Potential Source Regions Contributing to Seasonal Variations of Black Carbon Aerosols over Anantapur in Southeast India

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ABSTRACT

Continuous measurements of black carbon (BC) mass concentration performed at Anantapur [14.62°N, 77.65°E, 331 m asl], a suburban location in southeast India, using an Aethalometer from January to December, 2010, are analyzed and discussed here. The annual mean BC mass concentration ([BC]) was 3.03 ± 0.27 µg/m^3 for the above study period. The sharp morning (fumigation) peak occurs between 07:00 and 08:00 h almost an hour after the local sunrise while a broad evening (nocturnal) peak is at ~21:00 h with a minimum in noon hours (14:00–16:00 h). The seasonal mean values of [BC] are 5.05 ± 0.51 µg/m^3 in the winter, 3.77 ± 1.23, 1.55 ± 0.51, and 2.33 ± 0.82 µg/m^3 in the summer, monsoon and postmonsoon seasons, respectively. High BC values tend to occur when the wind is directed from the 180–225° sector, which may be well defined by the geographical location of the observation site. During the winter, the trajectory air mass pathways originated through north or central India with significant advection of continental aerosols arriving before the measurement region, results in an enhanced [BC]. Whereas in the monsoon season, the pristine marine air mass from the oceanic environment led to decrease in the concentration of BC. Comparison of monthly mean variations in AOD at 500 nm and black carbon aerosols is observed to be positive with poor correlation coefficient of 0.42. The ratio of BC/PM$_{2.5}$ varied from 1.3% to 7.2% with a mean value of 4.6% at Anantapur during the observation period and this ratio decreased with decreasing Ångström exponent (alpha).

Keywords: BC aerosol; Potential sources regions; AOD; Ångström exponent; PM$_{2.5}$.

INTRODUCTION

Knowledge of the distribution and sources of Black Carbon (BC) is essential in understanding its impact on radiative forcing and the establishment of a control strategy. A significant fraction of atmospheric aerosols is comprised of carbonaceous materials, which are often categorized as elemental or black/graphitic carbon (EC/BC) and organic carbon (OC). These forms of carbon play an important role in climate change, either contributing to or offsetting atmospheric warming (Hansen and Sato, 2001; Menon *et al.*, 2002). BC has attracted special attention because of its

contribution to radiative heating (Myhre *et al.*, 1998; Jacobson, 2001). The Intergovernmental Panel on Climate Change (IPCC) has estimated that the global mean clear-sky radiative forcing of BC is 0.23 ± 0.25 W/m^2 (IPCC, 2007), which is approximately half the value of Methane, the second most important greenhouse gas after Carbon dioxide. BC aerosols absorb solar radiation and are the second largest contributor to global warming, after greenhouse gases. On a global basis BC contributes ~0.5 W/m^2 to radiative forcing (Jacobson, 2001). BC may also have regional climatic impacts. For example, Menon *et al.* (2002) have suggested that the high concentrations of soot over India and China are responsible for a trend toward increased flooding in the south and drought in the north. It is estimated that the reduced atmospheric transparency caused by high soot concentrations over India and China decreases agricultural productivity by 10–20% (Chameides *et al.*, 1999). Soot deposited on plant leaves also reduces plant productivity (Bergin *et al.*, 2001).

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Black carbon (BC) and elemental carbon (EC) in airborne particulate matter originate from the incomplete combustion of carbonaceous fuel (of fossil fuels, biomass and agricultural wastes and forest fires) (Salako *et al.*, 2012). BC is light absorbing carbon, is one of the key components of atmospheric aerosols that contribute to positive radiative forcing (Ramanathan *et al.*, 2001). Recently, atmospheric BC has received considerable attention because of its significant influence on climate change as well as its adverse effects on human health (Kim *et al.*, 2011). BC is chemically inert and mostly in the accumulation (submicron) size regime, and has a long atmospheric lifetime (of several days to weeks) depending on the meteorological conditions (Reddy and Venkataraman, 1999; Babu and Moorthy, 2002) and hence is amenable for long-range transport (Moorthy and Babu, 2006). The bulk of BC aerosols (~90%) resides in the $PM_{2.5}$ (particulate matter having diameter $\geq$ 2.5 μm) fraction (Viidanoja *et al.*, 2002; Cao *et al.*, 2009). Excessive $PM_{2.5}$ concentrations have been shown to negatively affect the respiratory and cardiac health of humans (e.g., Park *et al.*, 2002) and also reduce visibility. Coal and biomass burning, vehicle exhaust and industrial emissions all contribute to the ambient PM in Anantapur. Therefore, observations for OC and EC analysis are needed for Anantapur (Li *et al.*, 2012).

Observations on BC aerosols are reported mainly from the urban locations in the Indian region (Latha and Badarinath, 2003; Ramachandran and Rajesh, 2007; Safai *et al.*, 2007; Beegum *et al.*, 2009; Moorthy *et al.*, 2009). However, observations from the rural/remote locations are very scarce. In this paper, the author reports the surface BC observations from a tropical semi-arid location in Anantapur, a suburban region in southeast India, during January–December, 2010. This paper highlights the diurnal and seasonal variation of [BC] particularly in relation to changing meteorological conditions throughout the year. Variations of [BC] with AOD and near surface total aerosol mass concentration (Mt) were also discussed. In addition to this, the authors also focused on the possible sources of these aerosols from long range transport which have been studied using backward trajectory analysis.

STUDY AREA, SYNOPTIC PATTERNS AND LOCAL METEOROLOGY

The measurements have been carried out at the Department of Physics, Sri Krishnadevaraya University (SKU) campus; Anantapur [14.62°N, 77.65°E, 331 m above sea level]. Anantapur located in Southeast India represents a very dry semi-arid, rain shadow and continental region of Rayalaseema, Andhra Pradesh, India. Within a 50 km radius, this region is surrounded by a number of cement plants, lime kilns, slab polishing and brick making units (see Fig. 1 bottom panel). These industries, the national highways (NH 44 and NH 205) and the town area are situated in the north to southwest side of the sampling site. The study area is also at a relatively short distance from two major capital cities, about 200 km away from Bangalore [12.96°N, 77.56°E, 949 m asl] located in a direction south of the observation site and approximately 400 km away from Hyderabad [17.36°N,

78.34°E, 536 m asl] situated in a direction northwest of the measurement location. The site of study is located in the southwest part of the city and is close to two highways that surrounds the city. The production of BC is mainly due to the biomass burning together with heavy traffic (mainly diesel vehicles). The old part of the city has rather narrow streets responsible for heavy traffic in some areas of the city, especially at rush hours. During the winter, domestic heating (mainly wood, charcoal and diesel central heating) represents an additional important source of anthropogenic BC aerosols (Kumar *et al.*, 2011). On the other hand, forest fires, agricultural and biomass burning and transport of air masses from Asian desert dust (Jung *et al.*, 2010) represents an additional source of BC.

Fig. 1 shows the monthly mean surface wind speeds during the months of January and July, 2010, respectively over the Indian subcontinent. During January (top panel left side) shown as representative of the wind pattern during northeast monsoon (November–March), the winds are calm, from a north/northeasterly direction and are from the polluted northern hemisphere. In July (top panel right side) 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 the north/northeast.

The continental conditions prevailing at this site are responsible for large seasonal temperature differences, providing hot summers (March–May) and cool winters (December–February). Most of the rainfall occurs during the monsoon (southwest monsoon; June–September) and postmonsoon (northeast monsoon; October and November) seasons. During the year 2010, the annual rainfall in the study area was 805 mm (see Table 1) and had less rainfall when compared to adjoining places in the Rayalaseema region and other parts of the state, Andhra Pradesh. The rainfall during the southwest monsoon period is 619 mm, which forms more than 77% of the total annual rainfall, but only 186 mm for the northeast monsoon period, which forms 23% of annual rainfall for the study period. Daily mean wind speed (WS), wind direction (WD), air temperature (AT), relative humidity (RH) and total rainfall (RF) have been obtained from automatic weather station (AWS) which is installed in the observation site near to the department. The statistical data for the monthly mean prevailing meteorological conditions during the period of study over the experimental site is shown in Table 1. The monthly mean wind speed was noticed maximum during the month of June, which is around 3.4 m/s and minimum in February ~1 m/s followed by January which is about 1.1 m/s. Most of the winds are prevailing from a southwesterly direction (180–250° degrees) during the total observation period. The monthly average maximum (minimum) AT of about 34.2°C (26°C) was recorded in the month of April (August) and maximum RH (minimum) of around 71.0 (27.0%) was observed in August (April).
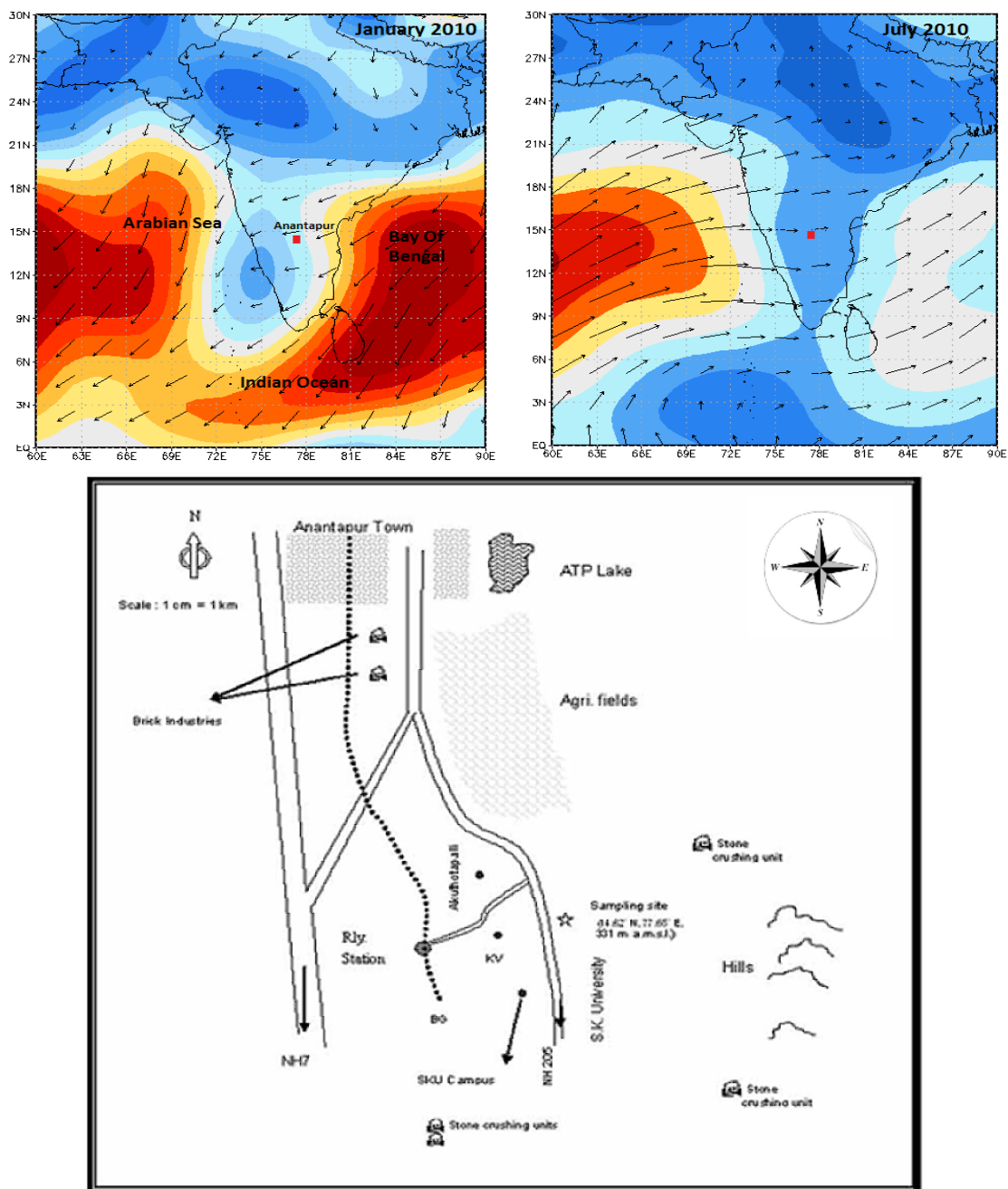
INSTRUMENTATION AND METHODOLOGY

Fig. 1. Monthly mean surface wind fields over Anantapur during two contrasting seasons, January, 2010 and July, 2010 (top panels), for which BC concentration is high and low, respectively. The bottom panel shows the city map of Anantapur showing the measurement location at Sri Krishnadevaraya University (SKU) indicated by ☆ and surrounding sources.

Measurements are carried out at ~12 m above ground level using an Aethalometer (Model AE-21, Magee Scientific, USA), a 10-channel Quartz Crystal Microbalance (QCM) Impactor (Model PC-2 of California Measurements Inc., USA), and a Multi-wavelength Solar Radiometer (MWR) during January–December, 2010. The Aethalometer is portable and easy to mount at any location, which is widely used for the real-time monitoring of BC aerosols in the atmosphere. The aethalometer use continuous filtration and a thermal optical transmission (TOT) technique to measure the mass concentration of BC in near- real- time (Hansen *et*

al., 1984) at two channels (wavelengths) 370 and 880 nm. The wavelength 880 nm represents the broad band wavelength response of the aethalometer detector. The instrument aspirates ambient air through an inlet at a preset flow rate where, the particles in the air impinge on its quartz fiber filter tape. [BC] was estimated by measuring the change in transmittance of this quartz fiber tape consequent to the deposition of the particles on it.

The attenuation (ATN) of the beam is a measure of the absorber mass. The [BC] was obtained from an incremental ATN between two measurements using the effective

Table 1. Statistical data of monthly mean meteorological parameters with ± 1σ variation observed at Anantapur during January–December, 2010.

Month	Wind speed (m/s)	Wind direction (deg)	Air temperature (°C)	Relative humidity (%)	Rainfall (mm)
January	1.1 ± 0.1	198.1 ± 6.0	27.0 ± 0.3	53.2 ± 1.4	22.0
February	1.0 ± 0.1	207.4 ± 10.4	28.1 ± 0.5	39.8 ± 0.8	0.0
March	1.6 ± 0.2	225.4 ± 11.3	30.6 ± 0.2	29.2 ± 1.1	0.0
April	1.5 ± 0.2	223.1 ± 11.2	34.2 ± 0.2	27.0 ± 1.4	12.0
May	3.0 ± 0.5	254.9 ± 12.7	32.3 ± 0.4	30.0 ± 2.4	59.0
June	3.4 ± 0.3	265.4 ± 12.0	30.1 ± 0.4	52.7 ± 1.7	53.0
July	2.8 ± 0.2	287.1 ± 12.0	27.6 ± 0.2	64.2 ± 1.5	219.0
August	3.3 ± 0.4	276.2 ± 12.0	26.0 ± 0.2	71.0 ± 1.9	187.0
September	2.1 ± 0.2	251.5 ± 12.0	26.7 ± 0.2	65.0 ± 1.0	121.0
October	2.1 ± 0.3	243.8 ± 12.2	27.2 ± 0.2	60.0 ± 1.6	39.0
November	1.4 ± 0.1	206.6 ± 10.3	26.8 ± 0.2	67.1 ± 1.6	59.0
December	1.1 ± 0.1	215.5 ± 8.0	27.1 ± 0.3	61.0 ± 1.1	9.0

specific mass absorption cross-section of the black carbon deposited on the filter (16.6 m^2/g for 880 nm channel), area of the sample spot, and the flow rate. As [BC] is inversely proportional to the absorption coefficient, a higher value of absorption would yield low BC concentrations. It is to be noted that hematite (iron oxide, Fe_2O_3) is the other strong absorber found commonly in atmospheric aerosol (Pinnick *et al.*, 1993; 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). More details of the instrument and error budget were given elsewhere (Hansen *et al.*, 1984; Babu and Moorthy, 2002).

The AE21 was operated with a sharp-cut cyclone inlet (Kenny *et al.*, 2000) equipped with an insect and rain guard at a flow rate of 3 L/min making the cut-point ~3.2 μm. Five minute averaging times were used. The instrument advances to a new filter spot when the total attenuation is equivalent to a BC loading of 1250 ng/m^3 (Dutkiewicz *et al.*, 2009), and it was operated with a 1 to 10 dilution option to minimize frequent filter advances. Each time the filter advances, concentration measurements are missed for three sampling intervals. These are normally filled by interpolation. Recently, there have been several papers that indicate under certain conditions the Aethalometer does not have a constant response curve because as the filter loads, multiple scattering and shadowing effects impact the readings (e.g., Weingartner *et al.*, 2003; Arnott *et al.*, 2005). This partially manifests itself as a step change in [BC] resulting at each filter change with over determined [BC] values for a clean filter and under determined [BC] values for loaded filters. The magnitudes of the effect depend on the nature of the particulate matter (PM) and the wavelength of the channel; it is greatest for fresh soot particles and less for mixed aged ambient aerosols. For the configuration used in this study, the uncertainty in [BC] was in the range 0.040–0.060 μg/m^3. The limitations and uncertainties of aethalometer in BC measurements and corrections needed are well documented in the recent literature (Weingartner *et al.*, 2003; Arnott *et al.*, 2005; Schmid *et al.*, 2006).

The QCM was used to measure the total PM mass (M_t) as well as size segregated (m_{ci}) PM mass concentration, and mass size distribution (dm/dr) of aerosols in the size range

between 0.05 and ≥ 25 μm over 10 channels. It operated at a flow rate of 0.24 L/min and sampling was done at nearly hourly intervals, manually, when ambient RH < 80%. Typical sampling duration varied from 5 to 6 minutes. More details of the instrument, data analysis and error budget are reported by other authors (Pillai and Moorthy, 2001; Madhavan *et al.*, 2008; Kumar *et al.*, 2009b) in the Indian region. The MWR measures the spectral solar flux as a function of time (airmass) from which the spectral aerosol optical depths (AOD) are deduced 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 in 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; Kumar *et al.*, 2009a, Balakrishnaiah *et al.*, 2011).

RESULTS AND DISCUSSION

Diurnal Variations of BC Mass Concentration

Diurnal variations of BC are very important in understanding the role of mesoscale atmospheric processes and the effect of local human activities. The diurnal variation of the atmospheric boundary layer (ABL) height and its structure is known to influence the surface BC concentrations (Moorthy *et al.*, 2003; Nair *et al.*, 2007). The average diurnal variation in [BC] is shown in Fig. 2 over Anantapur during January–December 2010, which shows significant well defined diurnal variation with gradual buildup during early morning hours with a sharp peak around 07:00–08:00 h, an afternoon minimum (14:00–16:00 h) and an evening increase attaining a broad peak around ~21:00 h. These patterns can be explained by a combination of emission sources and local meteorology.

The gradual buildup of the morning [BC] is attributed to the combined effects of fumigation effect (Stull, 1998; Fochesatto *et al.*, 2001; Babu *et al.*, 2002; Beegum *et al.*, 2009) of the boundary layer (the so-called fumigation effect) and a morning increase in anthropogenic activities (Safai *et al.*, 2007; Kumar *et al.*, 2011) in a rural environment as the observation site is surrounded by villages, industries and agricultural fields. The sampling location basically as rural;
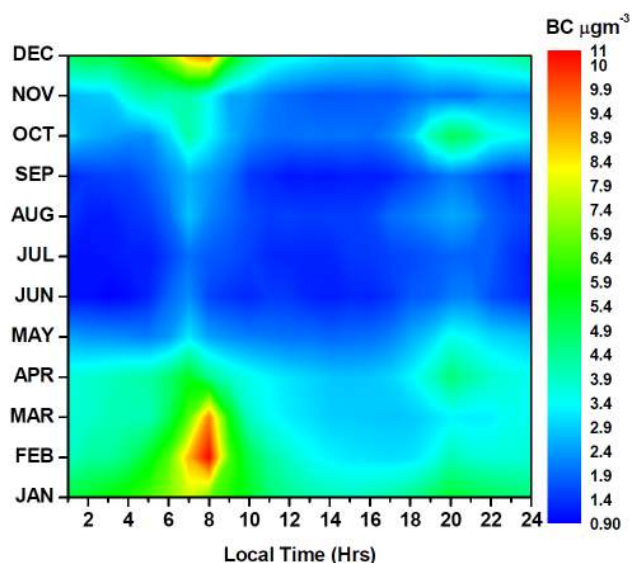
Fig. 2. Diurnal variations in BC mass concentration for the study period from Jan–Dec 2010 at Anantapur.

however, this is not really the case. Anantapur is a city of over 500,000 people, with industry and major highways. In addition, apparently, residences can be significant sources of BC due to residential wood combustion for cooking and heating. This is relevant because there can be significant local contributions to the BC mass concentration. Moreover, the cause of the morning peak in the diurnal cycle is attributed to fumigation due to an increasing mixing layer (Devara and Raj, 1983; Alapattu *et al.*, 2009; Mahalakshmi *et al.*, 2011) which brings polluted air from aloft down to the surface. This is a reasonable assumption in a remote area with little to no local emissions. However, an alternative explanation, in a location such as Anantapur, is that the morning peak is due to the morning rush hour traffic coupled with increased wood combustion for morning heating and cooking at Anantapur residences. This is supported by the fact that the morning peak increases in winter when one would expect increased residential wood combustion for heat. And also soon after sunset, the surface inversion (see Babu *et al.*, 2002; Safai *et al.*, 2007; Beegum *et al.*, 2009) begins to form, trapping more primary pollutants related to BC.

The identical diurnal variations in [BC] were also reported by several earlier researchers at the same observation site (Kumar *et al.*, 2011), other continental, coastal and urban locations reported by Allen *et al.* (1999), Babu and Moorthy (2002) and Beegum *et al.* (2009), respectively. An opposite scenario has been observed with high value in the afternoon and low in the early morning hours over Nainital [29.4°N, 79.5°E, 1958 m asl], a high altitude remote location in the central Himalayas (Dumka *et al.*, 2010); Sinhagad [18°21′N, 73°45′E, 1450 m asl], a high altitude station in the Western Ghats (Raju *et al.*, 2011); and Mt. Norikura in Japan (Nishitha *et al.*, 2007). It is mainly due to the mountainous (hilly) nature of the sampling site and the nearby populated valley; mountain/valley winds would also influence the BC diurnal variations in those above sites. It is noted that the [BC] may not be totally lost from the atmosphere, but only

is redistributed over a large spatial extent by the boundary layer dynamics (Chen *et al.*, 2001). The daytime dilution of [BC] is advantageous from the health perspective making the ambient air cleaner. But, as BC gets transported to higher heights, their radiative forcing values increases compared to the values when they are near the surface (Haywood and Shine, 1997).

Annual and Seasonal Variations of BC Concentration

The annual variations of BC mass concentration during January–December 2010 was shown in Fig. 3 (left panel). The histograms represent the monthly mean [BC] and the vertical lines passing through them are the errors in the estimation of monthly mean BC values. The BC mass concentration varied from 0.2 μg/m^3 (recorded in July 2011) to 14 μg/m^3 (recorded in January 2011) μg/m^3 with an annual mean of 3.03 ± 0.27 μg/m^3 for the period of study. Annual variations suggest large concentration (4.11 ± 0.38 μg/m^3) of BC during dry months (December–May) and low concentrations (1.94 ± 0.20 μg/m^3) during the monsoon (June–September) followed by the postmonsoon season (October and November). The highest monthly mean [BC] during the study period was observed in the month of January (5.25 ± 0.82 μg/m^3) and the lowest was observed in July (1.45 ± 0.28 μg/m^3). Over the observation site, December–May is generally under the influence of continental air masses while during June–November the air mass changes to clean marine associated with monsoons (from a southwest and northeast direction). This is clearly shown in the site map by including the change in the synoptic wind patterns derived from NCEP/NCAR reanalysis satellite data (Fig. 1).

Pollution loading over this station has a maximum in the winter month of January followed by the summer and postmonsoon months and minimum in the monsoon month of June/July as seen in Fig. 3 (left panel). There is a sudden drop in the pollutants loading from May to July in the study area. Pollutant concentrations remained low during southwest monsoon months and build up again in the postmonsoon with a high during the winter months. In winter months, calm or weak winds increase the concentration of pollutants at ground level where stronger winds carry the pollutants far away from the source and dilute them (Munn and Hirt, 1976). More discussion on annual mean BC concentrations measured at Anantapur (Fig. 3 (left panel)) in comparison with [BC] along with ± 1σ deviation or their range from distinct environments of India and other parts of the world are discussed in the sections to follow.

The hourly mean seasonal variations in [BC] for different seasons on a diurnal scale are shown in Fig. 3 (right panel). This shows a primary (fumigation peak) maximum around 07:00–08:00 h and a less prominent secondary maximum (nocturnal peak) in the late night hours between 21:00 h. The highest mean value of diurnal [BC] (about 10.5 μg/m^3 during morning hours) is observed in the winter season. 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–June in the summer months and starts decreasing from July onwards, which shows that the diurnal peak slightly shifts with season. Minimum [BC]
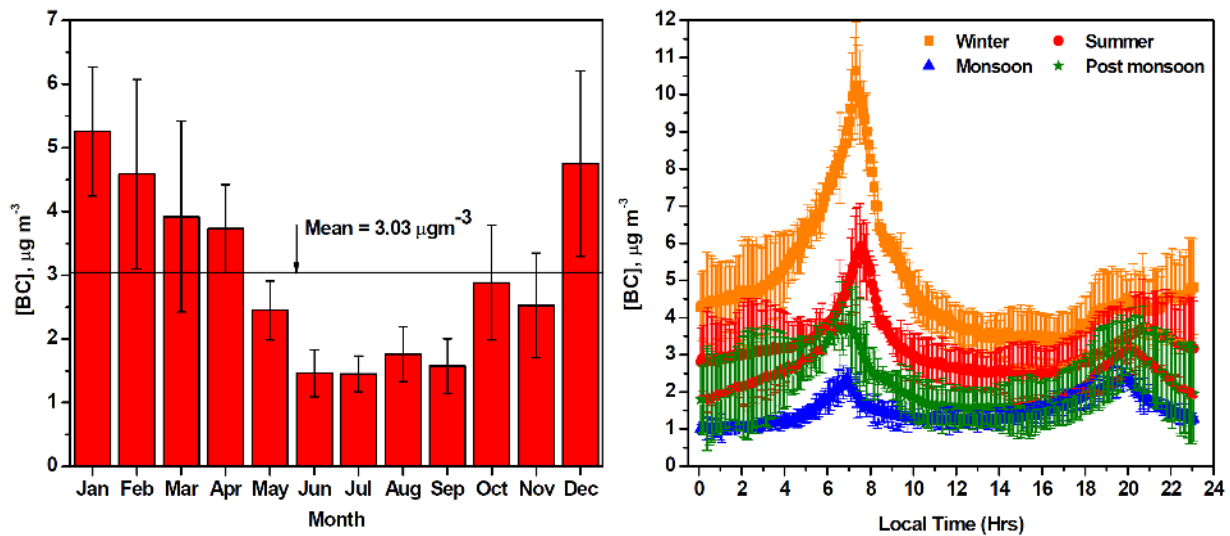
Fig. 3. Annual and seasonal (left and right panels, respectively) variations in BC concentration during the study period from Jan–Dec 2010.

was noticed in daytime between 10:00–16:00 h. The similar diurnal variations in all seasons suggest that the diurnal variation of BC is mainly governed by the diurnal evolution of ABL. Seasonal variations in [BC] mainly occurs because of differences in the extent of contraction and expansion of the ABL as a result of differential solar heating of the earth's surface as well as differences in sources and strengths of production in different seasons (Kunhikrishnan *et al.*, 1993). Gradual increase in [BC] from around ~16:00 h is due to increased production of BC aerosols and gradual formation of a surface based inversion opposing vertical mixing in the atmosphere (Kunhikrishnan *et al.*, 1993, Stull, 1998). On the basis of the observations using low-altitude wind profilers measurements at a nearby tropical Indian station, Gadanki [13.5°N, 79.2°E, 380 m asl], about 250 km away from Anantapur located in a direction southeast of the observation site, a low value of ventilation coefficient (the ventilation coefficient (VC) is given by VC = Z_iU, where Z_i is the mixed layer height; and U is the average wind velocity in the mixed layer) was reported during early morning and late night hours and high values during daytime (Krishnan and Kunhikrishnan, 2002). In another study over Gadanki, Krishnan and Kunhikrishnan (2004) reported that the ABL height attains maximum during the summer period and minimum during the winter. Recently, similar observations on mixed ABL height have been reported by Mahalakshmi *et al.* (2011) over an urban area of Hyderabad [17.36°N, 78.34°E, 536 m asl], India during 2008.

The seasonal mean values of BC mass concentrations for the year 2010 at Anantapur showed maximum of 5.05 ± 0.51 µg/m³ during the winter, followed by 3.77 ± 1.23, 2.33 ± 0.82 µg/m³ in summer, postmonsoon seasons, respectively and 1.55 ± 0.51 µg/m³ in the monsoon (see Fig. 3 (right panel)). Since [BC] is mostly associated with anthropogenic sources (biomass and fossil fuel burning) their concentrations increases at times when local generation of aerosols is prominent. In addition to waste burning, wood and scrubs are also burnt at early morning and night hours by several

people to keep themselves warm during cold winter months. During the winter season, [BC] increased by 4% from the annual mean. This has been attributed to prevailing meteorological conditions like low wind speed and low ventilation coefficient (Mahalakshmi *et al.*, 2011). Increase in pollutant concentrations during summer has been attributed to transport of air masses from the northwest regions (Badarinath *et al.*, 2010). During the monsoon, BC concentrations decreased to 52% from the annual mean as a result of the washout by precipitation and transport of clean air from the ocean. The weakening of convection, due to cloudy skies and mechanical turbulence due to wind shear, results in highly reduced concentrations in the residual layer (Babu *et al.*, 2002).

Frequency Distribution of BC Mass Concentration

Frequency distribution of BC mass concentrations for different seasons at Anantapur is shown in Fig. 4. The total 5 minutes averaged data samples are divided into finer mass interval of 1 µg/m³ for all seasons during the study period and the corresponding statistical data have been included in Table 2. During the winter season, 60% of the BC mass concentrations lie within the range of 2–5 µg/m³ and 26% between 5–8 µg/m³ and the lowest of 2% lie within the range less than 2 µg/m³. Whereas in the monsoon, 80% of [BC] lie in the range of < 2 µg/m³ and 19% of BC levels between 2–5 µg/m³. In fact, out of the all seasons, the number of days showing maximum BC concentrations > 10 µg/m³ were observed in the winter. During the summer, 68% of BC mass concentrations lies between 2–5 µg/m³ and only 1% for the range of > 10 µg/m³. An almost similar trend was observed during the post monsoon season. This again indicates that the overall dominance of higher BC concentrations (fine aerosol) is noticed during the winter and lower concentrations during the monsoon.

Influence of Wind Direction and Rainfall on [BC]
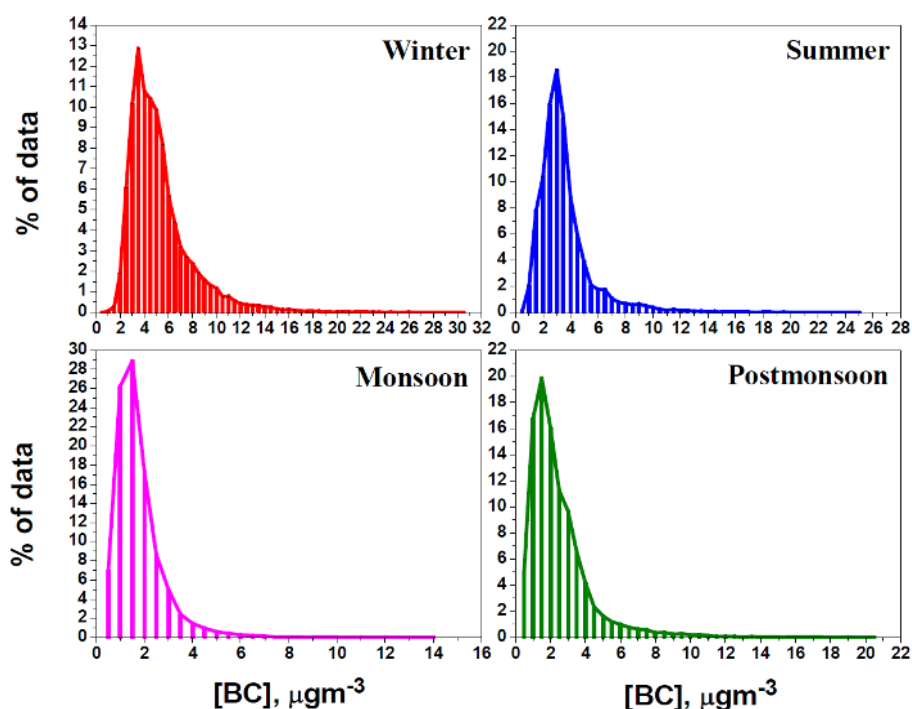
Fig. 5 shows the monthly mean variations in [BC] with

Fig. 4. Percentage occurrence of BC mass concentrations for different seasons in distinct size ranges measured at Anantapur during 2010.

Table 2. Percentage of occurrence of BC mass concentration ($\mu g/m^3$) for different ranges in various seasons at Anantapur during 2010.

BC range	% of data			
($\mu g/m^3$)	Winter	Summer	Monsoon	Postmonsoon
< 1	0.07	2.16	33.19	21.75
1–2	2.22	18.20	46.22	35.91
2–3	16.24	34.47	13.66	20.89
3–4	23.59	23.66	3.89	10.71
4–5	20.27	9.76	1.57	3.98
5–6	13.85	3.86	0.71	2.23
6–7	7.56	2.83	0.38	1.41
7–8	5.05	1.48	0.16	0.97
8–9	3.47	1.24	0.08	0.64
9–10	2.49	0.86	0.05	0.49
> 10	5.21	1.47	1.00	1.03

Fig. 5. Variations in [BC] with wind direction over Anantapur during the period of study, from Jan–Dec 2010.

wind direction. It is depicted in Fig. 5 that higher BC values tend to occur when the wind is directed from the 180–225° sector which may be explained by the geographical location of the observation site (see Fig. 1). It is pertinent to mention that most of small scale industries (not major industries), stone crushing industries, brick making units and two major National Highways (NH 44 & NH 205) are located in this direction. This results in emissions of fine aerosol pollutants which are transported towards the observation site due to the favorable winds. In addition to this, the levels of biomass burning and other anthropogenic activities are distinctly higher in the southwesterly region, so the air masses coming from these associated directions might bring more BC aerosols to the observation site.
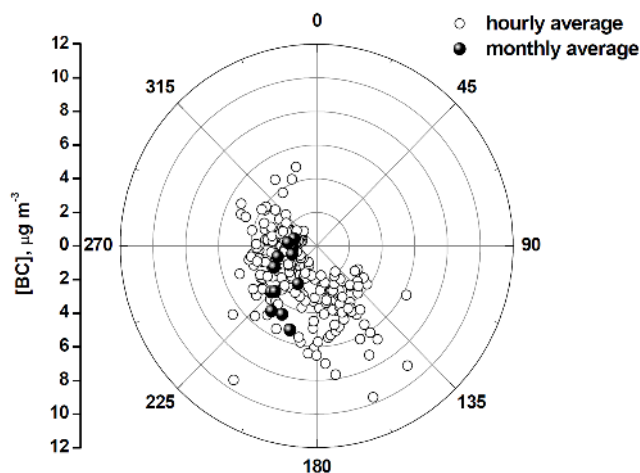
Several studies pointed out that rainfall could be an efficient sink for BC aerosols (Ogren *et al.*, 1984; Harrison *et al.*, 1997). Being in the submicron size range, chemically inert and hydrophobic (in its nascent form), BC has a long atmospheric life time (Babu and Moorthy, 2002), and wet removal is the main mechanism for BC from the atmosphere (Kohler *et al.*, 2001). In the lower atmosphere, the average life time of BC is generally from 7–10 days during dry weather conditions and 3–5 days during wet seasons (Reddy and Venkataraman, 1999). We thus tried to interpret this process with the BC concentration variation at Anantapur. It may be assessed that maximum rainfall occurs during the

monsoon and the post monsoon seasons at this location (see Fig. 6). As a result, lower BC concentration is found to occur during this period. Results of rainfall with BC co-variation illustrate that there is a clear trend between them with a regression coefficient of –0.72 (not shown in figure).

Potential Source Regions: Long Range Circulation

In general, BC particles are produced mainly by anthropogenic activities such as fossil fuel burning (from industries and automobiles) and biomass burning (for agriculture and domestic applications). However, natural sources such as forest fires also contribute to the BC mass concentrations are relatively much less. In the fine/submicron regime and chemically inert, BC particles can be transported both horizontally and vertically long distances by prevailing winds (Wolff, 1984) and can be found in substantial amounts even in the regions situated far away from potential sources. As such, BC aerosols could be used as good tracers for the transport of polluted air masses. In order to investigate the role of long-range transport in causing the observed temporal changes at Anantapur, we have computed the 7-day isentropic air mass back trajectories for all the days on which BC was measured using Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://www.arl. noaa.gov/ready/hysplit4.html) (Draxler and Rolph, 2003). Fig. 7 shows the back trajectories (once per day), each trajectory starts at 00:00 local time (LT) and arriving at Anantapur, at an altitude of 200 m agl. The 7-day period was considered in view of the atmospheric lifetime of BC (Reddy and Venkatraman, 1999). Even though, the trajectory analyses are limited in absolute accuracy (Stohl, 1998), they are quite useful for determining the potential pathways of aerosols.

To investigate the potential source regions, we separated the trajectories into different clusters, to ascertain the primary pathways. The main criteria of clustering are to minimize the variability between the trajectories within a cluster and to maximize the variability between the clusters. The cluster-mean trajectories and their percentage contribution to the total were calculated for each season and these are shown in Fig. 7. The retrieved cluster-mean trajectories are identified and grouped into three distinct source regions, namely, the Central India (CI), the Arabian Sea (AS) and the Indian Ocean (IO), mainly depending on the regions bounded by the Indian subcontinent. It is evident from Fig. 7 that the BC that the cluster-mean trajectories distinctly change their orientation and geographic locations of their over pass with season. Subsequently, BC mass concentrations are tagged with the trajectories belonging to a common cluster and are averaged to obtain average BC values for each cluster and these are presented in Table 3 along with standard errors. Here, it is to be noted that we are just discussing the possible source regions and not attempting to quantify the contribution from different regions.

As clearly seen in Fig. 7, during the winter, the trajectory clusters have a very long continental overpass from CI through northerly/northwesterly India before arriving Anantapur. These trajectories would thus be conducive for the significant advection of continental aerosols to the measurement region. The observed high value of [BC] at the experimental site is the consequence of these. During the summer, the trajectory air mass pathways bring major pollutant contribution from southwesterly/northeasterly sectors and a less number of cluster-mean trajectories transported from a southerly direction. A majority of these trajectories (> 70%) originated from the Arabian Sea (AS) and from the Indian Ocean (IO), which might transport anthropogenic aerosols along with the coarse sea salt aerosols, because these trajectories have a vast traverse across the oceanic regions where the wind speeds were moderately high (see Fig. 7). This would result in mixed type of aerosols and this explains moderate values of BC concentration. A lesser number of trajectories (< 30%) arriving at the measurement location from northern India bring a small contribution of anthropogenic aerosol component, and as such a moderate value of [BC] was noticed. The trajectories arriving in the monsoon season mostly originated from the AS traversing over western coastal regions of India, are purely oceanic in nature, which brings in relatively pristine marine air mass
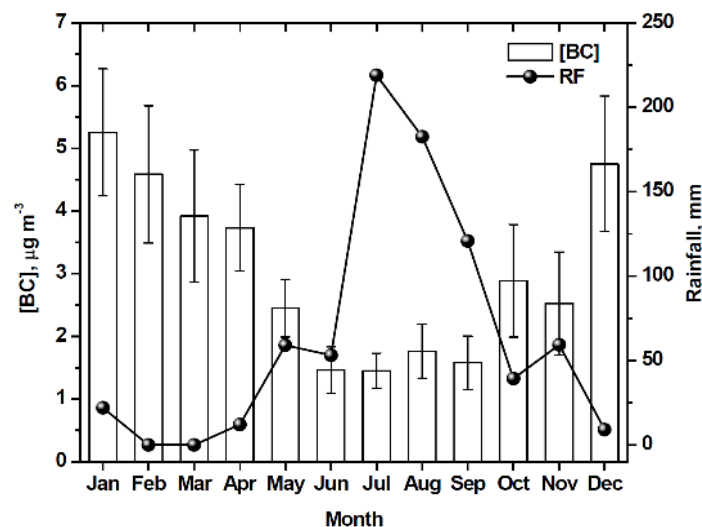

Fig. 6. Monthly mean variations in [BC] and rainfall (RF) at Anantapur during 2010.
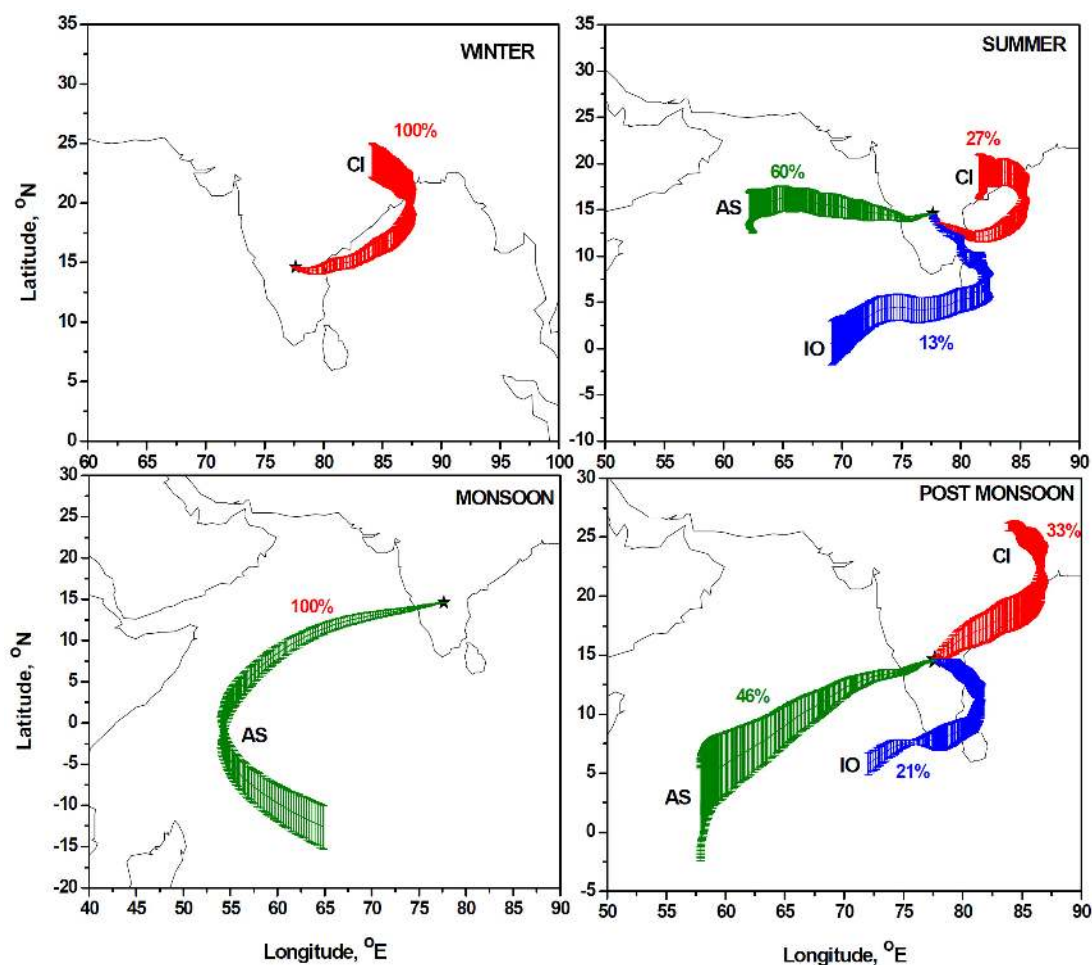
Fig. 7. Cluster-mean trajectory analyses with source regions arriving at the observation site (indicated by ★) at 200 m agl in different seasons for the study period from Jan–Dec 2010 (AS-the Arabian Sea, IO-the Indian ocean and CI-the Central India).

Table 3. Average BC concentrations (μg/m³) in different seasons according to the cluster-mean trajectories.

Season	Potential source regions		
	Central India	Arabian Sea	Indian Ocean
Winter	4.82 ± 0.19	-	-
Summer	3.98 ± 0.51	4.09 ± 0.17	3.26 ± 0.47
Monsoon	-	1.52 ± 0.05	-
Postmonsoon	2.08 ± 0.14	3.26 ± 0.27	1.64 ± 0.13

towards the observation site. They are mostly characterized by smaller amounts of accumulation mode aerosols and larger amount of coarse mode sea salt as the marine aerosols generally have a coarse mode associated with sea spray. This would result in lower values of [BC] during the monsoon. In contrast to this, in the postmonsoon season we have significant advection from northern India and from the west and east coasts of India. Only less than 60% of trajectories originate from the oceanic environment and remaining trajectories are from the mainland of India. This results in the seasonal mean [BC] during the postmonsoon being significantly higher than that during the monsoon and lower that that in the summer.

Percent of BC in PM₂.₅

Fig. 8 summarizes the fraction of daily average [BC] in PM$_{2.5}$. The horizontal line represents the mean BC/PM$_{2.5}$ ratio and hollow circles indicates monthly BC/PM$_{2.5}$ ratio for the entire sampling period. The BC/PM$_{2.5}$ ratio varied from 1.3% to 7.2% with a mean value of 4.6% during the observation period. Tripathi *et al.* (2005) reported BC as 7 to 15% of total suspended particulate (TSP) at Kanpur [26.47°N, 80.33°E, 142 m asl], India. Latha and Badarinath (2005) presented BC as 7% of TSP at Hyderabad, India, and Safai *et al.* (2007) reported BC as 2.3% of TSP for Pune [18°32′N, 73°51′E, 559 m asl], India. Cao *et al.* (2009) reported BC as 1.6 to 15.6% for individual PM$_{2.5}$ samples collected at Xi'an [34.2°N, 108.98°E, 400 m asl], China. In suburban regions of Europe and North America, BC contributes about 5% of TSP (Ramanathan and Crutzen, 2003). TSP contains more geological material than PM$_{2.5}$, so these lower BC fractions are expected. Venkatachari *et al.* (2006) reported higher BC fractions, 13% and 11% of PM$_{2.5}$ mass at two sites in New York City. The BC abundance in PM$_{2.5}$ is affected by the mixture of pollution sources and meteorological conditions. At Anantapur, higher BC/PM$_{2.5}$ ratio in the winter months may be influenced by biomass
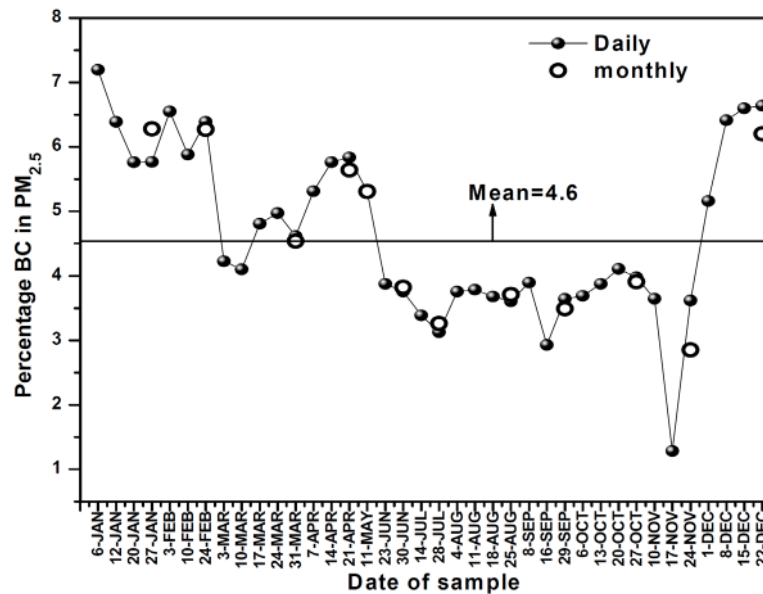
Fig. 8. Fraction of daily average BC in $PM_{2.5}$. The horizontal line represents the average BC for the entire monitoring period and the solid and hollow circles indicates daily and monthly $BC/PM_{2.5}$ ratios during the study period from Jan–Dec 2010.

burning. Whereas, the lower $BC/PM_{2.5}$ ratio in the monsoon is due to Asian and other fugitive dust, which increases $PM_{2.5}$ with non-carbonaceous material, thereby decreasing the $BC/PM_{2.5}$ ratio (Zhang *et al.*, 2002).

Variation of [BC] with AOD_{500} and $BC/PM_{2.5}$ Ratio with Ångström Exponent (Alpha)

A simultaneous estimate of monthly mean column aerosol optical depth (AOD) at 500 nm derived from a MWR and surface [BC] is shown in Fig. 9(a). AOD_{500} is found to increase with [BC] just for the two periods from September to October and from November to December, whereas in the other months this trend is not observed. Correlation between variations in AOD at 500 nm and BC aerosols is observed to be positive (not shown in Figure) and becomes poor at high BC concentrations with a correlation coefficient of 0.42. The correlation coefficient is not very high because of other (non-BC aerosols) contributors to AOD (Babu and Moorthy, 2002). It may be noted that AOD measurements were limited due to cloudy sky conditions and were carried out during day time only. Even though AOD is column measurements and BC is surface observations, both can be correlated. This excludes aerosol layers aloft, so that the variation in the surface can be taken as representative of the variations in the column. This observation highlights that over land, the abundance of BC would change the sign of forcing (Haywood and Shine, 1997). The effect of change in $BC/PM_{2.5}$ ratio on the Ångström exponent (alpha) is shown in Fig. 9(b). As BC being in the accumulation regime, increase in its fractional abundance may reflect in alpha. The Fig. 9(b) gives a clear picture of high values of $BC/PM_{2.5}$ ratio with low alpha values and vice-versa.

Comparison of BC Concentrations with Other Locations

BC mass concentrations measured in different land locations varying from clean, natural to highly polluted,

urban sites from various parts of the world are discussed in this section and are listed in Table 4. This comparison, though not comprehensive, is aimed at illustrating the nature of variations in [BC] that occurs in different environments. At Anantapur, the one-year average BC mass concentration was 3.03 μg/m^3, with a standard deviation of 0.27 μg/m^3, accounting for 4.6% of $PM_{2.5}$ on average. Anantapur's one year average BC concentrations are more than three times at Nainital [29.4°N, 79.4°E, 1938 m asl], NCO-P [27.97°N, 86.82°E, 5079 m asl] and Nam-Co [30.92°N, 91.12°E, 4718 m asl], less than two times those reported from Ahmedabad [23.03°N, 72.5°E, 49 m asl], Hyderabad [17.36°N, 78.34°E, 536 m asl], Karachi [24°51′N, 67°02′E, 11 m asl], Seoul [37°27′N, 126°57′E, 135 m asl] and four and seven times less than those at Mumbai [19.38°N, 72.83°E, 312 m asl], Xi'an [34.23°N, 108.88°E, 410m asl], Kanpur [26.47°N, 80.33°E, 142 m asl], Beijing [39°55′N, 116°25′E, 45 m asl] and Delhi [28.36°N, 77.05°E, 278 m asl], Lahore [31°32′N, 74°22′E, 235 m asl], respectively. It is noticed from Table 4 that BC mass concentrations measured from Trivandrum [8.44°N, 76.92°E, 35 m asl] and Gadanki [13.5°N, 79.2°E, 380 m asl], had comparable [BC] with Anantapur. Bangalore [12.96°N, 77.56°E, 949 m asl] and Pune [18°31′N, 73°55′E, 560 m asl] had average BC levels close to those in Guangzhou [23.13°N, 113.27°E, 98 m asl] and also BC concentrations at Kanpur are similar with those reported at Xi'an. Only Lahore seems to have higher average BC levels.

SUMMARY AND CONCLUSIONS

The present work addresses the diurnal and seasonal variability of black carbon aerosol mass concentration observed over a one-year period from January to December, 2010 at a semi-arid station of Anantapur, using a dual channel (370 and 880 nm) rack mount Aethalometer. All the parameters investigated in the paper show well defined
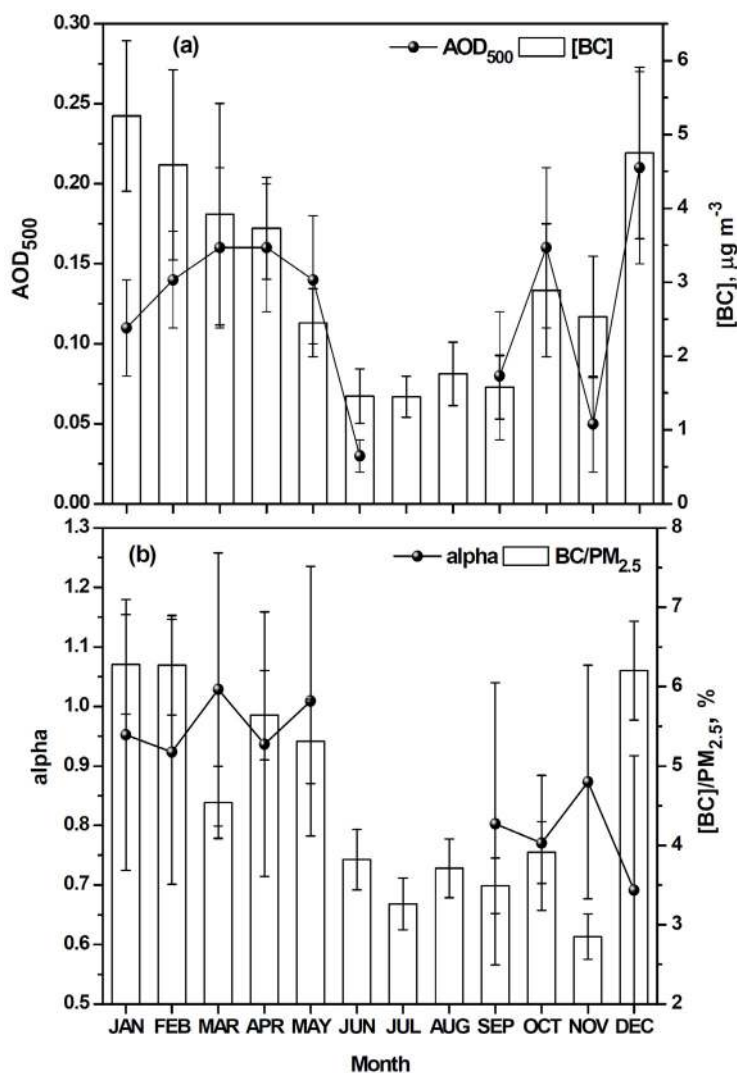
Fig. 9. Variations in monthly mean BC mass concentration ($\mu g/m^3$) and AOD_{500} (top panel), $BC/PM_{2.5}$ ratio (%) and Ångström exponent (alpha) (bottom panel).

diurnal, seasonal as well as significant intra-annual variations.

The annual mean BC concentration is 3.03 ± 0.27 $\mu g/m^3$ and varied between 0.2 to 14 $\mu g/m^3$. The seasonal BC concentrations for the year 2010 at Anantapur showed a maximum of 5.05 ± 0.51 $\mu g/m^3$ during the winter, followed by 3.77 ± 1.23, 2.33 ± 0.82 $\mu g/m^3$ in summer, postmonsoon seasons, respectively and 1.55 ± 0.51 $\mu g/m^3$ in the monsoon. The seasonal variation of black carbon aerosols showed a high concentration during winter season followed by the premonsoon and postmonsoon seasons and low concentration during the monsoon season. It can be explained by changes in emission sources and variability in meteorological conditions, especially increased domestic heating with wood, lower wind speeds, and shallower mixed layer depths in winter. BC mass concentration are found to show diurnal variations with two peaks, one in the morning (fumigation peak) between 07:00 and 08:00 h and the other in the evening (nocturnal peak) at 21:00 h. The peaks in the morning and evening hours are attributed to the vehicular combustion arising from the rush hour traffic, increasing in the local anthropogenic activities for cooking and heating and the

boundary layer dynamics. [BC] concentrations decreases substantially and the diurnal minimum is attained in the afternoon hours (14:00–16:00 h).

The 7-day isentropic backward trajectories for all the seasons during the study period indicated the possibility of transport of a significant fraction of fine mode (nucleation plus accumulation) aerosol from the central and northern parts of India during the winter leading to an increase in BC aerosol concentration, while the transport of major coarse mode aerosol from the Arabian Sea during the monsoon season indicated a decrease in BC concentrations and were carried away by the wind (~5 m/s). A clear inverse relationship is noticed between BC concentrations and rainfall. The entire rainfall for the year occurs over Anantapur during the southwest monsoon season (June–September) and this large rainfall (exceeding 500 mm) efficiently removes aerosols resulting in low values of BC. It is depicted that higher BC values in winter tend to occur when the wind is directed from the 180–225° sector which may be explained by the geographical location of the observation site, and in addition, the winds are from polluted north during

Table 4. BC mass concentration measured by an Aethalometer (880 nm) at different land locations in India and Asia.

Location	Type of location	Observation period	Mean BC ± σ/ BC mass range ($\mu g/m^3$)	Reference
Anantapur, India	Semi-arid, Rural	Jan.–Dec. 2010	3.03 ± 0.27	Present Study
Nainital, India	High altitude	Dec. 2004	1.36 ± 0.99	Pant *et al.*(2006)
NCO-P*, Nepal	High altitude	Mar. 2006–Feb. 2008	0.91 ± 1.13	Marinoni *et al.* (2010)
Nam-Co, Tibet	High altitude	Jul. 2006–Jan. 2007	0.82 ± 0.71	Ming *et al.* (2010)
Delhi, India	Urban, Industrialized	Mar.–May. 2006	3–27	Beegum *et al.* (2009)
Pune, India	Urban, Industrialized	Jan.–Dec. 2005	4.1	Safai *et al.* (2007)
Ahmedabad, India	Urban, Industrialized	Sep. 2003–Jun. 2005	0.21–10.2	Ramachandran and Rajesh (2007)
Mumbai, India	Urban, Industrialized	Jan.–Mar.1999	12.4 ± 5.1	Venkatraman *et al.* (2002)
Hyderabad, India	Urban	Jan. 2004	1.5–11.2	Latha *et al.* (2004)
Bangalore, India	Urban, Continental	Nov. 2001	4.2	Babu *et al.* (2002)
Kanpur, India	Urban, Continental	Dec. 2004	6–20	Tripathi *et al.* (2005)
Gadanki, India	Rural	Apr.–Nov. 2008	1–4	Gadhavi and Jayaraman (2010)
Trivandrum, India	Coastal, Urban	Aug. 2000–Oct. 2001	0.5–8.0	Babu and Moorthy (2002)
Beijing	Urban	Jul. 1999–Sep. 2000	8.7–10.1	He *et al.* (2001)
Guangzhou	Urban	Jul. 2006	4.7	Verma *et al.*(2010)
Xi'an, China	Urban	Sep. 2003–Aug. 2005	14.7 ± 9.5	Cao *et al.* (1009)
Lahore, Pakistan	Costal, Urban	Nov. 2005–Jan. 2006	21.7	Husain *et al.* (2007)
Karachi, Pakistan	Urban	2006–2007	5.9	Dutkiewicz *et al.* (2009)
Seoul, Korea	Urban	Jun. 1994	4.86–9.86	Kim *et al.* (1999)

* NCO-P: Nepal Climate Observatory-Pyramid.

this season. The surface boundary layer is shallow during the winter resulting in confinement of aerosols in a lesser volume. The BC/$PM_{2.5}$ ratio varied from 1.3 to 7.2% with a mean value of 4.6% at Anantapur during the observation period. Monthly mean variations in AOD at 500 nm and black carbon aerosols were observed to be positive and become poor at high BC concentrations with an overall correlation coefficient of 0.42. Monthly mean variation of BC/$PM_{2.5}$ ratio was observed to be decreased with decreasing Ångström exponent (alpha).

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