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# Characteristics of Black Carbon Aerosol at an Educational Site in Southern India

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## Abstract

Black carbon (BC) aerosol formed from incomplete combustion of fossil fuel and biomass combustion is collected at an educational institute, Madurai Kamaraj University, Madurai located in Southern India. The sampling is done using an Aethalometer (AE-31) from January to March (2015 to 2017) to study the effect of meteorological parameters and anthropogenic activities during the transition from winter to summer. BC identified from a variety of sources showed strong diurnal variation with two peaks, first one at early morning hours and the ot. her one at evening hours, but this peak timings varied with the seasons. It is observed that biomass combustion and tobacco smoke at the study site dominates other sources. Raman spectral analysis showed strong organic source peak.

***Keywords:*** Black carbon; Educational Institution; Tobacco smoke; Raman Spectrum

## 1. Introduction

Aerosols are quite ubiquitous; airborne particles from volcanic eruptions, cigarette smoke, smoke from power generation, and salt particles formed from ocean spray (Pramod *et al.* 2011). Incomplete combustion of fossil fuel, biofuel, and biomass result in the formation of organic carbon (OC), and black carbon (BC) aerosols (Ramachandran and Rajesh, 2017). BC is one among the most important constituent of ambient particulate matter and it remains inert in the atmosphere based on its predominant sub-micron size and chemical structure. Globally emitted BC aerosols from anthropogenic activities were estimated from bottom-up approaches, and are reported to be 7500 Gg/year, but with a large uncertainty range of 2000–29000 Gg/year (Bond *et al.*, 2013 and Kumar *et al.*, 2015). BC aerosols accumulate in the tropospheric region and disturb the radiative balance with its strong solar radiation absorption characteristics (Bond *et al.*, 2013, Jacobson, 2014). Due to its efficient light absorbing nature, BC exerts a positive direct radiative forcing on Earth's climate between +0.08–1.27 W/m$^2$ (Bond *et al.*, 2013). BC aerosols stay in the atmosphere for a very short duration (few days to weeks). During the atmospheric process, freshly formed BC aerosols are usually hydrophobic, while aged particles turn into hygroscopic with the coating of other pollutants. According to Bond and Bergstrom (2006), mass absorption cross-section of freshly formed uncoated BC

ranges between (4 to 8) $m^2/g$ whereas due to the pollutants coated BC results in the increase of mass absorption cross-section. Different source coating results in different wavelength absorption (Bond and Bergstrom, 2006). A hematite form of iron oxides which is an important composition of aerosols absorb visible light at wavelengths < 600nm (Sokolik and Toon, 1999; Yang *et al*., 2009). Studies conducted by Gadhavi and Jayaraman (2010) and Tiwari *et al*. (2014) during winter using aethalometer found that 20–40% and ~6% of the BC originates from biomass combustion measured at 370 nm. The probable major source of BC can be identified by the value of the absorption angstrom exponent (AAE).

BC is a major part of particulate matter, with a size less than 2.5 microns ($PM_{2.5}$) (Paliwal *et al*., 2016). Raman Spectroscopic analysis of particulate matters (PM) gives information about its composition. The Raman spectroscopic analysis on PM depends on the vibrational modes exerted by them (Ivleva *et al*., 2007). The scattered light produced from PM, which depends upon the lattice arrangement shows specific wavelength shifts. These shifts can be used to identify different composition. The Graphitic (G) band and Distorted (D) band produced by carbonaceous particles reveals information about the type and sources. According to Ivleva *et al*., 2007, the shapes of the bands helped to understand about the humic like substance from diesel soot. Temperature history and reactivity of the soot samples also can be determined from the bands (Carrara *et al*., 2009). Mertes *et al*. (2004) used quartz fiber filters for capturing Elemental carbon (EC) aerosol, which is a refractory constituent. Quantitative Raman spectroscopy study on EC showed Raman intensities between 510 $cm^{-1}$ and 1736 $cm^{-1}$. EC exhibits near similar trends in seasonal variations as BC aerosols. The absorption coefficient of EC is the essential input parameter for direct radiative forcing calculation.

In this experimental study, continuous measurement of black carbon aerosols in relationship with the meteorological parameters is conducted at a semi-arid site. The study is carried down from 2015 to 2017 to understand the light attenuation and absorption properties of aerosols during the transition between winter to summer months (January to March). The boundary layer dynamics, sunrise and sunset timings, meteorological conditions and black carbon concentration vary significantly during the transition between the winter and the summer months at the study site. This study helps to comprehend the optical properties and identify the major sources of BC during winter and post winter month.

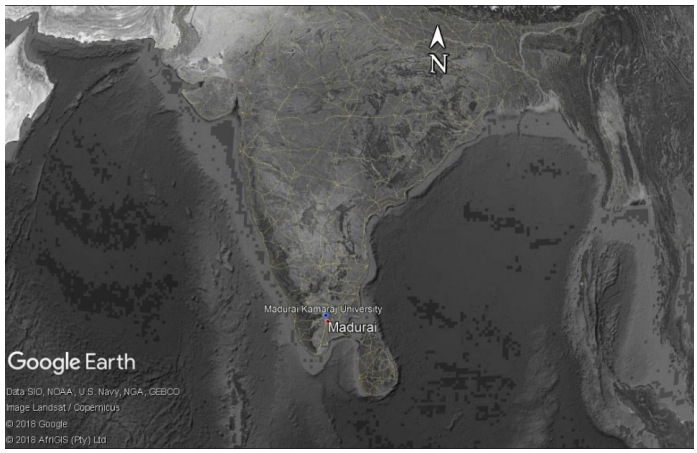


**Figure 1**. Google Earth imagery showing the study site Madurai Kamaraj University (9.94° N, 78.01° E) located at an semi-arid location in Southern India

## 2. Materials and Methods

*2.1. Study Site*

Madurai (9°91'N, 78°54'E), located 130 m above mean sea level is one among the emerging mega city. It is the second largest and one among the most densely populated city in the southern India covering a surface area of 52.8 km$^2$ and a population of 3.2 million in 2011 while it was 0.25 million in 2001, excluding the ~0.1 million floating population. The sampling was carried down at Highway cum vegetation covered rural location in Madurai (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) (Figure 1). During sampling the outdoor air temperature ranged from 22 to 31 °C with a relative humidity range of 62–90%. Windspeed are generally <5 m/sec and frequently from the northeast and southwest directions (Table 1).

*2.2 Instrumentation*

Black carbon mass concentration is measured using seven-wavelength (370, 470, 520, 590, 660, 880 and 950 nm) based AE-31 model Aethalometer (Magee Scientific Corporation, USA). The aerosols are collected using an inlet tube and a pump which operates at a stable airflow rate about 3 L/min with a time interval of 5 min. Aethalometer depends on filter based technique to measure the variability in light attenuation due to particles deposited onto a filter.

In the aethalometer, the measured attenuation is linearly proportional to the BC mass deposit. Hence, by using the wavelength dependent calibration factors, the light attenuation absorption coefficient is converted into BC mass concentration (Hansen, 2005 and Yang *et al.*, 2009). The babs in relation to the change in attenuation are given below as:

$$b_{abs}(\lambda)=\frac{1}{CR(ATN)}\times\frac{\Delta ATN(\lambda)}{\Delta t}\times\frac{A}{Q} \qquad \text{Eq.1}$$

where, $b_{abs}$ is the absorption coefficient, ΔATN, the ratio of the intensities of incoming light to the remaining light after passing through an optical path, for sensing beam and the reference beam respectively, $s_{atn}=\frac{14625}{\lambda}$, the specific attenuation cross section; A, the spot area and Q, the volume of air passed through the filter. The parameters C and R (ATN) were correction factors for minimizing the uncertainty in the readings associated with Aethalometer. The light absorption measured at 880 nm is considered to represent the effect of BC (Bodhaine, 1995) as there were no other major aerosol species which exhibits an absorption at that wavelength. The absorption angstrom exponent (AAE) is a measure of spectral dependence of aerosol absorption (Park *et al.*, 2006; Sandradewi *et al.*, 2008; Ajtai *et al.*, 2011; and Srivastava *et al.*, 2012).

$$b_{abs}(\lambda)=\beta\lambda^{-AAE} \qquad \text{Eq.2}$$

where, β is the particle loading, a constant and λ is the wavelength. Using the power law equation (2) and by performing a linear regression of ln ($b_{abs}$) and ln (λ) (Kirchstetter *et al.*, 2004) AAE is calculated. BC concentration measured using ATN at 880 nm channel with a $\sigma_{atn}$ value of 16.6 m$^2$/g is considered standard (Nair *et al.*, 2007; Beegum *et al.*, 2009; Gadhavi and Jayaraman, 2010; Kumar *et al.*, 2011; Dumka *et al.*, 2013). The calculation of Delta-C suggest-

**Table 1**. Monthly mean values of Air temperature, Windspeed, Relative Humidity, Sunrise, Sunset timings and Ventilation coefficient at the study site during the study period.

| Months | Air temperature (°C) | Windspeed (m/sec) | Relative Humidity (%) | VC (m$^2$/sec) | Sunrise time (LT) | Sunset time (LT) |
|---|---|---|---|---|---|---|
| January | 27 ± 1 | 3.3 ± 1.2 | 71 | 3.42 | 6:34 | 18:07 |
| February | 28 ± 2 | 2.7 ± 3.0 | 62 | 3.10 | 6:32 | 18:22 |
| March | 31 ± 1 | 1.9 ± 0.8 | 65 | 2.04 | 6:15 | 18:28 |

ed by Allen *et al*. (2004) serve as an indicator of wood burning particles, but not a direct quantitative measurement of mass concentrations.

$$DeltaC = BC_{(370nm)} - BC_{(880nm)}$$

Thermo electrically cooled charge coupled device based HORIBA Jobin Yvon LabRam HR 800 spectrometer is used to study the micro-Raman spectrum. A 1800 grooves $mm^{-1}$ and 2400 grooves $mm^{-1}$ holographic grating is used to equip the spectrograph. The power of He-Ne (633 nm) and mixed gas laser line (488 nm) is 17 and 55 mW respectively. The size of the laser spot on the sample is approximately 1 μm in diameter. The collection time of the spectrum is 3 seconds and a 40X objective is used to focus and collect the scattered signal.

## 3. Results and Discussion

*3.1 Diurnal variation of BC concentration*

Continuous BC aerosol mass concentration sampling is carried out for 24 hours per day during the months January to March during the years 2015 to 2017 and their mean concentration values are calculated for those months using equation 1. The boundary layer dynamics, sunrise and sunset timings, meteorological conditions and black carbon concentration vary significantly during the transition between the winter and the summer months at the study site. Figure 2 shows the monthly mean diurnal variation of BC aerosols with vertical bars denoting the standard deviation. It is evident from the graph that for all the three months, sharp peaks are observed during the 6:00 to 9:00 hours and 19:00 to 22:00 hours. The BC concentration decrease gradually during noon hours and a stable BC mass concentration is observed.

The sunrise and sunset timings during January to March is shown in table 1. The time interval between sunrise and sunset is the length of the day (hours) and the remaining interval is the length of the night (hours) (Ramachandran and Rajesh, 2017). According to Ramachandran and Rajesh (2007), as a result of fumigation effect, during the early morning sunrise hours the first peak in diurnal BC mass concentration is observed. Early morning hour's solar heating causes lifting of aerosol particles which are confined to the surface during the night hours. The early morning concentration peak timing varies with season, because the change in the sunrise timings during different seasons is directly proportional to the peak timings (Latha and Badarinath, 2005; Tripathi *et al*., 2005; Safai *et al*., 2007; Nair *et al*., 2007). According to Ramachandran and Rajesh (2017), as a result of fully evolved boundary layer and increased solar radiation during the noon hours, there is a decreased BC concentration during those hours. Decrease in boundary layer height and increased cooking and other anthropogenic activities during the evening causes the second peak (19:00 to 22:00 hours).

BC aerosol concentration in the study site is analyzed using the absorption angstrom exponent (AAE) analysis (Equation 2). The AAE values showed that the values ranged from 0.9 to 1.8 with the average value of 1.2. According to Kirchstetter *et al*. 2004, and Herich *et al*. 2011, the values of AAE less than or equal to 1.0 indicates the dominance of fossil fuel burning and the value around 1.5 represents BC mixed with larger sized mineral dust particles, whereas the AAE for soot particles evolved from biomass combustion ranged from 1.5 to 3.0. As the study site is located away from the polluting urban environment, industrial and brick kiln sources impacts are very low. Therefore, the most common known sources emitted are from fossil fuel (ff) and biomass wood burning (wb) at the study site. An emission inventory was done for this study site based on the sources of BC concentration and from the study it was concluded that the two most dominating sources were biomass and fossil fuel combustion (Bhaskar *et al*., 2018). Emissions from the brickkiln and forest fire were very low compared to the biomass and fossil fuel sources because the study site was completely isolated from such activities. Tobacco smoke emissions were included in the study.

Figure 3(a) show the diurnal variation with consistent changes in BC concentration contributed from fossil fuel sources ($BC_{ff}$) and wood burning sources ($BC_{wb)}$. During the study period, $BC_{ff}$ contributes in larger account to the BC throughout January with peek values during

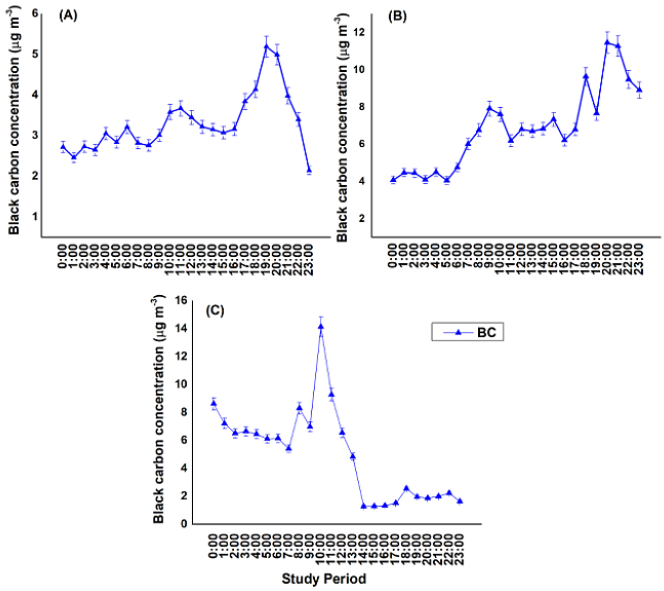


**Figure 2.** Hourly mean diurnal variation of BC concentration measured during the study period

late night hours extending up to the early morning hours. Figure 3(b) shows the percentage contribution of ($BC_{ff}$) and ($BC_{wb}$) during day and night hours. The percentage contribution of $BC_{ff}$ during January is due to the increased flow of heavy tonnage vehicles (HTV) containing essentials for the Pongal, a local festival and also due to the increased population reaching university after the vacation days (extending from second week of December up to first week of January). The number of vehicles crossing the study site ranges from 1000 to 5000 per day during January, while the vehicular population range changes from 500 to 1000 per day during February and March. During February and March months biomass contribution $BC_{wb}$ to the total BC is found higher during both morning and evening hours. The contribution of $BC_{wb}$ is higher by a factor of 2–4 than $BC_{ff}$ contributions The percentage contribution of $BC_{wb}$ is high during February, because of an increased usage of biomass combustion including the burning of crop residue (mostly sugarcane residue) (Bhaskar *et al.*, 2018) and the decreased flow of vehicles. The number of rainfall events during the study period plays a vital role in the decreased BC concentration compared during March, because the total number of rainfall events in January and February combined is 21 while in March alone is 30 to 45 events per year. The rainfall events recorded in March is mostly during the daytime hours. A pronounced evening peak in $BC_{wb}$ throughout the March was due to the significant and dominant wet biomass combustion activity, while during night to early morning hours lot of fossil fuel contribution is recorded.

To evaluate the dominance of $BC_{wb}$, Delta-C analysis is carried out and the results are plotted in Figure 4 and it shows a strong diurnal variation. According to Hopke *et al.* (2011), if wood combustion smoke is present then Delta-C values are positive, if not, then the values would be negative and the negative values might be from the filter between consecutive sampling, during which the desorption of UV absorbing semi-volatile organic species occurs. The changes Delta-C values during January are found to be negative in day and night hours indicating the dominance of $BC_{ff}$ activities, while during February and March there is an increased positive Delta-C during day time. The gradual buildup of early morning Delta-C values during February month likely corresponded with an increase of residential wood burning emissions after sunrise.

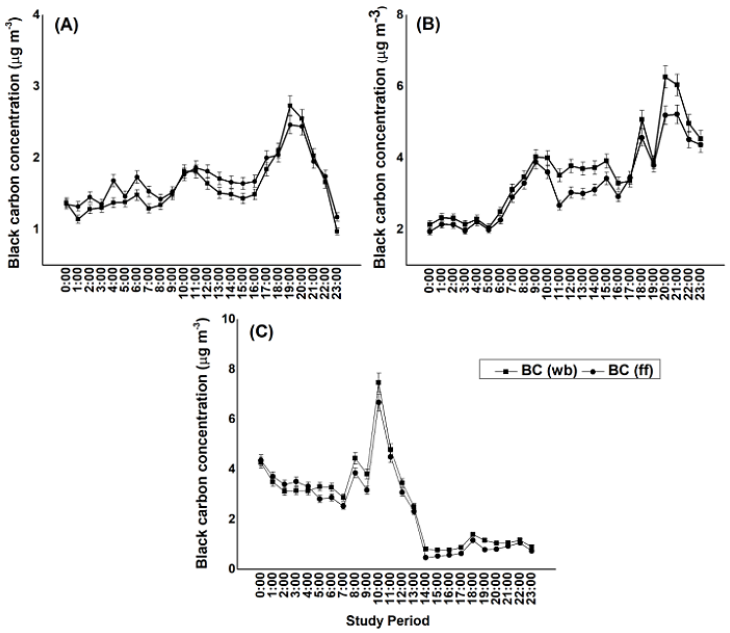


**Figure 3(a).** Hourly mean diurnal variation of BC concentration contributed from $BC_{ff}$ and $BC_{wb}$ (a) January (b) February and (c) March

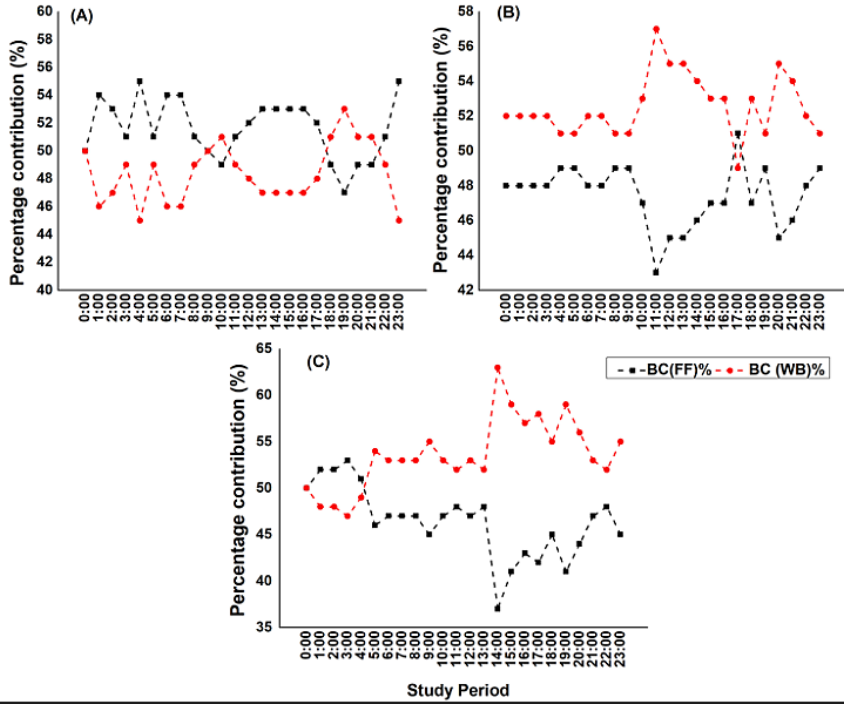


**Figure 3(b)**. Mean percentage contribution of $BC_{ff}$ and $BC_{wb}$ to the total BC concentration (a) January (b) February and (c) March

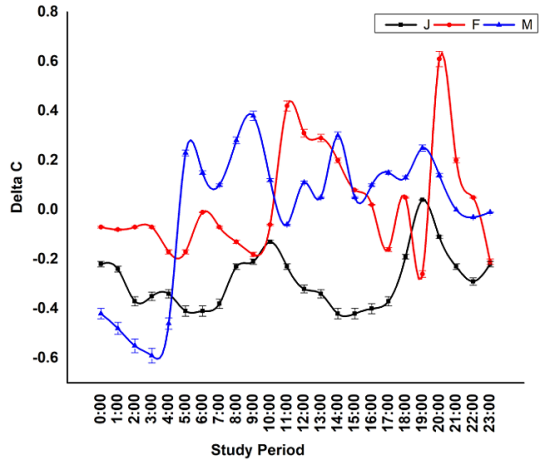


**Figure 4.** Delta-C analysis for the study period

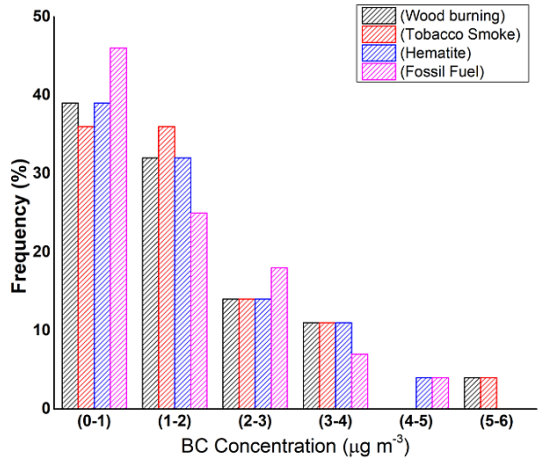


**Figure 5(a).** Frequency distribution (%) of BC concentration from different sources

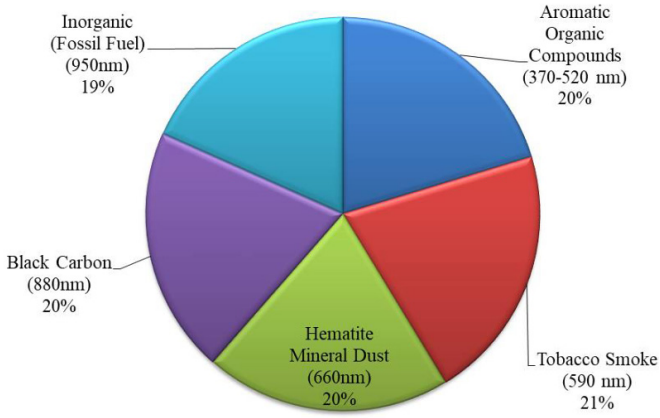


**Figure 5(b).** Pie chart showing the BC concentration from different sources measured at different wavelength

*3.2 Contribution of different sources*

Sources other than fossil fuel and biomass contributing to the total BC concentration which includes Tobacco smoke and Hematite mineral dusts are recorded at different wavelengths 590nm and 660nm, respectively using the aethalometer and analyzed. The preferences of wavelengths for such sources were obtained from the aethalometer manual (Hansen, 2005). During the study period the BC from different sources are classified and it ranged from (0.22 to 8.69) μg $m^{-3}$ and their frequency distribution based on the concentration is shown in figure 5(a). From the frequency distribution, higher concentration of BC >5 μg $m^{-3}$ are purely from wood combustion and tobacco smoke. Figure 5(b) shows the percentage distribution of BC from different sources. From the results it is observed that, at Madurai Kamaraj University the percentage contribution from tobacco smoke dominates over the other sources. Tobacco smoke contributes with a major share of 21%, 21% and 23% during the months of January, February and March respectively. Diurnal variation of BC tobacco smoke (BCts) contribution to the total BC is analyzed and plotted (Figure 6). Table 2 shows the studies from other sites.

*3.3 Molecular Analysis*

This study is based upon the aerosol morphology, its light absorbing characteristics and the nature of internally mixed soot particles (Liousse *et al*., 1993; Bond and Bergstrom, 2006). The ability of BC aerosols to absorb light arises from the molecular structure which is characterized by the valence electrons in the π- orbitals. Small energy gaps between bonding and anti-bonding orbitals allows the BC aerosols to absorb visible light. Based on the Raman Spectrum (RS) mode analysis of Ivleva *et al.* (2007), G band occurrence at 1576 to 1585 $cm^{-1}$ confirms the graphitic nature (hexagonal) of the observed samples, as black carbon aerosols are graphitic in nature and the occurrence of 1325 $cm^{-1}$ shows the non-hexagonal structure that represents the organic substance present in the sample. For the samples collected during February and March the Raman intensity for D band is high, confirming the dominance of organic sources (Figure 7).

BC mass concentration of different anthropogenic sources, including fossil fuel ($BC_{ff}$), wood burning ($BC_{wb}$) and from other sources are investigated during the years 2015 to 2017 at an educational institution located in Southern India. Higher BC concentration is observed during winter (January-February) and first month of summer (March) when compared to other monsoon and post monsoon months at the study site. The major findings and results obtained from the study are
ummarized as follows:

**Table 2.** Black Carbon concentration over different study locations

| Location | Location Type | Duration | BC (μg/$m^3$) | Reference |
|---|---|---|---|---|
| Mumbai | Urban, Industrial | January to March 1999 | 12.5 | Venkatraman *et al.* (2005) |
| Hyderabad | Urban, Semi-arid | January to December 2003 | 10.0 | Latha and Badrinath (2005) |
| Anantapur | Rural, Semi-arid | January to December 2010 | 5.05 | Reddy *et al.* (2012) |
| Agartala, Tripura | Rural, Continental | September 2010 to September 2012 | 17.8 | Guha *et al.* (2015) |
| Madurai | Urban, Semi-arid | January to March 2015 to 2017 | 1.62 | This study |

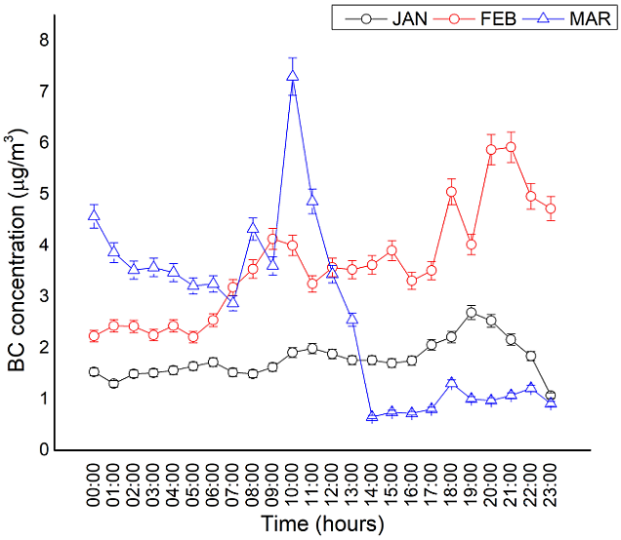


**Figure 6.** Hourly mean diurnal BCts concentration during the study period.

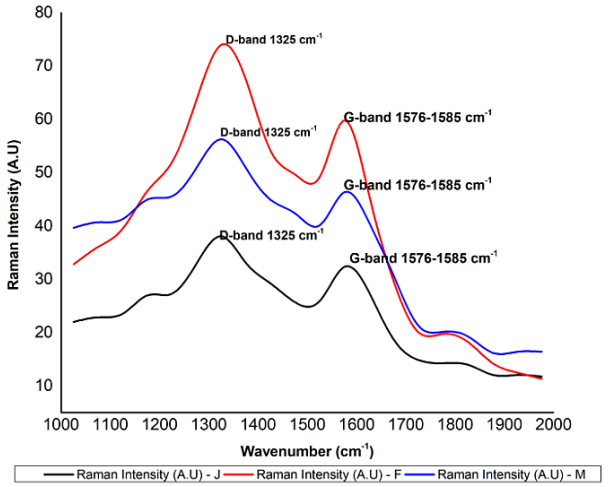


**Figure 7.** Raman spectral analysis of samples collected during the study period.

BC, exhibits a strong monthly variability with 8.69 µg $m^{-3}$ during February and a low 0.22 µg m-3 during March. Higher BC during January and February arise due to shallow winter atmospheric boundary layer associated with low wind speeds, and significant increase in the amount of fossil fuel combustion and biomass burning, while lower BC concentration during March months are due to wet removal of aerosols and elevated boundary layer height.

Due to the diurnal changes in strength of BC emission sources and boundary layer height, the BC mass concentrations exhibit strong diurnal variation. During the noon hours, the BC, $BC_{ff}$, and $BC_{wb}$ mass concentration values are high throughout the study period because of the fully evolved boundary layer and reduced anthropogenic activities. The contribution of $BC_{wb}$ was higher by a factor of 2–4 than $BC_{ff}$ and dominates throughout the day, which indicates persistent anthropogenic activities. In addition, contribution from tobacco smoke ($BC_{ts}$) at the study site dominates over the $BC_{ff}$ and $BC_{wb}$ contributions. Highest $BC_{ts}$ (23%) and $BC_{wb}$

(22%) contribution is observed during March.

Using LabRam HR 800 system coupled with 325 nm excitation laser lines, the Raman mapping of sample was carried out. The Raman spectral analysis shows the domination of BC aerosols produced from biomass sources during the study period.

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