Source Impact Analysis Using Char-EC/Soot-EC Ratios in the Central Indo-Gangetic Plain (IGP) of India

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ABSTRACT

This study measured the carbonaceous aerosols in the atmospheric fine particulate matter (fPM; $\text{dia.} < 2.5 \mu m$, $n = 102$ samples) above an urban location (Prayagraj) on the central Indo-Gangetic Plain (IGP) for 1 year (December 2016–November 2017). During the study period, the fPM mass concentrations exhibited very high variability, ranging from 22 to 367 (avg. ± SD: 149 ± 87) $\mu g m^{-3}$, and approximately 40% of the annual average consisted of carbonaceous aerosols (organic carbon [OC], water-soluble OC [WSOC], and elemental carbon [EC]). Furthermore, several diagnostic ratios (EC/fPM, OC/EC, char-EC/soot-EC, and WSOC/OC) indicated that the characteristics of the carbonaceous aerosols significantly differed between seasons, with emission sources, seasonal meteorology, and atmospheric chemistry driving the variation in abundance. Specifically, our trajectory analysis revealed an association between northwesterly air masses originating in northwestern India and higher concentrations of fPM and carbonaceous aerosols at the study site, which mainly occurred during the post-monsoon season and winter. However, anthropogenic emissions from local sources dominated the fPM and carbonaceous aerosols in summer and the monsoon season, during which we also observed the influence of air masses of mixed (continental/marine) origin. To identify the sources of the carbonaceous aerosols and evaluate their contributions, we analyzed the char-EC/soot-EC ratio in combination with the OC/EC ratio. Cross plots of these ratios identified biomass burning and fossil-fuel combustion as the largest sources during winter and the post-monsoon season, and summer and the monsoon season, respectively. Our results demonstrate the advantages of utilizing the char-EC/soot-EC ratio rather than the OC/EC ratio as a tracer of these two sources in the IGP region.

Keywords: Indo-Gangetic Plain, Carbonaceous aerosols, OC/EC ratio, Char-EC/Soot-EC ratio, Secondary organic aerosol

1 INTRODUCTION

Carbonaceous aerosols, viz., organic aerosol (OA) and elemental carbon (EC), have been observed as one of the ubiquitous constituents in the atmosphere, playing an active role in environmental health and climate (Penner et al., 1991; Haywood and Boucher, 2000; Jacobson, 2001; Ramanathan et al., 2001; Tiwari et al., 2016; Pal et al., 2018). Depending on the size range of particulates, sometimes a large portion of their total mass is comprised of the carbonaceous species. Furthermore, contributing negligibly in remote areas, they can contribute up to 80% by mass of fine aerosols ($< PM_{2.5}$) over the polluted urban region (Saxena and Hildemann, 1996; Putaud et al., 2004; Hand et al., 2012; Brooks et al., 2019). Organic aerosols can be produced directly by their emissions sources like biomass/biofuel burning (BB) and fossil-fuel combustion (FFC) (referred to as primary organic aerosol [POA]) as well as can be formed through physicochemical transformations (widely referred to as secondary organic aerosol [SOA]) (Finlayson-Pitts and Pitts, 2000; Gelenček, 2004; Rajput et al., 2014). A fraction of organic carbon (OC) is water-soluble and referred to as water-soluble OC (WSOC). The utility of WSOC to serve as a surrogate...
for SOA and/or their origin from BB emission (≤20%) has been suggested previously (Sannigrahi et al., 2006; Snyder et al., 2009; Singh et al., 2017). In sharp contrast to OC, EC is primary in origin and emitted from incomplete combustion activities such as biomass/biofuel burning, vehicular exhausts, and coal combustion (Gelencsér, 2004; Jimenez et al., 2009).

Unlike subject to alteration of OC/EC ratio under atmospheric aging, the char-EC/soot-EC ratio has been found relatively stable, and therefore suggested previously to serve as a robust tracer of their source origin (Han et al., 2007, 2010). Han et al. (2007, 2009, 2010, 2016) have extensively investigated the ratio of char-EC/soot-EC for source identification at different locations in China. However, most of the previous studies conducted over the Indian region have found the utility of OC/EC ratio (and sometimes molecular tracers) to characterize the BB and FFC emissions owing to higher contribution of OC from its predominant source of BB emission (Ram and Sarin, 2010; Pachauri et al., 2013; Rajput and Sarin, 2014; Rajput, 2018; Singh et al., 2018). Using various receptor modeling tools, source apportionment studies have revealed that BB and FFC contribute up to 60% of fine aerosols in urban areas of the upper IGP region (Singh et al., 2017; Jain et al., 2021a, b). In view of the above-mentioned studies, we decided to investigate the plausibility of identifying sources based on char-EC/soot-EC in conjunction with OC/EC ratio at a less explored site located in the Indo-Gangetic Plain (IGP). The present study assesses the atmospheric abundance of carbonaceous aerosols (OC, WSOC, char-EC, soot-EC) in fine particulate matter (fPM, i.e., dia. < 2.5 µm) over a period of 1 year from an urban site at Prayagraj located in the central part of IGP. One of the major objectives of this study is to assess the suitability of char-EC/soot-EC ratio in conjunction with OC/EC for tracing out their emissions from BB and FFC sources in IGP region. Furthermore, concentration-weighted trajectories (CWTs) and cluster analysis were also performed to assess the dominant source region(s) of carbonaceous aerosols that impacted the receptor site during different seasons.

2 METHODOLOGY

2.1 Description of Sampling Location

The sampling site Prayagraj (25°54′N, 81°85′E, 98 m a.s.l.) is situated in the central part of Indo-Gangetic Plain. The Prayagraj is considered a holy city owing to several temples and confluence point of rivers, the Ganga and the Yamuna (Fig. 1). The city covers an area of 70.5 km² and the district has a population density of 1,086 persons km⁻² (as per the census 2011). Over 100 million people from various states of India gather at Prayagraj city in the months of January and February to celebrate the festival of Kumbh. The visible major anthropogenic sources of air pollutants in

Fig. 1. Figure showing sampling location at Prayagraj in the central region of Indo-Gangetic Plain (IGP).
the city include coal-powered thermal power plants and vehicular emissions (both gasoline- and diesel-based). In the outskirts of the city, biofuel burning is carried out for cooking purpose around the year and also for heating purpose during the winter months. Furthermore, from October through February (post-monsoon and wintertime), under the northwesterly wind system, the receptor site is located downwind (~1000 km) of the source region of major agricultural biomass-burning emissions (Rajput and Sarin, 2014).

2.2 Meteorology

The IGP region can be categorized under a humid subtropical climate. The annual mean temperature of Prayagraj is about 26°C. January is recorded as the coldest and May as the warmest month (as per data set from the Indian Meteorological Department [IMD]; Fig. 2). Depending on the prevailing meteorology, four seasons have been studied as winter (December–February), summer (March–June), monsoon (July–September) and post-monsoon (October–November). Except for summer months, relative humidity usually remains more than 50% over the region. Annual average precipitation centers at ~900 mm, of which about 70% occurs during the monsoon period. In terms of wind speed, post-monsoon and winter seasons are characterized by calm weather condition. Furthermore, a shallower boundary layer height restricts the dispersion of pollutants, and thus, post-monsoon and winter seasons are associated with usually polluted events reflected by the lowest visibility.

2.3 Sampling and Chemical Analysis

Aerosol (hereinafter represented as fine particulate matter [fPM] with dia. < 2.5 µm) sampling was conducted using a customized low-volume sampling (LVS) unit which was operated at a continuous flow rate of 30 L min−1 on a pre-baked 47 mm Pallflex® Tissuquartz 2500QAT-UP microfiber filters. Such sampling units have been used to collect fPM in previous studies (Leck

![Fig. 2. Typical meteorological parameters (monthly average) during the study period over IGP (source: www.imd.gov.in).]
and Persson, 1996; Granat et al., 2002; Tiwari and Kulshrestha, 2019). 8-h continuous sampling (09:00–17:00 h) was conducted twice a week (almost on every third day) at the sampling site starting from 1 December 2016–30 November 2017. The sampler was mounted 1.5 m above the surface on the terrace of a building. In order to determine the fPM mass loading, each filter was kept in a desiccator (partially filled with activated silica gel) for ~24 h before measuring initial and final weights using a high-sensitivity microbalance. Filter samples were stored properly at ~4°C in a deep-freezer prior to the analysis. Field blanks were also collected during each month to assess the quality control of the data. The method detection limit (MDL) of the sampler was observed to be 0.54 µg m⁻³ with the uncertainty (as calculated from replicate samples) in the range of 8–10%. A total of 102 samples have been collected from the study site over the 1-year period covering all four seasons, viz., winter, summer, monsoon, and post-monsoon.

Desert Research Institute (DRI) Carbon Analyzer (Model 2001A) has been utilized to quantify the OC and EC using Interagency Monitoring of PROtected Visual Environments (IMPROVE-A) protocol. The thermal/optical reflectance (TOR) method was used for the determination of OC and EC (Chow et al., 2007; Han et al., 2007). A filter punch of 0.5 cm² area was taken from the sample and placed on the analyzer’s sample port. Due to step-wise heating of sample, differential volatilization of OC in four fractions occurs: OC1 at 120°C, OC2 at 240°C, OC3 at 480°C and OC4 at 580°C in a pure He condition whereas three fractions of EC volatilizes at higher temperatures: EC1 at 580°C, EC2 at 740°C and EC3 at 840°C in a mixture of 98% He + 2% oxygen. Pyrolyzed organic carbon (OP) is determined using the optical method as the reflectance reaches its initial value soon after the injection of mixture gas (at 580°C). This procedure corrects the overestimation of EC through continuous monitoring of reflectance during analysis. While the volatilization of OC occurs in the presence of pure He and catalyst MnO₂ at lower temperatures, EC is evolved in the presence of He gas and 2% oxygen at higher temperatures for their final conversion into CO₂. The difference between EC1 and OP is calculated as char-EC and the addition of EC2 and EC3 is defined as soot-EC (Han et al., 2010). Auto-calibration was run after every 8 samples. For the analysis, methane (CH₄) is used as the calibration gas. The repeatability error of OC and EC while analyzing the fPM was found to be in the range of 8–12%, which is within the acceptable limit (Han et al., 2007). The quality control and quality assurance (QC & QA) procedure has been followed as described previously in Cao et al. (2003).

About one-fourth of each filter (3.1 cm²) was used to analyze WSOC. For the analysis, each filter sample was sonicated and extracted thrice with 25 mL (= 10 + 10 + 5 mL) of Milli-Q water in a sterilized glass centrifuge tube. Sonication was done by placing sample tubes in an ultrasonic bath every time at 1 hertz for 30 min (each cycle) for extracting water-soluble fractions completely. Finally, all sample extracts of a particular sample were pooled and then filtered through a 0.2 µm pore-size nylon syringe filter and subsequently analyzed on a total organic carbon (TOC) analyzer (Model TOC-LCPH E200 ROHS; Shimadzu). Potassium hydrogen phthalate (KHP) and sodium carbonate solutions were used to calibrate the instrument. Each sample was injected thrice and analyzed on TOC analyzer to achieve a coefficient of variation (CV) < 3%. A small amount of the extracted sample was also used to analyze K⁺ ions using ion chromatography (IC). The detailed principle and working of IC have been already discussed in previous studies (Tiwari and Kulshrestha, 2019; Mishra and Kulshrestha, 2020). Several field blanks (10% of collected samples of each season) and lab blanks were also analyzed along with the batch of samples for QC & QA.

2.4 Secondary Organic Carbon (SOC) Estimation

Secondary OC was estimated using the EC-tracer method (Turpin and Huntzicker, 1995; Cabada et al., 2004; Srivastava et al., 2018). This method assumes that the minimum OC/EC ratio involves OC contribution only from the primary emission source (Castro et al., 1999; Pavuluri et al., 2011). Accordingly, the minimum OC/EC ratio can be used for the estimation of primary OC (POC) and SOC. In this study, SOC was calculated for each season separately using the following equations:

\[
POC = EC \times \left( \frac{OC}{EC} \right)_{\text{min}}
\]
SOC = OC – POC  

where \((OC/EC)_{\text{min}}\) represents the minimum value of OC/EC ratio for each season. This value is 1.0 in winter, 0.9 in summer, 0.6 in monsoon and 1.1 in post-monsoon season.

3 RESULTS AND DISCUSSION

3.1 Temporal Variability of fPM Mass Concentration

Temporal variability of fPM mass concentration is shown in Fig. 3(a). The concentrations of fPM varied from 22–367 µg m\(^{-3}\) during the entire study, with an annual mean of 149 ± 87 µg m\(^{-3}\) at the site (Prayagraj, central IGP). The annual average fPM concentration was observed to be ~3.7 times higher as compared to the National Ambient Air Quality Standard values (NAAQS; annual average PM\(_{2.5}\): 40 µg m\(^{-3}\)). It can be noticed that fPM concentrations were higher during winter and post-monsoon, followed by that in summer, and lowest in the monsoon season (Fig. 3(a)). The data set revealed that monthly averaged fPM concentration was maximum for November (317 µg m\(^{-3}\)) whereas minimum for August (47 µg m\(^{-3}\)). Our observation on higher concentrations of atmospheric particulates in the month of November is in good agreement with...
the previous studies (Ram and Sarin, 2010; Ram et al., 2012; Pachauri et al., 2013; Singh et al., 2017; Rajput et al., 2018). Those studies have addressed the predominant impact through long-range transport of large-scale agricultural rice straw (hundreds of millions of tons) burning emissions occurring in the northwestern part of the IGP region (Sawlani et al., 2018). Frequent precipitation during the monsoon period from July to September is attributable to wet scavenging of ambient aerosols (Singh et al., 2017). During the peak winter months of December and January, the fPM averaged at 226 and 200 µg m⁻³, respectively. Moderate levels of fPM concentrations were observed during the summer months from March to June, with the lowest mean value in April (99 µg m⁻³). These observed values are nearly comparable to previous studies conducted at different sites of IGP (Kulshrestha et al., 1998; Ram and Sarin, 2010; Satasangi et al., 2012; Singh et al., 2017). The variability in the abundance pattern of ambient aerosols was found to be highly dependent on emission sources, seasonal meteorology, and atmospheric chemistry, which has been discussed in the following sections.

3.2 Temporal Variability of Carbonaceous Aerosols

The annual mean concentration of OC, EC and WSOC in fPM was observed to be 27.3 ± 18.7, 15.1 ± 13.6 and 13.9 ± 10.5 µg m⁻³, respectively. A landscaping exercise was done for comparison in the spatial distribution of mass concentrations of OC, EC and WSOC from this study and studies reported earlier from different parts of India (Table 3). Temporal variabilities in the carbonaceous aerosol species, viz., OC/fPM, EC/fPM, OC/EC, char-EC/soot-EC and WSOC/OC ratio are shown in Figs. 3(b, c, d, e). Overall, OC/fPM varied from 0.02–0.61 (annual avg. ± SD: 0.18 ± 0.11). Likewise, EC/fPM also exhibited a significant variability, ranging from 0.01–0.26 (0.10 ± 0.06). In other words, the OC and EC annually averaged mass fraction (%) in fPM were 18% and 10%, respectively (Figs. 3(b, c)). On average, the total carbonaceous aerosol (TCA = OC × 1.6 + EC) at the present site was found to contribute about 39.4% of the fPM (annual mean: 149 µg m⁻³; Fig. 3(a)). The characteristic ratios of OC/EC and char-EC/soot-EC varied from 0.6–22.2 (2.8 ± 2.8) and 0.1–7.5 (0.7 ± 1.6), respectively, during the entire study period. The WSOC/OC ranged from 0.03–1.00 (0.61 ± 0.29). Furthermore, to understand better the seasonal abundance of carbonaceous aerosols and their characteristic ratios, it is necessary to look at the impact of long-range transport of aerosols through air-mass analyses during different seasons.

3.3 Concentration-weighted Trajectories and Cluster Analysis

In the present study, air-mass backward trajectories (AMBTs) were simulated utilizing the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model with an input of Global Data Assimilation System (GDAS) meteorological data at an atmospheric height of 500 m above mean sea level (Draxler and Rolph, 2003). Due to the longer residence time of fine particulates, 7-day AMBTs were retrieved for each sampling date, the exception being in the winter season wherein 5-day AMBTs were taken into account owing to poor ventilation coefficient during the season. Furthermore, cluster analysis of the fPM data set has been performed for each of the four seasons (Figs. 4(a, b, c, d)). Briefly, the seasonally segregated trajectories were clustered with measured fPM mass concentration based on a pre-existing algorithm of the nearest angle-distance function (Sirois and Bottenheim, 1995). Moreover, we have also carried out the CWT analysis, which is also a receptor model used widely to understand the spatial distribution of potential source regions of air pollutants (Bansal et al., 2019; Rai et al., 2020). In CWT analysis, based on the stretch of AMBTs, the geographic region is firstly sub-divided into a set of grid cells (grid resolution: 0.5° × 0.5°). Subsequently, the CWT method determines the pollutant’s concentration values in each grid cell based on integrated information and calculation of the average concentration of pollutant and air-mass residence time of AMBTs in the cell (Ghosh et al., 2015; Bansal et al., 2019). The outputs of CWT and cluster analysis for fPM during each season are shown in Figs. 4(a, b, c, d).

In the present study, trajectories were identified into two major clusters in each season except in the summer, wherein clustering the AMBTs into three groups explained the dominant pathways of aerosols arriving at the study site (Figs. 4(a, b, c, d)). The trajectory clusters show that the mean pathways have a distinctly different seasonal pattern. For example, during wintertime, fPM showed association with two major clustered trajectories, originating from the northwestern part of India.
and contributed equally (50%). Cluster #1 represents the influence of long-range transport from northwest regions extending from Afghanistan and Pakistan (Fig. 4(a)). However, Cluster #2 is representative of regional transport from upwind IGP (Fig. 4(a)). Summer months have witnessed the influence of air masses associated with three major clusters. Cluster #1 comprised 36% of AMBTs from the western side of the study site and originated over Gulf countries and traversed through the Arabian Sea (Fig. 4(b)). Both Clusters #2 and #3 comprised equally (32% of AMBTs) and followed a tract of southeastern (the Bay of Bengal and eastern states) and northwestern upwind region, respectively (Fig. 4(b)). The monsoon period has experienced by and large the influence of southwesterly air masses that originated over the northern Indian Ocean (Fig. 4(c)). Nearly 54% of air masses traverse through the Arabian Sea, whereas the remaining 46% of the air masses have a signature from the marine atmospheric boundary layer (MABL) over the Bay of Bengal (Fig. 4(c)). The lowest concentrations have been observed during the monsoon period attributable to deeper boundary layer height, high wind speed and reduced anthropogenic activities (particularly BB). Thus, the IGP experiences influence of air masses of mixed origins (continental/marine) during the summer and monsoon seasons. In the post-monsoon season, the highest mass concentration of fPM was found to be associated with Cluster #1, indicating that most of the fine aerosols were transported from northwest direction (53%) that includes states of Punjab, Haryana, Delhi and other upwind IGP regions and the northern part of Pakistan, while 47% of fPM was transported with easterly wind system as predicted by Cluster #2 (Fig. 4(d)).

Fig. 4. CWT and cluster trajectory analysis at the sampling site during (a) winter, (b) summer, (c) monsoon and (d) post-monsoon season (figures are software-based and do not represent political map).
Fig. 4. (continued).

The higher concentrations of fPM in this study during post-monsoon and winter seasons and the prevailing northwesterly wind system suggest that local sources in conjunction with long-range transport are responsible for degrading air quality over the study region.

3.4 Seasonal Variability of Carbonaceous Aerosols

Seasonally averaged values of fPM, OC/fPM, EC/fPM, OC/EC, WSOC/OC, and char-EC/soot-EC ratio are represented in Figs. 5(a–f) and Table 1. The seasonally averaged fPM concentration followed the trend: post-monsoon $(257 \pm 107 \mu g m^{-3}) >$ winter $(193 \pm 60 \mu g m^{-3}) >$ summer $(130 \pm 44 \mu g m^{-3}) >$ monsoon $(53 \pm 24 \mu g m^{-3})$ (Fig. 5(a)). The OC/fPM ($\approx 0.20$; Fig. 5(b)) exhibited insignificant seasonal variability in this study (Fig. 5(b)). The EC/fPM exhibited a distinctly different seasonal variability pattern with highest concentration in winter and lowest during post-monsoon: winter $(0.14 \pm 0.05)$ > summer $(0.09 \pm 0.06) >$ monsoon $(0.08 \pm 0.07) >$ post-monsoon $(0.06 \pm 0.02)$ (Fig. 5(c)). The characteristic ratio of OC/EC showed the following pattern: monsoon $(4.0 \pm 3.1) >$ post-monsoon $(3.1 \pm 1.7) >$ winter $(1.4 \pm 0.4)$ (Fig. 5(d)). Observed values of OC, EC and OC/EC have been compared with previous studies conducted at various sites of India (Table 2) and were found to be in close range of the sites located in IGP. Furthermore, the seasonally averaged WSOC/OC ratio varied in the following manner: monsoon $(4.0 \pm 2.2) >$ winter $(3.1 \pm 2.2) >$ post-monsoon $(3.0 \pm 1.6) = $ summer $(2.3 \pm 1.4) >$ monsoon $(1.3 \pm 1.1)$ (Fig. 5(f)). The data set of each season were subjected to statistical one-way ANOVA in order to assess the significant difference in the seasonally averaged mass concentration/ratio of analyzed parameters (Table S1).
### Table 1. Seasonal record in mass concentration statistics of different carbonaceous fractions in ambient aerosols from Prayagraj (this study).

| Parameters     | Winter (n = 30) | Summer (n = 36) | Monsoon (n = 22) | Post-monsoon (n = 14) |
|----------------|-----------------|-----------------|------------------|------------------------|
|                | Range           | Avg. ± SD       | Range            | Avg. ± SD              | Range            | Avg. ± SD              |
| fPM            | 83–350          | 193 ± 60        | 67–275           | 130 ± 44               | 22–122           | 53 ± 24                |
| OC             | 13.0–75.7       | 37.4 ± 16.7     | 3–93             | 25.8 ± 19.8            | 1.1–29.4         | 10.3 ± 7.0             |
| EC             | 10.5–60.4       | 26.9 ± 13.2     | 1–72             | 12.4 ± 12.7            | 0.6–11.2         | 3.9 ± 3.1              |
| OC/PM          | 0.07–0.39       | 0.20 ± 0.08     | 0.02–0.58        | 0.20 ± 0.13            | 0.03–0.44        | 0.20 ± 0.11            |
| EC/PM          | 0.05–0.23       | 0.14 ± 0.05     | 0.01–0.26        | 0.09 ± 0.06            | 0.01–0.26        | 0.08 ± 0.07            |
| OC/EC          | 1.0–2.6         | 1.4 ± 0.4       | 0.9–22.2         | 3.1 ± 3.5              | 0.6–10.3         | 4.0 ± 3.1              |
| Char-EC/soot-EC| 0.3–11.3        | 3.1 ± 2.2       | 0.1–6.5          | 2.3 ± 1.4              | 0.1–4.5          | 1.3 ± 1.1              |
| WSOC           | 10.4–21.5       | 13.2 ± 3.1      | 13.3 ± 3.1       | 6.5 ± 2.2              | 6.6 ± 2.2        | 7.9 ± 1.1              |
| Avg. ± SD      | 21.5 ± 6.6      | 10.4 ± 2.3      | 11.5 ± 2.6       | 6.6 ± 2.2              | 7.9 ± 1.1        | 20.5 ± 13.4            |
| SD            | 6.5             | 2.3             | 2.2              | 1.1                    | 1.1              | 2.0                    |
| Range          | 23.3            | 4.5             | 5.1              | 15.5                   | 15.5             | 13.4                   |

The ANOVA has been carried out using the SPSS (version 20) software. The results were considered significant for a p-value < 0.05. All the parameters, including fPM, EC/PM, OC/EC, WSOC/OC, and char-EC/soot-EC ratio were found statistically different, with the exception being the OC/PM ratio. A significant statistical seasonal difference in various carbonaceous aerosol components is attributable to the varying contribution from emission sources, meteorological conditions (like rainfall vs. drier period, low wind speed vs. high wind speed, shallower vs. deeper boundary layer height), and atmospheric chemistry (e.g., secondary aerosol formation).
Fig. 5. Seasonal variability (avg. ± 1σ) of (a) fPM, (b) OC/fPM, (c) EC/fPM, (d) OC/EC, (e) WSOC/OC and (f) Char-EC/soot-EC.

3.5 Source Identification

Previous studies have reported distinct ratios of OC/EC and char-EC/soot-EC for different source identification, as represented in Table 3. To identify probable sources of carbonaceous aerosols at the study site in each season, we have constructed a cross plot employing the mean value of existing data sets of OC/EC and char-EC/soot-EC ratios for various sources, viz., biomass burning, biofuel burning, coal combustion, and vehicular emissions (Fig. 6). The mean value of OC/EC and char-EC/soot-EC (taken from existing literature as shown in Table 3) was reported to be 6.0 ± 4.6 and 4.8 ± 2.2 for biomass burning, 3.3 ± 0.9 and 5.4 ± 2.8 for biofuel burning, 4.3 ± 1.9 and 1.3 ± 0.8 for coal combustion and 1.3 ± 0.5 and 0.6 ± 0.4 for vehicular emissions, respectively. Since both of these ratios have been established to trace the sources of carbonaceous aerosols (Table 3), we have coupled them to utilize as reference values to better understand the dominant sources (Fig. 6) (Han et al., 2010; Ram et al., 2012; Pani et al., 2019; Kumar et al., 2021b). Apart from char-EC, the K+ ions are also predominantly used to trace out biomass burning emissions (Cheng et al., 2013). Thus, a regression analysis of OC/EC and char/soot-EC with K+ was conducted for each season (Fig. S1). The ratio analysis of the winter season has shown that there is an influence of both biomass and fossil-fuel emissions (Fig. 6(a)). However, a strong correlation of K+ with char/soot-EC (r = 0.85) as compared to OC/EC (r = 0.22) with higher mean K+ value (7.1 µg m⁻³) clearly indicated the dominance of biomass-burning sources in winter months (Fig. S1(a)). Biomass burning was also observed as a major source in post-monsoon season owing to the strong correlation of char/soot-EC ratios with K+ (r = 0.92) with higher average levels of K+.
Table 3. Literature-based emission characteristics of various sources based on OC/EC and char-EC/soot-EC ratios.

| Study area          | OC/EC | Char-EC/soot-EC | Sources          | Reference            |
|---------------------|-------|-----------------|------------------|----------------------|
| Xi’an, China        | 4.6   | 4.4             | Biomass burning  | Han et al. (2010)    |
| Haikou, China       | 2.2   | 7.1             | Biomass burning  | Liu et al. (2018)    |
| Shanghai, China     | 11.3  | 2.8             | Biomass burning  | Wang et al. (2019)   |
| Xi’an, China        | 2.6   | 7.3             | Biofuel burning  | Han et al. (2016)    |
| Raipur, India       | 3.9   | 3.4             | Biofuel burning  | Sahu et al. (2018)   |
| Texas, U.S.         | 1.9   | 1.1             | Vehicular emission| Chow et al. (2004)   |
| Texas, U.S.         | 1.1   | 0.4             | Vehicular emission| Chow et al. (2004)   |
| Hong Kong           | 1     | 0.7             | Diesel exhausts  | Cao et al. (2006)    |
| Texas, U.S.         | 2.6   | 0.4             | Coal combustion  | Chow et al. (2004)   |
| Daihai area, China  | 6.4   | 1.8             | Coal combustion  | Han et al. (2008)    |
| Shanghai, China     | 3.8   | 1.7             | Coal combustion  | Cao et al. (2013)    |

Fig. 6. Cross plots of OC/EC and char-EC/soot-EC for identification of seasonal emission sources during (a) wintertime, (b) summer, (c) monsoon, and (d) post-monsoon. The data set from this study are shown by circles whereas the literature-based source values of OC/EC and char-EC/soot-EC are shown by star symbols (for source values, reference is made to Table 4).

(5.3 µg m⁻³) (Fig. S1(d)). However, no correlation of K⁺ with OC/EC (r = –60) can be attributed to the greater presence of SOA in the post-monsoon season. This further relates to the observations reported by previous studies on the long-range transport of paddy-residue burning emissions from upwind IGP (Rajput et al., 2016). During summer and monsoon season, it appeared to have a predominant impact of fossil-fuel (vehicular emission and coal combustion) sources with lower mean K⁺ values (2.2 and 1.5 µg m⁻³, respectively) (Figs. 6(c) and (6(d))). Furthermore, K⁺ in fPM remained strongly correlated with char/soot EC as compared to OC/EC in all the seasons, which
clearly established that the char/soot-EC ratio is a better indicator for source identification as compared to OC/EC at the study site (Fig. S1) in ambient air owing to the influence of differential scavenging pattern (of OC and EC) and SOA formation (Boreddy et al., 2018). Pani et al. (2019) have also found char/soot-EC ratio as a good indicator of BB and FFC sources over urban sites. Furthermore, the studies using receptor modeling approaches such as positive matrix factorization (PMF) and chemical mass balance (CMB) with the incorporation of molecular markers would be more helpful in accurately quantifying the source contribution over the Prayagraj site.

3.6 Source Variability and Secondary Organic Aerosol Formation

SOA can be formed primarily by photochemistry or aqueous-phase reactions of volatile organic compounds (VOCs) (Seinfeld and Pandis, 2016). The SOC/OC ratio was found to be highest during monsoon (0.7 ± 0.2), followed by summer (0.6 ± 0.3), post-monsoon (0.5 ± 0.3) and lowest in winter (0.3 ± 0.1). The seasonal variability trend of the POC/OC ratio is quite opposite to that of SOC/OC ratio (Fig. 7). Both POC and SOC fractions were found to exhibit significantly high ($p < 0.05$) seasonal differences. The mass fraction of SOC is always larger than the contribution by POC, the exception being the observation during wintertime when POC/OC ratio is higher than the SOC/OC ratio. Summing up, a distinctly different and opposite seasonal trend for primary and secondary fractions of OA over central IGP has been found in this study (Fig. 7).

4 CONCLUSION

In this study, we assessed the temporal variability of the atmospheric fPM ($d_{ia} < 2.5 \mu m$) as well as the mass concentrations and key ratios (OC/fPM, EC/fPM, OC/EC, char-EC/soot-EC, and WSOC/OC) of the associated carbonaceous aerosol species above Prayagraj (on the central IGP) for a period of 1 year. The major findings are as follows.

1. We observed significant seasonality in the variability of fPM, and the mass concentrations and ratios (excluding OC/fPM) of the carbonaceous aerosols, which constituted ~40% of the annual fPM mass concentration at the sampling site.

2. Our trajectory cluster and CWT analyses revealed an association between higher concentrations of fPM and carbonaceous aerosols at the study site, which mainly occurred during the post-monsoon season and winter, and prevailing northwesterly air masses. However, anthropogenic emissions from local sources dominated the fPM and carbonaceous aerosols in summer and the monsoon season, during which we also observed the influence of air masses of mixed (continental/marine) origin.

3. Utilizing the char-EC/soot-EC ratio in conjunction with the OC/EC ratio and $K^+$ mass concentration, we identified biomass burning as the primary source of carbonaceous aerosols.
during the post-monsoon season and winter but fossil-fuel combustion as the predominant contributor during summer and the monsoon season.

4. BB and FFC sources of the fPM-bound carbonaceous aerosols above the central IGP can be traced using the char-EC/soot-EC ratio.

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