Long-Term Variation in Carbonaceous Components of PM$_{2.5}$ from 2012 to 2021 in Delhi

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Abstract

Carbonaceous species [organic carbon (OC), elemental carbon (EC), elemental matter (EM), primary organic carbon (POC), secondary organic carbon (SOC), total carbon (TC), and total carbonaceous matter (TCM)] of PM$_{2.5}$ were analyzed to study the seasonal variability and long-term trend of carbonaceous aerosols (CAs) in megacity Delhi, India from January, 2012 to April, 2021. The average concentrations (± standard deviation) of PM$_{2.5}$, OC, EC, TC, EM, TCM, POC and SOC were 127 ± 77, 15.7 ± 11.6, 7.4 ± 5.1, 23.1 ± 16.5, 8.2 ± 5.6, 33.3 ± 23.9, 9.3 ± 6.3 and 6.5 ± 5.3 µg m$^{-3}$, respectively during the sampling period (10-year average). The average CAs accounted for 26% of PM$_{2.5}$ concentration during the entire sampling period. In addition, the seasonal variations in PM$_{2.5}$, OC, EC, POC, SOC, and TCM levels were recorded with maxima in post-monsoon and minima in monsoon seasons. The linear relationship of OC and EC, OC/EC and EC/TC ratios suggested that the vehicular emissions (VE), fossil fuel combustion (FFC) and biomass burning (BB) are the major sources of CAs at megacity Delhi, India.

Keywords PM$_{2.5}$ · OC · EC · TCM · Carbonaceous aerosols

Atmospheric aerosols are complex in their chemical composition (Weagle et al. 2018), varying spatially and temporally (Sharma et al. 2021a) and have different effects on local as well as regional air quality (Belis et al. 2013; Jain et al. 2020), visibility, Earth’s radiation budget, global climate (Ramanathan et al. 2001) and human health (Pope and Dockery 2006; Dockery and Stone 2007; Pope et al. 2009; Fuzzi et al. 2015; Lehtimäki et al. 2015; Gauderman et al. 2015; Velali et al. 2016). Studies reveal that the PM$_{2.5}$ was responsible for more than 3 million premature deaths per year worldwide (Jerrett 2015; Lehtimäki et al. 2015). PM$_{2.5}$ consists of organics [organic carbon (OC) and elemental carbon (EC)], elements, sea salt, secondary aerosols, etc. The constituents of carbonaceous species of PM$_{2.5}$ have an important impact on regional and global climate along with Earth’s atmospheric system (Ramanathan et al. 2001). Therefore, identifying and quantifying the carbonaceous aerosols (CAs) in PM$_{2.5}$ is essential to design the air quality management strategies to control PM$_{2.5}$ mass loading in the ambient air through targeted action (Waked et al. 2014).

Carbonaceous aerosols are the significant sources of atmospheric particulate matter (PM)/aerosols, containing up to 70% of aerosols mass loading (Kanakidou et al. 2005) and its major sources are biomass burning (BB), fossil fuel combustion (FFC) and biogenic emissions (Claeys et al. 2004; Venkataraman et al. 2005). Several studies have been conducted on CAs and their probable sources in urban (Ram and Sarin 2010; Mandal et al. 2014; Sharma et al. 2018; Jain et al. 2020), rural, remote (Begam et al. 2017) and high altitude atmosphere (Kaushal et al. 2018; Sharma et al. 2021a, b) of the Indian region on a short-term basis, but a few studies have been carried out on the long-term trend (Sharma et al. 2018; 2021a). In this paper, we reported the seasonal and annual variability of OC, EC, elemental matter (EM), primary organic carbon (POC), secondary organic carbon (SOC) and total carbonaceous matter (TCM) of PM$_{2.5}$ at an urban site of Delhi, India from January, 2012 to April, 2021.
Materials and Methods

PM$_{2.5}$ samples (n = 805) were collected periodically (2 samples/week) at the observational site of CSIR-National Physical Laboratory (28° 38’ N, 77° 10’ E; 218 m amsl), New Delhi from January, 2012 to April, 2021 [except July–December, 2012 and Covid-19 lockdown period (lockdown periods: 25 March, 2020–31 May, 2020; unlock periods: 1 June, 2020–31 August, 2020)]. The sampling site is a typical urban environment surrounded by heavy roadside traffic and an agricultural institute in the southwest direction (Sharma et al. 2018). Vehicular traffic, FFC, BB, secondary aerosols, minerals and soil dust are the major source of aerosols of Delhi and surrounding areas (Sharma et al. 2014). During 2020–2021, the total number of registered vehicles in Delhi was 11.8 million (Delhi Statistical Handbook 2021). The meteorology of Delhi is divided into four different seasons: winter (January–February), summer (March–May), monsoon (June–September) and post-monsoon (October–December). Winter months are chilly (temperature: ~ 3°C) whereas, the summers are generally scorching and dry (temperature: 47°C).

A fine particle sampler (APM 550, Envirotech, Delhi, IN; having a flow rate of 1 m$^3$ h$^{-1}$ with accuracy ± 2%) was used to collect the PM$_{2.5}$ samples on pre-baked quartz filters (size: 47 mm) for 24 h. The quartz filters were weighed before and after the sampling to determine the gravimetric mass concentration of collected PM$_{2.5}$ samples. OC and EC analysis of PM$_{2.5}$ samples was performed by a Thermo-optical carbon analyzer (DRI 2001A, Atmoslytic Inc., Calabasas, CA, USA) using IMPROVE-A protocol (Chow et al. 2004). OC/EC carbon analyzer is working on the preferential oxidation of OC and EC at different temperatures plateaus (140, 280, 480 and 580°C; for OC1, OC2, OC3 and OC4, respectively) in pure He and three temperature plateaus (580, 740 and 840°C for EC1, EC2 and EC3, respectively) in 98% He + 10% O$_2$ gas (Chow et al. 2004). A proper punch of ~0.536 cm$^2$ area of the filter was cut and analyzed in triplicate along with field blank filters (>50 samples during the sampling period). The standard calibration for peak area verification was performed daily using 5% CH$_4$ + balance helium gas (before and after sample analysis). Calibration of the OC/EC analyzer was performed by 4.8% of CO$_2$ + balance He gas along with known amounts of KHP (potassium hydrogen phthalate) and sucrose solution. In the present case, repeatability errors of OC (detection limit: 0.54 µg m$^{-3}$) and EC (detection limit: 0.21 µg m$^{-3}$) analysis were estimated as 3%–7% (Sharma et al. 2021a). Statistical data analysis of chemical species of PM$_{2.5}$ was performed by chi-square method using Monte Carlo statistics (non-parametric test) (Sharma et al. 2018). TCM is computed by the addition of organic matter (OM = 1.6 × OC) and EM (EM = 1.1 × EC) of PM$_{2.5}$ (Malm et al. 2004; Turpin and Lim 2001; Srinivas and Sarin 2014). The effective carbon ratio (ECR) of CAs provides the information about the formation of SOC in the atmosphere which is calculated as SOC/(POC + EC). SOC cannot be measured directly by a Carbon analyzer, therefore, SOC was computed by the EC tracer method (Castro et al. 1999) using the following equations:

\[ \text{POC} = [\text{OC}/\text{EC}]_{\text{min}} \times \text{EC} \] (1)

\[ \text{SOC} = \text{OC} − \text{POC} \] (2)

Results and Discussion

Figure 1 represents the time series plots of OC, EC and TC of PM$_{2.5}$ along with PM$_{2.5}$ concentration from January, 2012 to April, 2021. The annual average concentrations of PM$_{2.5}$, OC, EC, TC, EM, TCM, POC and SOC with standard deviation (±SD) were 127 ± 77, 15.7 ± 11.6, 7.4 ± 5.1, 23.1 ± 16.5, 8.2 ± 5.6, 33.3 ± 23.9, 9.3 ± 6.3 and 6.5 ± 5.3 µg m$^{-3}$, respectively during 2012–2021 (Table 1). During the study, a non-significant decreasing trend in annual concentration of PM$_{2.5}$ (y = −1.596x + 135.3; $R^2 = 0.18$) was recorded, whereas the significant decreasing trend in annual concentration of OC was recorded (y = −0.484x + 18.4; $R^2 = 0.49$), EC (y = −0.502x + 10.2; $R^2 = 0.51$) and TC (y = −0.986x + 28.6; $R^2 = 0.54$) were recorded (Fig. S1; in supplementary information). In 2017, the highest annual average concentration of PM$_{2.5}$ was noted as 143 ± 7 µg m$^{-3}$, whereas the minimum concentration of PM$_{2.5}$ was obtained in 2021 as 109 ± 53 µg m$^{-3}$ which are exceeding the National Ambient Air Quality Standards (annual: 40 µg m$^{-3}$). The higher concentrations of OC (19.3 ± 13.9 µg m$^{-3}$), EC (11.4 ± 7.5 µg m$^{-3}$) and TCM (43.3 ± 30.1 µg m$^{-3}$) were observed in 2013. OC, EC and TCM contributed to ~12%, ~5.5% and ~25%, respectively during 2012–2021 (10-year average). Jain et al. (2020) reported similar percentage contributions of OC (~12%), EC (~5.5%) and TCM (~25%) to PM$_{2.5}$ at Delhi, whereas Mandal et al. (2014) reported higher percentage contributions of OC (28%) and EC (9%) to PM$_{2.5}$ in an industrial area of Delhi.

The seasonal variations in the level of OC, EC, TC, TCM, POC and SOC of PM$_{2.5}$ and their percentage contribution to PM$_{2.5}$ and seasonal differences are summarized in Table 2. The highest concentrations of PM$_{2.5}$, OC, EC, TC, POC and SOC were recorded during post-monsoon seasons followed by winter, summer and monsoon seasons (Table 2; Fig. S2, in supplementary information). The higher concentrations of PM$_{2.5}$ and its carbonaceous species during
**Fig. 1** Time series plot in mass concentrations of PM$_{2.5}$, OC, EC and TC during 2012–2021 (the data from July–December, 2012 and the lockdown periods in 2020 were not included)

**Table 1** Annual average concentrations of carbonaceous species in PM$_{2.5}$ (OC, EC, TC, TCM, EM, POC and SOC) in Delhi

| Year | PM$_{2.5}$ ($\mu$g m$^{-3}$) | OC (µg m$^{-3}$) | EC (µg m$^{-3}$) | TC (µg m$^{-3}$) | TCM (µg m$^{-3}$) | EM (µg m$^{-3}$) | POC (µg m$^{-3}$) | SOC (µg m$^{-3}$) | ECR | OC/EC | EC/TC |
|------|----------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----|-------|-------|
| 2012 | 133±92                     | 18.7±10.6       | 10.1±6.4        | 28.8±16.8       | 41.0±23.7       | 11.1±7.0        | 5.2±4.0         | 13.4±8.5        | 0.27 | 1.85  | 0.35  |
| 2013 | 136±91                     | 19.3±13.9       | 11.4±7.5        | 30.6±21.1       | 43.3±30.1       | 12.5±8.3        | 7.0±6.2         | 12.3±8.1        | 0.34 | 1.79  | 0.37  |
| 2014 | 113±96                     | 16.6±14.5       | 9.5±8.4         | 26.1±22.8       | 37.0±32.3       | 10.5±9.2        | 5.8±4.7         | 10.7±9.4        | 0.35 | 1.75  | 0.36  |
| 2015 | 123±65                     | 13.8±9.1        | 6.0±3.3         | 19.8±12.2       | 28.7±17.9       | 6.6±3.6         | 7.1±3.9         | 6.7±5.8         | 0.52 | 2.31  | 0.30  |
| 2016 | 138±58                     | 14.5±13.2       | 4.9±3.8         | 19.4±16.7       | 28.6±25.0       | 5.4±4.2         | 7.0±5.4         | 7.5±6.4         | 0.64 | 2.97  | 0.25  |
| 2017 | 143±70                     | 17.0±11.7       | 6.5±3.8         | 23.5±15.1       | 34.4±22.5       | 7.2±4.2         | 9.5±5.5         | 7.5±7.3         | 0.47 | 2.60  | 0.28  |
| 2018 | 124±70                     | 13.4±9.5        | 6.8±4.4         | 20.1±13.5       | 28.8±19.6       | 7.5±4.9         | 7.5±4.9         | 5.9±5.7         | 0.43 | 1.97  | 0.34  |
| 2019 | 129±76                     | 15.8±14.2       | 7.0±5.1         | 22.8±19.3       | 32.9±28.3       | 7.7±5.6         | 7.8±5.7         | 8.0±7.7         | 0.47 | 2.27  | 0.31  |
| 2020 | 117±68                     | 14.2±11.0       | 6.2±4.6         | 20.4±15.4       | 29.6±22.5       | 6.8±5.1         | 7.7±5.7         | 6.5±5.9         | 0.53 | 2.31  | 0.30  |
| 2021 | 109±53                     | 14.0±8.9        | 5.9±3.6         | 19.9±12.3       | 28.9±18.0       | 6.5±4.0         | 9.6±5.8         | 4.5±4.4         | 0.33 | 2.38  | 0.30  |
| Average | 127±77                     | 15.7±11.6       | 7.4±5.1         | 23.1±16.5       | 33.3±23.9       | 8.2±5.6         | 9.3±6.3         | 6.5±5.3         | 0.43 | 2.22  | 0.32  |

± Standard deviation at 1σ (n=805 samples for 10 years); The data from July–December, 2012 and the lockdown periods in 2020 were not included.
Table 2  Seasonal variations of carbonaceous species in PM$_{2.5}$ and their percentage contribution in Delhi

| Species     | Seasons               | Winter (W) (n = 180) | Summer (S) (n = 216) | Monsoon (M) (n = 196) | Post-Monsoon (PM) (n = 213) | Seasonal difference |
|-------------|-----------------------|----------------------|----------------------|------------------------|-----------------------------|---------------------|
| PM$_{2.5}$ (µg m$^{-3}$) | 158 ± 64              | 95 ± 35              | 67 ± 28              | 190 ± 92               |                             | 62.9$^a$ 90.3$^b$  − 32.4$^a$ 27.4$^a$  − 95.3$^b$  − 122.7$^a$ |
| OC (µg m$^{-3}$)      | 20.5 ± 11.1           | 9.5 ± 4.1            | 6.5 ± 2.9            | 25.6 ± 14.3            |                             | 11.0$^a$ 14.0$^a$  − 5.1$^a$ 3.0$^b$  − 16.1$^a$  − 19.1$^a$ |
| EC (µg m$^{-3}$)      | 9.4 ± 5.1             | 4.8 ± 2.6            | 2.9 ± 1.6            | 10.9 ± 6.3             |                             | 4.5$^a$ 6.5$^a$  − 1.5$^b$ 2.0$^b$  − 6.0$^a$  − 8.0$^a$ |
| TC (µg m$^{-3}$)      | 29.8 ± 15.5           | 14.3 ± 6.4           | 9.4 ± 4.2            | 36.4 ± 19.7            |                             | 15.5$^a$ 20.5$^a$  − 6.6$^a$ 4.9$^a$  − 22.1$^a$  − 27.1$^a$ |
| TCM (µg m$^{-3}$)     | 43.0 ± 22.5           | 20.5 ± 9.1           | 13.5 ± 6.1           | 52.9 ± 28.8            |                             | 22.6$^a$ 29.5$^a$  − 9.8$^a$ 6.9$^a$  − 32.4$^a$  − 39.3$^a$ |
| EM (µg m$^{-3}$)      | 10.3 ± 5.6            | 5.3 ± 2.8            | 3.2 ± 1.7            | 12.0 ± 6.9             |                             | 5.0$^a$ 7.1$^a$  − 1.7$^b$ 2.1$^b$  − 6.6$^a$  − 8.8$^a$ |
| POC (µg m$^{-3}$)     | 11.9 ± 6.1            | 6.1 ± 3.0            | 3.5 ± 1.8            | 13.1 ± 7.2             |                             | 5.9$^a$ 8.4$^a$  − 1.2$^b$ 2.5$^b$  − 7.1$^a$  − 9.6$^a$ |
| SOC (µg m$^{-3}$)     | 8.5 ± 6.7             | 3.4 ± 1.9            | 2.9 ± 1.7            | 12.4 ± 8.9             |                             | 5.1$^a$ 5.6$^a$  − 3.9$^b$ 0.5$^b$  − 9.0$^a$  − 9.5$^a$ |
| ECR                   | 0.43 ± 0.27           | 0.37 ± 0.21          | 0.55 ± 0.31          | 0.56 ± 0.28            |                             | 0.06$^a$ −0.12$^a$  −0.13$^a$ −0.18$^a$  −0.19$^a$  −0.01$^b$ |
| OC/EC                 | 2.3 ± 0.7             | 2.1 ± 0.6            | 2.2 ± 0.8            | 2.5 ± 0.8              |                             | 0.2$^b$ 0.1$^b$  −0.2$^b$  −0.1$^b$  −0.4$^b$  −0.3$^b$ |
| EC/TC                 | 0.32 ± 0.17           | 0.34 ± 0.14          | 0.31 ± 0.12          | 0.30 ± 0.15            |                             | −0.02$^b$ 0.01$^b$ 0.02$^b$  −0.03$^b$  −0.04$^b$ 0.01$^b$ |
| OC (%)                | 12.9 ± 4.0            | 10.8 ± 4.9           | 10.7 ± 5.2           | 13.9 ± 6.6             |                             | 2.1$^a$ 2.2$^a$  −1.0$^b$ 0.1$^b$  −3.1$^a$  −3.2$^a$ |
| EC (%)                | 6.0 ± 2.2             | 5.6 ± 3.2            | 4.9 ± 3.1            | 6.1 ± 3.7              |                             | 0.4$^b$ 1.2$^b$  −0.1$^b$ 0.7$^b$  −0.5$^b$  −1.3$^b$ |
| TC (%)                | 18.9 ± 5.5            | 16.4 ± 6.9           | 15.6 ± 8.0           | 20.0 ± 9.9             |                             | 2.6$^a$ 3.4$^a$  −1.1$^b$ 0.8$^b$  −3.6$^a$  −4.5$^a$ |
| TCM (%)               | 27.3 ± 8.0            | 23.6 ± 11.1          | 22.5 ± 11.3          | 29.0 ± 14.2            |                             | 3.9$^a$ 4.8$^a$  −1.7$^b$ 0.9$^b$  −5.6$^a$  −6.5$^a$ |

The data from July–December, 2012 and the lockdown periods in 2020 were not included.

± Standard deviation at 1σ

$^a$Significantly different (at $p < 0.05$)

$^b$Significantly not different (at $p < 0.05$)
post-monsoon and winter seasons could be the source strength of PM$_{2.5}$ and prevailing meteorological conditions (temperature, relative humidity, wind speed, wind directions, etc.) as well as the lowering of mixing height of the boundary layer at the sampling site. Regional and long-range transportation of pollutants (from crop residue burning) towards the sampling site also arises from the north-western states of IGP (Punjab, Harayana) of India (Ravindra et al. 2022). Sharma et al. (2021b) also experienced similar seasonal variations of PM$_{10}$ and their chemical constituents in megacity Delhi based on long-term observations (2010–2019). During all the seasons, the POC value is recorded higher than the SOC level with an ECR value < 1.0 (0.3 to 0.6) which indicates the higher contribution of POC over SOC (Table 2).

Monthly average (pooled estimate of each month of 2012–2021) values of PM$_{2.5}$, OC, EC, TC and TCM from January, 2012 to April, 2021 are represented in Fig. 2. The highest monthly average concentrations of PM$_{2.5}$, OC, EC and TCM were 226 ± 71; 28.9 ± 12.2; 12.3 ± 6.8 and 59.8 ± 25.7 µg m$^{-3}$, respectively during December (post-monsoon) (Fig. 2), whereas the minimum monthly average levels of PM$_{2.5}$, OC, EC and TCM were 64 ± 23; 6.1 ± 2.4; 2.5 ± 1.3 and 12.4 ± 4.8 µg m$^{-3}$, respectively during July (monsoon). Several researchers (Mandal et al. 2014; Bisht et al. 2015; Jain et al. 2020; Sharma et al. 2021b) have reported similar monthly variations in carbonaceous species (OC, EC, TC and TCM) of PM$_{2.5}$ at Delhi and IGP of India (Ram and Sarin 2010; Srinivas and Sarin 2014; Khaiwal et al. 2021). The monthly concentration of PM$_{2.5}$ has also exceeded the National Ambient Air Quality Standards (NAAQS; 24 h: 60 µg m$^{-3}$; annual: 40 µg m$^{-3}$) of India.

Generally, FFC and BB fuel burning (dungcake, crop residue and wood-burning) contribute appreciably to OC and EC particulates (Ram and Sarin 2010; Begum et al. 2011). A positive linear correlation between OC and EC is usually considered as emissions from the same combustion sources (Salma et al. 2004; Ram et al. 2010; Sharma et al. 2014; Jain et al. 2017). Whereas, a weakly correlated OC and EC suggest the influence of secondary organic aerosols (SOA) formed through the gas-to-particle conversion of volatile organic compounds (VOCs) (Begum et al. 2004, 2006). In the present study, significant positive linear relationships between OC and EC were observed during winter ($R^2 = 0.66$), summer ($R^2 = 0.74$), monsoon ($R^2 = 0.62$) and post-monsoon ($R^2 = 0.67$) seasons (Fig. 3) in Delhi, indicating the influence of VE or BB (or both).

The OC/EC ratio has been applied extensively for decoding the sources of CAs around the globe (Novakov et al.
The higher OC/EC value (between 4 and 12) is generally used for BB (Szele et al. 2006), whereas, the OC/EC ratio between 1.4 to 4 are used (between 0.3 to 1 used for diesel vehicles) for VE along with BB (mixed type) (Amato et al. 2009; Salameh et al. 2015; Ram et al. 2010). In this study, the average OC/EC ratio of PM$_{2.5}$ were 2.3 ± 0.7, 2.1 ± 0.6, 2.2 ± 0.8 and 2.5 ± 0.8 during winter (range 1.1–5.7), summer (range 1.0–3.9), monsoon (range 1.2–5.9) and post-monsoon (range: 1.1–6.8) seasons, respectively (Table 2). The average OC/EC mass ratio evidenced that the FFC and BB are the dominant sources of CAs at the study site. The seasonal value of the EC/TC ratio also support the emissions of these sources over Delhi. Stable carbon and nitrogen isotopic analysis of PM$_{2.5}$ also evidenced that the VE and BB are the major sources of PM$_{2.5}$ in Delhi (Sharma et al. 2015, 2017, 2022; Jain et al. 2020) have applied the receptor models on long-term chemical species of PM$_{2.5}$ and reported that BB and VE were the major sources of PM$_{2.5}$ in Delhi. The average ECR values (Tables 1 and 2) of PM$_{2.5}$ demonstrated the abundance of POC (in the present case ECR < 1.0) over SOC and has more warming effects.

Several researchers (Jain and Sharma 2020; Singh et al. 2020; Sokhi et al. 2021; Saharan et al. 2022) have reported the reduction in concentrations of gaseous (NO, NO$_2$, CO, SO$_2$, O$_3$, NH$_3$, benzene, toluene, etc.) and particulate (PM$_{2.5}$ and PM$_{10}$) pollutants in Delhi, India, and globe during covid-19 lockdown period as compared to pre-lockdown period due to restricted activities. The sampling period of PM$_{2.5}$ also falls under the 4 lockdown, and 3 unlock periods in Delhi during 2020 (Table S1; in supplementary information). Therefore, analysis of PM$_{2.5}$ and its carbonaceous species (OC, EC, TC, and OC/EC) are essential to understand the levels of CAs during this period. During 4 lockdown periods (25 March 2020 to 31 May 2020), the PM$_{2.5}$ samples were not collected at the study site due to restricted entry into the laboratory. However, PM$_{2.5}$ sample collection was resumed from the unlock-1 period, i.e., 1 June 2020 onwards (Table S1; in supplementary information). Figure 4 shows the time series plots of PM$_{2.5}$, OC, EC, and TC concentrations from pre-lockdown to unlock period (January 2020 to December 2020) in Delhi. The average concentrations of PM$_{2.5}$, OC, EC, and TC were 125, 13.6, 6.6 and 21.1 µg m$^{-3}$, respectively during the pre-lockdown period, whereas the average concentrations of PM$_{2.5}$, OC, EC, and TC were 68, 6.0, 3.0 and 9.1 µg m$^{-3}$ during the unlock-1 period when only government offices were opened with limited capacity (Table S1; in supplementary information). The average reduction in mass concentrations of PM$_{2.5}$ (reduced by 45%) and its carbonaceous components OC (reduced by 55%), EC (reduced by 53%), and TC (reduced by 54%) were ~50% when compared with pre-lockdown to unlock-1 period. Also, the OC/EC ratio is increased up to ~50% (2.0 to 3.1), however the sources of CAs still arises from VE and FFC. Singh et al. (2020) reported >40% reduction in PM$_{2.5}$ concentration over Delhi during lockdown period in comparison to unlock period. In contrast, Saharan et al. (2022) reported a 35% reduction in PM$_{2.5}$ concentration during the lockdown period compared with the pre-lockdown period in Delhi during 2020. Several researchers in India and the globe reported a similar reduction in pollutants levels (Kotnala et al. 2020; Jain and Sharma 2020; Sokhi et al. 2021).

The present study demonstrates the seasonal variability and long-term trends with high loading of carbonaceous species (OC, EC, TC, EM, TCM, POC, and SOC) in PM$_{2.5}$ over megacity Delhi, India, which is further contaminating the ambient air quality of the region and impact on the level of short-lived climate-forcing pollutants. Seasonal variations in mass concentrations of PM$_{2.5}$, OC, EC, POC, SOC, and TCM were recorded maximum in post-monsoon and minimum in monsoon seasons. The study suggested that the VE, FFC, and BB are the major sources of CAs at megacity Delhi, India. During covid-19 unlock periods, the OC/EC ratio is increased up to ~50% (2.0 to 3.1), which shows the sources of CAs arises from VE and FFC. In urban areas, the number of vehicles, industries, and influence of human activities was increasing with time and expected to increase soon, which is believed to augment the abundance of TCM over the region. The CAs have a significant impact on atmospheric chemistry, climate, and environmental transport systems. Hence, there is a need to take necessary mitigation measures to control/cut down the emissions of carbonaceous aerosols from various sources.
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Author Contributions  The experimental design and the first draft were prepared by SKS. Sample collections and chemical analysis were carried out by SKS, RB, AR, MR, and TKM. All authors read and reviewed the manuscript before communication.

Declarations

Conflict of interest  There is no conflict of interest among the authors or with any external agency.

References

Amato F, Pandolfi M, Escrig A, Querol X, Alastuery A, Pey J, Perez N, Hopke PK (2009) Quantification of road dust resuspension in an urban environment by the multilinear engine: a comparison with PMF2. Atmos Environ 43:2770–2780
Begam GR, Vachaspati CV, Ahammed YN, Kumar KR, Reddy RR, Sharma SK, Saxena M, Mandal TK (2017) Seasonal characteristics of water-soluble inorganic ions and carbonaceous aerosols in total suspended particulate matter at a rural semi-arid site, Kadapa (India). Environ Sci Pollut Res 24(2):1719–1734
Begum BA, Kim E, Biswas SK, Hopke PK (2004) Investigation of sources of atmospheric aerosol at urban and semi-urban areas in Bangladesh. Atmos Environ 38:3025–3038
Begum BA, Akhter S, Sarkar L, Biswas SK (2006) Gravimetric analysis of air filters and quality assurance in weighing. Nuclear Sci Appl 15:36–41
Kotnala G, Mandal TK, Sharma SK, Kotnala RK (2020) Emergence of blue sky over Delhi due to coronavirus disease (COVID-19) lockdown implications. Aerosol Sci Eng 3:228–238

Lelieveld J, Evans JS, Fnais M, Giannadaki D, Pozzer A (2015) The contribution of outdoor air pollution sources to premature mortality on a global scale. Nature 525:367–371

Malm WC, Schichtel BA, Pitchford ML, Ashbaugh LL, Eldred RA (2004) Spatial and monthly trends in speciated fine particles concentration in the United States. J Geophys Res Atmos 109:D03S30

Mandal P, Saud T, Sarkar R, Mandal A, Sharma SK, Mandal TK, Basin JK (2014) High seasonal variation of atmospheric C and particulate concentrations in Delhi, India. Environ Chem Lett. https://doi.org/10.1007/s10311-013-0438-y

Novakov T, Andreae MO, Gabriel R, Kirchstetter TW, Mayol-Bracero OL (2000) Origin of carbonaceous aerosols over the tropical Indian Ocean: biomass burning or fossil fuels? Geophys Res Lett 27:4061–4064

Pope CA, Dockery DW (2006) Health effects of fine particulate air pollution: lines that connect. JAPCA 56:709–742

Pope CA, Ezzati M, Dockery DW (2009) Fine-particulate air pollution and life expectancy in the United States. N Engl J Med 360:376–386

Ram K, Sarin MM (2010) Spatio-temporal variability in atmospheric abundances of EC, OC and WSOC over Northern India. J Aerosol Sci 41(1):88–98

Ram K, Sarin MM, Tripathi SN (2010) One-year record of carbonaceous aerosols from an urban location (Kanpur) in the Indo-Gangetic Plain: characterization, sources and temporal variability. J Geophys Res. https://doi.org/10.1029/2010JD014188

Ramanathan V, Crutzen PJ, Kiehl JT, Rosenfeld D (2001) Aerosols, climate, and the hydrological cycle. Science 294(5549):2119–1214

Ravindra K, Singh T, Mandal TK, Sharma SK, Mor S (2022) Seasonal variations in carbonaceous species of PM$_{1}$, aerosols at an urban location situated in Indo-Gangetic Plain and its relationship with transport pathways, including the potential sources. J Environ Manag 303:114049

Saharan US, Kumar R, Tripathy P, Sateesh M, Garg J, Sharma SK, Mandal TK (2022) Drivers of air pollution variability during second wave of COVID-19 in Delhi, India. Urban Clim 41:101059

Salameh D, Detournay A, Prey J, Perez N, Liguri F, Sarada D, Bove MC, Broto P, Cassola F, Massabò D, Latella A, Pillon S, Formenton G, Patti S, Armengaud A, Piga D, Jaffrezo JL, Bartzis J, Tols E, Prati P, Querol X (2015) PM$_{2.5}$ chemical composition in five European Mediterranean cities: a 1-year study. Atmos Res 155:102–117

Salma I, Chi XG, Maenhaut W (2004) Elemental and organic carbon in urban canyon and background environments in Budapest, Hungary. Atmos Environ 38:2517–2528

Sharma SK, Mandal TK, Saxena M, Rashmi R, Sharma A, Gautam R (2014) Variation of OC, EC, WSOC and trace metals of PM$_{2.5}$ in Delhi. J Atmos Sol Terr Phys 113:10–22

Sharma SK, Mandal TK, Shenoy DM, Bardhan P, Srivastava MK, Chatterjee A, Saxena M, Saraswati SBP, Ghosh SK (2015) Variation of stable carbon and nitrogen isotopes composition of PM$_{10}$ over Indo-Gangetic Plain of India. Bull Environ Contam Toxicol 95(5):661–669

Sharma SK, Agarwal P, Mandal TK, Karaparakar SG, Shenoy DM, Peshin SK, Gupta A, Saxena M, Jain S, Sharma A, Saraswati (2017) Study on ambient air quality of megacity Delhi, India during Odd-Even Strategy. Manan 32(2):155–165

Sharma SK, Mandal TK, Sharma A, Jain S, Saraswati (2018) Carbonaceous species of PM$_{2.5}$ in megacity Delhi, India during 2012–2016. Bull Environ Contam Toxicol 100:695–701

Sharma SK, Banoo R, Mandal TK (2021a) Seasonal characteristics and sources of carbonaceous components and elements of PM$_{10}$ (2010–2019) in Delhi, India. J Atmos Chem 78:251–270

Sharma SK, Mukherjee S, Choudhary N, Rai A, Ghosh A, Chatterjee A, Vijayan N, Mandal TK (2021b) Seasonal variation and sources of elemental carbon by thermal/optical reflectance measurement in a coastal area of south China. J Geophys Res. https://doi.org/10.1029/2005JD006663

Chow JC, Watson JC, Chen LWA, Arnott WP, Moosmüller H (2004) Equivalence of elemental carbon by thermal/optical reflectance and transmittance with different temperature protocols. Environ Sci Technol 38:4414–4422

Claeys M, Wang W, Ion AC, Gellynck X, Maenhaut W (2000) Formation of secondary organic aerosols from isoprene and its gas-phase oxidation products through reaction with hydroperoxide. Atmos Environ 34(25):4093–4108

Delhi Statistical Handbook (2021) Registered vehicles in Delhi. Delhi govt. Delhi, www.des.delhibovt.nic.in

Dockery DW, Stone PH (2007) Cardiovascular risks from fine particulate air pollution. N Engl J Med 356:511–513

Fuzzi S, Baltensperger U, Carslaw K, Deccarsi S, Denier van der Gon H, Facchini MC, Fowler D, Koren I, Langford B, Lohmann U, Nemitz E, Pandis S, Riipinen I, Rudich Y, Schaap M, Slowik JG, Spracklen DV, Vignati E, Wild M, Williams M, Gilardoni S (2015) Particulate matter, air quality and climate: lessons learned and future needs. Atmos Chem Phys 15:8217–8299

Gauderman WJ, Urman R, Avol E, Berhane K, McConnell R, Rappaport E, Chang R, Lurmann F, Gilliland F (2015) Association of improved air quality with lung development in children. N Engl J Med 372:905–913

Jain S, Sharma SK, Choudhary N, Maswaliw R, Saxena M, Sharma A, Mandal TK, Gupta A, Gupta NC, Sharma C (2017) Chemical characteristics and source apportionment of PM$_{2.5}$ using PCA/APCS, UNMIX and PMF at an urban site of Delhi, India. Environ Sci Poll Res 24:14637–14656

Jain S, Sharma SK, Vijayan N, Mandal TK (2020) Seasonal characteristics of aerosols (PM$_{2.5}$ and PM$_{10}$) and their source apportionment using PMF: a four-year study over Delhi, India. Environ Poll 262:114337

Jalil S, Sharma SK, Vijayan N, Mandal TK (2020) Social and travel lockdown impact considering coronavirus disease (COVID-19) on air quality in megacities of India: present benefits, future challenges and way forward. Aerosol Air Qual Res 20(6):1222–1236

Jain S, Sharma SK, Choudhary N, Maswaliw R, Saxena M, Sharma A, Mandal TK, Gupta A, Gupta NC, Sharma C (2017) Chemical characteristics and source apportionment of PM$_{2.5}$ using PCA/APCS, UNMIX and PMF at an urban site of Delhi, India. Environ Sci Poll Res 24:14637–14656

Jain S, Sharma SK, Vijayan N, Mandal TK (2020) Seasonal characteristics of aerosols (PM$_{2.5}$ and PM$_{10}$) and their source apportionment using PMF: a four-year study over Delhi, India. Environ Poll 262:114337

Jerret M (2015) Atmospheric science: the death toll from air-pollution sources. Nature 525:330–331

Kanakidou M, Seinfeld JH, Pandis SN, Barnes I, Dentener FJ, Facchini MC, Dingenen RV, Everson B, Nenes ANCJSE, Nielsen CJ, Swietlicki E (2005) Organic aerosol and global climate modelling: a review. Atmos Chem Phys 5(4):1053–1123

Kaushal D, Kumar A, Yadav S, Tandon A, Attri AK (2018) Wintertime carbonaceous aerosols over Dhauladhar region of North-Western Himalayas. Environ Sci Poll Res 25:8044–8056

Kotnala G, Mandal TK, Sharma SK, Kotnala RK (2020) Emergence of blue sky over Delhi due to coronavirus disease (COVID-19) lockdown implications. Aerosol Sci Eng 3:228–238
of carbonaceous species and elements of PM$_{2.5}$ and PM$_{10}$ over the eastern Himalaya. Environ Sci Poll Res 28:51642–51656
Sharma SK, Karapurkar SG, Shenoy DM, Mandal TK (2022) Stable carbon and nitrogen isotopic characteristics of PM$_{2.5}$ and PM$_{10}$ in Delhi, India. J Atmos Chem 2:89. https://doi.org/10.1007/s10874-022-09429-0
Singh V, Singh S, Biswal A, Kesarkar AP, Mor S, Ravindra K (2020) Diurnal and temporal changes in air pollution during COVID-19 strict lockdown over different regions of India. Environ Pollut 266:115368
Sokhi RS, Singh V, Querol X, Finardi S, Targino AC, de Fatima AM, Pavlovic R, Garland RM, Massagué J, Kong S, Baklanov A (2021) A global observational analysis to understand changes in air quality during exceptionally low anthropogenic emission conditions. Environ Internat 157:106818
Srinivas B, Sarin MM (2014) PM$_{2.5}$, EC and OC in atmospheric outflow from the Indo-Gangetic Plain: temporal variability and aerosol organic carbon-to-organic mass conversion factor. Sci Total Environ 487:196–205
Szidat S, Jenk TM, Synal HA, Kalberer M, Wacker L, Hajdas K-G, Baltensperger U (2006) Contribution of fossil fuel, biomass burning and biogenic emissions to carbonaceous aerosols in Zurich as traced by $^{14}$C. J Geophys Res 111:D07206
Turpin BJ, Lim HJ (2001) Species contributions to PM2.5 mass concentrations: revisiting common assumptions for estimating organic mass. Aerosol Sci Technol 35(1):602–610
Velali E, Papachristou E, Pantazaki A, Choli-Papadopoulou T, Argyrou N, Tsourouktsoglou T, Lialiari S, Constantinidis A, Lykidis D, Lialiari TS, Besis A, Vouts a D, Samara C (2016) Cytotoxicity and genotoxicity induced in vitro by solvent extractable organic matter of size-segregated urban particulate matter. Environ Pollut 218:1350–1362
Venkataraman C, Habib G, Figueren-Fernandez A, Miguel AH, Friedlander SK (2005) Residential biofuels in south Asia: carbonaceous aerosol emissions and climate impacts. Science 307:1454–1456
Waked A, Favez O, Alleman LY, Pi ot C, Petit JE, Delaunay T, Ver linden E, Golly B, Besombes JL, Jaffrezo JL, Leoz-Garziandia E (2014) Source apportionment of PM$_{10}$ in a north-western Europe regional urban background site (Lens, France) using positive matrix factorization and including primary biogenic emissions. Atmos Chem Phys 14:3325–3346
Weagle CL, Snider G, Li C, van Donkelaar A, Philip S, Bissonnette P, Burke J, Jackson J, Latimer R, Stone E, Abboud I (2018) Global sources of fine particulate matter: interpretation of PM2.5 chemical composition observed by SPARTAN using a global chemical transport model. Environ Sci Technol 52(20):11670–11681

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