

# Spatial and seasonal variations of atmospheric particulate carbon fractions and identification of secondary sources at urban sites in North India

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Received: 2 February 2015 / Accepted: 23 April 2015  
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**Abstract** An intensive measurement campaign was undertaken to characterize eight fractions of organic carbon (OC) and elemental carbon (EC) in particulate matter (PM) at four urban sites with different pollution characteristics during summer, post-monsoon, and winter at Kanpur, India. Speciation samplers were used to collect particulate samples on quartz filters followed by analysis of OC and EC using Interagency Monitoring of Protected Visual Environments (IMPROVE)-based thermal/optical reflectance (TOR) method. Based on 24-h average results at each site, the highest levels of OC and EC were observed during winter as  $96.7 \pm 26.9$  and  $31.8 \pm 9.8 \mu\text{g}/\text{m}^3$  at residential site and traffic site, respectively. The levels of OC at residential sites during winter appeared to be more than twice of that during summer. The site close to the road traffic had the least value of OC/EC, as  $1.77 \pm 0.28$  during post-monsoon, and the site influenced by emissions of domestic cooking and heating had the highest value of OC/EC, as  $4.05 \pm 0.79$  during winter. The average abundances of OC1, OC2, OC3, OC4, OP, EC1, EC2, and EC3 in total carbon (TC) at all sites for three seasons were 10.03, 19.04, 20.03, 12.32, 10.53, 33.39, 3.21, and 1.99 %, respectively. A sharp increase in levels of OC1 and EC1-OP during winter at two residential sites revealed that biomass burning could be a significant contributor to carbonaceous aerosols. From the

application of EC-tracer method, it was observed that contribution of secondary organic carbon (SOC) to PM mass increased from 5 % during post-monsoon to 16 % during winter at residential sites and from 2 % during post-monsoon to 7 % during winter at traffic sites. Therefore, it could be inferred that increase in primary emissions coupled with unfavorable meteorological conditions could cause particle agglomeration and hygroscopic growth, leading to unpleasant pollution episode during winter.

**Keywords** Elemental carbon · Indo-Gangetic basin · Biomass burning · POC · SOC · EC-tracer method

## Introduction

The fast-growing population, urbanization, industrialization, and transportation system have led to unprecedented air pollution problems and related public health concerns in urban cities of developing countries (Butler et al. 2008; Liu et al. 2013). Particulate matter (PM) is of primary concern and chemical characterization of PM is of particular interest, due to its significant role in reduction of atmospheric visibility through formation of haze, human health effects, and climate change (Pongkiatkul and Kim Oanh 2007; Behera et al. 2011). Identification of sources of PM and understanding of the atmospheric processes responsible for present PM levels are essential for developing effective control policies in any urban environment.

The sum of organic carbon (OC) and elemental carbon (EC), known as total carbon (TC) or carbonaceous aerosol, is one among the major components of PM that contributes 20–50 % to PM mass in urban environments (Chow et al. 1993; Tiwari et al. 2013). These carbonaceous aerosol species are associated with many urban environmental problems

Responsible editor: Gerhard Lammel

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starting from local health effects and visibility problems to regional and global climate change (Ramanathan et al. 2005; Aggarwal et al. 2013). From the analytical point of view, OC and EC are defined with the thermal methodology, in which OC and EC are volatilized or oxidized on different temperature ranges. EC is emitted mainly from primary sources through incomplete combustion, whereas OC is released from both primary sources and secondary transformation to form secondary organic carbon (SOC) through atmospheric chemical conversion processes from the organic precursors (Hegde and Kawamura 2012; Saud et al. 2013). OC contains a large number of organic compounds, of which a significant fraction is potentially mutagen or carcinogen, such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) (Cao et al. 2004; Behera and Sharma 2010).

The levels of atmospheric OC and EC concentrations in large Indian cities have increased dramatically in recent years (e.g., Pavuluri et al. 2011; Pachauri et al. 2013a, b; Srivastava et al. 2014). This is also a fact that OC and EC from Indian cities have direct contributions to the Atmospheric Brown Clouds (ABCs), and as a result, the role of ABCs in atmospheric radiative and chemical properties is pronounced in the South Asian region (Reddy and Venkataraman 2002; Ramanathan et al. 2005). The continuous increase in concentration levels of PM in the Indo-Gangetic basin (IGB), India (latitudes 22° 30' N to 31° 30' N and longitudes 73° 30' E to 89° 0' E), one of the hotspots of ABCs, is a matter of concern (Sharma and Maloo 2005; Singh and Dey 2012). The trends of drastic decrease in atmospheric visibility coupled with occurrence of frequent fog formation and unfavorable meteorological conditions are observed in the IGB (De et al. 2005; Tiwari et al. 2011). The reduction in atmospheric visibility besides being unhealthy conditions result in delays/cancellation of trains and flights, putting a large population into inconvenience (Singh and Dey 2012; Liu et al. 2013).

During recent years, there is a spurt of studies on OC and EC of different PM sizes (suspended particulate matter (SPM), PM with aerodynamic diameter (AED)  $\leq 10 \mu\text{m}$  (PM<sub>10</sub>) and PM with AED  $\leq 2.5 \mu\text{m}$  (PM<sub>2.5</sub>)) in the IGB and other parts of India (Rengarajan et al. 2007; Ram and Sarin 2010; Pachauri et al. 2013a; Pavuluri et al. 2011; Aggarwal et al. 2013; Ram et al. 2010; Joseph et al. 2012; Tiwari et al. 2013). However, there are still knowledge gaps for more insights into the fractional characterization of OC and EC, source attributions in terms of primary and secondary origins for existing levels of OC and EC, their spatial and seasonal variations. To address the above concerns for research on carbonaceous aerosols, this current study presents the fractional characterization of OC and EC in a large Indian city based on seasonal and spatial distributions. The specific objectives of the study were as follows: (1) to assess qualitative influence of pollution sources on PM<sub>10</sub>, OC, and EC at four sampling sites during three different seasons; (2) to understand the spatial and seasonal

variations of eight carbon fractions (OC1, OC2, OC3, OC4, OP, EC1, EC2, and EC3); and (3) identification of secondary sources responsible for existing levels of OC in the atmosphere through EC-tracer method. To achieve these objectives, this study conducted an intensive measurement campaign at four sampling sites with different pollution characteristics during summer, post-monsoon, and winter seasons at Kanpur (latitude 26° 28' N and longitude 80° 24' E; Fig. 1), a representative city of the IGB region and Indian urban agglomeration.

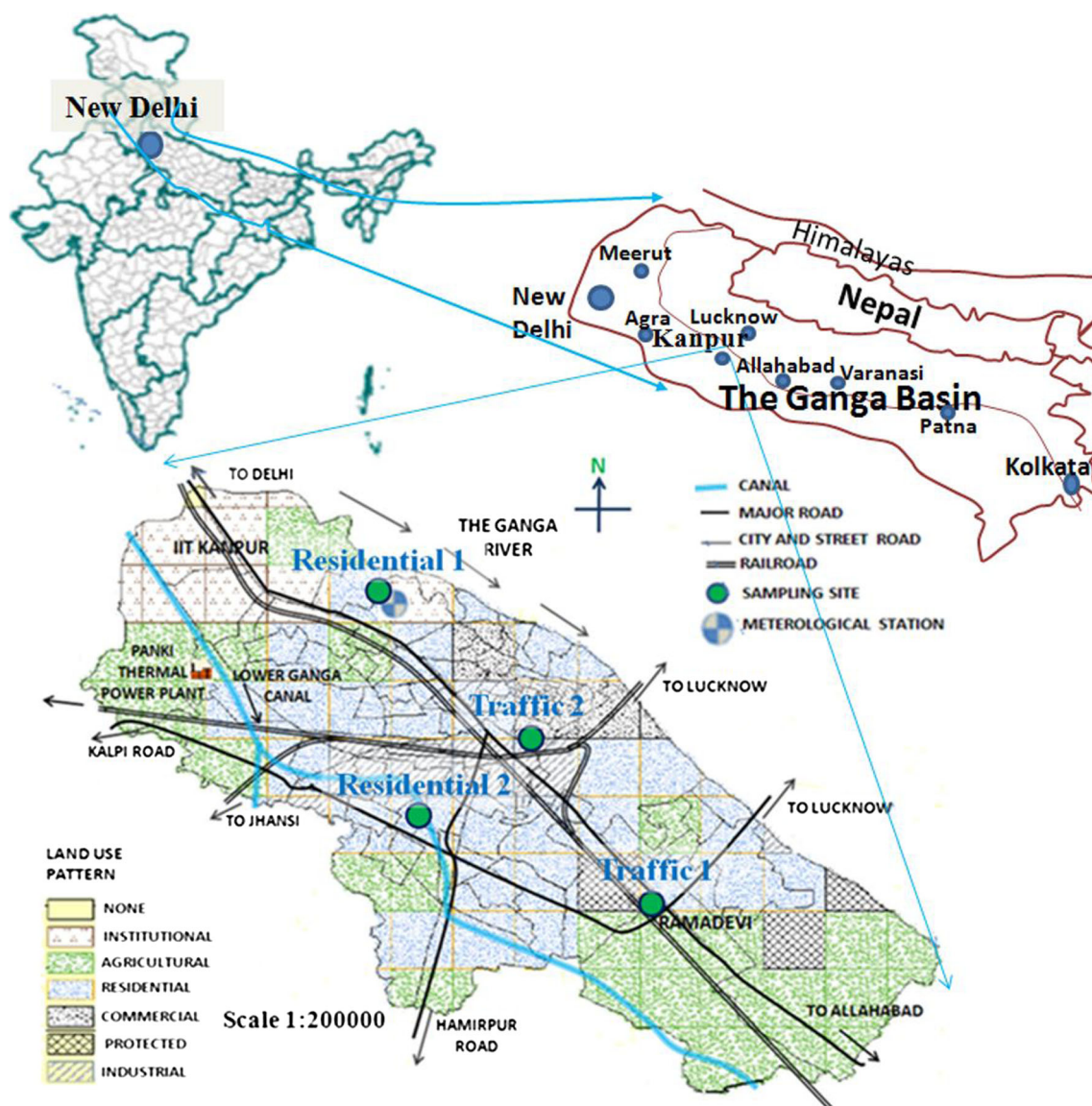
## Methodology

### Study area and measurement campaign

The study area, Kanpur, is a large industrial city (population ~4.0 million with annual growth rate of 3 % and area ~300 km<sup>2</sup>) in North India that represents the patterns of urban agglomeration, weather condition, and atmospheric seasonal variability in the IGB region. The emission source activities responsible for PM pollution can broadly be classified into the following: transport (motor vehicles), commercial, industrial, domestic, fugitive, etc. Garbage burning is practiced frequently during evening hours (more in winters) at several places of the city which could significantly contribute to PM pollution. During winter, agricultural residue burning at the peripheral areas of the city is also a common practice which could contribute to PM mass in the city. The detailed characteristics of the IGB region and study area are described elsewhere (Behera and Sharma 2010; Behera et al. 2011).

This extensive ambient air measurement campaign for sampling of PM was conducted at four sampling sites in the study area during April 2007 through February 2008 (Fig. 1). There were two residential and two traffic sites of air quality measurement. The pollution activities in two residential sites (referred as residential 1 and residential 2) were dominated by domestic cooking (fuels: liquefied petroleum gas (LPG), kerosene, coal and wood) restaurants, garbage burning, diesel generators (DG) operated during power-cut, and vehicles. Out of two traffic sites (referred as traffic 1 and traffic 2), traffic 2 was more close to roads, compared to traffic 1. Traffic 2 was located in the city center with higher pollution activities, compared to three other sites. Besides vehicles, other PM sources in the vicinity of sites at traffics 1 and 2 were domestic and commercial cooking, garbage burning, DG sets, etc.

The durations of sampling were as follows: summer (April to June 2007), post-monsoon (October to November 2007), and winter (December 2007 to February 2008). Partisol<sup>®</sup> Model 2300 4-channel speciation samplers (Thermo Fisher Scientific Inc., USA) were deployed to collect PM samples through an inlet at a flow rate of 16.5 l per minute (LPM) that



**Fig. 1** The Indo-Gangetic basin (IGB) (known as the Ganga basin in the map), India, and study area (Kanpur city) with land-use patterns and sampling sites

removes particles with AED greater than  $10\ \mu\text{m}$ ; the remaining particles were collected on the filter that represented  $\text{PM}_{10}$ . Quartz fiber filters of 47-mm diameter (Whatman grade QM-A) were used for collection of particles. Before sampling, quartz filters were heated at  $900\ ^\circ\text{C}$  for 6 h to remove background organic matter on filter paper. One field blank and laboratory blank were kept for every ten filters for further quality assurance and quality control (QA-QC) analysis. The filters were pre-conditioned and post-conditioned at temperature  $\sim 22\ ^\circ\text{C}$  and RH  $\sim 40\%$  in controlled desiccators for 24 h.  $\text{PM}_{10}$  mass concentration was determined gravimetrically by weighing the quartz filters before and after sampling using a digital microbalance (Mettler–Toledo MX–5, USA) with a sensitivity of 0.001 mg. Before and after particle collection, the filters were stored in Petri dishes (lined with aluminum foil), sealed with Teflon tape.

### Meteorology

During sampling period, meteorological parameters (temperature, relative humidity, rainfall, wind speed, and wind direction) were recorded through an automatic weather monitoring system (WM251 Envirotech, New Delhi) mounted on the roof of the sampling site (Fig. 1). It was programmed to collect data at 1-min interval and store them for later downloading. Prevailing wind direction was northeast and northwest during summer, northwest and northeast during post-monsoon, and west and northwest during winter. The daily average ambient temperature was observed as  $35.37 \pm 5.09\ ^\circ\text{C}$  during summer,  $20.51 \pm 5.22\ ^\circ\text{C}$  during post-monsoon, and  $13.37 \pm 4.58\ ^\circ\text{C}$  during winter. The corresponding relative humidity was  $48.04 \pm 18.48\%$  (summer),  $61.04 \pm 17.58\%$  (post-monsoon), and  $67.23 \pm 16.37\%$  (winter). Wind speed was  $1.4 \pm 0.76\ \text{m/s}$



during summer,  $0.72 \pm 0.61$  m/s during post-monsoon, and  $0.58 \pm 0.52$  m/s during winter. The weather conditions were very cold and calm during winter season (21.4 % calm). Overall, it was observed that the atmosphere during winter was more stable than during summer and post-monsoon.

### Characterization of eight carbon fractions

Eight fractions, five of OC and three of EC were analyzed using a DRI Model 2001A Thermal/Optical Carbon Analyzer according to the IMPROVE TOR protocol (Chow et al. 2001; Fung et al. 2002). In this methodology, a  $0.495\text{-cm}^2$  punch aliquot of a sample quartz filter was heated stepwise at temperatures of  $140^\circ\text{C}$  (OC1),  $280^\circ\text{C}$  (OC2),  $480^\circ\text{C}$  (OC3), and  $580^\circ\text{C}$  (OC4) in a nonoxidizing helium (He) atmosphere, and  $580^\circ\text{C}$  (EC1),  $740^\circ\text{C}$  (EC2), and  $840^\circ\text{C}$  (EC3) in an oxidizing atmosphere of 10 % oxygen in a balance of He. During heating of sample, carbon dioxide ( $\text{CO}_2$ ) was formed through oxidation of carbon present in the sample and then, the sample was reduced to methane ( $\text{CH}_4$ ) for quantification with a flame ionization detector. During the analysis, darkening of the filter deposit occurs in the pyrolysis of organic carbon into black carbon. This darkening is monitored by reflectance of a  $633\text{ nm}$  He-Ne laser light. In the process of addition of oxygen, the original and pyrolyzed black carbon combusts and the reflectance increases. The amount of carbon measured after oxygen is added until the reflectance achieves its original value, which is reported as optically detected pyrolyzed carbon (OP) (Cao et al. 2004, 2006). The eight carbon fractions, OC1, OC2, OC3, OC4, EC1, EC2, EC3, and OP, are reported separately in the data sheet. The IMPROVE protocol defines OC as  $\text{OC1} + \text{OC2} + \text{OC3} + \text{OC4} + \text{OP}$ , and EC as  $\text{EC1} + \text{EC2} + \text{EC3} + \text{OP}$  (Cao et al. 2006).

The uncertainty in estimation of OC and EC was the sum of percentage of uncertainty during sample handling in the field and laboratory (sampling) and uncertainty during analysis (instrument). The percentage of uncertainty was in the range from 10–15 %. Field and laboratory blank filters were analyzed for blank corrections for all fractions of OC and EC. It was observed that the overall blank concentrations from the quartz filters for OC and EC were  $0.37 \pm 0.2$  and  $0.0 \pm 0.02\text{ }\mu\text{g}/\text{cm}^2$ , respectively. The experimental data from measurement campaign were pre-screened before interpretation. OC and EC concentrations are reported after subtracting the blank concentrations.

### Estimation of POC and SOC using EC-tracer method

The EC-tracer method is widely used to estimate primary organic carbon (POC) and secondary organic carbon (SOC) (Strader et al. 1999; Yuan et al. 2005; Behera and Sharma 2010). The fundamental concept of EC-tracer method is based on the hypothesis as follows: if no SOC is formed in the

atmosphere, the representative ratio of OC to EC at sources should remain unchanged in the ambient air. In other words, if measured ratio of OC to EC (in ambient air) exceeds the representative ratio of primary OC/EC (at the source levels), then the additional OC is considered to be of secondary origin, i.e., SOC.

Ambient POC concentration can be expressed as the sum of OC from noncombustion sources and OC from combustion sources as Eq. (1):

$$\text{POC} = a + b\text{EC}_{\text{measured}} \quad (1)$$

where  $a$  represents OC from noncombustion sources (i.e., biogenic, soil and road re-suspended, long-range transport, etc.);  $b$  is the ratio of OC to EC at an emitting source (i.e.,  $(\text{OC}/\text{EC})_{\text{pri}}$ );  $b\text{EC}_{\text{measured}}$  represents OC from combustion sources (e.g., traffic, heating).

OC from combustion sources can be estimated from measured EC concentrations (in ambient air) and by assuming that from each combustion source, the emission ratio of primary OC and primary EC is relatively constant over space and time, implying  $\text{OC}_{\text{combustion}} = ((\text{OC}/\text{EC})_{\text{pri}}) \times \text{EC}_{\text{measured}}$  (Cabada et al. 2002; Saylor et al. 2006; Behera and Sharma 2010). The value of  $a$  can be estimated as the intercept of a straight line fit between  $\text{OC}_{\text{measured}}$  ( $y$ -axis) and  $\text{EC}_{\text{measured}}$  ( $x$ -axis).

OC from secondary formation (SOC) in carbon mass can be estimated as the difference between the measured OC ( $\text{OC}_{\text{measured}}$ ) and POC as per Eq. (2):

$$\text{SOC} = \text{OC}_{\text{measured}} - \text{POC} \quad (2)$$

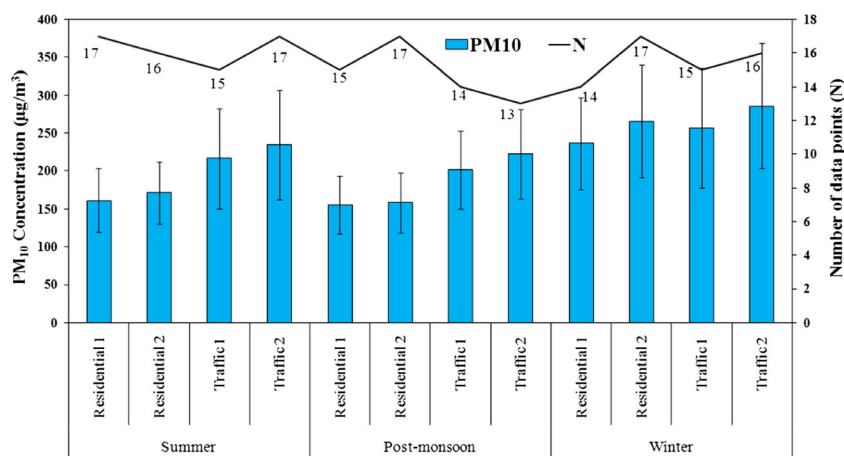
In this study,  $(\text{OC}/\text{EC})_{\text{pri}}$  of emitting sources for sampling sites were derived from activity data of the primary emission inventory of  $\text{PM}_{10}$ , reported by Behera et al. (2011). Hence,  $(\text{OC}/\text{EC})_{\text{pri}}$  were considered as 1.85, 1.95, 1.35, and 1.25 at residential 1, residential 2, traffic 1, and traffic 2, respectively. It was assumed that these primary ratios were constant irrespective of seasons. In other words, the SOC estimation during three different seasons would vary as per the measured concentrations of OC and EC during summer, post-monsoon and winter seasons.

## Results and discussion

### Overall results of PM, OC, and EC

Figures 2 and 3 show the overall results of  $\text{PM}_{10}$ , OC, EC, and OC/EC at four sampling sites on the basis of 24-h observations during summer, post-monsoon, and winter. Traffic 2 was the most polluted site with average  $\text{PM}_{10}$  levels as  $234.8$  and  $285.6\text{ }\mu\text{g}/\text{m}^3$  during summer and winter, respectively. The levels in  $\text{PM}_{10}$  during post-monsoon were lower than that of summer at four sites. The site residential 1 was less polluted in

**Fig. 2** Overall results of PM<sub>10</sub> in  $\mu\text{g}/\text{m}^3$  during the measurement campaign. The secondary y-axis contains number of data points (*N*) at the concerned sites. The error bars in PM<sub>10</sub> concentrations are standard deviations of respective data sets. Values are on the basis of 24-h sampling data

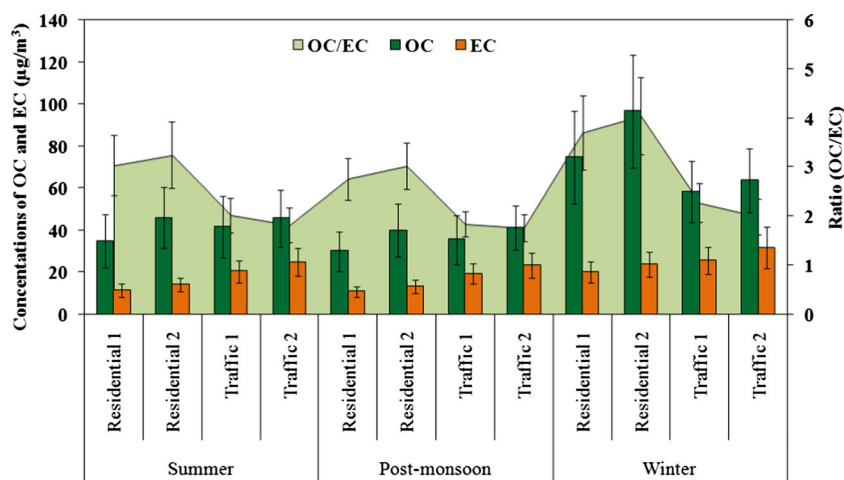


comparison to other sites during three seasons. Overall, 24-h PM<sub>10</sub> mass concentrations ranged from  $91.4 \mu\text{g}/\text{m}^3$  at residential 1 during post-monsoon to  $426.8 \mu\text{g}/\text{m}^3$  at traffic 2 during winter. It was observed that the levels of PM<sub>10</sub> exceeded 24-h Indian air quality standard of  $100 \mu\text{g}/\text{m}^3$  100 % of time (except at residential 1, with ~90 % exceedances during post-monsoon) (<http://www.cpcb.nic.in>). During winter, the highest levels of OC and EC concentration were observed at residential 2 and traffic 2, respectively. The lowest levels of OC and EC concentration were observed at residential 1 during post-monsoon. To assess the overall difference between seasons, a paired *t* test with unequal variance was performed for PM, OC, and EC at four sites using Minitab 15 English. From the results of *t* test, it was found that PM showed significant variation between summer and winter at 95 % level of confidence. However, PM during summer did not show any significant variation with concentration during post-monsoon. Similar to PM, OC, and EC showed statistically significant difference between summer and winter at four sites.

The reason for lower ambient levels of PM, OC, and EC during post-monsoon could be due to washout of pollutants

through occurrence of rain during monsoon. In our earlier study (Shukla and Sharma 2008), in the same study area, we observed lower concentrations of particulate ions and elements during post-monsoon. This should be noted that the measurement campaign was conducted just after heavy rainfall observed in monsoon season. In checking the severity of pollution at four sites, it appeared that OC levels during winter increased by 39 to 113 %, compared to concentration levels in summer. At the same time, EC increased by 26 to 75 %, and PM increased by 19 to 55 % during winter compared to concentration levels in summer. Steep increase in OC levels during winter is a matter of concern, as that indicates the extents of SOC formation in the atmosphere. The 24-h average OC/EC ratio varied from 1.77 at traffic 2 during post-monsoon to 4.05 at residential 2 during winter. OC/EC ratios were higher at two residential sites than levels at two traffic sites. Out of two traffic sites, traffic 2 had smaller OC/EC ratio than traffic 1. This could be due to the location of traffic 2, which was closer to the roads than traffic 1 and impacted by dominating EC emissions from vehicles. In addition to unfavorable meteorology with less mixing height and dispersion, the reasons for increasing levels of OC and EC in winter might be due to

**Fig. 3** Overall results of OC and EC in  $\mu\text{g}/\text{m}^3$  during the measurement campaign. The secondary y-axis contains OC/EC values of respective data sets. The error bars are standard deviations of respective data sets. Values are on the basis of 24-h sampling data



increase in combustion activities of domestic heating (burning of wood, coal, etc.) to sustain life during cold days. Biomass and agriculture residue burning could also be the reason for higher levels of OC and EC. The unfavorable meteorology with lower wind speed, more calm conditions and smaller boundary layer height during winter than in summer could synergize with more pollution activities to disproportionately shoot up the levels of OC both in terms of emissions and atmospheric formation. Similar trends of higher levels of OC during winter were observed by previous studies (e.g., Samara et al. 2014; Zhu et al. 2014).

Among the four sites, relatively high concentration of OC at residential 2 could be due contribution from OC dominating sources such as, domestic cooking, hotel and restaurant cooking and garbage burning near the sampling site. The higher concentration of EC at traffic 2 could be due to predominance of vehicular traffic source close to the sampling site. Table 1 presents ambient levels of OC and EC observed in other Asian regions. It can be clearly seen that the pollution status of OC and EC in Kanpur is similar to other parts of Asian region. Moreover, the levels of carbonaceous aerosols in the region of South Asia (Dhaka, Lahore, and Kanpur) are a matter of concern, and implementation of policies to reduce their levels is the need of the hour. We also compared the seasonal distributions of OC and EC with some other studies conducted in Asian cities (He et al. (2001) for Beijing, Cao et al. (2003) for Guangzhou, Ye et al. (2003) for Shanghai,

Ohta et al. (1998) for Sapporo, Lee and Kang (2001) for Hong Kong). These studies observed the trends similar to this study with higher OC and EC concentrations during winter than summer.

### Spatial distributions of eight carbon fractions

The importance of studies on characterization of eight carbon fractions on the basis of IMPROVE TOR protocol have been reported by several researchers that can help in assessing the carbon abundances, differentiating sources between gasoline and diesel vehicles, differentiating various geological dust, and estimating light extinction of OC and EC (Chow et al. 2001; Gu et al. 2010; Chuang et al. 2014). Figure 4 shows the 24-h levels of eight carbon fractions at four sampling sites. During three seasons, the average abundances of OC1, OC2, OC3, OC4, OP, EC1, EC2, and EC3 in TC at all sites were 10.03, 19.04, 20.03, 12.32, 10.53, 33.39, 3.21, and 1.99 %, respectively. From the spatial distributions of eight carbon fractions, three fractions of OC (OC2, OC3, and OC4) were observed to be the highest at residential 2, and for all three fractions of EC (EC1, EC2, and EC3), traffic 2 experienced the highest concentrations.

The above results confirm that activities like domestic and commercial cooking (characterized by incomplete combustion) could have a major share of OC in TC levels at residential sites, whereas vehicular emission had more control on EC

**Table 1** Comparison of results from this study with other Asian regions

Location	Study period	Size	OC ( $\mu\text{g}/\text{m}^3$ )	EC ( $\mu\text{g}/\text{m}^3$ )	OC/EC	Reference
Chongju, Korea <sup>a</sup>	1995	PM <sub>2.5</sub>	5.5	5.4	1.0	Lee and Kang (2001)
Beijing, China	2002	PM <sub>2.5</sub>	36.7	15.2	3.5	Dan et al. (2004)
Guangzhou, China	2002	PM <sub>2.5</sub>	22.6	8.3	2.7	Cao et al. (2003)
Lahore, Pakistan <sup>b</sup>	2007–2008	PM <sub>2.5</sub>	85.7	13.5	6.2	Stone et al. (2010)
Xi'an, China <sup>c</sup>	2003	PM <sub>2.5</sub>	102.0	21.6	4.7	Cao et al. (2007)
Agra, India <sup>d</sup>	2010–2011	PM <sub>2.5</sub>	53.6	9.5	5.6	Pachauri et al. (2013b)
Delhi, India	2010–2011	PM <sub>2.5</sub>	54.1	10.1	5.2	Tiwari et al. (2013)
Mumbai, India <sup>e</sup>	2007	PM <sub>2.5</sub>	34.1	8.3	4.1	Joseph et al. (2012)
Himalaya, Nepal	1999–2000	PM <sub>10</sub>	3.4	0.5	7.1	Carrico et al. (2003)
Manora Peak, India	2004	TSP	4.9	0.9	5.4	Rengarajan et al. (2007)
Dhaka, Bangladesh	2001	TSP	45.7	22.0	2.1	Salam et al. (2003)
Lahore, Pakistan	1992–1993	TSP	76.9	17.6	4.4	Smith et al. (1996)
Kanpur, India <sup>f</sup>	2007–2008	PM <sub>10</sub>	50.8	19.9	2.5	This study

TSP total suspended particles

<sup>a</sup> Average of fall and winter

<sup>b</sup> Results from the lower range

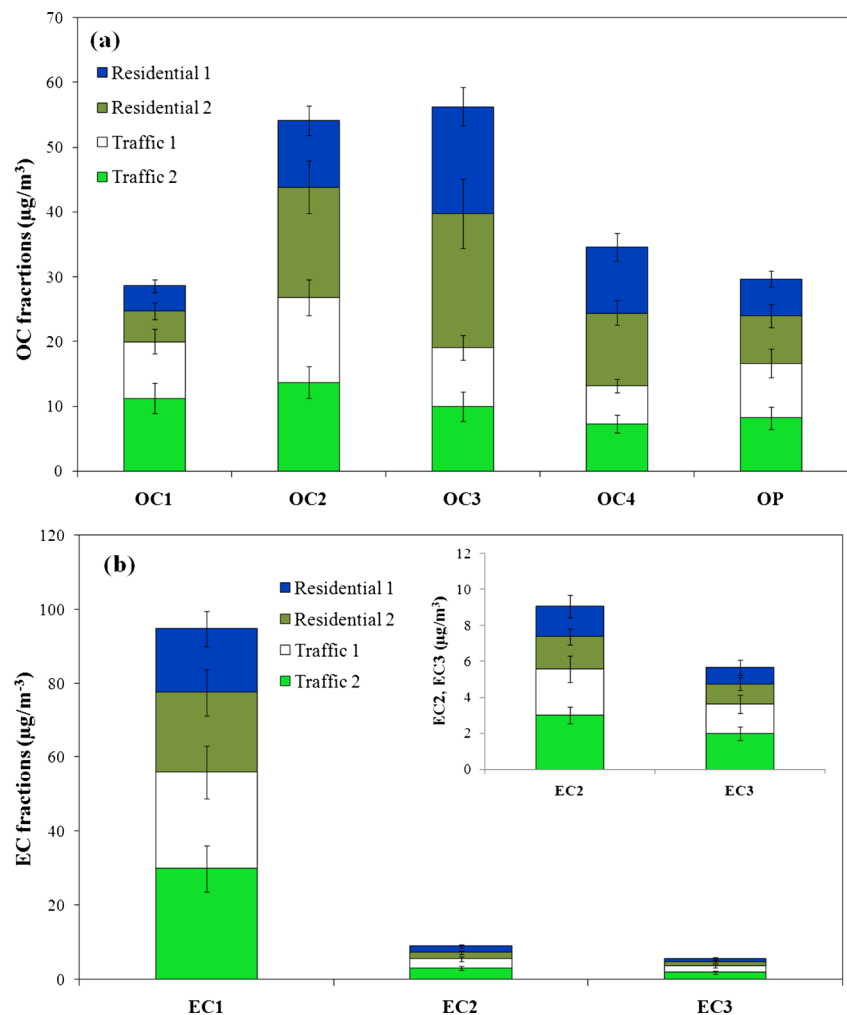
<sup>c</sup> Results from winter season

<sup>d</sup> Average of three sites

<sup>e</sup> Average of all sites during three seasons

<sup>f</sup> Average of all sites during three seasons

**Fig. 4** Spatial distribution of eight carbon fractions: **a** OC fractions and **b** EC fractions, with zoom-in figure of EC2 and EC3. The error bars are the respective standard deviations of the data sets



concentration in TC levels at traffic sites. In the past, several studies (e.g., Han et al. 2010; Samara et al. 2014) have also reported that sites influenced by vehicular traffic had a larger fraction of EC than that of OC. The level of OC1 was observed to be the highest at traffic 2, as  $11.2 \mu\text{g}/\text{m}^3$  followed by traffic 1, as  $8.8 \mu\text{g}/\text{m}^3$ . The percentage contribution of OC1 to TC varied from 6.0 % at residential 2 to 14.5 % at traffic 2. The low temperature ( $140^\circ\text{C}$ ) volatile component of OC (i.e., OC1), is fresh primary emission of OC, was higher at traffic sites as emissions from vehicles are directly received. Earlier study, Zhu et al. (2014) have also observed higher levels of OC1 at a tunnel site than at other urban sites.

From spatial distribution point of view, the contribution of OC2 in TC varied from 17.1 % at traffic 2 to 21.8 % at residential 2. Similarly, contributions of other carbon fractions to TC were as follows: OC3: 12.9 % at traffic 2 to 26.8 % at residential 2; OC4: 8.8 % at traffic 1 to 16.7 % at residential 1. It is clearly seen that as one goes away from the sources, the fraction of higher OC (OC2, OC3, and OC4) increase proved that SOC is formed on the way of transportation of emissions from the sources to the receptor sites. Thus, higher percentage

of OC2, OC3, and OC4 at residential sites can be attributed to the SOC.

Regarding EC fractions, EC1: 27.6 % (of TC) at residential 2 to 38.9 % at traffic 2; EC2: 2.3 % at residential 2 to 3.9 % at traffic 2; and EC3: 1.4 % at residential 2 to 2.6 % at traffic 2. This clearly indicated that EC2 and EC3 were emitted dominantly from vehicles running on the roads close to the sampling sites. This should be noted that sum of both EC2 and EC3 is known as the indicator for soot particles that are normally emitted from combustions from fossil fuels (Han et al. 2010; Lim et al. 2012). The existing levels of EC2 and EC3 found at residential sites can be attributed to the emissions transported from the roads located near to the sampling sites.

The abundance of OP in TC varied from 9.2 to 12.5 % among four sites. The study by Yang and Yu (2002) observed that OP is a nearly a subset of water-soluble organic carbon (WSOC) content, and WSOC accounts for 13–66 % of OP. Therefore, a substantial amount of water-soluble polar compounds are expected to exist in the atmosphere of the study area. Previous study of Ram et al. (2012) at a site in the study area had reported that WSOC/OC ratio varied from 0.21 to

0.65. With the limitation of our study, i.e., without the measurement of WSOC concentrations, it is difficult to explain the reasons on variations of OP concentrations. Joseph et al. (2012) reported the ranges of various ratios of carbon fractions: TC/PM<sub>2.5</sub> 37–44 %; OC1/TC 3.8–4.3 %; OC2/TC 24–32 %; OC3/TC 20–22 %; OC4/TC 10–14 %; EC1/TC 29–36 %; EC2/TC 2–3 %; and EC3/TC 0.4–1.0 %. The results of our study for individual mass contribution of OC4, EC1, EC2, and EC3 to TC mass were in similar range that of Joseph et al. (2012). However, the difference in OC2 and OC3 with respect to TC at Kanpur and Mumbai can be attributed to site characteristics and particulate size. In our study, the contribution of OC1 to TC was higher than the study at Mumbai, and this could be due to comparatively higher levels of VOCs prevailing sources (e.g., biomass/garbage burning) in the ambient air of Kanpur. The percentage contribution of TC to PM mass in our study (28.9 to 39.4 %) was lower than the study at Mumbai for the reason that our study was on PM<sub>10</sub> and the Mumbai study was on PM<sub>2.5</sub>; the similar variation in EC to PM ratio is attributable to variation in particle size.

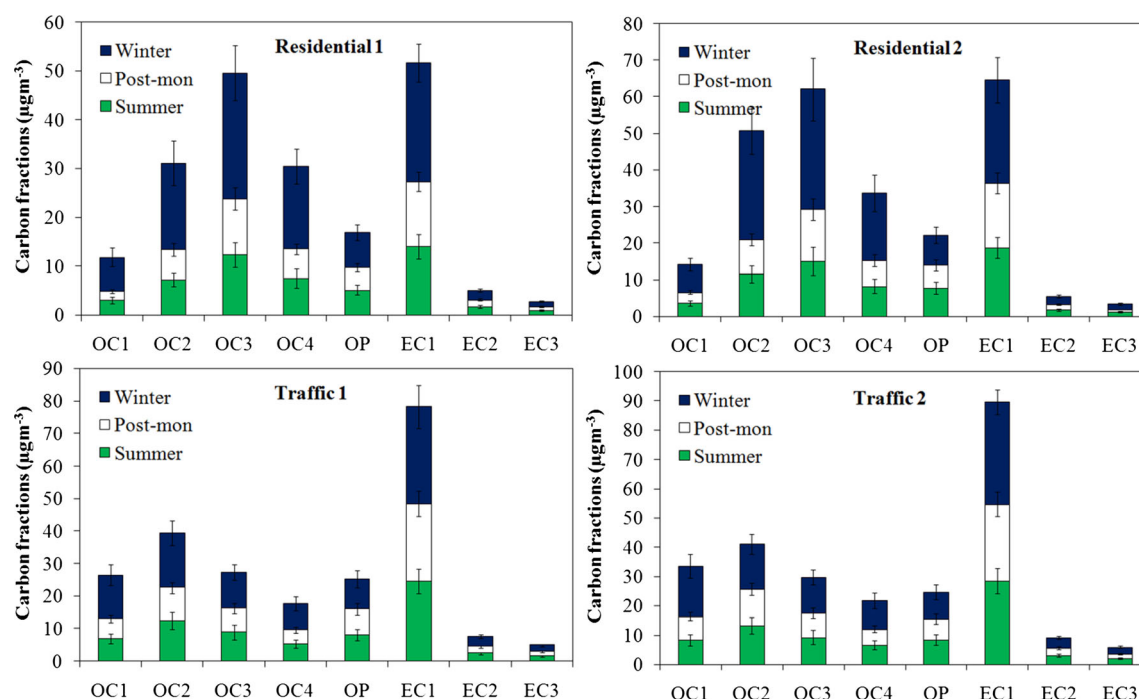
### Seasonal variations of fractions of OC and EC

Figure 5 shows the seasonal distributions of eight carbon fractions at four sampling sites. On the basis of 24-h observations, the levels of eight carbon fractions varied from minimum (during post-monsoon) to maximum (during winter), the minimum and maximum concentration were as follows: (1) OC1 1.83  $\mu\text{g}/\text{m}^3$  at residential 1 to 17.23  $\mu\text{g}/\text{m}^3$  at traffic 2, (2) OC2 6.18  $\mu\text{g}/\text{m}^3$  at

residential 1 to 29.83  $\mu\text{g}/\text{m}^3$  at residential 2, (3) OC3 7.26  $\mu\text{g}/\text{m}^3$  at traffic 1 to 32.78  $\mu\text{g}/\text{m}^3$  at residential 2, (4) OC4 4.27  $\mu\text{g}/\text{m}^3$  at traffic 1 to 18.48  $\mu\text{g}/\text{m}^3$  at residential 2, (5) OP 4.59  $\mu\text{g}/\text{m}^3$  at residential 1 to 9.30  $\mu\text{g}/\text{m}^3$  at traffic 1, (6) EC1 13.24  $\mu\text{g}/\text{m}^3$  at residential 1 to 34.97  $\mu\text{g}/\text{m}^3$  at traffic 2, (7) EC2 1.43  $\mu\text{g}/\text{m}^3$  at residential 1 to 3.54  $\mu\text{g}/\text{m}^3$  at traffic 2, and (8) EC3 0.57  $\mu\text{g}/\text{m}^3$  at residential 2 to 2.38  $\mu\text{g}/\text{m}^3$  at traffic 2. These results showed that wider range of concentrations of OC fractions were observed compared to EC fractions. It suggests rapid changes in OC fraction due to formation of SOC with time.

To assess the overall difference between seasonal concentrations, a paired *t* test with unequal variance was performed for OC1, OC2, OC3, OC4, OP, EC1, EC2, and EC3 at four sampling sites using Minitab 15 English. It was observed that levels of OC1, OC2, OC3, OC4, OP, EC1, EC2, and EC3 showed significant seasonal variations at 95 % confidence level ( $P < 0.05$ ) at all four sites between summer and winter. However, there was no significant variation between the levels in summer and post-monsoon except for OC1 at residential 1, EC3 at residential 3. Therefore, further analyses of eight carbon fractions among sites during summer and winter was undertaken.

The winter to summer concentration ratio of OC1 was more than 2.2 at two residential sites and about 1.8 at two traffic sites. It indicated that pollution from transported biomass burning could have greater impact on residential sites compared to traffic sites. Cao et al. (2005) and Gu et al. (2010) suggested the role of biomass burning in increasing levels of OC1. The possibility for transportation of emissions from the agricultural residue burning sites to city areas has been



**Fig. 5** Levels of eight carbon fractions during summer, post-monsoon, and winter at four sampling sites. The error bars are the respective standard deviations of the data sets



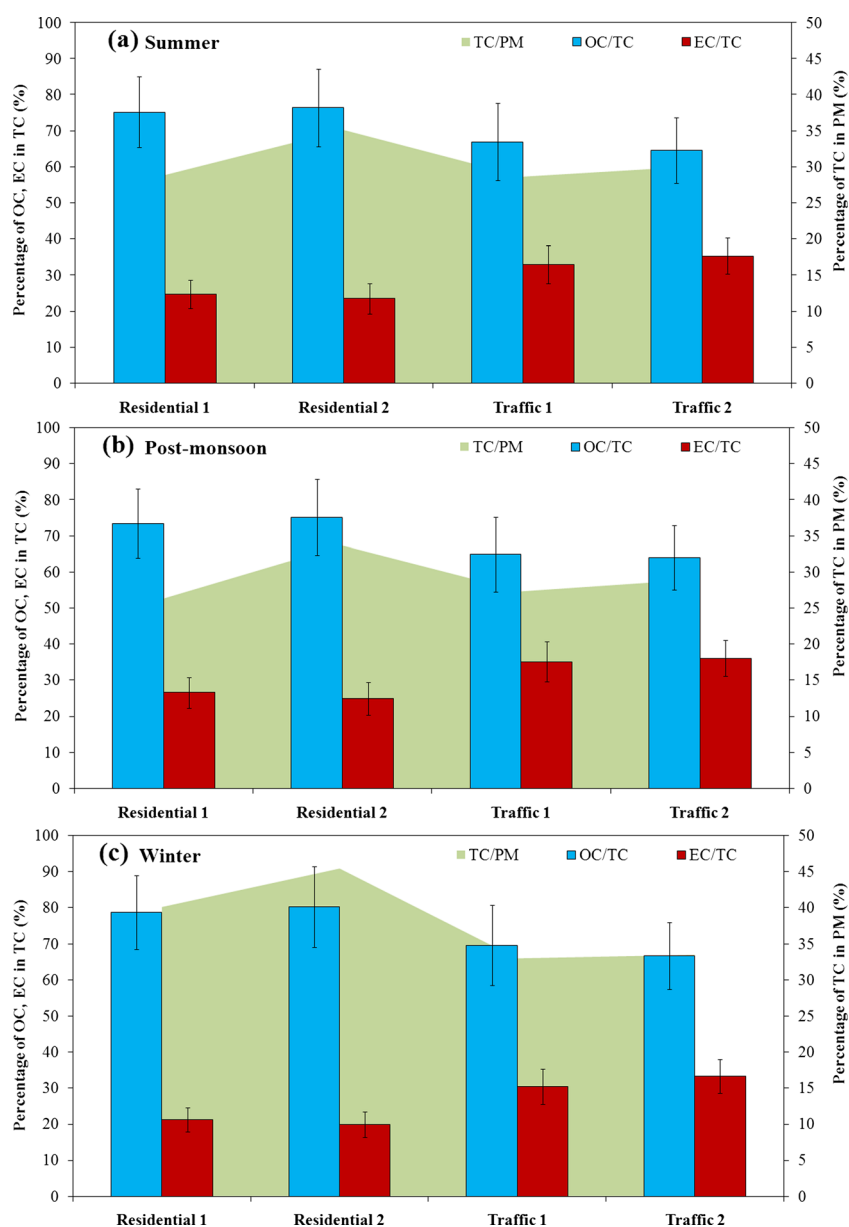
described in Overall results of PM, OC, and EC with respect to prevailing meteorology. To assess the possibility of biomass burning emissions on air quality at residential sites during winter, we used another indicator of biomass burning (i.e., char content=EC1-OP) (Han et al. 2010; Chuang et al. 2014). The winter to summer concentration ratio of EC1-OP was 1.9 and 1.3 at residential sites and traffic sites, respectively. Therefore, it could be inferred that biomass burning had significant impact on ambient levels of carbonaceous aerosols during winter of the measurement campaign. The winter to summer concentration ratio of OC2, OC3, and OC4 was more than 2.0 at two residential sites and less than 1.5 for two traffic sites. This indicated that the increase in emissions of domestic heating (combustion activities to get rid of cold) and coal combustion for more energy demand during winter could trigger to more emissions

of OC2, OC3, and OC4. Finally, from the seasonal variation observations, it was revealed that increase in primary emissions in the vicinity of the sampling sites and biomass burning emissions, coupled with stable atmospheric conditions made winter more critical than summer and post-monsoon.

### Contributions of OC and EC to TC and PM mass

The contributions of OC to TC (Fig. 6) indicate that residential sites contributed more OC to TC than the traffic sites. Specifically, during summer, the contributions of OC to TC varied from 64.6 % at traffic 2 to 76.5 % at residential 2. Similarly, the values varied from 63.9 % at traffic 2 to 75.2 % at residential 2 during post-monsoon and from 66.7 % at traffic 2 to 80.2 % at residential 2 during winter. In the same explanation

**Fig. 6** Mass contributions of OC and EC to TC and TC to PM<sub>10</sub> at four measurement sites during **a** summer, **b** post-monsoon, and **c** winter. The error bars are the respective standard deviations of the data sets



mentioned above, it could be inferred that the results were clear evidences for increase in pollution activities, specifically biomass burning during winter that could lead to surge in OC levels in the study area.

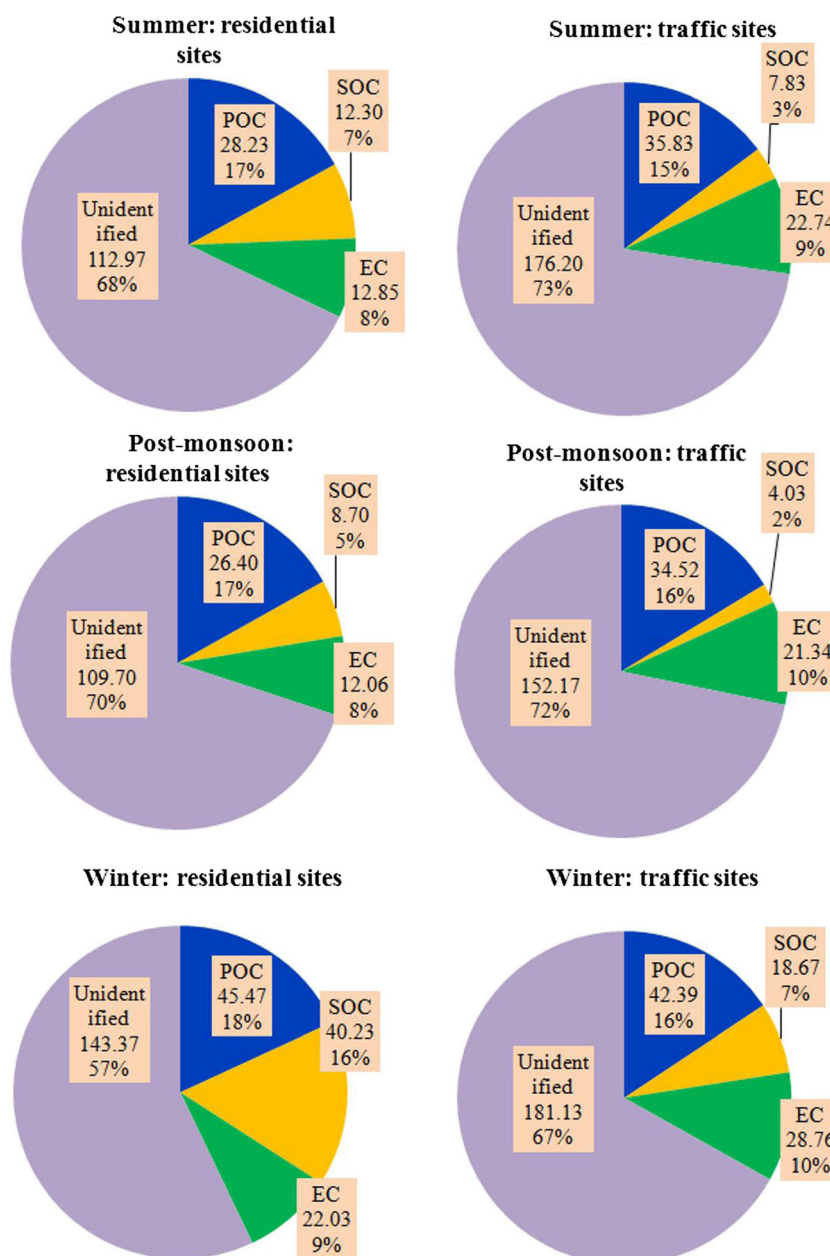
The contributions of EC to TC had more influences at traffic sites than that of residential sites. These trends clearly indicated that vehicles were largely responsible for ambient levels of EC. The contributions of EC to TC varied from 23.5 % at residential 2 to 35.4 % at traffic 2 during summer, from 24.8 % at residential 2 to 36.1 % at traffic 2 during post-monsoon, and from 19.8 % at residential 2 to 33.3 % at traffic 2 during winter. Hence, gradual increase in contribution of OC to TC took place during winter, compared to that during summer at residential sites. On the other hand, sharp decrease in

contributions of EC to TC took place during winter, compared to that during summer at residential sites. An interesting trend was obtained at traffic sites with significant decrease in contributions of EC to TC during winter, compared to that during summer. Therefore, it could be argued that the role of biomass burning including agricultural residue burning with increase in emission activities (responsible for primary and secondary OC levels) were paramount during winter season.

### Primary and secondary source contributions

Figure 7 shows the mass contributions of POC, SOC, and EC to ambient levels of  $PM_{10}$  at residential and traffic sites during summer, post-monsoon, and winter, and the estimations of

**Fig. 7** Concentrations and mass contributions of POC, SOC, and EC to  $PM_{10}$  at two types of sites during summer, post-monsoon, and winter. The numbers mentioned in figure represent the mass concentration ( $\mu g/m^3$ ) of respective component, whereas % represents the percentage contribution of respective component to PM mass



POC and SOC were based on the semi-empirical approaches described in [Methodology](#) section. On average, the contribution of POC to PM mass at residential sites varied from 17 % during post-monsoon to 18 % during winter, and at traffic sites from 15 % during post-monsoon to 16 % during winter. Similarly, the contribution of EC to PM mass at residential and traffic sites increased significantly during winter as compared to summer and post-monsoon. These results suggested that significant increase in primary emissions during winter took place due to more energy requirement and biomass burning. Hence, increase in primary emissions along with unfavorable meteorological conditions (poor dispersion) could cause the rise in ambient levels of EC and POC. The contribution of SOC to PM mass at residential sites varied from 5 % during post-monsoon to 16 % during winter, and at traffic sites from 2 % during post-monsoon to 7 % during winter. The disproportionate significant increase in SOC concentration during winter is a matter of concern, as this could lead to decrease in atmospheric visibility in the region.

To examine the reason behind the significant increase in SOC concentration during winter compared to summer and post-monsoon, the reasons could be prevailing temperature and mixing height. The atmospheric chemistry for formation of SOC is driven through formation of reactive organic gases (ROGs) in two predominant processes as follows: (1) production of condensable organic compounds through oxidation reaction and (2) subsequent nucleation and condensation (Na et al. 2004; Li et al. 2013). In general, radicals such OH, NO<sub>3</sub>, and O<sub>3</sub> are required in order for oxidation to occur, and there is greater formation of these radicals in summer than in winter due to higher actinic fluxes observed in summer (Seinfeld and Pandis 1998; Strader et al. 1999). However, this fact may not always remain the same, as possibility of multiple effects of temperature on SOC formation might happen in the atmosphere at higher temperature. For example, the vapor pressures of the secondary species also increase with temperature, driving a greater percentage of the total mass into the gas phase (Gray et al. 1986; Strader et al. 1999). As a result, there is a decrease in the fraction of the total species in the aerosol phase. Strader et al. (1999) also observed decrease in SOC formation with increase in temperature during their field experiment in San Joaquin Valley, USA. In addition to the above facts on prevailing temperature, mixing height could be another factor for such seasonal trends of SOC levels. The lower mixing height during winter could possibly allow accumulation of SOC precursors and the acceleration of SOC formation (Strader et al. 1999; Behera and Sharma 2010). In other words, particle agglomeration and ageing could take place during winter leading to increase in SOC levels. Overall, it could be inferred that increase in primary emissions coupled with unfavorable meteorological conditions make the atmosphere more conducive during winter for higher particle growths through the process of nucleation, condensation, agglomeration and ageing.

## Conclusions

This paper presents the findings of an intensive field study that was conducted at the urban sites in North India to examine the spatial and seasonal trends in levels of particulate carbon fractions and insights into formation of SOC. From the experimental results, it was observed that the highest levels of OC occurred during winter at a residential site, whereas the highest levels of EC occurred at a traffic site. The percentage of average mass of TC in PM mass was the highest during winter that varied from 32.9 % at a traffic site to 45.4 % at a residential site, indicating that winter is the critical season for higher levels of carbonaceous aerosols. The influence of source characteristics near to the measurement sites was clearly seen through the measured values of OC/EC. The site close to the road traffic had the least value of OC/EC; however, the site influenced by emissions of domestic cooking and heating had the highest value of OC/EC. The role of biomass burning on pollution loads of OC and EC was established through a sharp increase in levels of OC1 and EC1-OP during winter at two residential sites. The results from EC-tracer method in segregating the concentrations of POC, SOC, and EC showed that the contribution of SOC to PM mass increased from 5 % during post-monsoon to 16 % during winter at residential sites and from 2 % during post-monsoon to 7 % during winter at traffic sites. This study finds that increase in primary emissions along with unfavorable meteorological conditions (low temperature and poor dispersion) during winter could enhance the process of nucleation, condensation, agglomeration and ageing, leading to higher levels of carbonaceous aerosols in the study area.

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