Supplementary Material for

Sources and Radiative Impact of Carbonaceous Aerosols Using Four Years High Resolution Ground-Based Measurements Over the Central Himalayas

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\textbf{Supplementary Text – S1, S2}

\textbf{Supplementary Tables – Table S1, S2}

\textbf{Supplementary Figures – Figure S1, Figure S2, Figure S3, Figure S4, Figure S5, Figure S6, Figure S7a, b, Figure S8}

\textit{Supplementary Text}

S 1. Description of OC segregation methods

The basic principle involved in this EC tracer method can be expressed by the following two equations:

\begin{equation}
POC = [OC/EC]_{pri} \times EC + c \quad (1)
\end{equation}

\begin{equation}
SOC = OC - POC \quad (2)
\end{equation}

where POC, SOC, OC, [OC/EC]_{pri} and c represent primary organic carbon, secondary organic carbon, organic carbon, the ratio of primary OC to EC and contribution from non-combustion sources/sampling artefacts, respectively (Cabada et al., 2004).

We have utilised four methods to get the [OC/EC]_{pri}. The first method referred to as ‘TEN’ implies calculating [OC/EC]_{pri} from the lowest ten percentiles of the observed OC/EC ratio. The second method which we refer to as ‘REG’ uses the regression slope of OC–EC plot as the
As opposed to ‘TEN’, this method may also represent the non-combustion sources since it considers the intercept in the above-mentioned regression plot while calculating POC. The third method, to be called ‘CAB’, determines the regression slope as in the former but from the dataset in which a period of high SOC formation is removed (Cabada et al., 2004). To ensure that the primary OC–EC ratio is dominated by the most probable period of primary OC–EC emissions and not from the periods of secondary emissions, or rain, we have employed an approach as suggested by Cabada et al. (2004). In this method, firstly, the dataset with rain affected period was removed, following which, in the remaining set, only the night-time (1900 to 0500 hours) data was used to avoid daytime periods of high secondary formations. Even in this filtered data set, we further confine our data only to the period of high correlation between eBC and CO ($r^2 > 0.5$, $p < 0.0001$). This criterion of correlation is used to confine the dataset to a period of most probable high primary emissions. The slope thus determined from this approach is used in evaluating POC and hence SOC. The fourth method called ‘MRS’ uses the Minimum Regression Square method to get $[\text{OC} / \text{EC}]_{\text{pri}}$ (Wu and Yu, 2016; Wu et al., 2019). In this method, the OC/EC corresponding to the minimum $r^2$ (between EC and a range of possible SOC values) represents the actual $[\text{OC} / \text{EC}]_{\text{pri}}$ (Wu and Yu, 2016). To account for the diurnal dynamics, the $[\text{OC} / \text{EC}]_{\text{pri}}$ for each hour in each month is calculated using MRS and then the minimum of these hourly $[\text{OC} / \text{EC}]_{\text{pri}}$ ratios is taken as the actual monthly value of the $[\text{OC} / \text{EC}]_{\text{pri}}$ (Wu et al., 2019).

**S 2 Method for segregation fossil fuel burning and biomass burning components in eBC**

The absorption coefficient ($b_{\text{abs}}$) at a given wavelength denotes light absorption by aerosols and is proportional to $\lambda^{-\alpha}$ where $\alpha$, depends on the emission sources i.e., fossil fuel or biomass. This relation can be employed for quantifying the source apportionment of ambient BC concentrations (Sandradewi et al., 2008). This method relies on two basic assumptions:
1. Total absorption coefficient ($b_{\text{abs, total}}$) at a given wavelength is simply the sum of absorption coefficient due to fossil fuel combustion ($b_{\text{abs, ff}}$) and biomass burning ($b_{\text{abs, bb}}$).

$$b_{\text{abs, total}} (\lambda) = b_{\text{abs, ff}} (\lambda) + b_{\text{abs, bb}} (\lambda)$$

(4)

2. Absorption from burning of fossil fuel and biomass follows different spectral dependencies given by,

$$\frac{b_{\text{abs, ff}} (\lambda_1)}{b_{\text{abs, ff}} (\lambda_2)} \left(\frac{\lambda_1}{\lambda_2}\right)^{-\alpha_{\text{ff}}}$$  \hspace{1cm} (5a)

$$\frac{b_{\text{abs, bb}} (\lambda_1)}{b_{\text{abs, bb}} (\lambda_2)} \left(\frac{\lambda_1}{\lambda_2}\right)^{-\alpha_{\text{bb}}}$$  \hspace{1cm} (5b)

There are no combustion industries in the mountainous Nainital region and therefore the first assumption is valid to a high degree for the current site as the emissions from other sources are not significant. The second assumption is met since the absorption due to biomass burning dominates in the lower (370-520 nm) wavelength range while that due to fossil fuel combustion is the dominant term in the higher (660-950 nm) wavelength (described in section 4.1.). To avoid absorption due to volatile organic compounds or other absorbing non-BC particles in the 370 nm channel, light absorption measurements are instead taken at $\lambda_1 = 470\text{nm}$ and $\lambda_2 = 950\text{nm}$ for source quantification (Zotter et al., 2017). Ångström exponents for fossil fuel ($\alpha_{\text{ff}}$) and biomass burning ($\alpha_{\text{bb}}$) are chosen to be 0.9 and 1.68 as recommended by Zotter et al. (2017).

Using equations 4, 5a and 5b, the concentrations $eBC_{bb}$ and $eBC_{ff}$ can be determined as:

$$eBC_{bb} = \frac{b_{\text{abs, bb}} (\lambda_2)}{MAC(\lambda_2)}$$  \hspace{1cm} (6)

$$eBC_{ff} = \frac{b_{\text{abs, ff}} (\lambda_2)}{MAC(\lambda_2)}$$  \hspace{1cm} (7)

where,
\[
b_{abs,bb}(\lambda_2) = \frac{b_{abs}(\lambda_1) - b_{abs}(\lambda_2)(\frac{\lambda_1}{\lambda_2})^{-\alpha_{ff}}}{(\frac{\lambda_1}{\lambda_2})^{-\alpha_{bb}} - (\frac{\lambda_1}{\lambda_2})^{-\alpha_{ff}}} \quad (8)
\]

\[
b_{abs,ff}(\lambda_2) = \frac{b_{abs}(\lambda_1) - b_{abs}(\lambda_2)(\frac{\lambda_1}{\lambda_2})^{-\alpha_{bb}}}{(\frac{\lambda_1}{\lambda_2})^{-\alpha_{ff}} - (\frac{\lambda_1}{\lambda_2})^{-\alpha_{bb}}} \quad (9)
\]

Importantly, for the segregation, \( b_{abs} \) values are converted to eBC mass concentrations using monthly mass absorption cross-section - MAC (950 nm) values for the present sites. These MAC values are not the same as the default ones used in the Aethalometer, but these are derived specifically for this site and wavelength using simultaneous EC measurements. Details for MAC determination and the monthly values can be found in Srivastava et al. (2021).

This method is applied in different studies which have demonstrated that it is reliable for estimating the different sources of eBC (Favez et al., 2010; Herich et al., 2011; Crilley et al., 2015; Zotter et al., 2017). The outcomes have also shown reasonable agreement with studies utilising radiocarbon isotope determination (Sandradewi et al., 2008; Zotter et al., 2017). For instance, Zotter et al. (2017) used data at eight different stations with different characteristics and showed that the source apportionment results from Aethalometer and \(^{14}\)C are well correlated and the fitting residuals exhibit only a minor positive bias of 1.6%. Further, considering the reliable performance of this method, it has also been used for source apportionment studies at several other sites in the world, like those at Athens (Kalogridis et al., 2018), Los Angeles Basin (Mousavi et al., 2018) and other sites in India (Kumar et al., 2020; Palle et al., 2021).
Table S1. Monthly values of [OC/EC]_{pri} as obtained from REG, TEN, CAB and MRS methods.

These four methods are defined in the section 4.2.1.

<table>
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<th>TEN</th>
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<th>MRS</th>
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<td>2.45</td>
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Table S2. Monthly values of site-specific mass absorption cross-sections (MAC) in m$^2$/g.

<table>
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Fig. S1. The diurnal variation in OC/EC ratio averaged for the four seasons during 2014-2017. Error bars represent one standard deviation from the mean.

Fig. S2. Diurnal variation of percentage contribution of fossil fuel (eBC$_{ff}$) and biomass burning (eBC$_{bb}$) components during 2014-17.
Fig. S3. Monthly variations in the diurnal amplitudes of eBC$_{ff}$ and eBC$_{bb}$ observed during 2014-2017. The percentage difference between them is also shown.

Fig. S4. The seasonal mean concentrations of eBC along with fossil fuel (eBC$_{ff}$) and biomass (eBC$_{bb}$) components during four seasons during 2014-2017. Percentage contributions of eBC$_{ff}$ and eBC$_{bb}$ are also shown.
Fig. S5. Correlation of POC and SOC concentrations (obtained from CAB, REG and TEN methods) with eBC, eBC_{bb}, eBC_{ff} and the boundary layer height during 2014–2017.
Fig. S6. POC and SOC from CAB, TEN and REG during 19 April – 4 May 2016.
Fig. S7. Aerosol subtype with altitude observed by CALIPSO near the study region encircled in red during (a) low fire (22 April) and (b) high fire (1 May) days.
Fig. S8. Average diurnal variation of POC, SOC, OC, EC, RH, wind speed (WS), temperature (T) and solar radiation (sol rad) during 2014–2017. OC, temperature, solar radiation and RH have been scaled down by factors of 3, 5, 150 and 15, respectively, to fit all plots in the same graph.
REFERENCES


Kumar, R.R., Soni, V.K. and Jain, M.K. (2020). Evaluation of spatial and temporal heterogeneity of black carbon aerosol mass concentration over India using three year measurements from


