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Status of Atmospheric Carbonaceous Matter in Various Locations of India

YASMEEN F. PERVEZ[1,*], SHAILENDRA K. KUSHAWAHA[1], S. NAIR[2] and SHAMSH PERVEZ[3]

[1]Department of Chemistry, Chhatrapati Shivaji Institute of Technology, Durg-491 001, India
[2]Department of Chemistry, Bhilai Institute of Technology, Durg-491 001, India
[3]School of Studies in Chemistry, Pt. Ravishankar Shukla University, Raipur-492 010, India

*Corresponding author: E-mail: dr.ypervez@gmail.com

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This article reviews the sampling locations, collection methodology and analysis of organic carbon (OC) and elemental carbon (EC), OC/EC ratio, total carbon, secondary organic carbon (SOC) from various sampling location methodology for collection and analysis in their respective sizes of particulate matter (PM) in selected cities of India. The carbonaceous aerosols consist of organic carbon (OC) and elemental carbon (EC) and combination of two is known as total carbon (TC). This paper reviewed numerous studies, which were carried out in various hotspot locations (namely residential, industrial, traffic intersections, rural, indoor environment *etc*.) of selected cities (Delhi, Agra, Kanpur, Pune, Mumbai and Ahmedabad) in India. The annual concentration of PM_{10} varied from 280.7 to 160 μg/m^3 and $PM_{2.5}$ varied from 145.59 to 88.89 μg/m^3, respectively. Carbonaceous analysis result showed that the annual concentration of organic carbon varied from 93 to 12.8 μg/m^3 and elemental carbon varied from 27 to 2.1 μg/m^3, respectively. OC/EC ratio in India (Delhi, Agra, Kanpur, Pune, Mumbai and Ahmedabad) varied from 3.28 to 17.2. In most of the cities OC/EC ratio exceeded 2, indicating the formation of secondary organic carbon (SOC). Secondary organic carbon is one of the major contributors of organic carbon and its contribution varied from 18 to 61 % in the selected location. The origin of carbonaceous aerosols were mostly from combustion processes.

Keywords: Particulate matter, Organic carbon, Elemental carbon, Total carbon.

INTRODUCTION

Atmospheric aerosols have attained considerable importance in present-day picture in urban cum industrial areas due to growing anthropogenic activities as well as in terms of their effects on human and climate. It plays an important role in climate change and atmospheric chemistry in the troposphere because of the direct effect on the radiative balance of the Earth's atmosphere by scattering or absorbing incoming shortwave radiation [1,2]. Recent interest has centered on fine particles ($PM_{2.5}$) and chemical species, particularly carbonaceous fractions [3], since they act as both air pollutants and climate agents [4-6]. Air pollution is recognized as one of the biggest problems now a days due to particulate matter which is of more concern for cities that are under industrialization in developing countries like India.

Carbonaceous aerosols are the single largest absorber of solar radiation and their heterogeneous reactions change the dynamics of atmospheric boundary layer which reduces visibility and also create health hazard [7]. They constitute a significant fraction in the fine particles ($PM_{2.5}$) and it could be accounted for up to 40 % of mass of $PM_{2.5}$ in urban atmosphere [8]. however, the rural atmosphere it accounted up to 77 % [9]. The rapid industrialization and urbanization in Asian countries required more energy consumption that lead to enhanced emission level of carbonaceous aerosols [10,11].

The carbonaceous aerosols are mainly composed of organic carbon (OC) as a major component, contributing up to 90 % and relatively low contribution 5-10 % from element carbon (EC), also called soot and black carbon (BC) [12].

Element carbon is a primary pollutant, emitted as by product of all combustion processes such as industrial emission, traffic, outdoor fires and household biomass fuels. It possesses a strong capability of absorbing solar radiation and is considered to play an important role in global climate change as it causes positive radiative forcing. It is listed as the second most important component of global warming after CO_2 [13,14]. Black carbon aerosols absorb sunlight in visible wavelengths, increasing inversely with wavelengths from the near infrared (1 μm) to the ultraviolet with a power law of 1. Their light absorbing characteristics has been implicated in regional atmospheric warming; changing Asian monsoon patterns and accelerated melting of the Himalayan glaciers. Elemental carbon directly released from primary anthropogenic source

through incomplete combustion [15]. Elemental carbon has been associated with an increase in mortality from lung cancer and other respiratory ailments [16].

Organic carbon refers to the molecule containing carbon hydrogen bond and represents a large variety of organic compounds, acids *etc*. Organic carbon is emitted directly or formed *in situ* in the atmosphere. It is a major component in scattering light and cooling the atmosphere. Organic carbon aerosols are emitted both from high and low temperature combustion system in primary and secondary forms; the later is produced by atmospheric oxidation of co-emitted VOC gases [17]. Organic carbon aerosols absorbed sunlight insignificantly at visible wavelengths, with the exception of brown carbon (BrC), a class of organic carbon, are caused by a mixture of primary and secondary particulate matter generated from engine exhaust, biomass burning and industrial processes, which strongly absorbs in the short visible wavelength, relative to black carbon.

Organic carbon emitted directly from sources in particulate form is considered as primary organic carbon, while secondary organic carbon produced from gas to particle conversion phenomenon through VOCs. Low vapour pressure, sufficient temperature and sunlight influence the formation of secondary organic carbon in atmosphere [18]. Organic carbon may contain polycyclic aromatic hydrocarbons (PAHs) and other organic constituents that have potential mutagenic and carcinogenic effects [19].

Black carbon and organic carbon occur together in particulate matter as soot, in the atmosphere, over this region alter net solar radiation flux to the earth's surface, the net effect of which may outweigh the regional warming effect of greenhouse gases [20]. However, soot also scatters solar radiation back into space, which may counteract the warming effect. Water soluble organic carbon may be mixed in liquid water, so changing its optical properties and affecting the creation and surface tension of cloud droplets [17].

The ratios of OC/EC have been used to indicate the presence of primary as well as secondary organic carbon [21]. The OC/EC ratios were also estimated which are used to interpret the emission and transformation characteristics of carbonaceous aerosols. Various researchers have studied different emission sources and assigned characteristic OC/EC ratios to them. OC/EC ratios of 1.0 to 4.2 for diesel and gasoline powered vehicular exhaust [22]. OC/EC ratios of 16.8 to 40.0 for wood combustion [23]. OC/EC ratios of 2.5 to 10.5 for residential coal smoke [24]. OC/EC ratios of 32.9 to 81.6 for kitchen emissions [25]. OC/EC ratios of 7.7 for biomass burning [26,27].

This paper reviews, the work carried out by various researchers in India. The primary objective of the paper is to review studies on the characterization and status of carbonaceous aerosols from selected locations in India.

EXPERIMENTAL

Study area: Cities chosen under this review, represent a wide spectrum of activities, socio-economic development, geophysical character, climate, sources of pollution, *etc*. Delhi, Kanpur and Agra are in Northern India with typical dry climate and mixture of industrial and commercial activities. Mumbai is coastal city, which experience land/sea breeze influencing dispersion of pollutants. Pune is upcoming city that have more of commercial/institutional activities than industrial. Ahmedabad, an urban city is located in a semiarid region of western India close to southeast boundary of great Thar Desert and northeast of Arabian sea.

Mandal *et al.* [28] carried out monitoring at CSIR R&D Centre, Naraina Industrial area, Phase I, Delhi. Sampler placed near to CSIR R&D centre to identify the probable source of carbonaceous aerosols in an industrial area of Delhi, India. Sharma *et al.* [29] collected the PM_{10} sample at sampling site of the CSIR National Physical Laboratory, New Delhi. The sampling site represents a typical urban atmosphere, surrounding by huge roadside traffic and agricultural field in the southwest direction, there are different small, medium and large industries in and around Delhi. Pipal *et al.* [30] collected Aerosol samples at sampling site of the Indian Institute of Tropical Meteorology, New Delhi, on the roof top of the building (15 m above the ground level). The major source of pollution are power plants, road transport, small – scale industries and domestic cooking. Ram *et al.* [31] studied Ambient aerosols (PM_{10}) Samples were collected at sampling site at Kanpur representing an Urban environment, is located in the central part of the Indo-Gangetic Plain. The sampler was setup on the third floor of the Environmental Engineering Laboratory (15 m above ground level) located inside the Indian Institute of Technology campus and upwind of the emission source from the city. The source of pollution are biomass burning emission, wood fuel burning for domestic use and vehicular exhaust. Kumari *et al.* [32] carried out sampling at Dayalbagh, a suburb situated in the north of Agra. The sampler was present on the roof of the Science Faculty building. The source of pollution at sampling site are vehicle emission and biomass burning. Kumari *et al.* [33] particulate matter sampling carried out at the urban residential site in Khandari, Agra. The sampling site was loated on the roof of a building (10 m above the ground) located in residential apartment complex. Taneja *et al.* [34] collected $PM_{2.5}$ samples from a roadside *i.e.*, Khandari site, Agra. The sampler was setup at the top (25 m above the ground) of the building roof at the road side. The sampling area is directly affected by heavy traffic emission and biomass combustion. Elizabeth *et al.* [35] carried out sampling at two hotspot sites residential (Khar) and industrial (Mahul) in Mumbai city. Satsangi *et al.* [36] collected particulate matter samples at the roof of Chemistry Department, Savitribai Phule Pune University (SPPU). The site is representative of greenery construction activities and major traffic junction which is one and half km away from the sampling site. Apart from this residential colony, shopping malls and colleges are also situated around the site. Rastogi and Sarin [2] monitored sampling site for aerosol collection which was located on terrace of an eight storey building (40 m above the ground level) in institute premises, PRL Ahmedabed. The site is ideal to study the chemical interaction among mineral aerosols (derived from desert area), sea salts (from the Arabian sea) and anthropogenic emission (due to rapid urbanization).

Table-1 shows the monitoring parameters and frame work of different locations of India.

TABLE-1
MONITORING PARAMETER AND FRAME WORK

	Sampling instrument	Flow rate	Sampling period (h)	Sampling frequency	Analytical instruments	Analytical method
PM_{10}	APM 541	1 m^3/h	12	4-10 days in a month	–	Gravimetric
	APM 460	1.12 m^3/h	8	4-5 days in a month	–	Gravimetric
	HVS	1 lpm	8	15 days during Jan.-Feb. 7 days during March-June	–	Gravimetric
$PM_{2.5}$	APM 550 r	1 m^3/h	12	8-10 days in a month	–	Gravimetric
	APM 550	16.6 lpm	24	–	–	Gravimetric
OC/EC	PM_{10} sampler	1m^3/h	12	4-10 days in a month	Carbon analyser	IMPROVE thermal optical reflectance protocol
	PM_{10} sampler	1 lpm	8	15 day during Jan.-Feb. 7 day during March-June	Carbon analyser	Thermal-optical transmittance protocol
	PM_{10} sampler	1.12 m^3/h	8	4-5 days in a month	Carbon analyser	USEPA method IMPROVE protocol with negative pyrolysis
	$PM_{2.5}$ sampler particulate collected on Quartz filter	16.6 lpm	24	–	Semi- continuous thermal/optical carbon analyser	NIOSH 5040 protocol

Organic carbon and elemental carbon in most of the countries are analyzed by Carbon Analyser (Atmoslytic Inc., USA) by IMPROVE thermal optical reflectance protocol (TOR) [37,38]. The principle of the analyzer is based on preferential oxidation of organic carbon and elemental carbon at different temperatures. The principal function of the analyzer is to pyrolyse the sample and then to char the organic carbon compounds into elemental carbon. The method analyzes for organic carbon fractions (OC1, OC2, OC3 and OC4 at 140, 280, 480 and 580 °C), pyrolyzed carbon fraction (OP) and elemental carbon fractions (EC1, EC2 and EC3 at 580, 740 and 840 °C), respectively. The IMPROVE protocol software defines OC as OC1 + OC2 + OC3 + OC4 + OP, EC as EC1 + EC2 + EC3 – OP and TC as OC + EC.

C-mat 5500 carbon analyzer (Strohlein, Germany) used for measurement of organic carbon concentrations, organic carbon was volatilized at 650 °C under nitrogen and catalytic converted to CO_2, while elemental carbon was brunt to CO_2 at 650 °C under oxygen. The formation of CO_2 was determined with a NDIR detector. The concentrations of organic carbon and elemental carbon concentrations strongly depended on the condition of the operational OC/EC splitting.

Aethalometer (Magee Scientific, USA) having flow rate of 2 lpm. The instrument provides the real time data of black carbon in the air stream. Real time indoor black carbon concentrations can be estimated using a MicroAeth Model AE-51 (Magee Scientific, Berkeley, CA) which was set at a flow rate of 50 mL/min.

Transmission OC/EC Lab Instrument (Sunset Laboratory, Forest Grove, USA, Model 2000) the samples were analyzed in two stages.

Stage I: An aliquot was quartz filter paper was initially heated stepwise up to temperature of 870 °C in a non-oxidizing helium atmosphere and then cooled at 550 °C.

Stage II: Then again heated at 870 °C in an oxidizing atmosphere (98 % He + 2 % O_2). The evolved carbon at each temperature step is oxidized to CO_2 and then reduced to CH_4 for quantification with flame ionization detector. The transmittance of light from He-Ne laser, through the filter punch, is continuously monitored and used for settings the OC/BC split line.

RESULTS AND DISCUSSION

The comparison of $PM_{2.5}$ and PM_{10} with the reported value in different location of India is presented in Table-2. In this study, the reported annual concentration of PM_{10} (280.7 μg/m^3) and $PM_{2.5}$ (145.49 μg/m^3) in Delhi are higher as compare to mega cities like Kanpur (PM_{10} = 160 μg/m^3), Ahmedabad ($PM_{2.5}$ = 139.4 μg/m^3), Pune (PM_{10} = 169.9 μg/m^3, $PM_{2.5}$ = 104.25 μg/m^3), Mumbai ($PM_{2.5}$ = 88.89 μg/m^3) (Fig. 1). The observed value of particulate matter are substantially higher than the annual standard values stipulated by Indian National Ambient Air Quality Standard (NAAQS) (40 μg/m^3 for $PM_{2.5}$ and 60 μg/m^3 for PM_{10}) and World Health Organization (WHO) (25 μg/m^3 for $PM_{2.5}$ and 50 μg/m^3 for PM_{10}), respectively. They also exceeded by far, limit value of standards of United State Environmental Protection Agency (USEPA) (15 μg/m^3 for $PM_{2.5}$) and European Union Standards (20 μg/m^3 for $PM_{2.5}$). The high concentrations of particulate matter in different cities of India may be due to vehicular activities, construction activities, biomass and fossil fuel combustion and industrial emission.

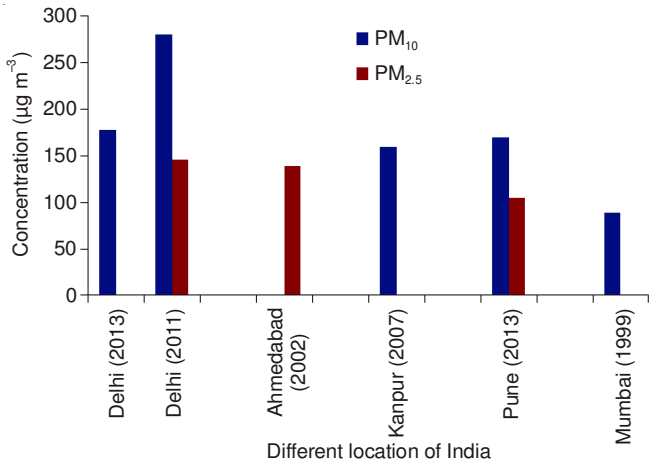

Fig. 1. Annual concentration (μg m^{-3}) of PM (PM_{10} and $PM_{2.5}$) in different locations of India

During winter higher concentration of PM_{10} (351.2 μg/m^3) were observed in Delhi and $PM_{2.5}$ (273 μg/m^3) were observed in Agra. This was compared with other studies conducted in

TABLE-2
COMPARISON OF DIFFERENT PARAMETERS IN VARIOUS STUDY PERIODS OVER INDIAN REGIONS

S. No.	Place	Study periods	Seasons	Sampling area	Concentration (μg/m^3)								OC/EC	Concentration (%)			
					PM_{10}	$PM_{2.5}$	OC	EC	TC	TCA	SOA	POC		OC	EC	TC	TCA
A: Annual																	
1	Delhi	Annual	2013	Urban	177.9	–	26.7	6.1	32.80	48.82	9.62	23.2	4.30	15.00	3.40	18.4	27.4
2	Delhi	Annual	2011	Urban	280.7	–	93.0	27.0	120.3	176.14	–	–	3.44	32.75	9.47	42.8	61.8
3	Delhi	Annual	2011	Urban	–	145.59	41.1	13.0	54.37	79.04	–	–	3.28	28.24	9.10	37.3	54.2
4	Ahmedabad	Annual	2002	Urban	–	139.40	12.8	2.1	14.90	22.58	4.00	–	8.30	9.18	1.50	10.6	16.1
5	Kanpur	Annual	2007-08	Urban	160.0	–	21.7	3.4	24.10	36.80	–	–	6.70	13.56	2.12	15.0	23.0
6	Pune	Annual	2013	Urban	169.9	–	33.4	2.4	35.80	55.84	22.47	14.0	17.20	19.66	1.41	21.0	32.8
					–	104.25	31.3	2.7	33.98	52.73	23.54	10.7	15.80	29.98	2.61	32.5	50.5
7	Mumbai	Annual	2007-08	Urban	–	88.89	32.7	7.7	40.40	60.02	–	–	3.99	36.80	8.60	45.4	67.5
B: Winter season																	
1	Delhi	Winter	2013	Urban	213.1	–	36.1	9.6	45.69	67.32	9.05	36.6	3.74	16.92	4.52	21.4	31.5
2	Delhi	Winter	2011	Urban	351.2	–	118.0	37.0	155.10	226.02	–	–	3.21	33.66	10.32	44.1	63.6
3	Delhi	Winter	2011	Urban	–	180.26	51.1	18.0	68.84	99.48	–	–	2.92	28.33	9.85	38.1	55.1
4	Delhi	Winter	2012	Urban	–	231.19	50.1	11.0	60.78	106.60	38.76	27.6	5.43	21.67	4.61	26.2	46.1
5	Agra	Winter	2012	Urban	–	165.42	70.0	9.5	79.49	121.40	48.16	26.5	13.70	42.29	5.76	48.0	73.3
6	Agra	Winter	2011-12	Urban	–	189.00	58.0	5.2	63.20	98.00	–	–	11.70	30.69	2.75	33.4	51.8
7	Agra	Winter	2009	Suburban	–	273.00	60.9	7.5	68.40	104.94	–	–	8.10	22.30	2.74	25.0	38.4
8	Kanpur	Winter	2007-08	Urban	205.5		40.4	4.9	45.30	69.24	–	–	8.70	19.66	2.38	22.0	33.6
C: Summer season																	
1	Delhi	Summer	2013	Urban	185.9	–	29.3	5.5	34.78	52.37	14.07	20.1	5.38	15.77	2.93	18.7	28.1
2	Delhi	Summer	2011	Urban	258.0	–	81.5	23.0	104.4	153.3	–	–	3.58	31.59	8.92	40.4	59.9
3	Delhi	Summer	2011	Urban	–	145.44	40.0	12.0	51.91	76.03	–	–	3.43	27.52	8.05	35.6	52.2
5	Agra	Summer	2011	Urban	–	55.30	18.2	3.2	21.40	32.32	7.20	12.9	5.40	32.91	5.78	38.6	58.4
6	Kanpur	Summer	2007-08	Urban	133.3	–	13.4	2.1	15.50	23.54	–	–	7.30	10.05	1.57	11.6	17.6
D: Monsoon season																	
1	Delhi	Monsoon	2013	Urban	134.7	–	14.7	3.4	18.07	29.85	5.37	12.7	4.39	10.92	2.48	13.4	22.1
2	Delhi	Monsoon	2011	Urban	122.9	–	39.3	11.0	50.71	74.26	–	–	3.49	31.93	9.27	41.2	60.2
3	Delhi	Monsoon	2011	Urban	–	78.67	20.0	6.3	26.29	38.29	–	–	3.65	25.40	8.00	33.4	48.6

India. Sharma *et al.* [29] have reported concentration of PM_{10} (213.1 μg/m^3), Ram *et al.*[40] reported concentration of PM_{10} (205.5 μg/m^3). Pipal *et al.* [29] reported concentration of $PM_{2.5}$ (180.26 μg/m^3); Taneja *et al.* [33] reported concentration of $PM_{2.5}$ (189 μg/m^3) in road side of urban area.

During summer and monsoon high concentration of PM_{10} (258 and 134.7 μg/m^3) were observed in Delhi [28,38], respectively. $PM_{2.5}$ (145.44 and 78.67 μg/m^3) were observed in Delhi [29]. The concentration of PM_{10} and $PM_{2.5}$ during the study were in the following order winter > summer > monsoon (Fig. 2). The contribution of PM_{10} and $PM_{2.5}$ was attributed to high emission from bursting of fire crackers during Diwali festival, Biomass burning and unfavourable meteorological conditions such as stability of atmosphere, slow dispersion and low-average mixing height during winter season as compared to monsoon and summer season. Higher concentration of PM_{10} during winter may be due to the combined effect of source strength and lower boundary layer height [40]. Generally during winter, the meteorology of Delhi is dominated by high pressure centred over Western China causing increased atmospheric stability, which in turn, allows less general circulation engulfing more stagnant air masses [40]. Additionally, lack of precipitation during winter may also reduce the potential of wet deposition and associated cleansing mechanism of the atmosphere.

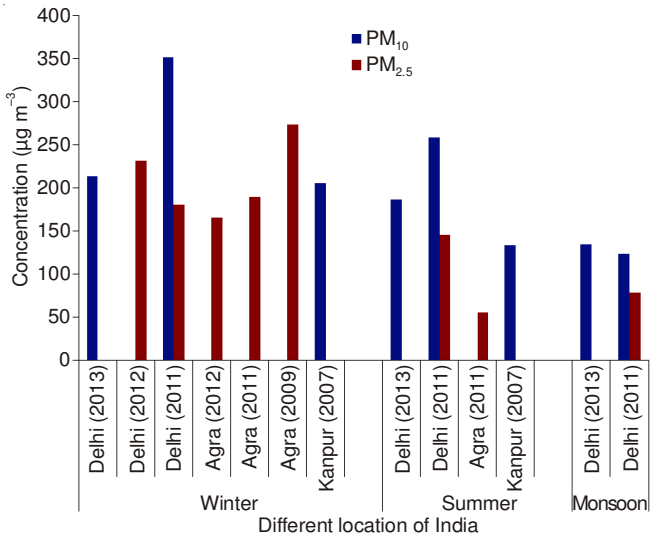

Fig. 2. Seasonal concentration (μg m^{-3}) of PM (PM_{10} and $PM_{2.5}$) in different locations of India

The elemental carbon and organic carbon concentration exhibit a large temporal variability during the studies and their annual mass concentration varied from 27 to 2.4 μg/m^3 and 93 to 12.8 μg/m^3, respectively (Table-2, Fig. 3). The comparison of mass concentration of elemental carbon and organic carbon in different location in India are presented in Fig. 4. The highest organic carbon and elemental carbon concentration (118 and 37 μg/m^3) was observed in Delhi (2011) during winter. The major variation in concentration of organic carbon and elemental carbon was observed during different season. Such as elemental carbon arise from the incomplete combustion of various kinds of organic material, including coal, oil, petrol, wood and other biomass [41]. The higher concentration of elemental

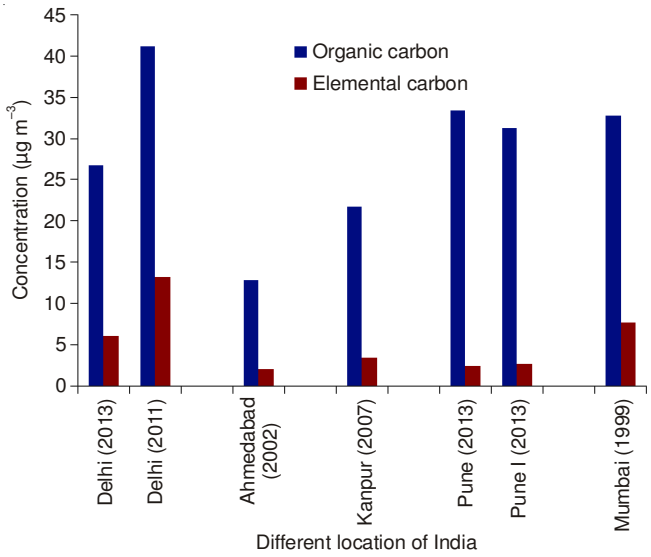

Fig. 3. Annual concentration (μg m^{-3}) of organic carbon and elemental carbon in different locations of India

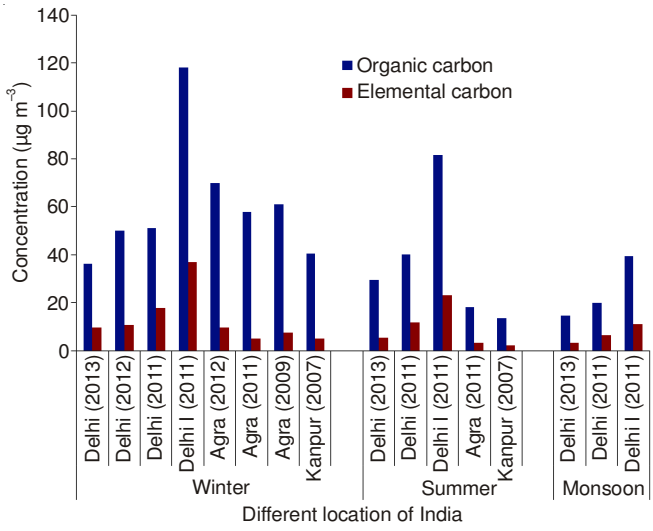

Fig. 4. Seasonal concentration (μg m^{-3}) of organic carbon and elemental carbon in different locations of India

carbon in winter are due to the long range transport of primary pollutants as well as local emission such as festival effect and burning of fossil fuel along with meteorological impact. The higher concentration of organic carbon in winter season leads to frequent accumulation of atmospheric pollutants in lower atmospheric layer. Hence, it may be concluded that the difference in values at the sites could be due to the variation in primary and secondary source of carbonaceous aerosol in different cities.

The ratio of OC/EC reported over a few Urban cities of India presented in Table-2, Fig. 5. Annual higher OC/EC ratio was observed in Pune (17.2), it may be inferred that the carbonaceous species in Pune was from all emission source excluding kitchen emission. Higher OC/EC ratio in winter observed in Agra (13.75) which suggest the presence of secondary organic aerosol. Higher OC/EC ratio in summer observed in Kanpur (7.3) suggests vehicular emission and biomass burning emission. Higher OC/EC ratio in summer observed in Delhi (4.39) suggests vehicular emission. Secondary organic aerosol plays

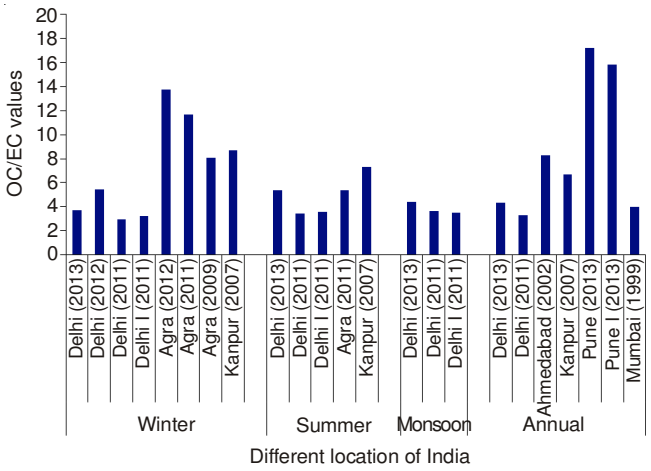

Fig. 5. Value of OC/EC in different locations of India

plays an important role on physical and chemical properties of atmosphere, relating to haze, visibility, climate and health. Secondary organic aerosol can be calculated by estimated by following formula on the basis of (OC/EC) ratio [42] as:

$$SOC = TOC - (OC/EC)_{minimum} \times EC$$

The minimum ratio of OC/EC is used to estimate secondary organic carbon.

Table-2 shows the value of secondary organic aerosol which varied from 48.16 to 9.5 μg/m^3 in winter, 14.07 to 7.2 μg/m^3 in summer, 5.3 μg/m^3 in monsoon, respectively in the study of different cities of India. Higher value of secondary organic aerosol during winter is due to VOC condensed into aerosol at lower temperature, as more residential combustion of coal and wood for space heating source in winter.

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