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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; Lelieveld 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 (Jerret 2015; Leliveld 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.

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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 south-west 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:

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

$$\mathrm{SOC} = \mathrm{OC} - \mathrm{POC} \quad (2)$$

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 (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 ± 70 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%, ~ 6% and ~ 26%, respectively to $PM_{2.5}$ concentrations 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, TCM, 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

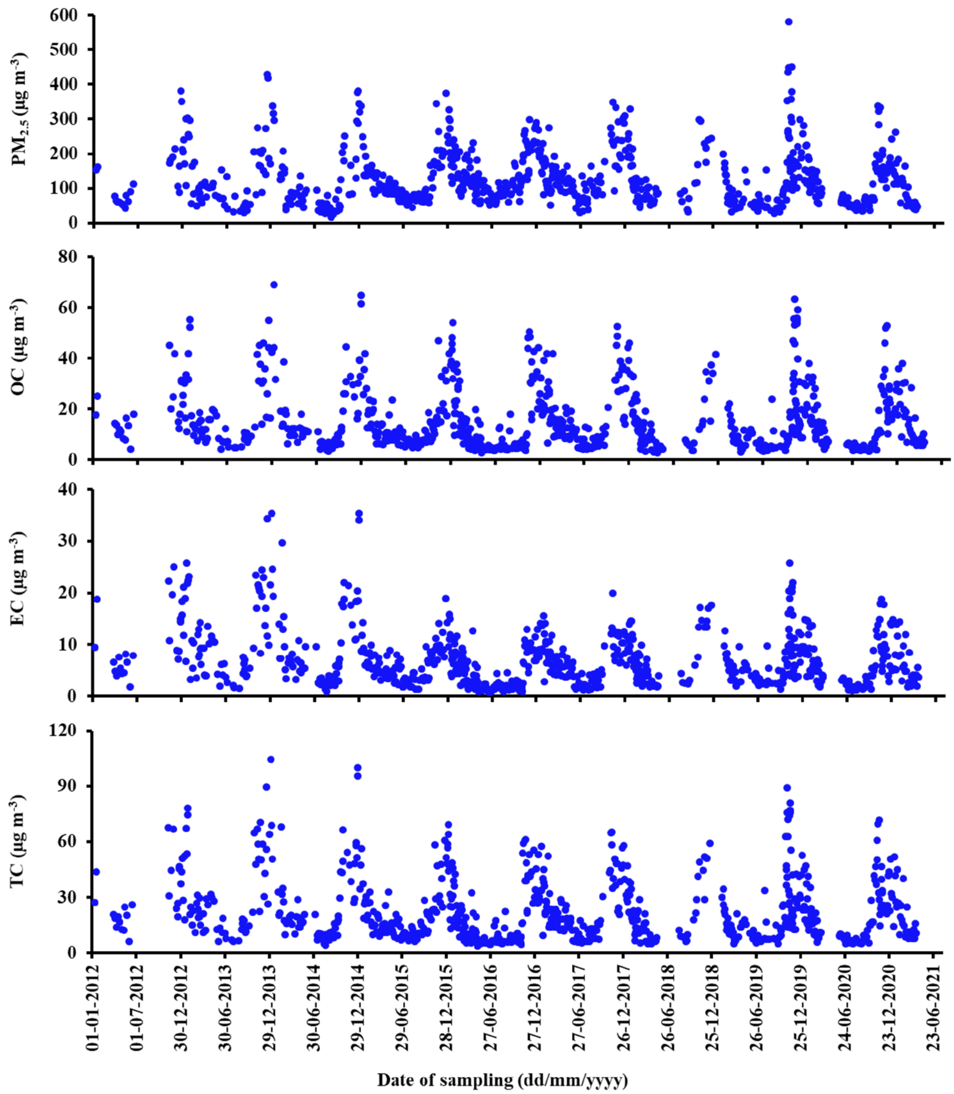


Fig. 1 Time series plot in mass concentrations of $PM_{2.5}$, OC, EC and TC during 2012–2021 (the data from July–December, 2012 and the lockdown periods in 2020 were not included)

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

Year	$PM_{2.5}$ (µg m^{-3})	OC	EC	TC	TCM	EM	POC	SOC	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	13.4 ± 8.5	5.2 ± 4.0	0.27 ± 0.16	1.85 ± 0.37	0.35 ± 0.12
2013	136 ± 91	19.3 ± 13.9	11.4 ± 7.5	30.6 ± 21.1	43.3 ± 30.1	12.5 ± 8.3	12.3 ± 8.1	7.0 ± 6.2	0.34 ± 0.31	1.79 ± 0.65	0.37 ± 0.13
2014	113 ± 96	16.6 ± 14.5	9.5 ± 8.4	26.1 ± 22.8	37.0 ± 32.3	10.5 ± 9.2	10.7 ± 9.4	5.8 ± 4.7	0.35 ± 0.20	1.75 ± 0.43	0.36 ± 0.15
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 ± 0.27	2.31 ± 0.59	0.30 ± 0.11
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 ± 0.35	2.97 ± 0.86	0.25 ± 0.12
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 ± 0.30	2.60 ± 0.73	0.28 ± 0.13
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 ± 0.29	1.97 ± 0.62	0.34 ± 0.12
2019	129 ± 96	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 ± 0.23	2.27 ± 0.48	0.31 ± 0.13
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 ± 0.32	2.31 ± 0.71	0.30 ± 0.12
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 ± 0.29	2.38 ± 0.75	0.30 ± 0.13
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 ± 0.27	2.22 ± 0.62	0.32 ± 0.12

± 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				Seasonal difference					
	Winter (W) (n = 180)	Summer (S) (n = 216)	Monsoon (M) (n = 196)	Post-Monsoon (PM) (n = 213)	W-S	W-M	W-PM	S-M	S-PM	M-PM
$PM_{2.5}$ (μg m^{-3})	158 ± 64	95 ± 35	67 ± 28	190 ± 92	62.9[a]	90.3[a]	− 32.4[a]	27.4[a]	− 95.3[a]	− 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[a]	− 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[a]	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[a]	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 $p < 0.05$)

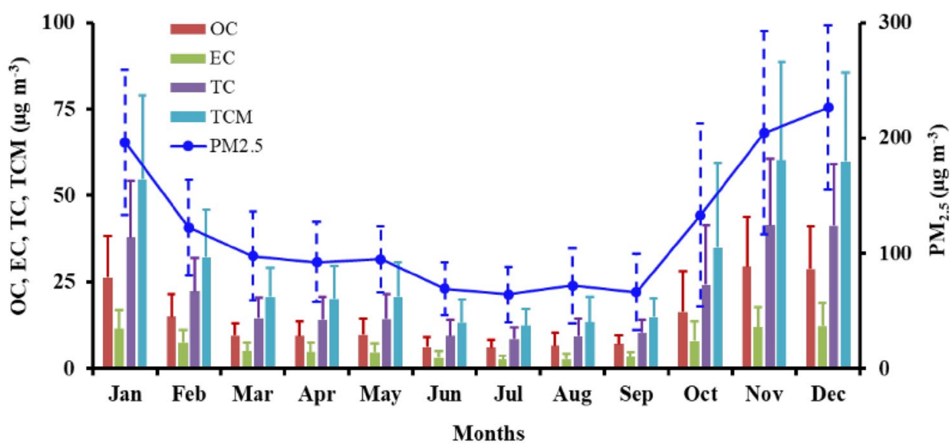
post-monsoon and winter seasons could be the source strength of $PM_{2.5}$ and prevailing meteorological conditions (temperature, relative humidity, wind speed, wind directions, etc.) as well as the lowering of mixing height of the boundary layer at the sampling site. Regional and long-range transportation 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.

Fig. 2 The monthly average variation in concentrations (error bar: ± SD) of $PM_{2.5}$, OC, EC, TC and TCM (pooled estimate of 2012 to 2021; the data from July–December, 2012 and the lockdown periods in 2020 were not included)



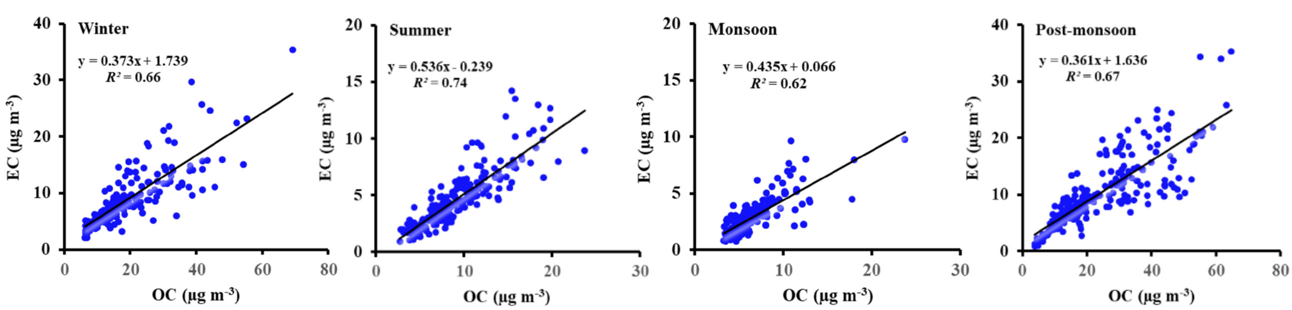


Fig. 3 Relationship of OC and EC (of $PM_{2.5}$) during winter, summer, monsoon and post-monsoon seasons (the data from July–December, 2012 and the lockdown periods in 2020 were not included)

2000; Cheng et al. 2006; Ram et al. 2010). The higher OC/EC value (between 4 and 12) is generally used for BB (Szidat 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 suggested 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.

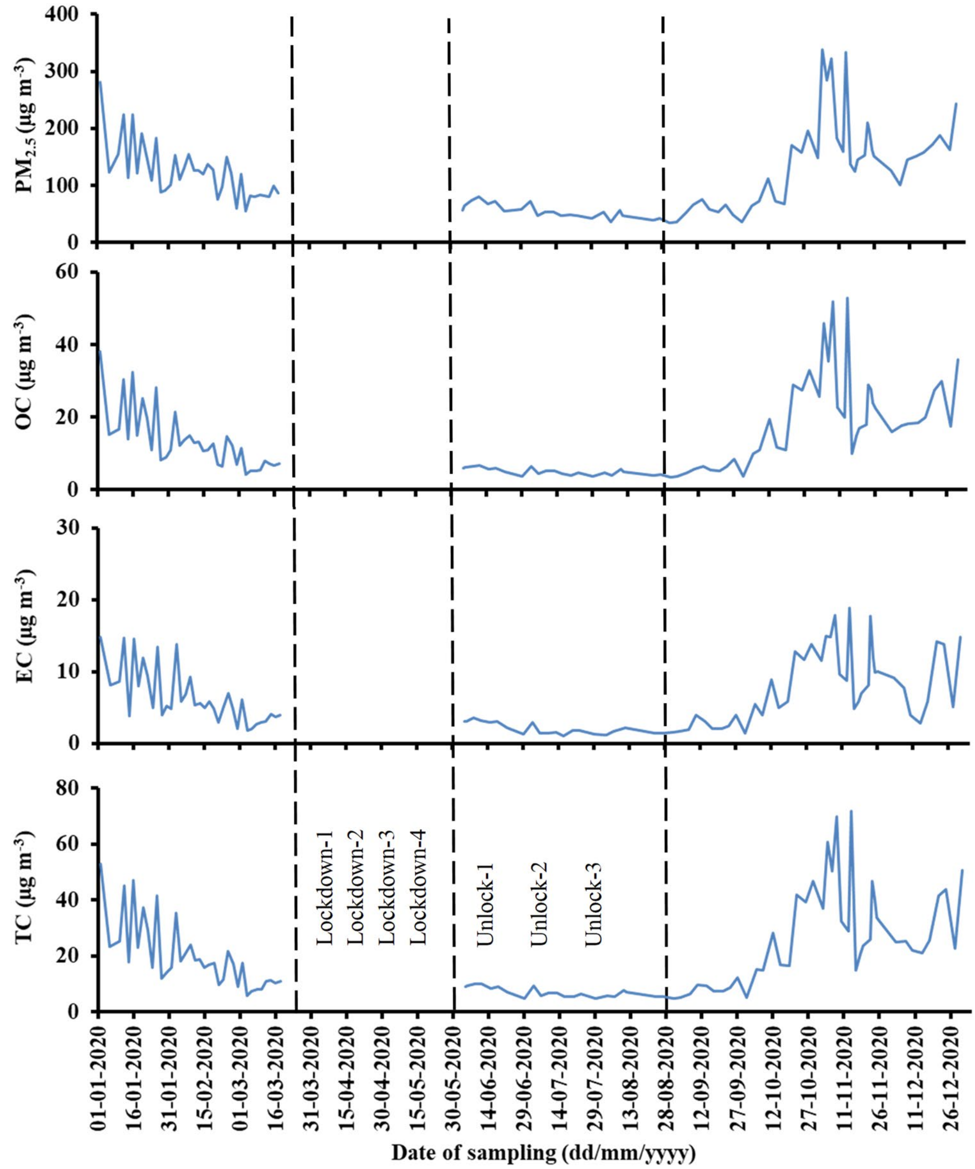


Fig. 4 Time series plots of $PM_{2.5}$, OC, EC and TC during per-lockdown, lockdown and unlock period during 2020 in Delhi (the data were not collected during the lockdown periods)

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s00128-022-03506-6.

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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.

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