Research Article

Seasonal variation of carbonaceous species in PM_1 measured over residential area of Delhi, India

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Abstract
Seasonal variation in the concentration of Organic Carbon (OC) and Elemental Carbon (EC) was investigated in PM_1 size fraction over a residential area in Delhi, India. The sampling was carried out using a cascade impactor from April 2018 to March 2019. The OC/EC content in the sample was measured by a carbon analyzer (Thermal/Optical, DRI 2001). The annual average concentration of OC and EC was found to be 28.01 ± 14.61 μg/m^3 and 10.40 ± 7.41 μg/m^3, respectively in PM_1 (139.52 ± 49.20 μg/m^3). The highest concentrations of OC and EC were observed in the post-monsoon and winter season, respectively. Average OC & EC concentrations in summer, monsoon, post-monsoon, and winter seasons were found to be 22.57 ± 3.72 & 5.51 ± 1.28, 11.70 ± 3.07 & 3.60 ± 0.39, 42.06 ± 7.10 & 11.72 ± 2.45 and 37.28 ± 14.45 & 18.52 ± 6.23 μg/m^3, respectively. Total carbonaceous material (TCM) accounted for approximately 40% of PM_1 concentration. The annual average OC/EC ratio was observed to be 2.69, which ranged between 1.77 to 5.39. Analysis of carbon subfractions reveals OC3 and EC1 as the dominant fraction suggesting fossil fuel combustion as dominant emission sources. The exposure risk estimated on the basis of inhalation dose was observed very high during the winter season which can adversely affect the health of people. This study also revealed that the emissions from the residential area are the key source of OC and EC, along with PM_1.

Keywords Elemental carbon · Fine mode · Coarse mode · Organic carbon · Residential area

1 Introduction

Air pollutants released from various types of activities comprise a complex mixture of large and small particles [1]. These particles may be of multiple size fractions, thus in recent times, researches have focused on particles of diameter between 0.01–100 μm [2]. The atmospheric particles having a size (aerodynamic diameter) of less than 2.5 μm ($PM_{2.5}$), and more than 2.5 μm are considered as fine mode (FMP) and coarse mode (CMP) particles, respectively [3]. Most of the anthropogenic activities produce particles with a diameter of ≤ 1 μm that dominate in the urban atmospheric environment [4–6]. A number of inorganic and organic chemical species are found associated with particles of various size fractions [7]. Among these chemical species, carbonaceous fractions of atmospheric aerosol are becoming the matter of great concern in the world because it takes part in the formation of smog, and degradation of visibility by various types of chemical reaction in the atmosphere [8]. Its adverse effects are not only limited to the local area, but also plays a significant role in global warming, climate change, and melting of glaciers [9, 10]. In addition to the various adverse environmental effects, it also affects human health in many ways [11].

The carbonaceous species of atmospheric aerosols are classified into Organic Carbon (OC), Elemental Carbon (EC) or Black Barbon (BC) [12, 13]. Generally, OC contributes 10–50%, while EC contributes only a minor portion of

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the total mass concentration of atmospheric PM in many regions of the world [2]. Further, OC and EC contribute approximately 90% and 5–10% to the total carbon (TC) in the ambient air [14–16]. EC is directly released into the ambient air as a primary pollutant, while OC is produced as a Primary Organic Carbon (POC) by combustion activities or as Secondary Organic Carbon (SOC) by photochemical reaction [17, 18]. OC consists of a number of organic compounds that may be of aromatic or aliphatic in nature, while EC is mainly composed of graphite-like structure [7, 19]. OC is scattering in nature, and it cools the atmosphere by reflecting sunlight, whereas EC absorbs sunlight and produces warming effects [20]. In the atmosphere, biomass burning, emissions from vehicular movement, incomplete combustion of fossil fuel, coal combustion and brick kiln are the important anthropogenic sources of carbonaceous species [21, 22]. In recent years, variation in mass concentrations, source identification, and adverse impacts of BC or EC, and OC have been broadly studied at various locations of the world [23–42]. A brief summary of the OC/EC study carried out in different parts of the world is presented in Table 1.

Delhi has been ranked as one of the highly polluted capital cities in the world [43–45]. Vehicular emissions, thermal power plants, road dust, and industries are the important sources of aerosol particles in Delhi [46, 47]. Smog formation which results in bad air, eye irritation and poor visibility is common phenomenon that can be observed during post-monsoon and winter seasons [48]. Further, the situation gets worse during the Diwali (festival dominated by fire crackers) period due to a very high pollution load [49]. During winter season, Air Quality Index (AQI) reaches up to hazardous/emergency level due to the increased incidence of agricultural waste burning in neighboring states [50]. Therefore, various researches have been done to examine the probable adverse impact of OC and EC on the atmospheric properties. It has been also reported that OC and EC account for higher mass percentage in PM_1 than in bigger PM size fractions [27, 51–54]. Further, the data available on OC and EC concentration in the PM1 size fractions in India are very limited to the researcher. Thus, the present study was carried out to examine the seasonal variation of OC and EC in PM_1 over a residential area in Delhi, India.

2 Materials and methods

2.1 Study area and sampling site

Delhi is the capital city of India, located (28.61°N, and 77.23°E) in the northern part of the country covers an area of 1483 km^2 (out of which 1114 km^2 falls in urban and 369 km^2 in rural areas) and 16.79 million population. Here, summer is very hot (45–48 °C), while winter is characterized by very low temperatures (1–2 °C) during the coldest months of December and January [55]. The sampling site (Janakpuri) represent residential area, and is located in the South–West part of Delhi. This area is far from the influence of any industrial activities. It is located near the busy outer ring road, which connects different parts of the city with regular traffic movement. The eatery, commercial shops, petrol pumps, and restaurants are also located in the nearby area. The location of the sampling site is shown in Fig. 1.

Table 1 Summary of important studies reported at different locations of the world

S. No	City	Country	Site	PM Size	OC (μg/m^3)	EC (μg/m^3)	OC/EC	Reference
1	Delhi	India	Residential	PM_1	28.01	10.4	2.69	Present Study
2	Delhi	India	Urban	$PM_{2.5}$	50.11	10.67	5.43	Pipal et al. [64]
3	Delhi	India	Industrial	$PM_{2.5}$	41.12	13.25	3.28	Mandal et al. [36]
4	Delhi	India	Urban	$PM_{2.5}$	51.1 ± 15.2	10.4 ± 5.5	5	Tiwari et al. [42]
5	Delhi	India	Urban	$PM_{2.5}$	37.73 ± 14.32	7.79 ± 3.73	5.86 ± 0.99	Bisht et al. [33]
6	Delhi	India	Urban	$PM_{2.5}$	23.2	10.7	2.2	Srivastava et al. [41]
7	Kanpur	India	Urban	$PM_{2.5}$	47 ± 21	7.7 ± 2.9	6.0 ± 1.3	Ram and Sarin [100]
8	Beijing	China	Industrial	$PM_{2.5}$	13.3 ± 14.0	3.3 ± 2.7	3.7	Wang et al. [25]
9	Tianjin	China	Industrial	$PM_{2.5}$	56.8 ± 33.9	8.9 ± 2.2	5.8	Li and Bai [26]
10	Xiamen	China	Urban	PM_{10}	20.5 ± 12.5	4.2 ± 2.0	5.3	Niu et al. [27]
11	Hong Kong	China	Residential	$PM_{2.5}$	12	3.4	3.5	Duan et al. [28]
12	Seoul	Korea	Urban	$PM_{2.5}$	4.1 ± 2.70	1.6 ± 1.0	2.9 ± 2.7	Park et al. [29]
13	Seoul	Korea	Urban	$PM_{2.5}$	10.2	4.1	2.4	Kim et al. [30]
14	Thessaloniki	Greece	Urban	PM_1	2.11	0.09	24.03	Samara et al. [31]
15	Alicante	Spain	Industrial	$PM_{2.5}$	2.5	1.0	2.8	Yubero et al. [32]

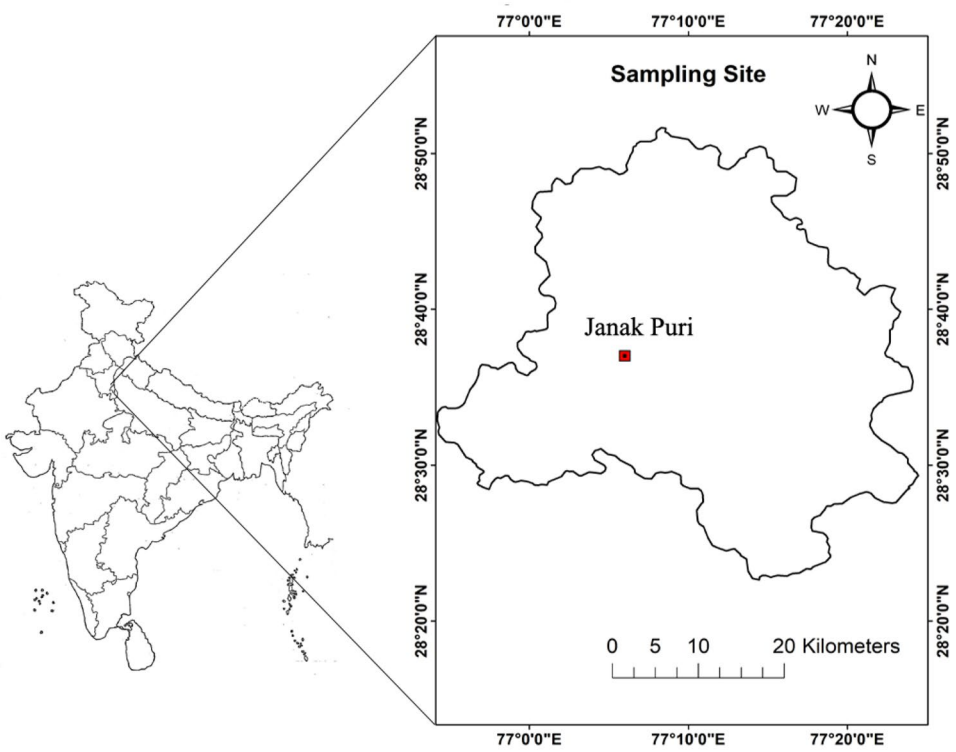

Fig. 1 Map showing location of the aerosol sampling site (Janakpuri, Delhi)

2.2 Aerosol sampling

The aerosol sampling was performed in the Janakpuri residential area from April 2018 to March 2019. Size fractioned aerosol particles were collected on Whatman Quartz Microfiber Filter (QMF) using a Dekati PM10 cascade impactor sampler. A total of thirty-six aerosol samples (three samples per month) were collected. Similar types of protocols have been observed to follow by other researchers [56–58]. The filter was also pre-heated in a muffle furnace at 700 ˚C for 5 h in order to lower the carbon contents (OC and EC) in the blank QMF [59]. The impactor was installed at the rooftop of a residential apartment, and operated with the flow rate of thirty liters per minute (30-LPM) for 24 h. The cascade impactor collects aerosol particles in 4 different size fraction (≥ 10 µm, 10–2.5 µm, 2.5–1.0 µm, and ≤ 1.0 µm). In this study, the aerosol particles deposited in ≥ 10 µm 10–2.5 µm, 2.5–1.0 µm, and ≤ 1.0 µm size range were represented as ≥ PM_{10}, $PM_{10-2.5}$, $PM_{2.5-1}$, and ≤ PM_1. To investigate the seasonal difference of OC/EC concentration, the entire period of study was further sub-divided into summer (April, May, June), monsoon (July, August, September), post-monsoon (October, November), and winter (December, January, February, March) seasons [19]. Temperature, Relative Humidity (RH), and Wind Speed (WS) during the sampling period were recorded with the help of Envirometer (Fisher Scientific) and illustrated in Fig. 2.

2.3 OC/EC analysis

A carbon analyzer (Thermal/Optical, DRI 2001) was used to measure EC and OC concentration in the aerosol sample collected on QMF. The instrument measures carbon contents present in the aerosol sample by heating the quartz filter in a step-by-step manner at different temperature ranges (140–840 °C) [60]. The carbon analyzer releases carbon contents in 8 different carbon subfractions such as OC1, OC2, OC3, OC4, EC1, EC2, EC3, and OP or pyrolyzed organic carbon (OCPyro). The OC subfractions are released in a non-oxidizing (Helium with 99.99% purity), while EC sub-fractions in the oxidizing atmosphere (98% He + 2% O_2). The OCpyro or OP is evolved when carrier gas flow is changed from non-oxidizing to the oxidizing atmosphere at 580 °C. After OC/EC analysis carbon analyzer measure OC, EC, and TC concentration according to the following equation:

$$OC = OC1 + OC2 + OC3 + OC4 + OCPyro \quad (1)$$

$$TC = OC1 + OC2 + OC3 + OC4 + EC1 + EC2 + EC3 \quad (2)$$

$$EC = TC - OC \quad (3)$$

The aerosol sampling method by the cascade impactor and experimental protocol for OC/EC analysis by the

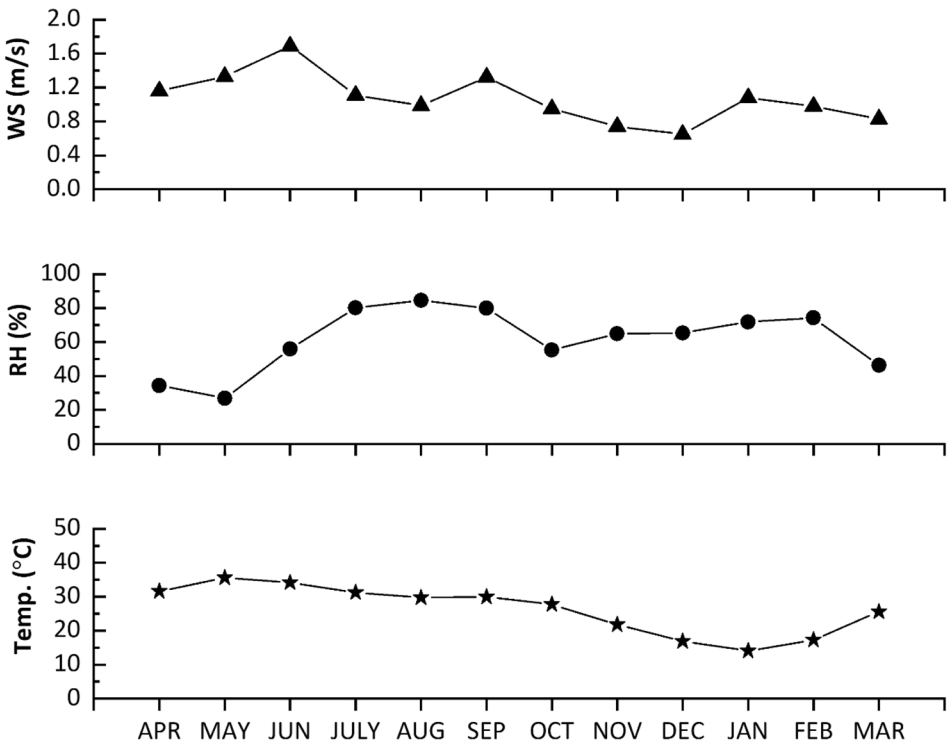

Fig. 2 Monthly variation of temperature (Temp), relative humidity (RH) and wind speed (WS) during 2018–2019 in Delhi

carbon analyzer (DRI 2001) has already been described in detail by Singh, Srivastava [61].

3 Results and discussion

3.1 Size segregated PM analysis

The average concentration of aerosol particles deposited in various size fractions during the entire study period is presented in Table 2. The particles accumulated in 4 different size fractions (≥ 10 µm, 10–2.5 µm, 2.5–1.0 µm and ≤ 1.0 µm) were again sub-divided into fine (≤ PM2.5) and coarse (≥ PM2.5) mode particulate matter (FMP & CMP). Further, Total Suspended Particles (TSP) were also estimated by adding up the mass of particles of first 4 different size fractions (≥ 10 µm + 10–2.5 µm + 2.5–1.0 µm + ≤ 1.0 µm). The annual average TSP concentrations were found to be 347.25 ± 132.49 µg/m^3. The highest deposition of aerosol particles was observed in the PM_1 among four different size ranges. Further, annual average concentrations of aerosol particles deposited in ≥ PM_{10}, $PM_{10-2.5}$, $PM_{2.5-1}$, and ≤ PM_1 were found to be 38.71 ± 14.55, 76.76 ± 24.37, 92.25 ± 61.20 and 139.52 ± 49.20 µg/m^3, respectively. Out of twelve different months, highest concentration of aerosol particles was observed in December whilst the lowest in July (Fig. 3).

A clear seasonal variation was observed in the mass concentration of aerosol particles deposited in all the 4 size ranges. Among 4 different seasons, the highest concentration of TSP was found during winter (473.46 ± 66.04 µg/m^3) while lowest in monsoon season (160.99 ± 26.12 µg/m^3). Further, average concentration of ≥ PM_{10}, $PM_{2.5-10}$, $PM_{1-2.5}$, and ≤ PM_1 during winter season were found to be 43.70 ± 5.58 µg/m^3, 85.77 ± 10.43 µg/m^3, 159.09 ± 8.94 µg/m^3, and 184.90 ± 10.87 µg/m^3, respectively. The season-wise TSP concentration was found in the order of monsoon < summer < post-monsoon < winter. Further concentration of PM_{10} and $PM_{2.5}$ were also calculated and compared with the National Ambient Air Quality Standard (NAAQS) of India. Here, the annual average concentration of PM_{10} (308.54 ± 121.25 µg/m^3) and $PM_{2.5}$ (231.77 ± 107.03 µg/m^3) was found approximately 5 times higher than the NAAQS prescribed limits (i.e., PM_{10} = 60 µg/m^3 and $PM_{2.5}$ = 40 µg/m^3). Various researchers have observed the higher concentration of $PM_{2.5}$ and PM_{10} than the maximum permissible limit set under the NAAQS in Delhi, India [62, 63]. The annual average concentrations of CMP and FMP particles were found to be 115.47 ± 36.20 and 231.77 ± 107.03 µg/m^3, respectively. FMP (67%) dominated the CMP (33%) during the entire period of study. The percentage contribution of FMP (73%) in TSP was observed more than the CMP (27%) during the winter season. Further, the dominance of FMP over CMP has been reported at various locations in Delhi by Hazarika [64].

Table 2 Monthly, annual and seasonal average (±Standard Deviation) in μg/m^3 of Particulate Matter (PM) deposited in four different size fractions, i.e. fine mode (FM), coarse mode (CM) and total suspended particles (TSP) during 2018–19 in Delhi

Month	≥ PM_{10}	$PM_{10-2.5}$	$PM_{2.5-1}$	PM_1	TSP	CMP	FMP
April	49.11 ± 7.27	72.22 ± 6.17	47.62 ± 6.65	133.02 ± 9.35	301.97 ± 39.97	121.33 ± 16.34	180.64 ± 60.39
May	51.23 ± 5.17	80.11 ± 14.50	71.05 ± 6.99	157.09 ± 9.22	359.48 ± 46.41	131.34 ± 20.42	228.14 ± 60.84
June	54.68 ± 3.88	129.63 ± 16.28	60.77 ± 5.02	161.22 ± 6.25	406.31 ± 52.31	184.33 ± 52.99	221.99 ± 71.03
July	23.99 ± 2.04	36.12 ± 6.80	24.11 ± 6.10	68.33 ± 11.14	152.55 ± 20.92	60.11 ± 8.58	92.44 ± 31.27
August	12.11 ± 2.75	42.22 ± 6.04	17.33 ± 1.63	65.11 ± 5.43	136.77 ± 24.44	54.33 ± 21.29	82.44 ± 33.79
September	15.22 ± 2.96	73.22 ± 12.59	25.11 ± 4.13	80.11 ± 12.59	193.66 ± 32.99	88.44 ± 41.01	105.22 ± 38.89
October	34.28 ± 4.67	69.22 ± 11.12	98.59 ± 9.21	121.55 ± 10.96	323.64 ± 37.75	103.50 ± 24.71	220.14 ± 16.24
November	49.05 ± 8.29	75.33 ± 9.53	126.12 ± 7.23	148.22 ± 7.17	398.72 ± 45.50	124.38 ± 18.58	274.34 ± 15.63
December	50.85 ± 6.50	91.07 ± 14.93	192.22 ± 10.86	220.23 ± 13.38	554.37 ± 80.62	141.92 ± 28.44	412.45 ± 19.81
January	45.65 ± 5.29	89.04 ± 11.13	180.54 ± 7.35	184.11 ± 11.21	499.34 ± 68.72	134.69 ± 30.68	364.65 ± 2.52
February	37.09 ± 4.65	67.55 ± 8.15	153.04 ± 8.08	192.02 ± 11.16	449.70 ± 72.28	104.64 ± 21.54	345.06 ± 27.56
March	41.22 ± 5.89	95.43 ± 7.52	110.56 ± 9.49	143.23 ± 7.72	390.44 ± 42.56	136.65 ± 38.33	253.79 ± 23.10
Annual	38.71 ± 14.55	76.76 ± 24.37	92.25 ± 61.20	139.52 ± 49.20	347.25 ± 132.49	115.47 ± 36.20	231.77 ± 107.03
Summer	51.68 ± 5.44	98.32 ± 12.32	59.82 ± 6.22	150.44 ± 8.28	355.92 ± 46.23	145.67 ± 33.85	210.26 ± 25.83
Monsoon	17.11 ± 2.58	50.52 ± 8.47	22.18 ± 3.95	71.18 ± 9.72	160.99 ± 26.12	67.63 ± 18.26	93.37 ± 11.42
Post-monsoon	41.67 ± 6.48	72.27 ± 10.33	112.36 ± 8.22	134.89 ± 9.06	361.18 ± 41.63	113.94 ± 14.76	247.24 ± 38.33
Winter	43.70 ± 5.58	85.77 ± 10.43	159.09 ± 8.94	184.90 ± 10.87	473.46 ± 66.04	129.48 ± 16.84	343.99 ± 66.46

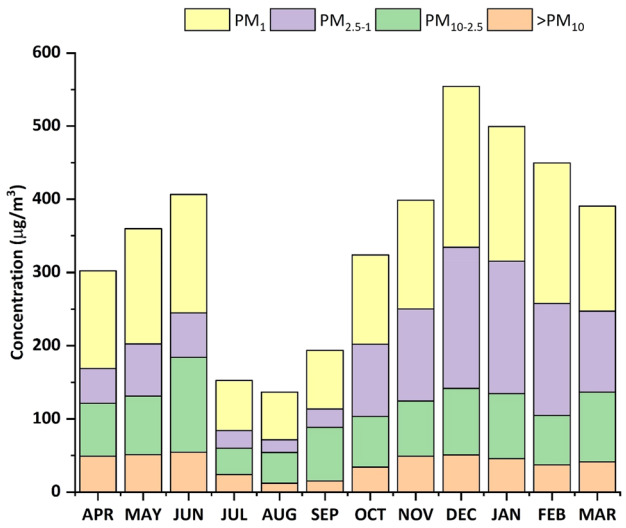

Fig. 3 The mass concentration (μg/m^3) of aerosol particles deposited in four different size fractions (≥ PM_{10}, $PM_{10-2.5}$, $PM_{2.5-1}$ and ≤ PM_1) during 2018–2019

3.2 Variation of OC/EC in PM_1

Monthly, annual and seasonal average concentration of OC and EC measured in PM_1 is presented in Table 3. The annual average concentration of OC and EC was found to be 28.01 ± 14.61 and 10.40 ± 7.41 μg/m^3, respectively. Here, OC and EC contributed 20.1 and 7.5% to PM1 mass concentration. Ram, Sarin [15] have reported almost similar contributions of TC (30–35%) to the TSP in Delhi. However, in another study by Panda et al., approximately 16% and 30% of TC in PM2.5 was reported in Delhi and Bhubaneshwar, respectively [65]. Increasing trends in the concentration of OC and EC in PM1 were noticed from October to December month. Figure 4 shows the comparative description of month-wise variation of OC and EC along with PM1. Among different months, highest concentrations of OC and EC were found in December (OC: 56.02 ± 5.80 and EC: 24.99 ± 5.15 μg/m^3), whereas the lowest value was observed in the month of August (OC: 9.03 ± 4.10 and EC: 3.37 ± 0.95 μg/m^3). A clear seasonal variation was observed in the OC and EC during the study period. The highest concentrations of OC and EC were observed in the post-monsoon and winter seasons, respectively. While, the minimum concentration of OC and EC were observed in the monsoon season due to rain washout.

The average OC & EC concentrations in summer, monsoon, post-monsoon, and winter seasons were found to be 22.57 ± 3.72 & 5.51 ± 1.28, 11.70 ± 3.07 & 3.60 ± 0.39, 42.06 ± 7.10 & 11.72 ± 2.45 and 37.28 ± 14.45 & 18.52 ± 6.23 μg/m^3, respectively. Overall, season-wise variation in the concentration of OC was observed in the order of post-monsoon > winter > summer > monsoon, while EC followed the trend of winter > post-monsoon > summer > monsoon. Tiwari et al. [66] have observed high concentration of OC (54 ± 39 μg/m^3) and EC (10 ± 5 μg/m^3) during winter season in Delhi. The high concentrations of OC (50.11 ± 11.93 μg/m^3) and EC (10.67 ± 3.56 μg/m^3) was also reported in Delhi as compared to Agra by Pipal [67]. In a study conducted from 2010–17 Sharma

Table 3 Monthly, annual and seasonal average (±Standard Deviation) in μg/m^3 units of Organic Carbon (OC), Elemental Carbon (EC), Total Carbon (TC), Primary Organic Carbon (POC), Secondary Organic Carbon (SOC), Organic Matter (OM), Elemental Matter (EM), Total Carbonaceous Matter (TCM) and OC/EC ratio in PM_1 during 2018–19 in Delhi

Month	OC	EC	TC	POC	SOC	OM	EM	TCM	OC/EC
April	23.04 ± 6.58	5.04 ± 2.18	28.09 ± 5.18	6.56 ± 2.84	16.49 ± 8.89	36.87 ± 10.53	5.55 ± 2.40	42.42 ± 8.90	5.39
May	26.04 ± 10.70	6.96 ± 3.37	33.00 ± 8.65	9.05 ± 5.94	16.99 ± 15.05	41.66 ± 17.12	7.66 ± 4.69	49.32 ± 14.31	4.56
June	18.64 ± 3.10	4.53 ± 0.83	23.17 ± 3.93	5.89 ± 1.08	12.75 ± 2.03	29.83 ± 4.96	4.98 ± 0.91	34.81 ± 5.87	4.12
July	11.02 ± 2.54	3.37 ± 0.97	14.39 ± 3.51	4.39 ± 1.26	6.63 ± 1.29	17.63 ± 4.06	3.71 ± 1.07	21.34 ± 5.13	3.30
August	9.03 ± 4.10	3.37 ± 0.95	12.40 ± 4.62	4.54 ± 1.43	4.49 ± 4.08	14.45 ± 6.56	3.84 ± 1.21	18.29 ± 6.90	2.75
September	15.05 ± 1.78	4.04 ± 0.72	19.09 ± 1.89	5.26 ± 0.93	9.79 ± 2.03	24.08 ± 2.84	4.45 ± 0.79	28.53 ± 2.93	3.81
October	37.04 ± 5.22	9.98 ± 2.55	47.02 ± 7.61	12.98 ± 3.32	24.06 ± 2.64	59.26 ± 8.35	10.98 ± 2.81	70.24 ± 10.96	3.70
November	47.08 ± 13.36	13.45 ± 7.15	60.53 ± 8.37	17.49 ± 11.93	29.59 ± 24.87	75.32 ± 21.38	14.80 ± 10.10	90.12 ± 12.49	4.46
December	56.02 ± 5.80	24.99 ± 5.15	81.02 ± 10.95	32.49 ± 6.70	23.53 ± 0.95	89.64 ± 9.28	27.49 ± 5.67	117.13 ± 14.94	2.27
January	38.03 ± 3.29	20.03 ± 5.27	58.06 ± 7.45	26.04 ± 6.85	11.98 ± 10.03	60.84 ± 9.21	22.04 ± 5.80	82.88 ± 9.54	1.96
February	34.06 ± 7.60	19.03 ± 2.14	53.09 ± 9.69	24.74 ± 2.78	9.32 ± 4.95	54.50 ± 12.16	20.94 ± 2.36	75.43 ± 14.46	1.77
March	21.02 ± 1.58	10.03 ± 3.29	31.05 ± 3.41	13.04 ± 5.77	7.98 ± 6.36	33.64 ± 2.52	11.03 ± 4.88	44.67 ± 4.89	2.24
Annual	28.01 ± 14.61	10.40 ± 7.41	38.41 ± 21.38	13.53 ± 9.63	14.48 ± 7.81	44.81 ± 23.38	11.44 ± 8.15	56.25 ± 30.74	2.69
Summer	22.57 ± 3.72	5.51 ± 1.28	28.09 ± 4.91	7.17 ± 1.67	15.41 ± 2.31	36.12 ± 5.95	6.06 ± 1.41	42.18 ± 7.26	4.10
Monsoon	11.70 ± 3.07	3.63 ± 0.39	15.30 ± 3.44	4.73 ± 0.46	6.97 ± 2.67	18.72 ± 4.91	3.96 ± 0.39	22.67 ± 5.26	3.25
Post-monsoon	42.06 ± 7.10	11.72 ± 2.45	53.77 ± 9.55	15.23 ± 3.19	26.83 ± 3.91	67.29 ± 11.36	12.89 ± 2.70	80.18 ± 14.06	3.59
Winter	37.28 ± 14.45	18.52 ± 6.23	55.81 ± 20.50	24.08 ± 8.10	13.20 ± 7.08	59.65 ± 23.12	20.37 ± 6.86	80.03 ± 29.75	2.01

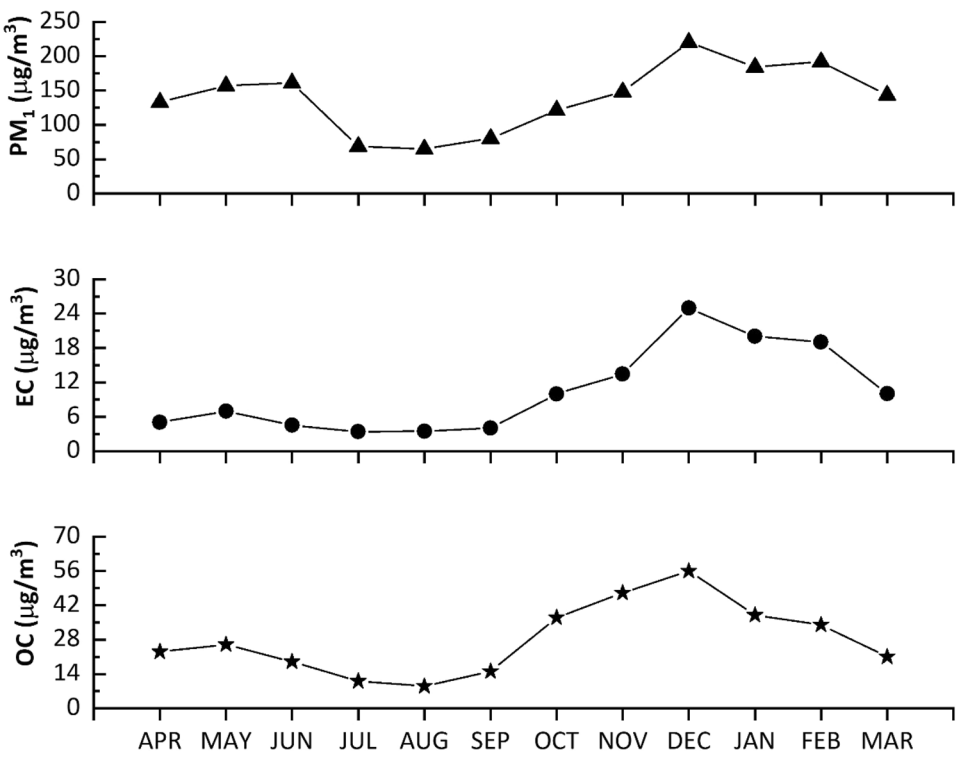

Fig. 4 Monthly variation of OC, EC, and PM_1 concentration (μg/m^3) during 2018–2019

[68] observed OC and EC concentration with the average value of 25.6 ± 14.0 and 8.7 ± 5.8 μg/m^3, respectively, in the urban area of Delhi. A comparative account of OC and EC concentration reported from other regions are given in Table 1. In the present study, high OC and EC concentrations in post-monsoon and winter seasons were attributed to the high emissions from agricultural waste burning, coal combustion and a huge display of fireworks during Diwali festival [13, 50, 69, 70]. Further, in addition to the strength of emission sources, favorable meteorological conditions such as wind speed, low temperature, stable atmosphere and low mixing height also contribute to the high concentration of OC and EC during winter [41, 71].

3.2.1 Carbon subfractions in PM_1

The season-wise variation of OC and EC subfractions are shown in Fig. 5. In this study, OC3 and EC1 were observed as the most abundant OC and EC subfractions, respectively, in all 4 seasons. The high concentration of OC1 was found during the post-monsoon season, which indicates the emission from biomass burning [72]. The high levels of OC2, OC3, OC4, OP, and EC1 were observed in the winter season, which suggests the emission from a combination of various sources such as automobiles, biomass burning, and coal combustion [73]. Further, a slight increase in the concentrations of EC2 and EC3 was observed during post-monsoon and winter seasons, which indicates the emission of these carbon subfractions from diesel vehicles [74]. Since the sampling site is situated near the busy road so emission from vehicular activities might have contributed slightly higher EC concentration in comparison to other seasons. A similar pattern of variation in different carbon subfractions was reported in Delhi [36, 75].

3.2.2 Relationship between OC, EC, and TC

The probable emission source of OC and EC can be predicted by their association with each other [76]. Significant linear correlation ($R^2 = 0.54$) was observed between OC and EC, which indicates that both carbonaceous species have a similar source of emission (Fig. 6). Here, OC/EC ratios were found in the ranges of 1.79–4.58 with an annual average value of 2.69. Generally, the OC/EC ratio of more than two (> 2.0) suggests the presence of secondary organic carbon (SOC) in the ambient air [77]. The higher OC/EC ratio indicates the influence of biomass burning, while a lower ratio indicates the emission from fossil fuel burning [78].

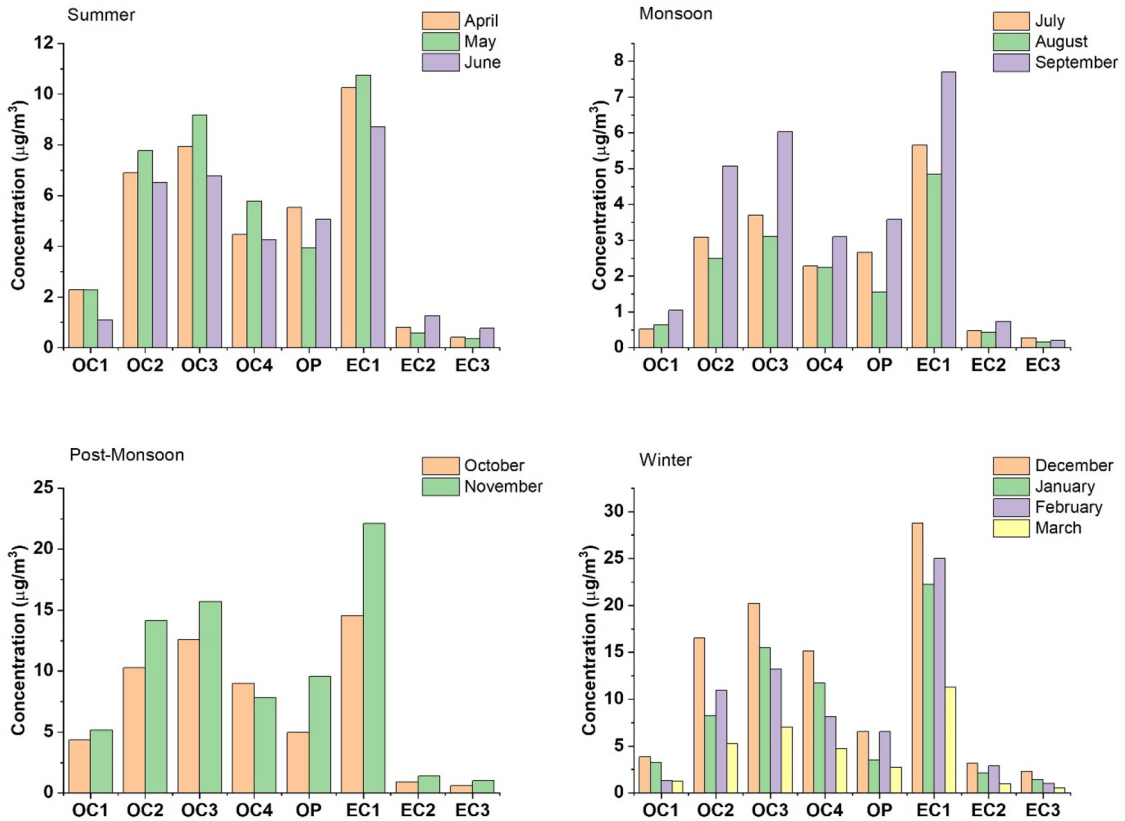

Fig. 5 Monthly and seasonal variation of various carbon subfractions (OC1, OC2, OC3, OC4, OP, EC1, EC2, EC3) during 2018–19

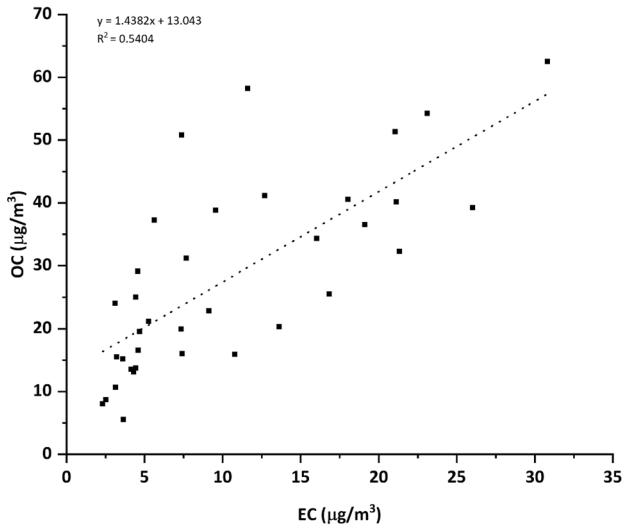

Fig. 6 Scatter plot between OC and EC during 2018–2019

A number of emission sources have been identified on the basis of different values of the OC/EC ratio. Watson et al. [79] have observed OC/EC ratio of 2.7, 1.1, 9.0, and 14.5 for biomass burning, coal combustion, vehicular emissions, and forest fires, respectively in Northwest Colorado, USA. Cao et al. [73] have also observed the OC/EC ratio of more than 12 and 4.1 for emissions from coal combustion and motor vehicle exhaust, respectively.

The probable sources of emission of OC and EC can also be identified by measuring the value of the EC/TC ratio of the aerosol sample. The EC/TC ratio between 0.1 to 0.2 and > 0.5 indicate emission from biomass burning and fossils fuel combustion, respectively. The high EC/TC ratio is very common in urban areas due to emission from vehicular exhaust [80]. Here, the EC/TC ratio was found to be more than 0.20 in all the 4 seasons that ranged from 0.20–0.36. Similar trends in the EC/TC ratio was observed by Kumar et al. [75] in the ambient air of Delhi. An increasing trend of the EC/TC ratio was observed from summer to winter season which is in conformity to the study conducted by Yttri et al. [80]. Salam et al. [81] have observed EC/TC ratio in the range of 0.28–0.39 from traffic and residential sites in Dhaka. Novakov et al. [82] have also observed an EC/TC ratio in the range of 0.1–0.2 from biomass burning during the Indian Ocean Experiment (INDOEX). Generally, the value of OC/EC or EC/TC ratio depends on the analytical method (Thermal/Optical) used to measure the OC and EC concentration [83]. Therefore, the OC/EC or EC/TC ratio can be different for the same sample if the analytical method will be different. Here, we have also observed a lower OC/EC ratio in PM_1 as compared to the value reported for $PM_{2.5}$ and PM_{10} by various researchers (Table 1). Therefore, our observed results also justify the previous finding that the OC/EC ratio increases with the increase in the aerodynamic diameter of the PM [84].

3.2.3 Estimation of SOC, ECR, and TCM

The EC tracer method was used to measure the secondary organic carbon (SOC) because it cannot be measured directly by a carbon analyzer [85]. According to this method, it is assumed that the emission of EC is primary in nature, and the value calculated by this procedure represents the true concentration of SOC [86, 87]. Therefore, SOC concentration was measured according to the following equations [88]:

$$SOC = (OC)_{total} - (OC)_{pri} \quad (4)$$

$$(OC)_{pri} = (OC/EC)_{min} \times (EC) \quad (5)$$

where $(OC)_{pri}$ is primary organic carbon (POC) and $(OC/EC)_{min}$ represents the minimum OC/EC ratio noticed among all aerosol samples collected during the study period. In the present study, $(OC/EC)_{min}$ value was taken as 1.5, which was observed in all 4 seasons. The OC/EC ratio is generally influenced by various factors, such as ambient temperature and different types of emission sources (Cabada et al. 2004). Therefore, the season-specific $(OC/EC)_{min}$ values were used for the estimation of SOC.

The measured concentrations of POC and SOC are given in Table 3. Here, the annual average concentration of POC and SOC in PM_1 were found to be 13.53 ± 9.63 and 14.48 ± 7.81 µg/m^3, respectively. The season-wise concentration of SOC was found to be 15.41 ± 2.31, 6.97 ± 2.67, 26.83 ± 3.91 and 13.20 ± 7.08 µg/m^3 in summer, monsoon, post-monsoon and winter seasons, respectively. Further, the concentration of SOC was observed higher than POC during summer, monsoon and post-monsoon, whereas POC (24.08 ± 8.10 µg/m^3) dominated the SOC in the winter season. Among the four different seasons, the formation of SOC was minimum during the monsoon. The low ambient temperature with a weak photochemical reaction might be a possible reason for the minimum concentration of SOC during the winter season [88]. Singh et al. [89] have observed the same trends of variation in POC and SOC concentration in Delhi. Further, Satsangi et al. [59] have found the high POC during winter with almost twice the value of SOC concentration, which is similar to our observed value of POC and SOC in the winter season. Low SOC formation during winter as compared to summertime was also observed in the ambient air of Delhi by Sharma et al. [40].

An Effective Carbon Ratio (ECR) gives information related to the formation of SOC in ambient atmosphere

[90]. Therefore, the ECR value was calculated according to the following equation:

$$ECR = SOC / (POC + EC) \quad (6)$$

Generally, the ECR value of > 1.0 indicates the high contributions of SOC over POC, while < 1.0 suggests the dominance of POC over SOC and more warming effects [91]. The ECR value ranged from 0.21 ± 0.10 to 1.78 ± 1.25 during the study period. Among different seasons, the minimum value of ECR was observed in winter (0.35 ± 0.12), while the maximum was found in summer (1.54 ± 0.28). In the present study, a similar trend in the variation of ECR value was observed, as reported in Pune [90]. The lower value of ECR (< 0.3) in the winter, and high value (> 0.7) during summer and monsoon season were observed by Safai et al. [90]. In the urban atmosphere, usually, the higher and lower value of ECR indicates the presence of more scattering and absorbing type of carbonaceous aerosols, respectively. The measured value of ECR is displayed in Fig. 7.

The concentration of total carbonaceous material (TCM) was measured according to the following equation.

$$TCM = OM + EM \quad (7)$$

where OM is organic matter (OM = 1.6 × OC), and EM is elemental matter (EM = 1.1 × EC). A conversion factor of 1.4 is commonly applied for the calculation of OM [80, 92]. However, a factor of 1.6 ± 0.20 and 1.1 was suggested earlier as a better conversion factor to estimate OM and EM concentration in an urban area, respectively [93, 94]. The annual average concentration of TCM was found to be 56.25 ± 30.74 μg/m^3. Further, TCM concentration in PM_1 was found 42.18 ± 7.26, 22.67 ± 5.26, 80.18 ± 14.06 and 80.03 ± 29.75 μg/m^3 during summer, monsoon, post-monsoon and winter seasons, respectively. Furthermore, the annual average percentage contribution of OM, EM, and TCM in PM_1 was found to be 32.12, 8.20, and 40.32%, respectively. A clear seasonal trend in the percentage contribution of TCM was observed, which accounted for 28.04, 31.85, 59.44, and 43.28% of PM_1 during summer, monsoon, post-monsoon, and winter seasons, respectively. The higher percentage of EM was observed in winter (11.02%), while OM was observed in post-monsoon (49.89%) season. The estimated concentrations of OM, EM, and TCM are presented in Table 3.

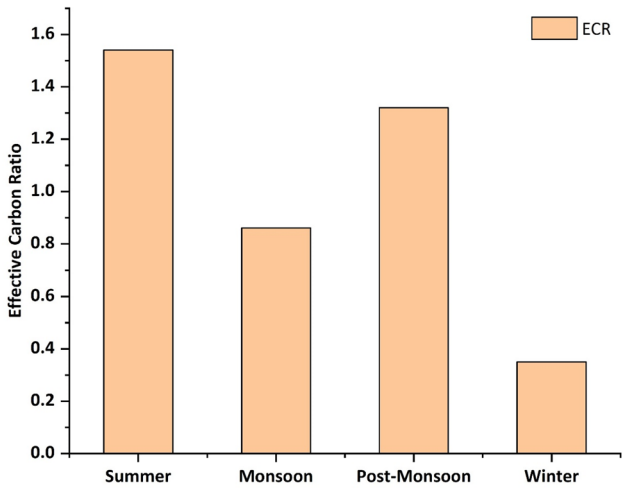

Fig. 7 Effective carbon ratio (ECR) during summer, monsoon, post-monsoon and winter season during 2018–19

3.3 Estimation of exposure risk to EC

A number of studies have been conducted to estimate the level of exposure of particulate matter and associated chemical species over a certain period of time [95–97]. The exposure risk is measured on the concept that how much dose of air pollutants is inhaled in the body at a particular atmospheric environment and the time spent in that place [98]. Therefore, the exposure risk or inhalation dose of EC is calculated according to the following equation [99].

$$\text{Inhalation Dose } (D) = \int Cp \times IR_{(\Delta t)} \times dt \quad (8)$$

where D is the average inhalation dose in microgram (μg), while C_p is the concentration of pollutants in micrograms per cubic meter (μg/m^3). Here, IR(Δt) is the breathing rate (1.62 or 0.027 m^3/min for male and female in the age range of 21–51 years) for mean short term exposure as suggested in the risk information by the United States Environmental Protection Agency [100]. In this equation, t is the exposure time of a person to air pollutants (i.e., minimum time spent by a person in outdoor activities), which is taken as 8 h for calculation of inhalation dose [99]. The estimated inhalation dose was found significantly high during winter and post-monsoon season, which could adversely affect the respiratory system. The annual average inhalation dose during the study period was found as 135 μg. Season-wise inhalation dose of EC was observed in the sequence of winter (240 μg) > post-monsoon (151 μg) > summer (71 μg) > monsoon (47 μg). Mahilang et al. [101] have observed the exposure of high value of inhalation dose of EC released in the ambient air during the firework activities. Dhaini et al. [102] have also reported the high death rate and mortality rate of 7.8–10% due to exposure to increased concentration of EC. Long term exposure to the high concentration of EC can cause complicated heart disease (acute myocardial infarction) and death of the people [96].

4 Conclusions

The present investigation carried out in a residential area has demonstrated a clear seasonal variation in the concentration of aerosol particles collected in different size ranges. The highest deposition of aerosol particles was observed in PM_1 among the four different size fractions of PM. Average concentrations of particles deposited in ≥ PM_{10}, $PM_{10-2.5}$, $PM_{2.5-1}$, and ≤ PM_1 were observed to be 38.71 ± 14.55, 76.76 ± 24.37, 92.25 ± 61.20 and 139.52 ± 49.20 μg/m^3, respectively. Further, annual average concentration of OC and EC measured in PM_1 size fraction was found to be 28.01 ± 14.61 and 10.40 ± 7.41 μg/m^3, respectively. Among 4 different seasons, the highest OC and EC concentrations were observed during the post-monsoon and winter, respectively. Slow wind speed and low ambient temperature has created favorable condition for these carbonaceous aerosol particles to remain suspended in the air for longer period of time. Overall, season-wise variation in the concentration of OC was observed in the order of post-monsoon > winter > summer > monsoon, while EC followed the trend of winter > post-monsoon > summer > monsoon. The OC/EC ratio was found in the range of 1.77–5.39 with an average value of 2.69. Among carbon's 8 subfractions OC2, OC3 and EC1 were found in highest concentrations indicating the mixing of various emission sources. The present study also concludes that the emissions from the residential area are a significant source of EC and OC. The rising level of these pollutants can affect the atmospheric properties and health of the people. The estimated inhalation dose of EC indicates the high exposure risk, which can adversely affect the health of people. Therefore, a better policy is needed to control the OC and EC, along with the high PM pollution in the residential area.

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Compliance with ethical standards

Conflict of interest There is no conflict of interest among the authors with this work.

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