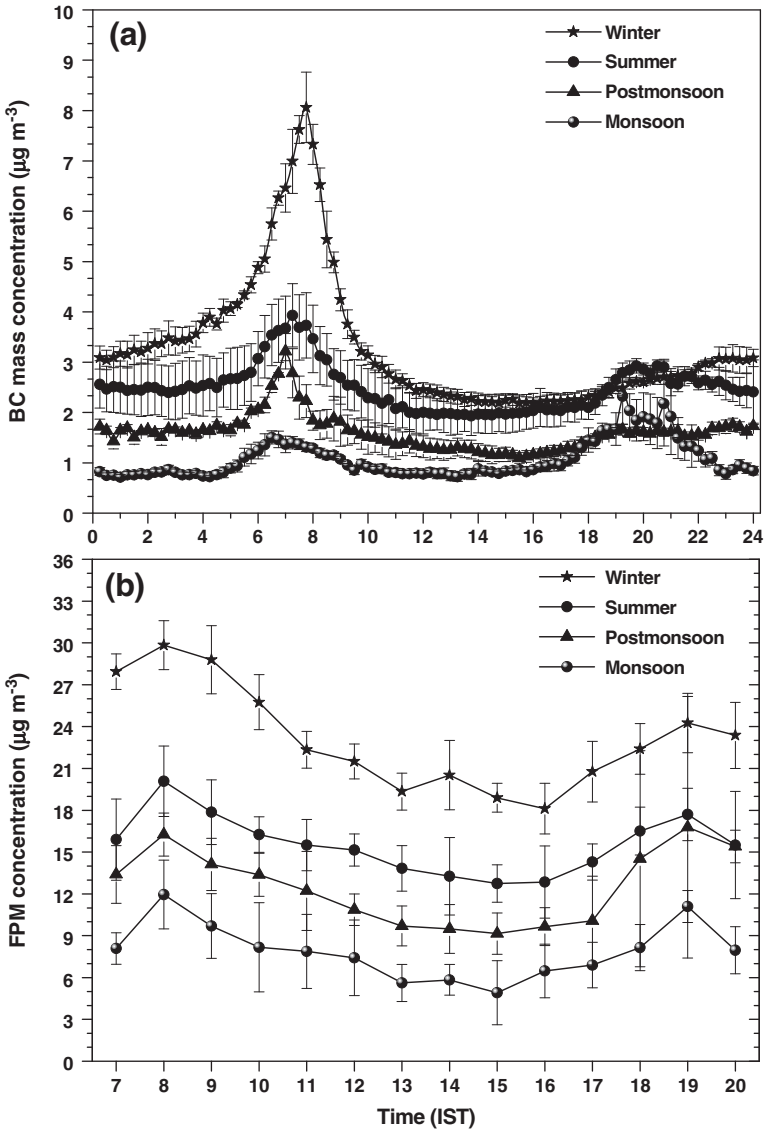

Fig. 4. Season-wise diurnal variations of (a) BC and (b) FPM mass concentrations as a function of time with $\pm 1\sigma$ deviation for the study period August 2006–July 2007.

$2.53 \pm 0.14\ \mu g\ m^{-3}$ (19.24 ± 2.31) during the afternoon hours at 16:00 h followed by an increasing trend after the local sunset with a nocturnal peak of $3.22 \pm 0.22\ \mu g\ m^{-3}$ (24.89 ± 1.83) at 19:00–22:00 h. However, the evening peak was not as dominant as the morning one. It is pertinent to mention that the second peak is slowly increasing from March to June in summer months and starts decreasing from July onwards. The diurnal mean BC mass concentration was around $2.89 \pm 0.24\ \mu g\ m^{-3}$ in winter, $2.50 \pm 0.22\ \mu g\ m^{-3}$ in summer, $1.33 \pm 0.12\ \mu g\ m^{-3}$ in post-monsoon and $1.04 \pm 0.11\ \mu g\ m^{-3}$ during the monsoon season. The prominence of this type of diurnal variation as well as the magnitude of BC mass concentration decreases during the summer months with morning peak value of $3.93 \pm 0.29\ \mu g\ m^{-3}$, noon value of $1.93 \pm 0.13\ \mu g\ m^{-3}$ and nocturnal peak value of $2.93 \pm 0.24\ \mu g\ m^{-3}$. The ambient BC mass concentrations were low in monsoon months and during the post-monsoon months practically no diurnal variability was observed in the near surface BC mass concentration.

Aerosol concentration is affected by the stability of boundary layer, which is active during the daytime due to surface temperature increase and stable at night (Stull, 1998). It has been shown that the nocturnal boundary layer is shallower than its daytime counterpart by a factor of about 3 (Kunhikrishnan et al., 1993). Also, as the wind speeds, in general, are lower during night the ventilation coefficient rapidly reduces, resulting in the confinement of aerosols during nighttime. This results in an increase in the BC mass concentration during early nighttime (Babu and Moorthy, 2002). The BC mass concentration reduces as the night advances due to reduction in anthropogenic activities and loss of particles closer to the surface by sedimentation. The gradual buildup of the BC mass concentration from the morning hours and the occurrence of peak between 07:00

and 09:00 h about an hour after the local sunrise during both winter and summer are attributed to the combined effects of fumigation effect of the boundary layer and morning increase in the anthropogenic activities in a rural environment. The fumigation effect in the boundary layer brings in aerosols from the nocturnal residual boundary layer in a short time after the sunrise (Stull, 1998). The increased solar heating as the day advances produces a deeper and more turbulent boundary layer which leads to a faster dispersion resulting in a dilution of BC near the surface (Babu and Moorthy, 2002).

4.3. Mass fraction of BC

The mass mixing ratio (F_{BC}) of BC to the composite aerosol is an important parameter in aerosol radiative forcing estimation (Babu et al., 2004). Even though BC contributes only a few percent (5–15%) to the total aerosol mass, it can produce significant radiative effects. At SKU we had simultaneous measurements of FPM aerosol mass loading from total mass concentration obtained using the QCM particle analyzer in conjunction with that of BC mass concentration for the period from August 2006 to July 2007 to understand the BC contribution in total fine particle mass concentration over the study area. The month-to-month variation of this fraction is shown in Fig. 5. The ratio of BC to total mass concentration is very high (>10%) during the winter months (December to February), 5 to 7% in summer months (March to May) and lowest (2 to 4%) during monsoon months (June to September). Analysis of FPM mass concentration and BC aerosol mass concentration suggests that the share of BC to FPM mass concentration observed to be ~15%. The results are in agreement with other studies reported in literature (Hess et al., 1998; Podgorny et al., 2000; Babu and Moorthy, 2002; Satheesh, 2002). Such large share of BC can have serious implications on surface and atmospheric radiative forcing Haywood and Shine (1997). A mere 6% of soot contributes 11% to the optical depth (Allen et al., 1999; Satheesh, 2002) a 35% reduction in total solar radiation over the ocean surface and an increase of ~50% in atmospheric heating (Podgorny et al., 2000). Over land the impact would be higher (Haywood and Shine, 1997).

Here a mention of the uncertainty in the derived BC mass-fraction is warranted. Extensive field studies using the aethalometer for BC measurements and their intercomparison with the other analytical techniques like thermal or thermal-optical analysis (TOA) methods or filter based light transmission methods have yielded excellent agreement (Kirchstetter and Novakov, 2007; Watson et al., 2005; Allen et al., 1999). Considering these along with our operational configuration of the aethalometer, we attribute an uncertainty of 10% (with a minimum of 100 ng m^{-3}) for the BC mass. This is also comparable to the uncertainty in the FPM mass concentration estimates using the QCM. Considering the uncertainties in the QCM measurements along with those of M_b, the overall uncertainties in the mass fraction would be ~15% (Babu and Moorthy, 2002). For the experimental configuration used, this fraction gives the fraction of BC that resides within the size regime of 2.5 μm. If we consider that the entire BC is confined to the submicrometer size and obtain the ratio of the measured M_b to the composite mass for the size bins 8, 9, and 10 of the QCM (which corresponds to sizes below 0.8 μm) then the mass fraction works out to ~10% (Moorthy and Babu, 2006).

4.4. Effect of wind speed on BC mass concentration

An attempt has been made to investigate the possible relation of BC to the meteorological parameters. Fig. 6 shows monthly average values for BC mass concentration versus wind speed and direction. Black carbon mass concentrations

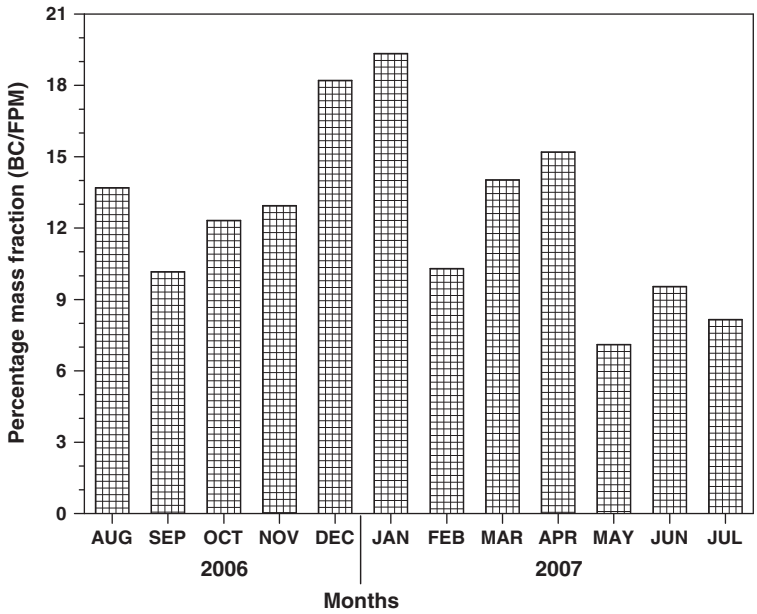

Fig. 5. Annual variation of BC to FPM aerosol mass concentration fraction during August 2006–July 2007.

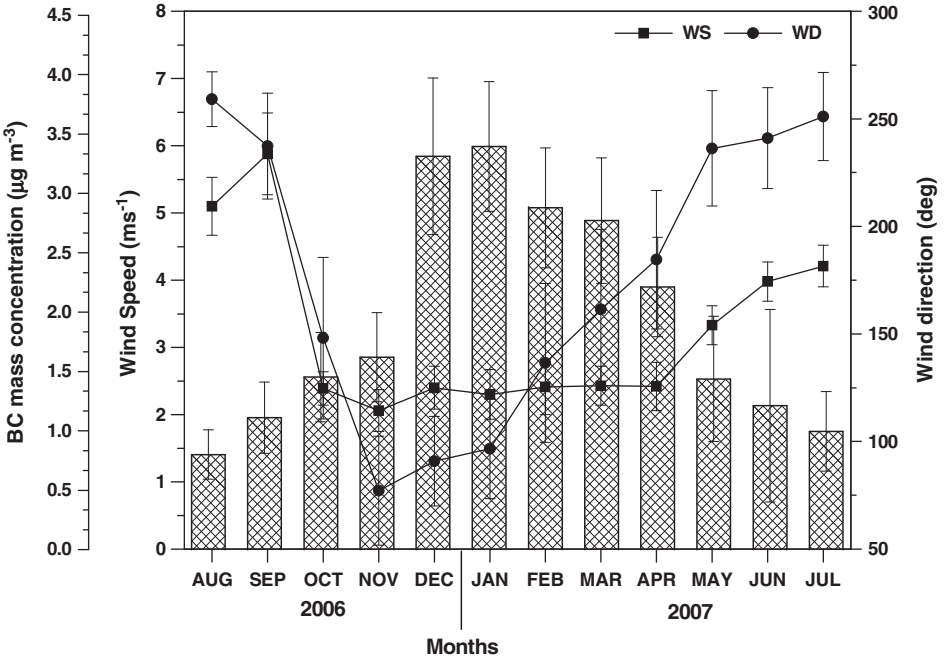

Fig. 6. Annual variations of BC concentration versus wind speed (WS) and wind direction (WD) at Anantapur for the study period. The vertical bars over the measurement points represent the standard error.

showed inverse relation with wind speed. An examination of the annual variation of monthly mean surface winds (Fig. 6) shows that during the months of July and August, the average winds were higher (5–6 m s^{-1}) compared to the other months. Hence the large decrease in the BC concentrations from May to October and during August can be very well attributed to the prevailing high wind speeds (5.4 m s^{-1}) (Saha and Despiau, 2009). The increase in wind speed causes an increase in the ventilation effects, thereby dispersing the aerosols in the ambient air and consequently causes a decrease in the observed BC concentrations.

Although there is a tendency of high BC concentrations associated with high wind speed, this association may be coincidental (as during daytime, both higher wind speed and more traffic density may occur), and moreover, high BC concentrations are not visible during all occurrences of high wind speeds. It is probable that due to higher wind speeds, BC produced from local sources are transported to other locations (Ramachandran and Rajesh, 2007). Similarly high shifts of mean wind speed values did not produce significant comparable shifts in BC concentrations. Increased wind speed leads faster dispersion and hence a dilution of BC nears the surface and vice-versa. Similar observations have been made in other studies in literature (Bhugwant et al., 2000). It was reasoned that a clear correlation between wind speeds and BC mass concentrations gives an indication of the proximity of BC sources at the measurement site, while a not so significant correlation shows that the BC originates from distant sources. It is observed that during high BC concentrations, wind patterns start shifting in direction from north easterlies to southwesterly (Fig. 2) where possible sources of BC are generated from agricultural burning, automobile exhaust and are concentrated near the measurement site (not transported to the other locations) due to low wind speeds (~1.2 m s^{-1}).

4.5. Regression analysis of BC mass concentration versus wind speed

As we know, wind speed is an important factor in affecting the daily variations of BC. The monthly average diurnal variations of BC mass concentration ([BC]) and wind speed for two typical seasons, summer and monsoon are shown in Fig. 7. The increase in wind speed causes an increase in the ventilation effects, thereby dispersing the aerosols in the ambient air and consequently causes a decrease in the observed BC concentrations. It is probable that due to higher wind speeds BC produced from local sources in Anantapur is transported to other locations. It was found from BC measurements made in urban sites in Ahmedabad (Ramachandran and Rajesh, 2007) and Toranto (Sharma et al., 2002) that low wind speeds lead to much less dispersion of BC and the traffic emissions remain concentrated around the emission site. We examine the relationship between the daily averaged BC mass concentrations and the prevailing wind speeds for the two respective periods: (1) November to February (winter), and (2) March to May (summer), which is shown in Fig. 8. Notwithstanding a fair amount of scatter, BC concentration decreases with increase in the wind speed observed in both cases. The correlation is better for the winter data ($R^2 = -0.72$), as compared to the later period (-0.63). Further the slope and intercept for the winter data were -1.18 and 5.78 (Fig. 8a), respectively and for the later period, these were -1.26 and 5.03 (Fig. 8b). The steeper slope and the higher intercept value for the winter data can be mainly attributed to the high BC concentrations observed, which are typical to the season. These fitting parameters indicate that with increase in wind speeds the BC mass concentrations would decrease as seen. A clear negative correlation between BC mass concentration and wind speed at Evans Avenue (urban, $R^2 = -0.77$) and Downsview

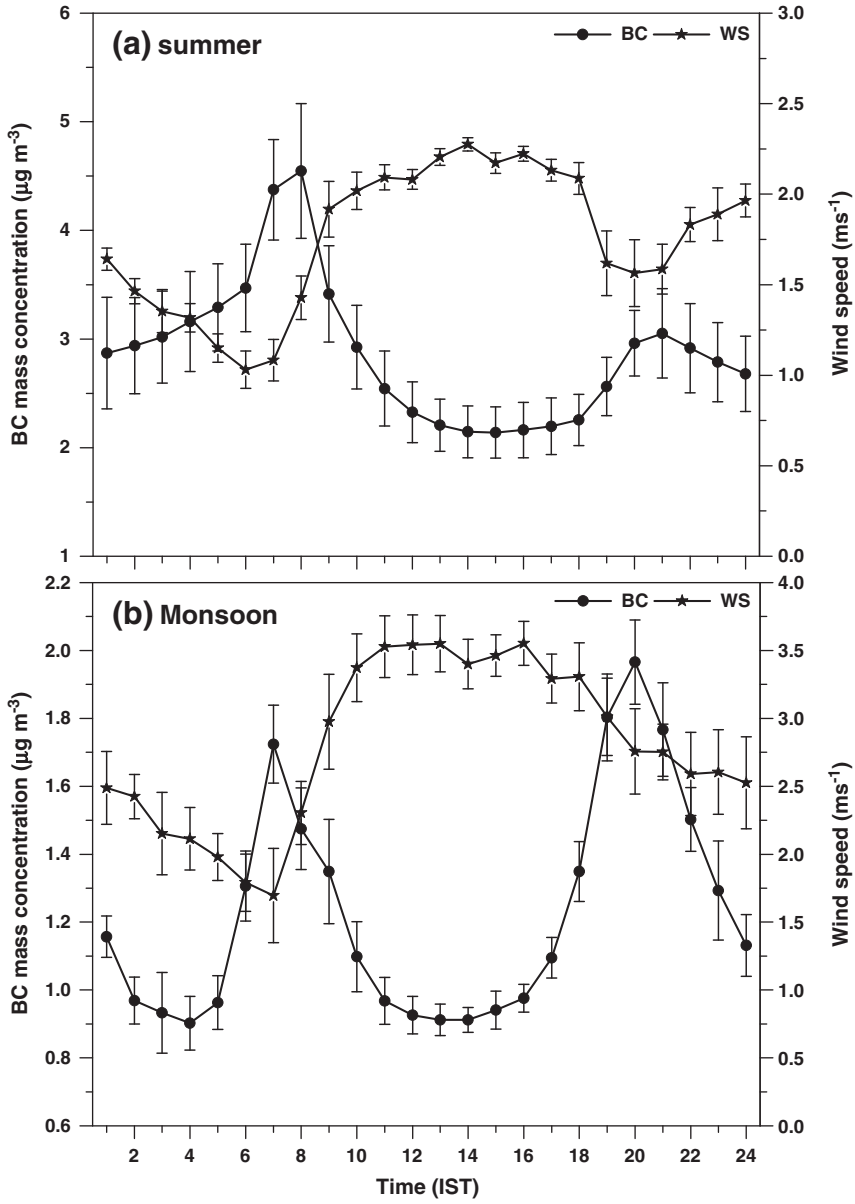

Fig. 7. Monthly average diurnal variation of BC mass concentration and wind speed for two typical seasons (a) summer and (b) monsoon during the entire study period.

(suburban, $R^2 = -0.64$) was found while in Egbert, a rural location ($R^2 = -0.1$) no clear pattern was found (Sharma et al., 2002). It was reasoned that a clear correlation between wind speeds and BC mass concentrations gives an indication of the proximity of BC sources at the measurement site, while a not so significant correlation shows that the BC originates from distant sources.

4.6. Back trajectory analysis

As our study region is not industrialized, the sources of BC would mainly be automobile exhaust and local domestic activities combined with BC advected from the inland regions. There are no combustion sources that are seasonal in nature. The advection, of course, has strong seasonal dependence as seen in earlier chapters. Several forest fires of moderate strength occur every year during the winter period over the forest land lying a few hundred kilometers north-east (upwind) of Anantapur, mainly due to the dry prevailing conditions and it is likely that a part of these emissions would be advected to this location by the favorable wind direction, during this season. To examine the role of long-range transport of BC to the measurement site, from the adjoining regions, 7-day airmass back trajectories were computed using Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT, http://www.arl.noaa.gov/ready/hysplit4.html) model of National Oceanic and Atmospheric Administration (NOAA) (Draxler and Rolph, 2003). Mass plots of these trajectories (arriving at 500 m a.g.l.

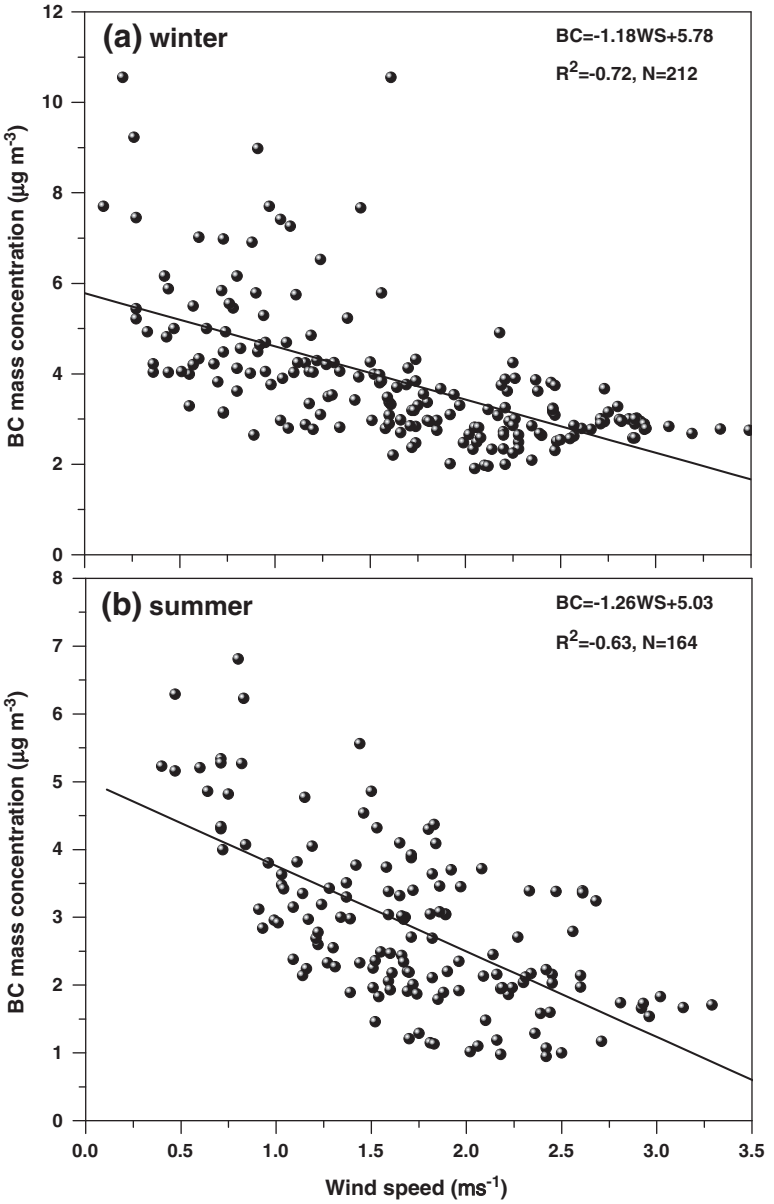

Fig. 8. Scatter plot for the BC mass concentration showing negative correlation with wind speed for two different seasons during the study period.

(above ground level) at SKU) for each season are shown in Fig. 9. In the figure, panels (a) and (c) show the contrasting seasons winter and monsoon seasons, while (b) and (d) show the periods of transition, summer and postmonsoon seasons. It can be seen that during winter season, when the [BC] values are high, most of the trajectories show advection either from the Indo-Gangetic plain (IGP) or from the East Asia across the Bay of Bengal. There is some significant advection from the west coast of India and Arabia also. In sharp contrast to this, during monsoon season all the trajectories are of oceanic nature and reach the observation site without any continental overpass. As such, [BC] values are the lowest of the year during this season.

During the transition periods, the trajectories show different types of advection with consequent impact on [BC]. During summer (transition from winter to monsoon), Fig. 9b shows that majority of the trajectories have a long history over the ocean and only less than 30% of the trajectories have a continental or close to continental history (either West Asia, east Coastal India or over Sri Lanka). Unlike this, in post-monsoon season we have significant advection from the IGP, central peninsular India, west coast of India and east coast of Africa. Only less than 25% of the trajectories are purely oceanic in nature. This results in seasonal mean BC during postmonsoon being significantly higher than that during summer, even though both are transition periods. Similar effects of long range transport of aerosols from the IGP to the east coast of India have been reported by Niranjan et al. (2004) and at Ahmedabad in western part of India by Ganguly et al. (2006). Corrigan et al. (2007) and Kaskaoutis et al. (2009) have also reported the impact of continental advection on aerosol concentration at

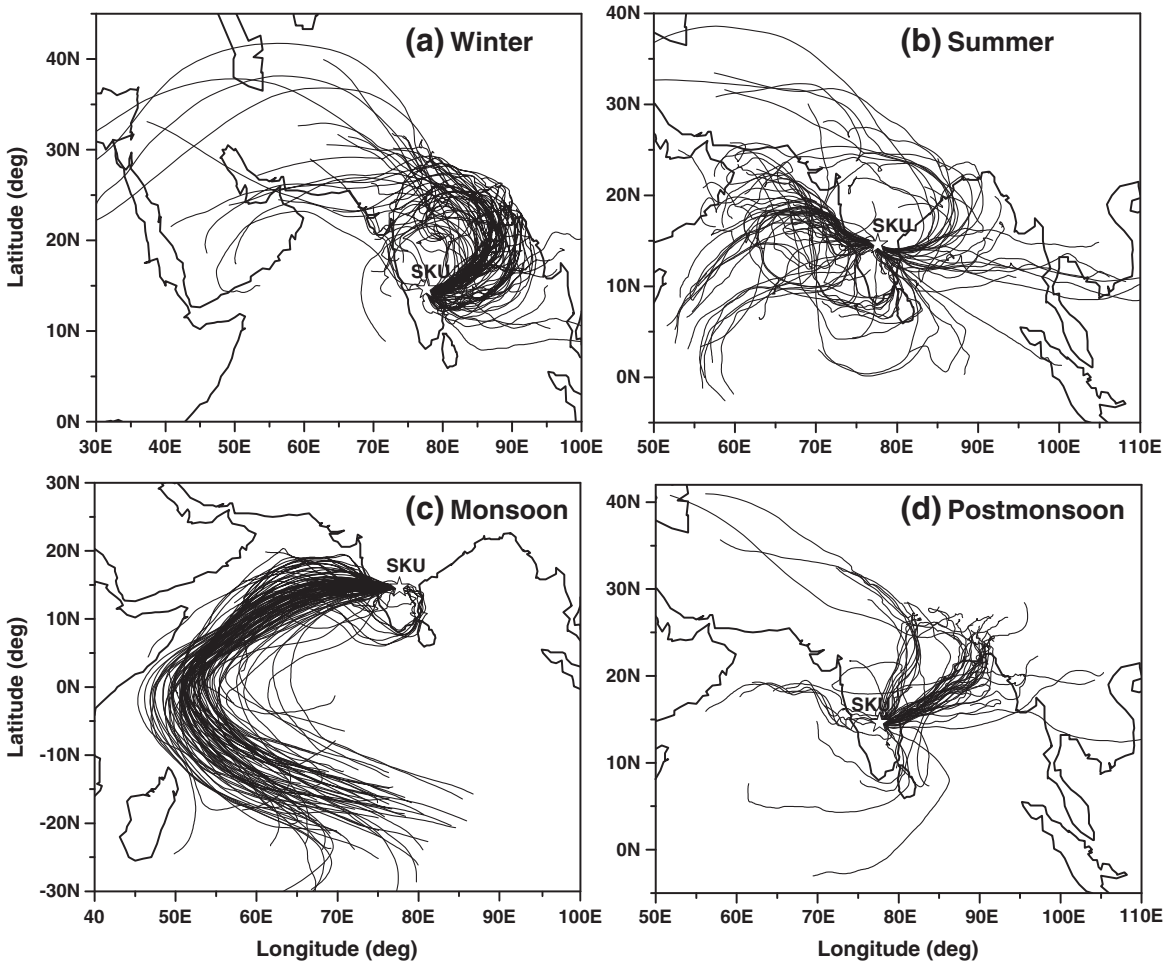

Fig. 9. Airmass plots of the back trajectories (arriving at 500 m a.g.l. (above ground level) at SKU) for each season. Panels (a) and (c) show the contrasting seasons, winter and monsoon, while (b) and (d) show the periods of transition, summer and postmonsoon.

Maldives and Hyderabad, respectively. All these indicate that long-range transport has an important role in determining the BC concentration at Anantapur.

4.7. Scatter plot of black carbon mass concentration with aerosol optical depth and total mass concentration

A scatter diagram of simultaneous estimates of aerosol optical depth (AOD) (τ_p) at 500 nm and surface black carbon (BC) mass concentration (M_b) is shown in Fig. 10 (top panel). Notwithstanding the fair amount of scatter (which is expected as BC is not the only contributor to τ_p), τ_p is found to increase with M_b. Correlation between variations in AOD at 500 nm and black carbon aerosols is observed to be positive ($R^2 = 0.62$) and becomes poor at high BC concentrations. The correlation coefficient is not very high because of other contributors to AOD. It may be noted that AOD measurements were limited due to cloudy sky conditions. Even though τ_p is aerosol column optical depth and BC is surface mass concentration, both can be correlated as τ_p measurements were carried out during day time. This excludes aerosol layers aloft, so that the variation in the surface can be taken as representative of the variations in the column.

In the light of the large share of BC to total fine particle mode mass concentration discussed in Section 4.3 and the positive correlation between τ_p and M_b, we examined the correlation between [BC] to total mass concentration. This is very important, particularly over land, as abundance of BC would change the sign of forcing (Haywood and Shine, 1997). A scatter plot of M_b against M_t using the simultaneous data and is shown in Fig. 10 (bottom panel). This database had 171 pairs of simultaneous data. The slope of the least square fitted line was used to estimate mean mass fraction for this period, which was found to be $7.9 \pm 0.2\%$. A significant positive correlation ($R^2 = 0.72$) is noticeable and also that BC is only one of the several possible constituents of atmospheric aerosols.

5. Conclusions

Black carbon aerosol mass concentration measurements made for a one year period from August 2006 to July 2007 over Anantapur, a semi-arid rural area located in southern India are analyzed. The methodology of [BC] measurement

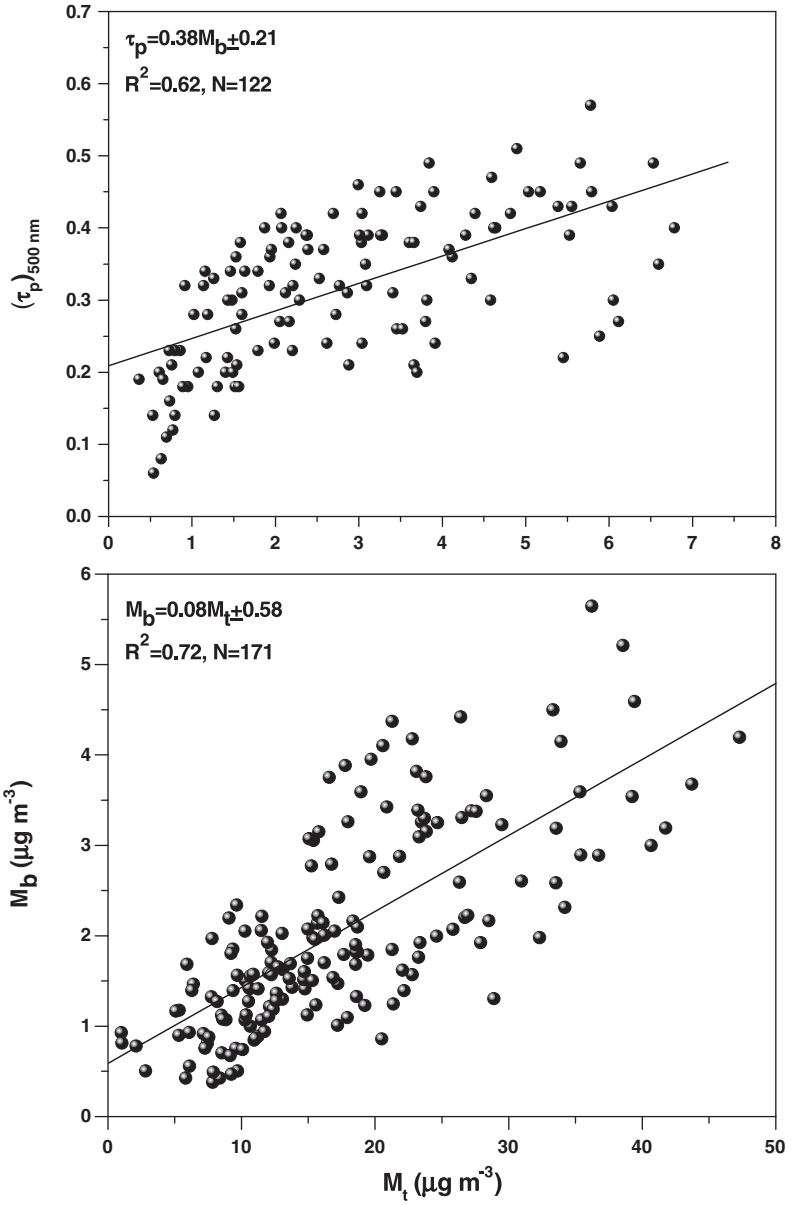

Fig. 10. Scatter diagram of BC mass concentration (M_b) versus (top panel) AOD ($\tau_{p\lambda}$) at 500 nm and (bottom panel) aerosol total mass concentration (M_t).

using an aethalometer is discussed in detail. The uncertainties in the measured [BC] are discussed. An absorption efficiency of 16.6 $m^2\ g^{-1}$ is used to convert the measured attenuation into BC mass concentrations. Seasonal variations of BC aerosol mass concentration showed high concentrations during dry (winter) season and low concentrations ($\lesssim$1 $\mu g\ m^{-3}$) during the monsoon season.

BC mass concentrations are found to show diurnal variations with two peaks, one between 07:00 and 09:00 h and the other between 19:00 and 21:00 h. The boundary layer is shallower during night than day and as the wind speeds are lower the ventilation coefficient rapidly reduces during night leading to confinement of aerosols in the night. This results in an increase in [BC] in the night. As the night advances BC mass concentration reduces due to both reduction in anthropogenic activities and loss of particles closer to the surface by sedimentation. The gradual rise of [BC] in the morning and occurrence of peak between 07:00 and 09:00 h are due to fumigation effect in the boundary layer and increase in the anthropogenic activities. The peaks in the morning and evening hours are attributed to the vehicular combustion arising from the rush hour traffic and the boundary layer dynamics.

BC contributes only a few percent (5–15%) to the total aerosol mass, which can produce significant radiative effects. The ratio of BC to total mass concentration is very high (>10%) during the winter months (December to February), 5 to 7% in summer months (March to May) and lowest (2 to 4%) during monsoon months (June to September). Considering the uncertainties in the QCM measurements along with those

of M_b, the overall uncertainties in the mass fraction would be ~15%. Increased wind speed leads faster dispersion and hence a dilution of BC nears the surface and vice-versa. It is observed that during high BC concentrations, wind patterns start shifting in direction from north easterlies to southwesterly where possible sources of BC are generated from agricultural burning, automobile exhaust and are concentrated near the measurement site (not transported to the other locations) due to low wind speeds (~1.2 m s^{-1}). The regression analysis between [BC] and wind speed indicates that, with increase in wind speeds the BC mass concentrations would decrease and vice-versa. A significant positive correlation (not high) is observed between BC mass concentrations and simultaneous estimates of AOD at 500 nm and total aerosol mass concentration which shows that BC is only one of the several possible constituents of atmospheric aerosols.

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