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Long-Term (2012–2021) Variation in Carbonaceous Aerosols of $PM_{2.5}$ at an Urban Site of Megacity Delhi Situated over Indo-Gangetic Plain of India †

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Abstract: A long-term (January 2012 to December 2021) study on carbonaceous aerosols of fine particulates ($PM_{2.5}$) was conducted over the megacity of Delhi, India, to evaluate their seasonal and yearly variations. During the entire study period, the observed annual mean levels (μg m^{-3}) of $PM_{2.5}$ and its carbonaceous components (OC, POC, SOC, EM, EC, TCM, and TC) were recorded as 126 ± 72, 15.6 ± 11.6, 9.3 ± 6.3, 6.4 ± 5.1, 8.2 ± 5.6, 7.3 ± 5.1, 33.2 ± 21.9, and 23.1 ± 16.5, respectively. On average, the CAs/TCM ratio accounts for 26% of $PM_{2.5}$ concentrations. During the monsoon (minimum) and post-monsoon (maximum) season, significant seasonal variability in $PM_{2.5}$ and its carbonaceous species (OC, EC, POC, SOC, and TCM) was observed. Based on the linear association (OC vs. EC) and ratios (OC/EC as well as EC/TC) of species, three significant sources of CAs (vehicular emissions (VE), fossil fuel combustion (FFC), and biomass burning (BB)) were identified.

Keywords: $PM_{2.5}$; aerosols; carbonaceous species; OC; IGP region

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1. Introduction

Carbonaceous aerosols (CAs) are the primary contributors of ambient particulates, accounting for up to 70% of fine aerosol mass [1], and they substantially influence atmospheric chemistry, climate, and human health [2]. Majorly, the emission of CAs occurs from fossil fuel combustion, biomass burning, and biogenic emissions [3]. Numerous studies have been carried out in the recent past on CAs of $PM_{2.5}$ and their potential sources in urban [4,5], rural, remote [6], as well as high-altitude atmospheres [7] of the Indian region with a yearlong dataset or less, but limited studies are available on a long-term basis. Therefore, to comprehend a more detailed knowledge of fine aerosols ($PM_{2.5}$) and their atmospheric processes and sources, the long-term research of CAs is crucial in the Indo-Gangetic plain (IGP) region of India. The seasonal and annual changes in $PM_{2.5}$ and its carbonaceous components, such as organic carbon (OC), primary organic carbon (POC), secondary organic carbon (SOC), elemental carbon (EC), total carbonaceous matter (TCM), and total carbon (TC), at the urban site of Delhi were reported in this paper, along with their possible sources.

2. Materials and Methods

Periodic sampling (2 samples a week) of $PM_{2.5}$ was performed from January 2012 to December 2021 at the CSIR-National Physical Laboratory (28°38′ N, 77°10′ E; 218 m above mean sea level). Delhi, the capital of India, is surrounded by different climatic zones and is considered one of the most polluted megacities in the world [8]. The meteorology of Delhi

demonstrates four distinct seasons, which are winter (January to February; temperature: ~3 °C), summer (March to May; temperature: 47 °C), monsoon (June to September; rain: 800 mm), and post-monsoon (October to December). A detailed description of the sampling location is available in our earlier publication [9].

$PM_{2.5}$ samples were collected on quartz filters (size: 47 mm) using a fine particle sampler (flow rate: 1 m^3 h^{-1}) for 24 h. A Thermo-optical carbon analyzer (Model: DRI 2001A) was used to measure OC and EC following the IMPROVE-A protocol [10]. Detailed operating procedures of the instrument are available in Sharma et al. [9]. The TCM concentration can be computed by the addition of OM (OM = 1.6 × OC; 1.6 factor for $PM_{2.5}$) and EM (EM = 1.1 × EC) of $PM_{2.5}$ [11,12]. The EC tracer approach [13] was used for determining the SOC (Equation (1)):

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

3. Results and Discussion

Figure 1 illustrates the annual changes in $PM_{2.5}$ and their carbonaceous components (OC, EC, and TC) during the study period (January 2012 to December 2021). In this long-term study, the annual mean (± standard deviation) concentrations (μg m^{-3}) of $PM_{2.5}$ and their carbonaceous components (OC, POC, SOC, EM, EC, TCM, and TC) were recorded as 126 ± 72, 15.6 ± 11.6, 9.3 ± 6.3, 6.4 ± 5.1, 8.2 ± 5.6, 7.3 ± 5.1, 33.2 ± 21.9, and 23.1 ± 16.5, respectively. Among the species, the contribution (with regard to annual mean level) to $PM_{2.5}$ was observed highest by TCM (~26%), followed by OC (~12%) and EC (~6%). Similar observations were also stated by Jain et al. [5], with ~25% of TCM, ~12% of OC, and ~5.5% of EC to $PM_{2.5}$ over Delhi.

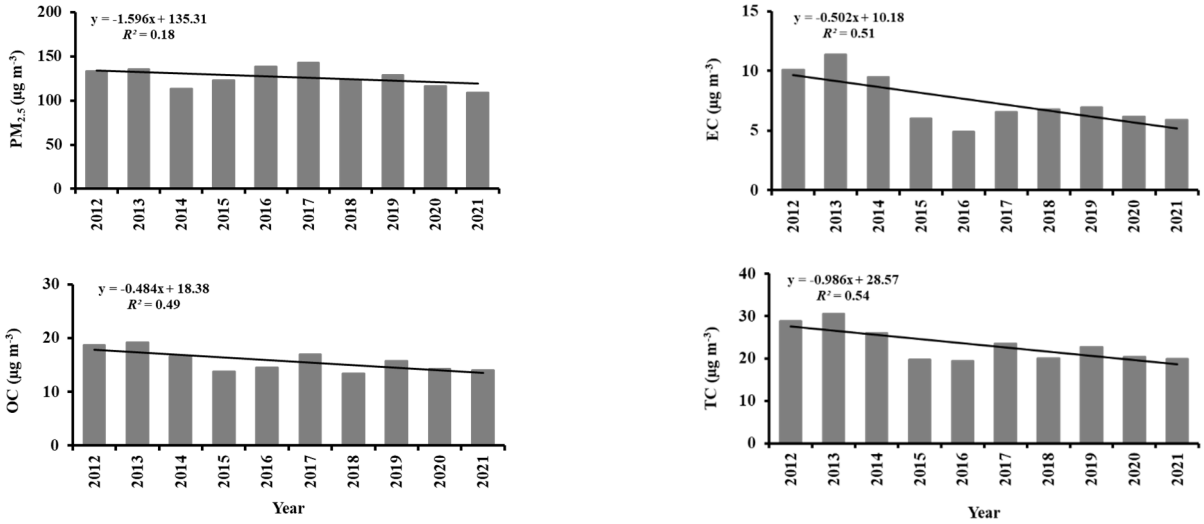

Figure 1. Annual trend in $PM_{2.5}$, EC, OC, and TC levels over Delhi.

Figure 2 illustrates the seasonal variability in $PM_{2.5}$ and its carbonaceous species (OC, EC, TC, and TCM). $PM_{2.5}$ and its CAs (OC, EC, TCM, POC, and SOC) were recorded as minimum and maximum during the monsoon and post-monsoon season, respectively (Table 1). This might be owed to the source intensity of $PM_{2.5}$, prevailing meteorological circumstances, and the long-distance movement of pollutants from Punjab and Haryana to the receptor location [8].

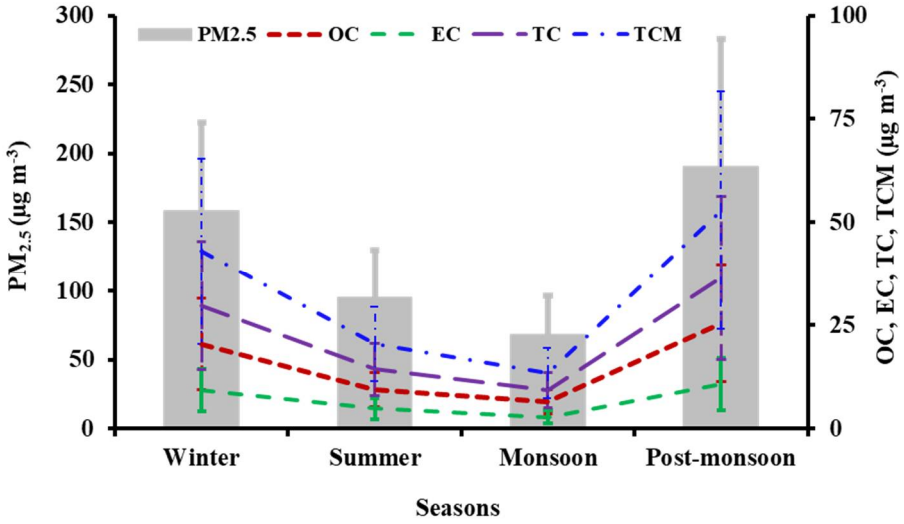

Figure 2. Seasonal mean of (error bar: ± SD) OC, EC, TC, and TCM of $PM_{2.5}$ in Delhi.

Table 1. Seasonal mean of $PM_{2.5}$ and their carbonaceous species (µg m^{-3}) in Delhi.

Species	Seasons			
	Winter	Summer	Monsoon	Post-Monsoon
$PM_{2.5}$	157 ± 64	94 ± 35	66 ± 28	189 ± 92
OC	20.6 ± 10.9	9.4 ± 4.3	6.7 ± 2.8	25.8 ± 14.2
EC	9.3 ± 5.2	4.9 ± 2.7	2.8 ± 1.5	10.8 ± 6.5
TC	29.7 ± 15.5	14.4 ± 6.5	9.5 ± 4.3	36.5 ± 19.4
POC	11.8 ± 5.9	6.2 ± 3.1	3.4 ± 1.7	13.2 ± 7.1
SOC	8.3 ± 6.6	3.5 ± 1.8	3.0 ± 1.6	12.3 ± 8.6
OC/EC	2.3 ± 0.7	2.1 ± 0.6	2.2 ± 0.8	2.5 ± 0.8

The mass ratio of OC/EC has been used to identify the possible sources of CAs in most studies [14,15]. The higher value of the OC/EC ratio that ranges from 4 to 12 indicates the dominance of biomass burning (BB) [15]. On the other hand, when the OC/EC ratio ranges between 1.4 and 4, it indicates the occurrence of VE together with BB [16]. During the study period, the observed average ratio of OC/EC of $PM_{2.5}$ were 2.3 ± 0.7, 2.1 ± 0.6, 2.2 ± 0.8, and 2.5 ± 0.8 during winter, summer, monsoon, and post-monsoon seasons, respectively (Table 1). From the observed OC/EC mass ratio, BB and FFC are the major sources of CAs over study site in Delhi. Jain et al. [5] have also reported that BB, FFC, and VE are the main sources of $PM_{2.5}$ in the megacity Delhi. Sharma et al. [17] investigated the stable carbon and nitrogen isotope of aerosols at a Delhi metropolitan location and revealed that the VE, BB, and FFC are the most likely main sources of aerosols in the city. These sources are also influenced by regional and long-distant transport of pollutants in Delhi.

4. Conclusions

In the present study of a 10-year long-term dataset, the annual and seasonal mean concentrations of $PM_{2.5}$ and all carbonaceous species vary significantly in the megacity Delhi. In total, about 26% of CAs account for the $PM_{2.5}$ mass concentration. A substantial variability in the seasonal concentrations of $PM_{2.5}$ and its associated Cas were observed as follows: post-monsoon > winter > summer > monsoon seasons. A linear relationship was observed between OC and EC, which demonstrates that pollutants are generated from similar combustion sources in Delhi, India. In addition, BB, VE, and FFC are the prominent sources of CAs of $PM_{2.5}$ suggested by the OC/EC, as well as EC/TC ratios. Thus, the observed results state the significant effect of CAs from different sources.

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Conflicts of Interest: The authors declare no conflict of interest.

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