Study on Diurnal and Seasonal Black Carbon and Aerosol Optical Depth Variation over a Semi-arid Region Madurai

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ABSTRACT

Equivalent black carbon (EBC) aerosols, produced from incomplete combustion processes of fossil fuel and biomass burning at a semi-arid site, is collected using aethalometer instrument from June-2014 to May-2016 to understand its absorption characteristics. The variation in EBC mass concentration was not only due to anthropogenic activities, but also due to changes in the atmospheric boundary layer height. The variation of EBC is highly influenced by the wind speed at the study site. Aerosol Optical Depth (AOD) has good correlation with rainfall and a poor correlation with EBC at the study site indicating the dominance of other aerosols at the location.

Keywords: *Equivalent black carbon aerosols, Aerosol Optical Depth, Absorption Angstrom Exponent and Planetary Boundary Layer height.*

1. Introduction

Black carbon (BC) aerosol is produced as primary particles from incomplete combustion processes such as fossil fuel and biomass burning (Udayasoorian et al., 2014; Kuniyal et al., 2016). Lifetime of BC aerosols depends on the meteorological conditions in an area and its average atmospheric residing time is low during wet seasons compared to dry seasons (Babu and Moorthy, 2001). Absorption and scattering of the solar radiation by BC causes "solar dimming effect" and it weakens the land-ocean temperature gradient resulting in the reduction of the Indian summer monsoon rainfall

(Ramanathan et al., 2005; Tiwari et al., 2016). BC causes radiative warming in the atmosphere and the Intergovernmental Panel on Climate Change (IPCC, 2007) has estimated that the global mean clear-sky radiative forcing of BC is 0.23 (±0.25) Wm^{-2} (Udayasoorian et al., 2014). As the sources are spatially and temporally varying, BC concentrations are highly region dependent. The concentration varies with the level of transportation density (Pandey and Venkataraman, 2014), biomass fuel used for cooking purposes (Saud et al., 2012) and level of industrialization (Ramachandran and Rajesh, 2007). These aerosols which are highly absorbing, depict strong spectral, seasonal and
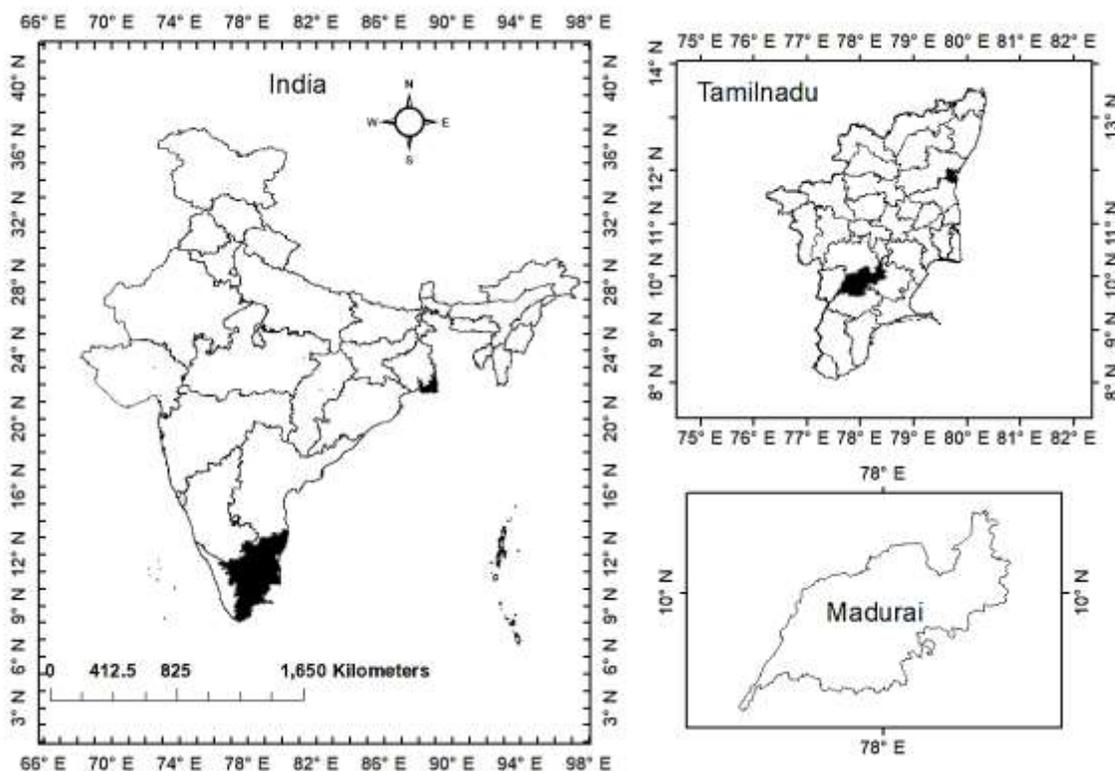
Figure 1: Location of the study region Madurai in India.

diurnal variability (Tiwari et al., 2016). There are many studies over India showing the seasonal and diurnal variation of the BC concentration at different types of environment (Badarinth et al., 2007; Udayasoorian et al., 2014; Kuniyal et al., 2016). Emissions from India contribute about 7 to 14% of global BC emissions (Bond et al., 2013; Klimont et al., 2015). Urban areas and rural areas are dominated by aerosols emitted from fossil fuel (Tiwari et al., 2009) and biomass burning respectively. BC aerosols are more effective in warming the atmosphere when emitted from fossil fuel combustion compared to the BC emitted from biomass combustion and it offsets the cooling effect of sulfate aerosols (Ramana et

al., 2010). Black carbon is denoted as mass equivalent black carbon (EBC) when obtained through the light absorption measurement technique (Petzold et al. 2013; Zotter et al. 2017). Aethalometer is the widely used instrument which uses light absorption (at seven different wavelengths ranging from 370 nm to 950 nm) measurement method and it is appropriate for long-term measurements (Zotter et al. 2017). To derive the contributions from biomass and fossil fuel combustion sources at a particular location, absorption Angstrom exponent (AAE) values obtained from Aethalometer are used. In this paper, the results of aerosol optical depth (AOD) and aerosol EBC measurements carried out for the period from

June 2014 to May 2016 to understand the absorption characteristics of EBC at the semi-arid site are presented.

2. Materials and Methods

2.1. Site description

Madurai (9°91'N, 78°54'E), the second largest and most densely populated tourist city in southern India covers a surface area of 52.8 km^2 and a population of 3.2 million in 2011 while it is 0.25 million in 2001, excluding the ~0.1 million floating population (Figure 1). Madurai city, experiencing rapid urbanization is located 130 m above the mean sea level (MSL) on the banks of river Vaigai. Based on Indian Meteorological Department (IMD) classification the entire study period is divided into four seasons; winter (January-February), summer (March-May), monsoon (June-September) and post monsoon (October-November-December). The seasonal average values of surface meteorological parameters such as temperature (T) in °C, wind speed (WS) in ms^{-1}, relative humidity (RH) in % are given in Table 1 and their diurnal variation are shown in Figure 2. Table 1 shows that the temperature and relative humidity were highest during summer and post-monsoon and minimum in winter and monsoon, respectively. The wind speed is higher in monsoon followed by post monsoon and minimum in summer. The maximum peak in temperature is seen during 1300-1500 hrs and minimum at 0600 hrs irrespective of the seasons. WS is higher in early afternoon to early evening

and is very low during late evening to early morning hours in all the seasons. The relative humidity is low during late afternoon to late night and maximum in the early morning hours. The total amount of rainfall received over the entire study period is 2480 mm with ~48% rain during the post monsoon season and almost 0% rainfall during winter. The measurement is carried down at different microsites in Madurai, which includes Highway cum educational site (Madurai Kamaraj University), residential site (Jaihindpuram and Southgate), sensitive site (Kamarajapuram), industrial site (Keerathurai), and traffic site (Simmakal, Kalavasal, and Periyar).
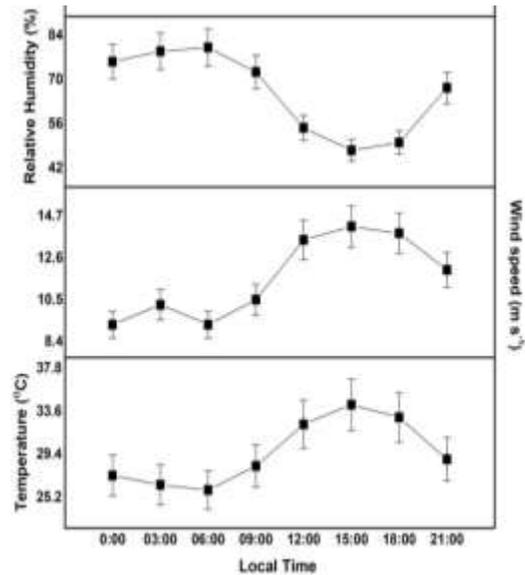
Figure 2: Seasonal averaged diurnal variation of meteorological parameters during the study period.

Table 1. Seasonal average values of air temperature (˚C), relative humidity (%), wind speed (ms⁻¹),planetary boundary layer height(m) and rainfall (mm) during the study period.

Seasons	Air Temperature (˚C)	Relative Humidity (%)	Wind Speed (ms⁻¹)	PBLH (m)	Rainfall (mm)
Winter	24 ± 0.35	62 ± 2.12	4 ± 0.35	915 ± 84.85	9
Summer	27 ± 0.86	57 ± 2.91	3 ± 0.28	1095 ± 127.18	342
Monsoon	26 ± 0.73	51 ± 2.87	6 ± 1.33	865 ± 44.68	951
Post-monsoon	25 ± 0.58	73 ± 5.21	4 ± 0.58	690 ± 121.79	1178

2.2. Measurement of EBC and aerosol optical depth (AOD)

Continuous and near-real time measurement of aerosol spectral absorption started in Madurai from June, 2014, using a seven wavelength Aethalometer (Model AE-31, Magee Scientific, USA; http://www.magescientific.com) (Hansen et al., 1984). A whole air inlet devoid of any cutoff is positioned at 8 m above the ground level is connected to the Aethalometer for aspirating ambient air (Vaishya et al., 2016). The instrument operated at a flow rate of 5 LPM and the absorption data temporal resolution was set to five minutes. Air flow is not dried prior to feeding to the instrument. The Aethalometer measures the attenuation of light that passes through a quartz fiber filter media, on which aerosol particles are deposited, and hence quantifies (1) the magnitude of absorption, in terms of absorption coefficient b_{abs} (Mm⁻¹), and (2) amount of absorbing aerosols, in terms of equivalent black carbon (EBC) mass

concentration (μgm^{-3}), using a conversion factor. Due to a non-linear relationship between light attenuation and surface loading of aerosols on the filter media, underestimation of b_{abs} and EBC occurs. A correction method proposed by (Weingartner et al., 2003) is used in this study to compensate for this 'loading effect'. The Absorption Angstrom exponent (AAE) is calculated from the b_{abs} values, assuming a power law dependency (Angstrom, 1964) of b_{abs} on the wavelength λ as represented in Equation 1:

$$b_{abs}(\lambda) = \beta\lambda^{-AAE} \quad \longrightarrow (1)$$

Where, β is a constant. In order to study and quantify the contributions from biomass burning sources and fossil fuel sources to EBC, time series analysis of the EBC data along with that of AAE, fossil fuel combustion percentage (FF%) and biomass burning percentage (WB%). FF% and WB% is defined as the ratio of EBC from Fossil fuel and biomass burning sources to

total EBC, respectively. The monthly MODerate resolution Imaging Spectroradiometer MODIS/Terra globally gridded Level 3 product (version 005) with 1 x 1 degree spatial resolution is obtained. (Dataset ID: MOD08_D3.005). AOD values which are above 0 and less than 1.0 are only considered because value greater than 1.0 would have resulted most likely due to cloud contamination (Indira et al. 2013; Ramachandran, 2007). More detailed information on algorithms for the retrieval of aerosol and cloud parameters is available at http://modis-atmos.gsfc.nasa.gov.

3. Results and Discussion

The mean EBC mass concentration in Madurai is 5.10 ± 2.40 µg m^{-3}. The mean EBC values of the semi-arid sites of southern India such as Hyderabad (Latha and Badrinath, 2005) and Anantapur (Reddy et al. 2012) showed values 4-10 µg m^{-3} and 3.03 µg m^{-3} similar to Madurai site, however the values are lower when compared with the other urban site, such as Kanpur (7.03 µg m^{-3}) and Delhi (6.70 ± 5.70 µg m^{-3}) and higher than Chennai (2.1 µg m^{-3}), observed by Tiwari et al. (2013); Nair et al. (2007) and Aruna et al. (2013). The mean seasonal values of EBC at Madurai are 8.97 ± 2.34 µg m^{-3}, 4.69 ± 2.14 µg m^{-3}, 6.70 ± 3.83 µg m^{-3} and 3.34 ± 2.53 µg m^{-3} during the winter, summer, monsoon and post-monsoon seasons, respectively. EBC levels are monitored at different locations in Madurai (Highway cum Educational site, Residential site, Sensitive site,

Industrial site, and Traffic site). The Traffic sites (9.26 ± 3.1 µg m^{-3}; Simmakal, Kalavasal, and Periyar) and Industrial site (7.37 ± 2.2 µg m^{-3}; Keerathurai) showed higher concentration with
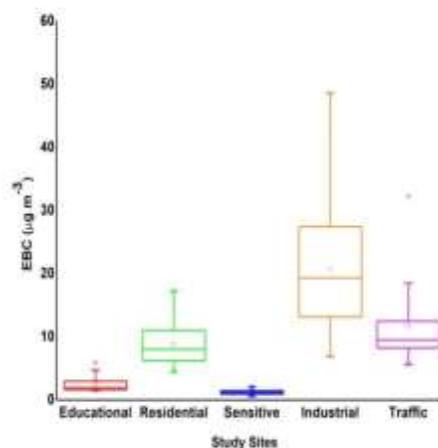
Figure 3: Box-whisker plot shows theequivalent black carbon concentration measured at different sampling sites.

The 5min EBC maxima extending to 46.88 µg m^{-3} and 30.26 µg m^{-3}, respectively. The EBC monitored at Residential sites (3.62 ± 0.53 µg m^{-3}; Jaihindpuram and Southgate), Highway cum Educational site (2.88 ± 0.35 µg m^{-3}; Madurai Kamaraj University) and Sensitive site (2.10 ± 0.30 µg m^{-3}; Kamarajapuram) showed lower concentration with their 5min EBC maxima extending to 16.91 µg m^{-3}, 10.95 µg m^{-3}, and 6.09 µg m^{-3}, respectively. The average values of EBC at each microsites are represented using a box plot (Figure3). In the box plot, there are vertical line (standard deviation), lower whisker (5th percentile), lower edge of the box (25th percentile), central line of the box (median),

filled circle (mean), upper edge of the box (75th percentile), and upper whiskers (95th percentile).
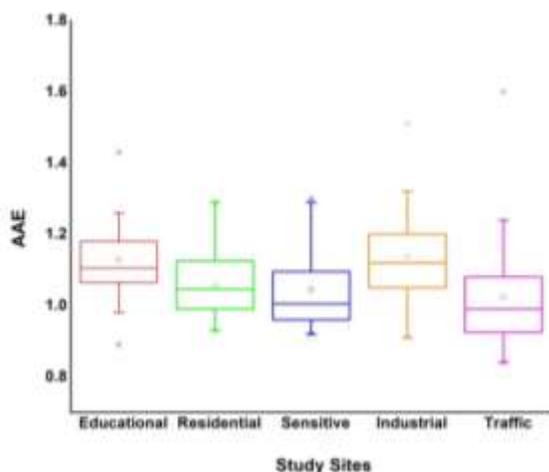
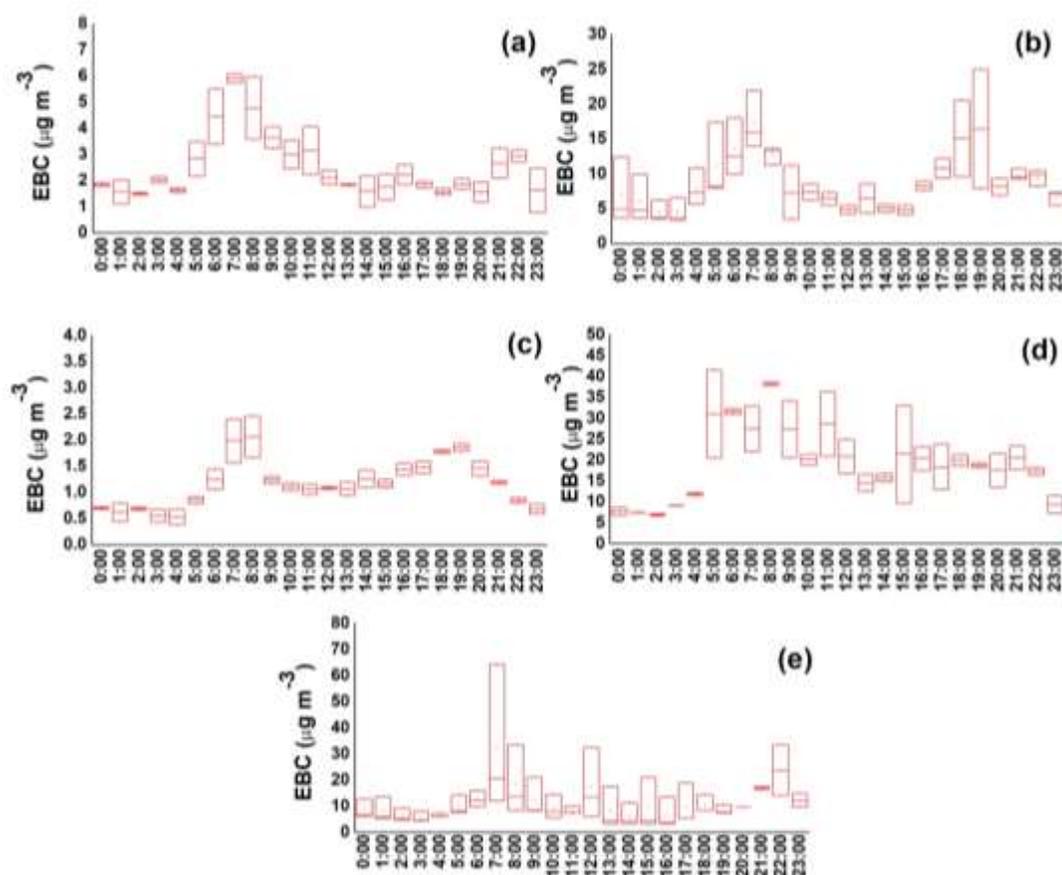**Figure 4: Box-whisker plot shows the absorption angstrom exponent calculated for different sampling sites.**

3.1. Source identification

With the values of AAE, the probable major source of EBC can be identified. According to Kirchstetter et al. (2004) and Herich et al. (2011), the values of α less than or equal to 1.0 indicates the dominance of fossil fuel and the value around 2 represents EBC mixed with larger sized mineral dust particles evolved from biomass combustion. The AAE values at the micro sites are investigated to identify the type and amount of local source's contribution to the EBC concentration. The average AAE values at the highway cum educational site (1.13), residential sites (1.05), sensitive site (1.04), industrial site (1.14), and traffic site (1.02) are plotted in the Figure4. It is found that industrial site and highway cum educational sites are least

dominated by biomass combustion sources while the other sites are dominated by fossil fuel sources.

3.2. Diurnal variation

According to Udayasoorian et. al. (2014) diurnal variations of EBC are important in understanding the effect of anthropogenic activities and the role of mesoscale atmospheric processes. EBC concentrations measured for an entire day at every 5 min interval are combined together into an average for each hour. The diurnal variation of EBC at each of the microsites are studied separately and their source contribution (WB% and FF%) is plotted in Figures 5&6. Highway cum vegetation covered location, Madurai Kamaraj University, located at the foothills of Nagamalai and adjacent to the Madurai-Theni National Highway NH-85 and is on the outskirts of the nearest populated city (about 12.8 km). The diurnal EBC values showed peak values starting at early morning hours 05:00 and extending to 10:00 (LT); whereas during the late night hours there is an another peak. The early morning peaks are caused by the rural background of the study site where most households depend on biomass combustion for all their basic needs (Bhaskar et al., 2018). During the late night hours, due to the increased flow of Heavy duty vehicles (HDV) on the highway results in the fossil fuel activity contribution (Figure 6a). Residential sites Jaihindpuram (Madurai East) and Southgate (Madurai West) are selected for

Figure 5: Box-whisker plot shows the hourly averaged diurnal variation of EBC concentration measured at different sampling sites (a) Educational site (b) Residential site (c) Sensitive site (d) Industrial site (e) Traffic site.

this study because of the presence of high amount of households at these locations (situated inside the city) compared to other locations (a cumulative of about 23198 households) and with limited amount of industrial activities. Most of the households use LPG for their cooking so the reduced amount of biomass combustion contributes to the total EBC measured at the study sites when compared to the educational site. The diurnal variation of EBC shows two peaks during the early morning hours (06:00 to 08:00) and late evening hours (17:00 to 19:00). Vehicular activities near the resident site dominate during those peak hours (Figure6b). A sensitive (school zone) site, Kamarajapuram is selected for its 4 schools and 1 college covering about 3km inside the city, with >10,000 students using non-fossil fuel based vehicles. This sensitive site has the lowest EBC concentration when compared to the other sites. This site also shows two peak concentrations which are dominated by FF%
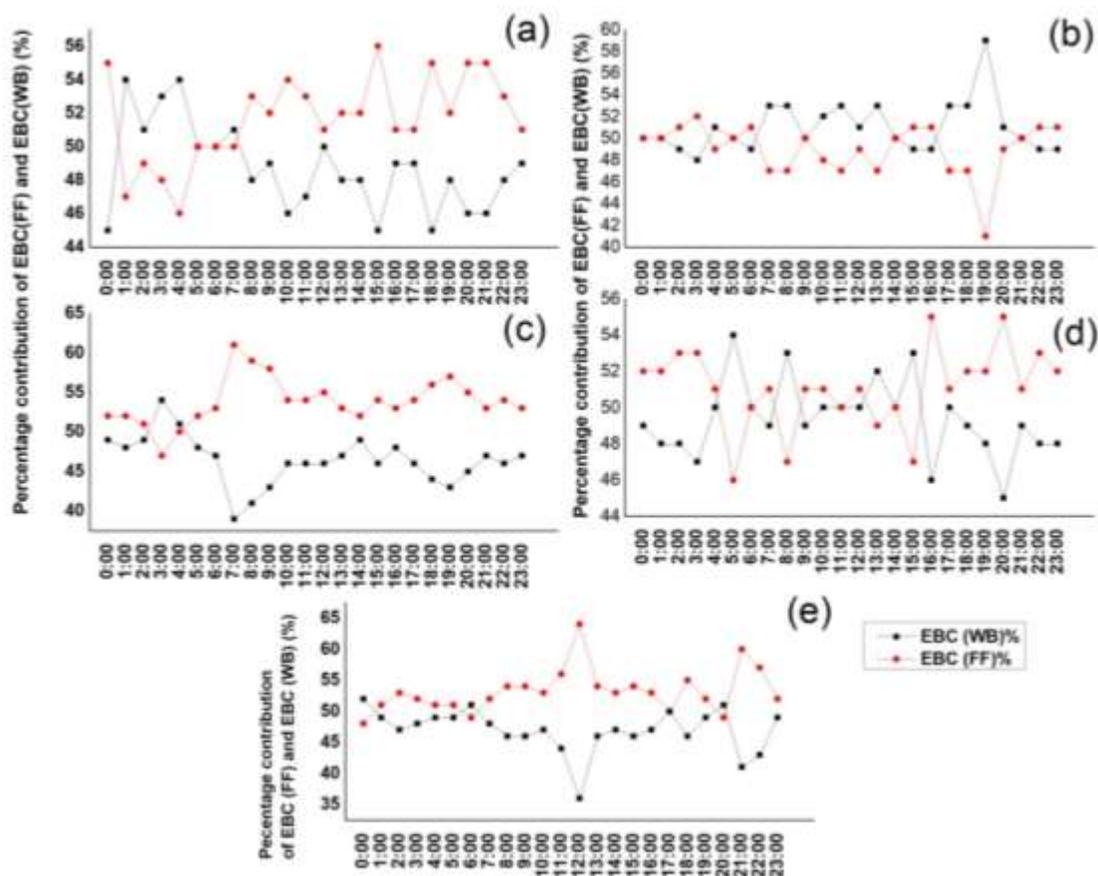
Figure 6: The percentage contribution of fossil fuel (FF) and biomass (WB) combustion source to the total EBC concentration measured at different sampling sites (a) Educational site (b) Residential site (c) Sensitive site (d) Industrial site (e) Traffic site on an hourly scale.

emitted from the HDV (school buses). No outside HDV vehicles are allowed during the daytime inside this site and due to this, reduced EBC concentration is observed at this study site. The minimum biomass contributions are from the school and college canteen where biomass is used as a major fuel for cooking free-of-cost food for the school students (Figure6c). Keerathurai with >20 paddy processing mills, each with huge boilers that depends on biomass as the primary fuel for processing the paddy is selected as an industrial site and their diurnal EBC variation is studied. During every Wednesday and Saturday, about a ton of wood is brought into at least 14 mills for the combustion during the rest of the weekdays. The diurnal EBC concentration shows more than two peaks and EBC from WB% dominates during the daytime while the FF% during the late night hours. The FF% is from the HDV vehicles which takes the processed paddy to the other locations only during the night hours (Figure 6d). The traffic sites, Simmakal, Kalavasal, and Periyar, with more than 40,000 vehicles

circulating cumulatively around these locations every day with some additional vehicles during the weekend days are considered for this study. The diurnal variation of EBC at these study sites shows two major peaks during early morning hours and during noon hours. The FF% contribution to EBC is dominant during almost all the study hours (Figure 6e).

The seasonal average values of AOD are high during summer (0.46 ± 0.11) and monsoon season (0.40 ± 0.04) and minimum during the winter (0.38 ± 0.04) and post monsoon season (0.37 ± 0.08). The values of AOD observed during the study period ranged from (0.41 to 0.60). Increased surface heating and varying wind speed causes vertical mixing of aerosols that result in increased AOD values during the summer season of the study period. Gas-to-particle conversion processes and weak generation of aerosols during the winter and post monsoon seasons causes reduced AOD (Yogesh et al., 2012; Indira et al., 2013) values during the study period. EBC mass concentration measured were analyzed and compared with the AOD values retrieved from MODIS Terra satellite data for the study period at Madurai The seasonal variation of AOD, the EBC mass concentration has been studied and it was found that there was a negative correlation between them. The correlation coefficient value found between them was 0.69. This negative correlation indicates that there were particles other than EBC, which may be with size larger than EBC could be found and that might influence the AOD in that locality.

4. Conclusions

The mean EBC mass concentration measured using Aethalometer AE-31 at Madurai during June-2014 to May-2016 is 5.10 ± 2.40 µg m^{-3}. The mean seasonal EBC values are 8.97 ± 2.34 µg m^{-3}, 4.69 ± 2.14 µg m^{-3}, 6.70 ± 3.83 µg m^{-3} and 3.34 ± 2.53 µg m^{-3} during the winter, summer, monsoon and post-monsoon seasons, respectively.

a) Major reasons for the variability in aerosol concentration with respect to different seasons are due to changes in i) amount of rainfall received, ii) synoptic wind patterns, iii) atmospheric boundary layer (ABL) height that varied as 913, 955, 1083, and 683 m, during winter, summer, monsoon and post-monsoon seasons respectively, iv) aerosol source strength and v) land use patterns.

b) The average diurnal variation of EBC shows two peaks, one in the early morning hours (Between 6:00 to 9:00 hours (LT)) and the second during evening hours (19:00 to 22:00 hours (LT)), while the concentration decreased gradually during noon hours and a stable EBC mass concentration is observed. Heavy duty vehicles are allowed in the city only during nighttime between 22:00 and 06:00, but there are public transport buses and other traffic vehicles running all the day.

c) The seasonal (Nighttime / Daytime) ratios are found to be 0.82, 1.0, 0.79 and 0.76 during the winter, summer, monsoon and post-monsoon, respectively. The increase in tourist activity results in the increased Nighttime EBC concentration during the summer season.

Understanding the sources of EBC, their geographical distribution and future changes are therefore important to improve climate modelling and would support the development of policies, exploring climate co-benefits of air pollution regulation controlling sources of BC.

Acknowledgements

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