Bulletin of Environmental Contamination and Toxicology (2018) 100:695–701
https://doi.org/10.1007/s00128-018-2313-9

# Carbonaceous Species of $PM_{2.5}$ in Megacity Delhi, India During 2012–2016

**S. K. Sharma[1,2] · T. K. Mandal[1] · A. Sharma[1] · Srishti Jain[1] · Saraswati[1]**

Received: 8 December 2017 / Accepted: 2 March 2018 / Published online: 7 March 2018


**Abstract**
Organic carbon (OC) and elemental carbon (EC) in $PM_{2.5}$ were estimated to study the seasonal and inter-annual variability of atmospheric total carbonaceous aerosols (TCA) at an urban site of megacity Delhi, India for 5 years from January, 2012 to December, 2016. The annual average (± standard deviation) concentrations of $PM_{2.5}$, OC, EC and TCA were 128 ± 81, 16.6 ± 12.2, 8.4 ± 5.8 and 34.5 ± 25.2 μg $m^{-3}$, respectively. During the study, significant seasonal variations in mass concentrations of $PM_{2.5}$, OC, EC and TCA were observed with maxima in winter and minima in monsoon seasons. Significant correlations between OC and EC, and OC/EC ratio suggested that vehicular emissions, fossil fuel combustion and biomass burning could be major sources of carbonaceous aerosols of $PM_{2.5}$ at the sampling site of Delhi, India.

**Keywords** $PM_{2.5}$ · Organic carbon · Elemental carbon · Total carbonaceous aerosols

Particulate matter (PM) has been recognized as a key pollutant due to its potential effects on local and regional air quality, visibility, earth's radiation budget and global climate (Fuzzi et al. 2015). There is an evidence that exposure to $PM_{2.5}$ leads to negative impacts on human health, including respiratory and cardiovascular diseases, allergies and premature mortality (Pope and Dockery 2006; Dockery and Stone 2007; Gauderman et al. 2015; Velali et al. 2016). Recent studies indicate that $PM_{2.5}$ was responsible for over 3 million premature deaths per year worldwide (Jerret 2015; Lelieveld et al. 2015) and this will be increased in the coming years if preventive measures are not taken strictly. Ambient $PM_{2.5}$ consists of organics, mineral dust, major and trace metals as well as sea salt and inorganic pollutants (Ram and Sarin 2010; Sharma and Mandal 2017). $PM_{2.5}$ containing carbonaceous aerosols has important effects on climate as well as earth's atmospheric system (Jacobson 2001). Therefore, quantification of total carbonaceous aerosols (TCA) in $PM_{2.5}$ is necessary to develop air quality improvement strategies to control and reduce ambient $PM_{2.5}$ concentrations through targeted action (Sharma et al. 2017b; Waked et al. 2014).

Organics are a major component of ambient aerosols, containing up to 40% of the fine aerosol mass (Jacobson et al. 2000; Kanakidou et al. 2005). The major source of TCA includes biomass burning, combustion of bio and fossil fuels and biogenic emissions (Venkataraman et al. 2005). The Asian continent has been inferred as a major source region of natural dust, pollution and biomass burning aerosols (Simoneit et al. 2004). Asian aerosols have a potential impact on tropospheric chemistry of the region as well as global climate forcing (Lawrence and Lelieveld 2010). Black carbon [also known as elemental carbon (EC)], organic carbon (OC) and sulphate aerosol particles significantly contribute to the atmospheric radiative forcing and climate change (Jones et al. 2005). Organic aerosols have potential to scatter sun's radiation, to reduce the hygroscopicity of inorganic species and cause variation in light scattering property of aerosols with change in relative humidity (Sjogren et al. 2007). In this context, the long-term study of carbonaceous aerosols is essential over the south Asian region. In this study, we report the seasonal and inter-annual variability in concentrations of OC, EC and TCA of $PM_{2.5}$ over a period of 5 years at an urban site of megacity Delhi, India. We have also highlighted the possible potential sources of OC, EC and TCA of $PM_{2.5}$ in megacity Delhi, India.

✉ S. K. Sharma
sudhir.npl@nic.in; sudhircsir@gmail.com

[1] CSIR-National Physical Laboratory, Dr. K S Krishnan Road, New Delhi 110 012, India

[2] Environmental Sciences and Biomedical Metrology Division, CSIR-National Physical Laboratory, Dr. K S Krishnan Road, New Delhi 110 012, India

## Materials and Methods

Delhi, the capital of India is surrounded by four different climatic zones (Himalayas in the north, central hot plains in the south, the Thar desert in the west and the Indo Gangetic plain in the east), which influence its semi-arid climate and is considered as one of the most polluted megacities in the world (Gupta et al. 2018). $PM_{2.5}$ samples were collected at CSIR-National Physical Laboratory, New Delhi (28°38′N, 77°10′E; 218 m amsl) from January, 2012 to December, 2016 (except July–December, 2012 due to malfunctioning of the instrument). The sampling site represents a typical urban environment with heavy roadside traffic all around and agricultural fields in the southwest direction. The area is under the influence of air mass flow from north-east to north-west in winter and from south–east to south–west during summer. A vehicular growth rate with around 9.70 million registered vehicles in 2015–2016 is quite alarming (Directorate of Economics and Statistics 2016). The temperature of Delhi varied from maximum (48°C) in summer (March–June) to a minimum (2°C) in winter (November–February). The average rainfall in Delhi during monsoon (July–September) was of the order of ~ 700–900 mm. A detailed description of sampling site is available in Sharma et al. (2016).

Ambient $PM_{2.5}$ samples (n = 387: 22 samples in 2012; 66 samples in 2013; 74 samples in 2014; 94 samples in 2015 and 131 samples in 2016) were collected on pre-combusted (at 550°C) and desiccator-stored quartz fiber filters (QM-A, Whatman, GE Healthcare UK Limited, Buckinghamshire, UK) by using a fine particle sampler (APM 550, Envirotech, Delhi, IN) at 10 m height above the ground level. Ambient air was passed through the quartz filter (47 mm) at a flow rate of 1 $m^3\ h^{-1}$ (accuracy ± 2%) for 24 h. The filters were weighed before and after the sampling in order to determine the mass concentration of $PM_{2.5}$. Each filter was used for OC and EC analysis in triplicate using OC/EC carbon analyzer (DRI 2001A, Atmoslytic Inc., Calabasas, CA, USA). The principle of the OC/EC analyzer is based on preferential oxidation of OC and EC at different temperatures (Chow et al. 2004). The principal function of the analyzer is to pyrolyze the sample and then to char the OC compounds into EC. The method analyses for OC fractions (OC1, OC2, OC3 and OC4 at 140, 280, 480 and 580°C), pyrolyzed carbon fraction (OP) and EC fractions (EC1, EC2 and EC3 at 580, 740 and 840°C), respectively (Chow et al. 2004). Detailed descriptions of the analytical methods, calibration procedures, etc. are available in our previous paper and reference therein (Sharma et al. 2015). Statistical analysis of $PM_{2.5}$, OC and EC data were carried out using standard recommended methods (Sharma et al. 2016). Statistically significant differences of chemical species of $PM_{2.5}$ on a seasonal basis were analyzed by Chi square method using Monte Carlo statistics (non-parametric test using SPSS software) (Datta et al. 2010). TCA is calculated as the sum of organic matter (OM = 1.6 × OC) and elemental carbon of $PM_{2.5}$ (Srinivas and Sarin 2014). A conversion factor of OC > 1.4 is suggested for urban / sub-urban aerosols (Zhang et al. 2005). The sampling site is considered as a typical urban location, hence we adopted 1.6 as a factor to convert OC to OM (1.6 × OC).

## Results and Discussion

Figure 1 shows the time series of $PM_{2.5}$, OC, EC and TCA from January, 2012 to December, 2016. The concentrations of $PM_{2.5}$ and its chemical constituents were highest during the months of December, 2013 and January, 2014 while minimum during the month of September, 2014. The annual average concentrations of $PM_{2.5}$, OC, EC and TCA with standard deviation (± SD) were 128 ± 81, 16.6 ± 12.2, 8.4 ± 5.8 and 34.5 ± 25.2 μg $m^{-3}$, respectively from January, 2012 to December, 2016 (Table 1). The highest annual average concentration of $PM_{2.5}$ was in 2013 (136 ± 91 μg $m^{-3}$), whereas the minimum concentration of $PM_{2.5}$ was in 2014 (113 ± 96 μg $m^{-3}$). Similarly, higher concentrations of OC (18.7 ± 10.6 μg $m^{-3}$), EC (10.1 ± 6.4 μg $m^{-3}$) and TCA (40.0 ± 23.2 μg $m^{-3}$) were in 2013. In the present case, the annual average concentrations of OC, EC and TCA contributed to ~ 13%, ~ 7% and ~ 27%, respectively to $PM_{2.5}$. Jain et al. (2017) reported the similar percentage contributions of OC (~ 14% of $PM_{2.5}$) and EC (~ 8% of $PM_{2.5}$) to $PM_{2.5}$ at Delhi, whereas Mandal et al. (2014) reported higher percentage contributions of OC (28% of $PM_{2.5}$) and EC (9% of $PM_{2.5}$) to $PM_{2.5}$ in an industrial area of Delhi.

The seasonal variations in mass concentrations of $PM_{2.5}$, OC, EC and TCA along with their seasonal differences are summarized in Table 2. During winter, the concentrations of $PM_{2.5}$, OC and EC were recorded more than twice as compared to the summer and monsoon seasons (Table 2). This may be due to the source strength of $PM_{2.5}$ and prevailing meteorological conditions at the sampling site. Significant lowering of mixing height of the boundary layer during the winter season may also contribute to the higher concentration of $PM_{2.5}$ (Datta et al. 2010). The highest percentage contributions of OC (13.9%), EC (6.3%) and TCA (28.6%) to $PM_{2.5}$ were recorded during winter season. Whereas during summer and monsoon seasons, more or less similar percentage contributions of OC (10%), EC (5%) and TCA (20%–22%) to $PM_{2.5}$ were observed (Table 2).

Monthly average variations in the concentrations of $PM_{2.5}$, OC, EC and TCA from January, 2013 to December, 2016 are depicted in Fig. 2. The highest monthly

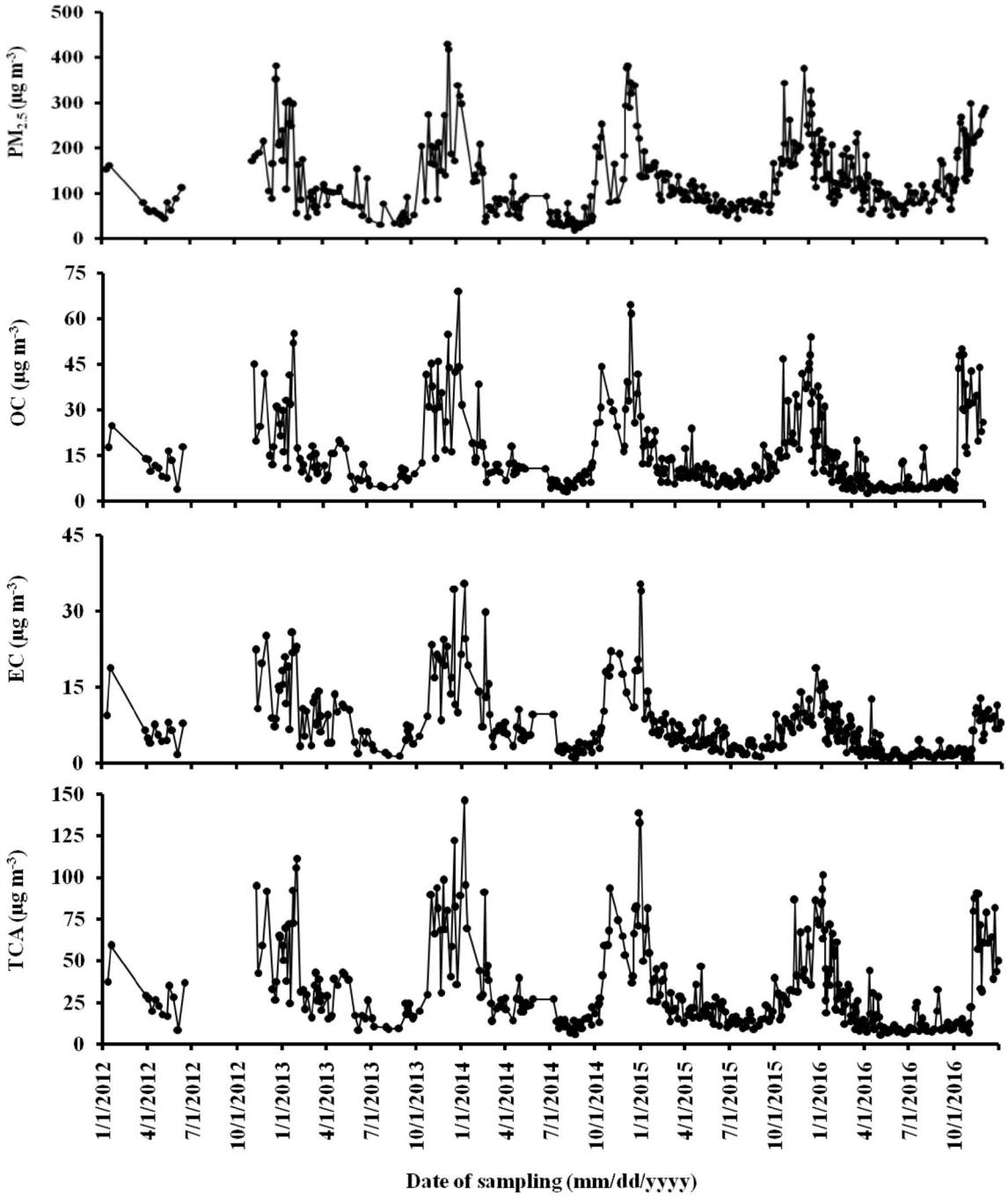


**Fig. 1** Temporal variability in mass concentrations of $PM_{2.5}$, OC, EC and TCA during 2012–2016

average concentrations of $PM_{2.5}$, OC, EC and TCA were $317 \pm 25.9$; $48.2 \pm 19.1$; $26.4 \pm 8.1$ and $104 \pm 45.9$ µg $m^{-3}$, respectively during the winter month of January, 2014 (Fig. 2). The minimum monthly average concentration of $PM_{2.5}$ was $34.4 \pm 15.7$ µg $m^{-3}$ in September, 2014. Concentrations of OC, EC and TCA were $4.8 \pm 0.4$; $1.5 \pm 0.2$ and $9.2 \pm 0.5$ µg $m^{-3}$, respectively in the monsoon month of August, 2013. Bisht et al. (2015) and Jain et al. (2017) also reported similar monthly variations in carbonaceous species (OC, EC and TCA) of $PM_{2.5}$ at Delhi. Mandal et al. (2014) reported significant monthly as well as seasonal variations in carbonaceous species of $PM_{2.5}$ in an industrial area of Delhi. Monthly as well as seasonal variations in mass concentrations of $PM_{2.5}$, OC, EC and TCA may be due to the source strength and prevailing meteorological conditions at the sampling site of Delhi, India. The changes in mixing

**Table 1** Annual average and percentage contributions of $PM_{2.5}$, OC, EC, TCA in Delhi

| Year | $PM_{2.5}$ ($\mu g\ m^{-3}$) | OC | EC | TCA | OC (%) | EC | TCA | OC/EC |
|---|---|---|---|---|---|---|---|---|
| 2012 | 133[b] ± 92 | 18.7[b] ± 10.6 | 10.1[b] ± 6.4 | 40.0[b] ± 23.2 | 14.1[b] | 7.6[b] | 30.1[b] | 1.85 |
| 2013 | 136[a] ± 91 | 19.3[a] ± 13.9 | 11.4[a] ± 7.5 | 42.2[a] ± 29.4 | 14.2[a] | 8.4[b] | 31.1[b] | 1.69 |
| 2014 | 113[a] ± 96 | 16.6[a] ± 14.5 | 9.5[a] ± 8.4 | 36.0[a] ± 31.4 | 14.6[a] | 8.4[b] | 31.8[b] | 1.75 |
| 2015 | 123[a] ± 65 | 13.8[a] ± 9.1 | 6.0[a] ± 3.3 | 28.1[a] ± 17.6 | 11.2[a] | 4.9[b] | 22.8[a] | 2.30 |
| 2016 | 134[a] ± 64 | 14.5[a] ± 13.2 | 4.9[a] ± 3.8 | 28.1[a] ± 24.6 | 10.8[a] | 3.7[a] | 21.0[a] | 2.96 |
| Mean | 128 ± 81 (17.4–429) | 16.6 ± 12.2 (2.7–69.1) | 8.4 ± 5.8 (0.78–35.3) | 34.5 ± 25.2 (5.21–145.9) | 13.0 | 6.6 | 27.4 | 2.11 |

± Standard deviation (n = 387 for 5 years); values in parentheses are ranges

[a]Significantly different ($p < 0.05$)

[b]Significantly not different ($p < 0.05$); (used Chi square: Monte Carlo method; at 95% confidence level)

**Table 2** Seasonal variations in $PM_{2.5}$, OC, EC, TCA concentrations and their percentage contribution

| Species | Season | | | Seasonal difference | | |
|---|---|---|---|---|---|---|
| | Winter (W) (n = 142) | Summer (S) (n = 131) | Monsoon (M) (n = 114) | W–S | W–M | S–M |
| $PM_{2.5}$ ($\mu g\ m^{-3}$) | 202.5 ± 76.5 | 99.7 ± 35.0 | 80.0 ± 42.6 | 102.8[a] | 122.5[a] | 19.7[a] |
| OC ($\mu g\ m^{-3}$) | 27.4 ± 13.7 | 9.3 ± 4.2 | 8.6 ± 6.6 | 18.1[a] | 18.8[a] | 0.7[a] |
| EC ($\mu g\ m^{-3}$) | 12.1 ± 7.0 | 4.9 ± 3.1 | 4.2 ± 4.0 | 7.2[a] | 7.9[a] | 0.7[b] |
| TCA ($\mu g\ m^{-3}$) | 56.0 ± 27.5 | 19.8 ± 9.5 | 18.0 ± 14.4 | 36.2[a] | 38.0[a] | 1.8[b] |
| OC (%) | 13.9 ± 6.6 | 10.3 ± 5.6 | 10.8 ± 7.4 | 3.6[a] | 3.1[a] | −0.5[b] |
| EC (%) | 6.3 ± 3.9 | 5.5 ± 3.8 | 5.3 ± 4.5 | 0.8[b] | 1.0[b] | 0.2[b] |
| TCA (%) | 28.6 ± 13.7 | 19.9 ± 12.6 | 22.5 ± 14.2 | 8.8[a] | 6.1[a] | −2.6[a] |

± Standard deviation

[a]Significantly different ($p < 0.05$)

[b]Significantly not different ($p < 0.05$)

height of the boundary layer during various seasons may also contribute to higher concentration of $PM_{2.5}$ and its carbonaceous species (Datta et al. 2010).

Emissions from vehicles and combustion of biomass (cow dung, agricultural waste and wood burning) are known to contribute significantly to atmospheric OC and EC (Ram and Sarin 2010). A significant linear correlation between OC and EC is usually indicative of similar sources like vehicular emissions or biomass burning (Salma et al. 2004; Ram et al. 2010; Sharma et al. 2014; Jain et al. 2017). Conversely, weakly correlated values of OC and EC suggest the presence of secondary aerosols and signify favorable conditions for gas-to-particle conversion of VOCs via photochemical atmospheric reactions (Begum et al. 2004, 2006). Significant positive linear relationships between OC and EC during winter ($R^2 = 0.57$; at $p < 0.05$), summer ($R^2 = 0.79$; at $p < 0.05$) and monsoon ($R^2 = 0.90$; at $p < 0.05$) seasons were observed (Fig. 3) at the sampling site, indicating a degree of influence from either vehicular emissions or biomass burning.

Several studies have indicated that the ratios of chemical composition of $PM_{2.5}$ may also give significant information about their sources (Novakov et al. 2000; Cheng et al. 2006; Ram et al. 2010). Fossil fuels (vehicular emission, industrial activities and small scale generators) could also be the dominant sources of OC and EC in the megacity like Delhi. During the study, the average ratio of OC/EC of $PM_{2.5}$ was 2.11 whereas annual average ratios of OC/EC were 1.85, 1.69, 1.75, 2.30 and 2.96 during 2012, 2013, 2014, 2015 and 2016, respectively (Table 1). The OC/EC ratio also depends on both the proximity of the emissions and relative weight of road traffic and biomass burning. The OC/EC ratio of road traffic emissions generally varies between 1.4 and 4 (Amato et al. 2009; Salameh et al. 2015). Large values (between 4 and 12) of this ratio are generally found for biomass burning emissions (Szidat et al. 2006). Average values of OC/EC proffer the evidences of emissions from vehicles along with biomass burning, accounting the dominant sources of carbonaceous species of $PM_{2.5}$ at the sampling site of Delhi. Analysis of stable carbon isotopic composition of $PM_{2.5}$ indicated that vehicular emissions and biomass burning are the major sources of PM over the Delhi region (Sharma et al. 2015; 2017a). Jain et al. (2017) has shown that biomass burning

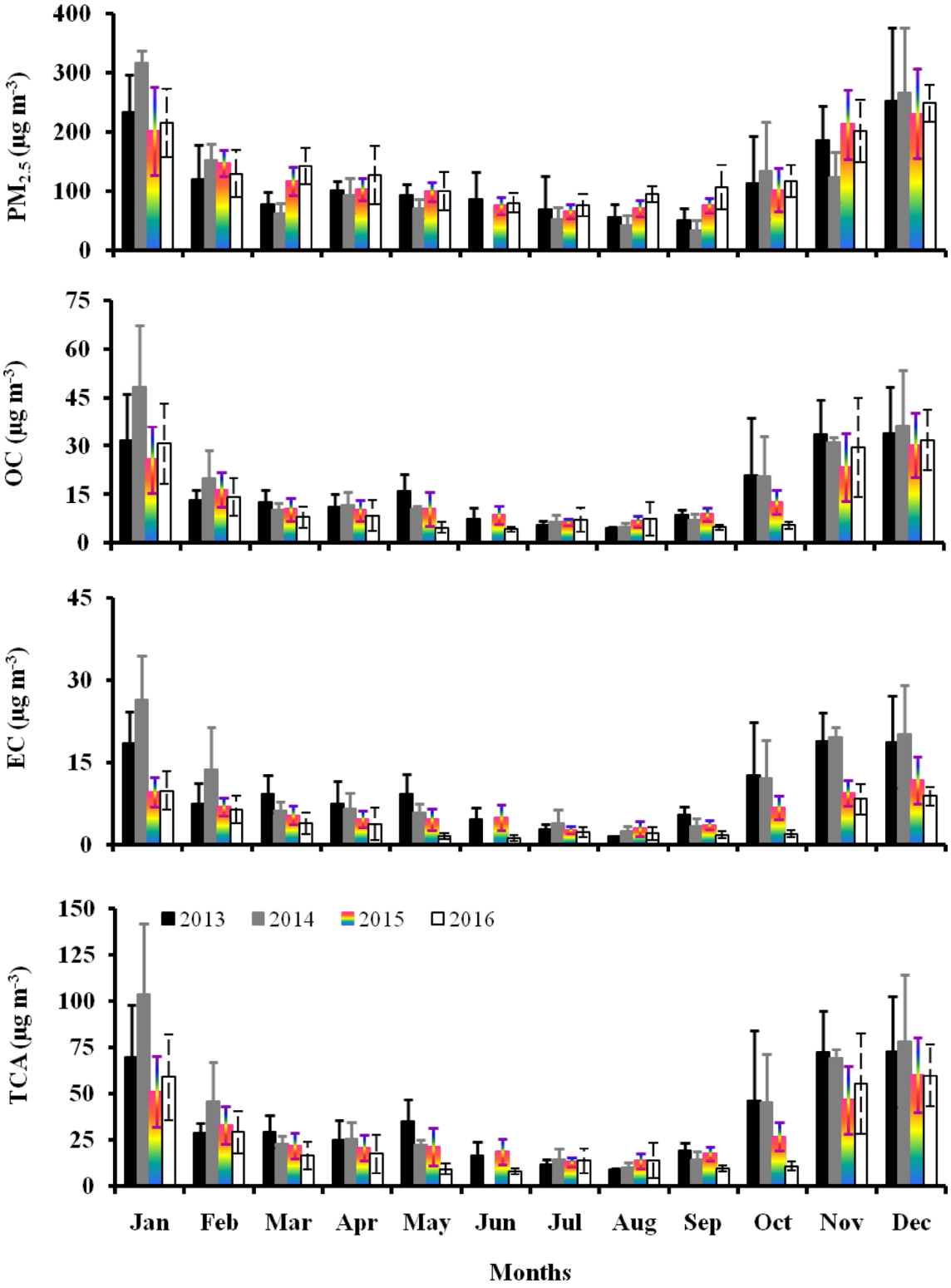


**Fig. 2** Monthly variation in concentrations of OC, EC and TCA of $PM_{2.5}$ along with $PM_{2.5}$ from 2012 to 2016

and vehicular emissions are the major sources of OC and EC of $PM_{2.5}$ in megacity Delhi. In urban areas, the number of vehicles, industries and influence of human activities are increasing with time and expected to increase in the near future, which is believed to augment the abundance of TCA over the region. The carbonaceous aerosols have significant

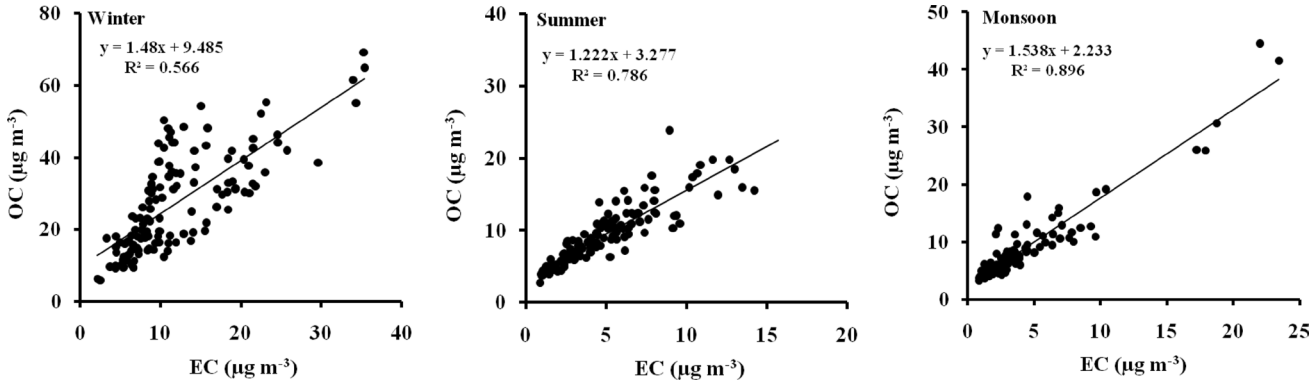


**Fig. 3** Scatter plots between OC versus EC of $PM_{2.5}$ during winter, summer and monsoon seasons in Delhi

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.

**Acknowledgements** The authors are thankful to the Director, CSIR-NPL, New Delhi and Head, Environmental Sciences & Biomedical Metrology Division (ES&BMD), CSIR-NPL, New Delhi for their encouragement. The authors also acknowledge the Ministry of Environment Forest and Climate Change (MoEFCC), New Delhi for providing partial financial support for this study (GAP-161632 Project).

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