

## Carbonaceous aerosols at an urban residential site in Agra

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Carbonaceous aerosols are an important constituent of the aerosols and may be broadly divided in two parts, viz. organic carbon (OC) and elemental carbon (EC). Carbonaceous aerosol concentration was determined at an urban residential site in Agra. Sampling of fine particles (PM<sub>2.5</sub>) was carried out during May to August 2011. The concentration of PM<sub>2.5</sub> at urban residential site was 55.3±17.4 µg m<sup>-3</sup>, which is within the prescribed limits. OC varied from 7.6 to 37.5 µg m<sup>-3</sup>, with an average of 18.2±6.4 µg m<sup>-3</sup>; EC ranged 1.2 - 9.4 with an average of 3.2±1.6 µg m<sup>-3</sup>; while total carbonaceous aerosols (TCA) varied from 13.6 to 69.4 µg m<sup>-3</sup>. On an average, TCA accounted for 64.9% of PM<sub>2.5</sub> mass implying that carbonaceous aerosol is a significant component of PM<sub>2.5</sub>. OC/EC ratio at the site was in the range of 3.5 - 11.9. OC/EC ratio indicates the contribution of both the primary and secondary sources.

**Keywords:** Carbonaceous aerosols, Particulate matter, Organic carbon, Elemental carbon, OC/EC ratio

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### 1 Introduction

Aerosols are solid or liquid particles suspended in the air. Aerosol affects the Earth's radiation budget and cloud formation, which in turn affect the climate. The epidemiological studies have demonstrated that the exposure of particulate matter (PM) is associated with the occurrence of acute respiratory infections, lung cancer and chronic respiratory and cardiovascular diseases<sup>1,2</sup>. Carbonaceous aerosol is a significant component in fine particles PM<sub>2.5</sub> (aerodynamic diameter less than 2.5 µm), accounting for up to 40% of PM<sub>2.5</sub> mass in urban atmosphere<sup>3</sup>. During the last 10–15 years, the carbonaceous aerosol has become one of the most intensely studied fields within the atmospheric sciences. The interest in atmospheric carbonaceous aerosol can be attributed to its postulated impacts on the global climate and on human health<sup>4-6</sup>.

Typically, two classes of carbonaceous aerosol commonly present in ambient air are organic carbon (OC) and elemental carbon (EC), which are large contributors to the fine particle burden in the urban atmosphere and heavily industrialized areas<sup>7</sup>. OC, containing polycyclic aromatic hydrocarbon and other components with possible mutagenic and carcinogenic effects, can be directly emitted from sources (primary OC) or produced from atmospheric reactions involving gaseous organic precursors

(secondary OC)<sup>8</sup>. EC, often named black carbon or soot carbon, may be the second most important component of global warming in terms of direct forcing, after CO<sub>2</sub> (Ref. 9). This is mainly attributed to the black carbon part of the carbonaceous aerosol, which absorbs solar radiation in the atmosphere.

Despite the evident significance of OC and EC in atmospheric and environmental process, the knowledge on their concentrations, sources and formation mechanisms is quite poor, especially in India. The Indo-Gangetic Plain (IGP), extending from north to east, is one of the most polluted regions in northern India with characteristic emissions from small scale industries, vehicular traffic and biomass burning (wood fuel and agriculture waste). Although, several studies have been carried out to study the optical properties of aerosols over northern India and to understand their impact on the radiation budget<sup>10-13</sup>, only few studies report carbonaceous species in this region especially urban and suburban<sup>14-21</sup>. OC has been reported only at Kanpur and Delhi. At Kanpur, OC and EC has been 36.6±17.1 and 6.2±2.0 µg m<sup>-3</sup>, respectively<sup>15</sup>; while in Delhi, OC and EC has been 54.0±39.0 and 10.0±5.0 µg m<sup>-3</sup>, respectively<sup>20</sup>.

The aerosol characterization studies, especially from urban areas in the semi-arid region are important in the context of enhanced emission levels and heterogeneous chemistry with the mineral dust.

Several authors have reported chemical composition of total suspended particulates in the Indian region<sup>22-24</sup>. Data on chemical characteristics of PM<sub>2.5</sub> is sparse in the Indian region.

In view of this, PM<sub>2.5</sub> concentration of carbonaceous species at the urban residential site is determined in the presented study. Further, an elucidation of primary and secondary sources is also attempted.

## 2 Experimental method

### 2.1 Sampling site

Particulate matter sampling was carried out at an urban residential site (UR) in Khandari, Agra. The sampling site was located on the roof of a building (10 m above the ground) located in residential apartment complex. There are about 2000 residences around the sampling site. Most of the families living in this area are using LPG for cooking purpose. Almost all families have either two or four wheelers for their commuting. In an estimate, it was found that 35-40% people are using four wheelers and the remaining are using two wheelers. The sampling site lies by the side of the road that carries mixed vehicular traffic of the order of 1000 vehicles in a day. The sampling site is a residential site, which lies about 800 meters from the National Highway 2 (Fig. 1). The traffic junction is upwind to the sampling site, that is, the wind direction is northwest, i.e. from sampling site to National Highway 2. Hence, aerosol composition is not influenced by high traffic density.

The climate of Agra is strongly influenced by the aeolian dust down blown from the Asian subcontinent and Thar Desert. Temperature ranges between 12-48°C (max) and 0.7-30°C (min), relative humidity 25-95%, and annual rainfall is concentrated only between June and September. Agra is known for glass and rubber industries. Industrial activities of Agra include rubber processing, engineering and a few ferrous casting industries based on natural gas. Apart from local sources, Mathura refinery and Firozabad glass industries are both situated at a distance of 40 km from Agra. The wind speed in Agra is mostly 1-2 ms<sup>-1</sup>.

### 2.2 Sample collection

Sampling was carried out during May to August 2011 at the urban residential site in Khandari, Agra, India using HVS Envirotech APM 550 Respirable Dust Sampler operated at a flow rate of 16.6 L min<sup>-1</sup> for 24 hrs on 47 mm quartz fiber filter. Before exposure, the quartz fiber filter was pre-heated in a muffle furnace at 800°C for 6 h to remove organic impurities. Before and after sampling, the filter was desiccated for 24 h, and then weighed on an electronic microbalance (Mettler) to determine the PM mass. Each filter was weighed at least three times before and after sampling, and the net mass was obtained by subtracting the pre-sampling weight from the post-sampling weights. After weighing, the filter wrapped in aluminium foil was sealed in polyethylene zip lock bags and stored in deep freezer at -4°C until the time of chemical analysis to prevent the evaporation of volatile components.

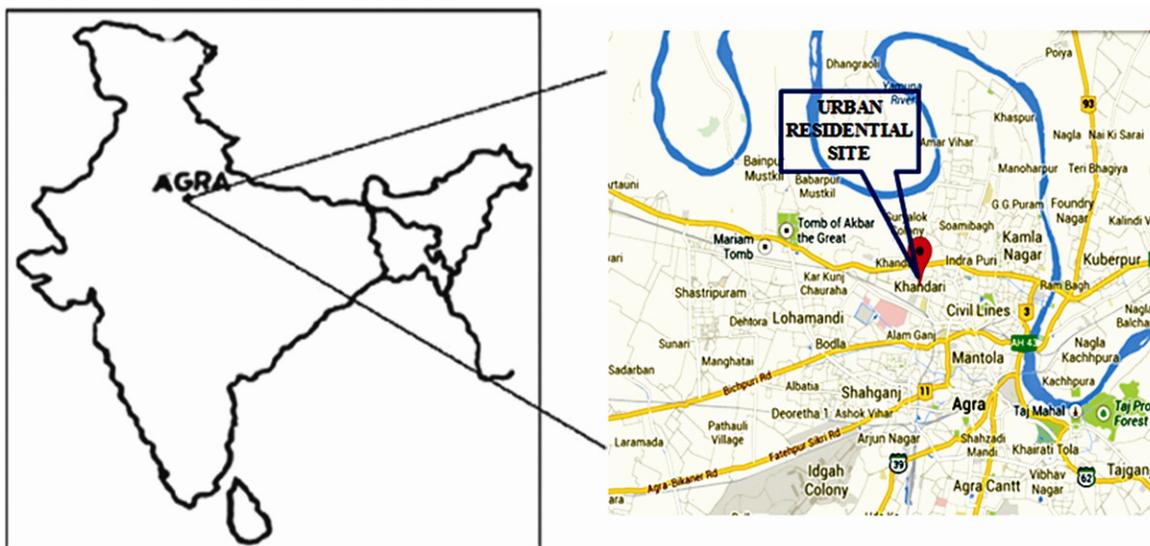


Fig. 1 — Map showing residential site

Laboratory blank filters ( $n = 4$ ) were collected to reduce gravimetric bias due to filter handling during and after sampling. Filters were handled only with tweezers coated with Teflon tape to reduce the possibility of contamination. After weighing, the samples were wrapped in aluminum foil and sealed in polyethylene zip-lock bags and stored in deep freezer at  $-4^{\circ}\text{C}$  until the time of analysis to prevent the degradation of organic compounds due to photo-oxidation. It was assumed that the collected particulate matter was uniformly distributed over the entire area of filters. Blank test was also monitored by using operational blanks (unexposed filters), which were processed with field samples. The blank filters were taken once a month. They were exposed in the field when the field sampling box was opened to remove and replace field samples. Field blank values were very low ( $0.2 \pm 0.1 \mu\text{g}$ ), typically below or around the method detection limits ( $0.28 \pm 0.1 \mu\text{g m}^{-3}$ , using  $3\sigma$  values of total procedural blank concentrations of the filter)<sup>16</sup>.

### 2.3 Carbonaceous aerosol analysis

A filter punch of  $1.5 \text{ cm}^2$  size was cut from the sample filter and analyzed for OC and EC concentration using Transmission OCEC Lab Instrument (Sunset Laboratory, Forest Grove, USA, Model 2000). The analysis was performed in two stages: (i) an aliquot of quartz filter first heated stepwise up to  $870^{\circ}\text{C}$  in a non-oxidizing He atmosphere, cooled to  $550^{\circ}\text{C}$ ; and (ii) then heated to  $870^{\circ}\text{C}$  in a oxidizing atmosphere (98% He + 2%  $\text{O}_2$ ). The evolved carbon at each temperature step is oxidized to  $\text{CO}_2$  and then reduced to  $\text{CH}_4$  for quantification with a flame ionization detector. The transmittance of light from a He-Ne laser, through the filter punch, is continuously monitored and used for setting the OC/EC split line, thereby, correcting for pyrolysis/charring during the first stage of the analysis. Details of analysis are represented in an earlier paper<sup>25</sup>.

Standardization of OC/EC Analyzer was carried out every day using sucrose solution ( $3.2 \mu\text{g}/\mu\text{L}$ ). A solution of  $10 \mu\text{L}$  gives  $32.0 \pm 1.8 \mu\text{g OC}$ . For quality control, the analyzer was calibrated using a blank punch of pre-heated Quartz Fiber Filter and standard sucrose solutions every day. Sampled quartz filters were also analyzed similarly for blank corrections. The overall blank concentrations from the quartz filters for OC and EC were  $0.5 \pm 0.2$  and  $0.0 \pm 0.02 \mu\text{g cm}^{-2}$ , respectively. These were subtracted from the measured OC and EC concentrations in the aerosol samples<sup>26</sup>.

### 3 Results and Discussion

The statistics for  $\text{PM}_{2.5}$  mass for urban residential site is presented in Table 1 and represented in Fig. 2. The level of  $\text{PM}_{2.5}$  at the urban residential site is  $55.3 \pm 17.4 \mu\text{g m}^{-3}$ , which is within the prescribed limits set for residential sites. National Ambient Air Quality Standard (NAAQS) has set a 24 hour average standard of  $\text{PM}_{2.5}$  mass concentration as  $60 \mu\text{g m}^{-3}$  for residential sites. Table 1 shows comparison of  $\text{PM}_{2.5}$  mass of urban residential (UR) site at Agra with that of other urban residential sites. It has been observed that the concentration of  $\text{PM}_{2.5}$  at Khandari, Agra are equal to the concentration at Ahmedabad and Xiangzhou (XZ), Zhuhai; and lower than Siming District, but it is higher than Baptist University (BU), Hong Kong and Seoul, Korea.

The measured abundances of OC and EC in this study are represented in Fig. 2. OC varied from  $7.6$  to  $37.5 \mu\text{g m}^{-3}$  with an average of  $18.2 \pm 6.4 \mu\text{g m}^{-3}$  while EC ranged  $1.2 - 9.4$  with an average of  $3.2 \pm 1.6 \mu\text{g m}^{-3}$ . In  $\text{PM}_{2.5}$ , OC was the dominant portion and contributed about 32.9% at the urban residential site.

Table 1 represents the comparison of OC and EC with some urban residential sites of Asia. OC concentration at the urban residential site (Agra) is equal to those observed at Ahmedabad (India) but

Table 1 — Comparison of  $\text{PM}_{2.5}$ , OC and EC at urban site

Site	Type	$\text{PM}_{2.5}$ , $\mu\text{g m}^{-3}$	OC, $\mu\text{g m}^{-3}$	EC, $\mu\text{g m}^{-3}$	OC/EC, $\mu\text{g m}^{-3}$	References
Agra, India	UR	55.3	$18.2 \pm 6.4$	$3.2 \pm 1.3$	5.4	Present study
BU, Hong Kong	UR	48.5	$8.4 \pm 6.3$	$4.4 \pm 4.7$	2.3	Cao <i>et al.</i> 2003 (Ref. 27)
XZ, Zhuhai, China	UR	59.3	$12.2 \pm 4.4$	$5.0 \pm 1.6$	2.5	Cao <i>et al.</i> 2003 (Ref. 27)
Seoul, Korea	UR	41.80	$10.89 \pm 0.94$	$6.98 \pm 0.58$	---	Park & Kim 2005 (Ref. 28)
Siming District, China	UR	72.12	$19.28 \pm 9.48$	$3.3 \pm 1.32$	5.8	Fuwang Z <i>et al.</i> 2011 (Ref. 29)
Ahmedabad, India	UR	55.7	$18.3 \pm 5.9$	$3.0 \pm 0.9$	6.2	Rengarajan <i>et al.</i> 2011 (Ref. 30)

\*UR= Urban residential

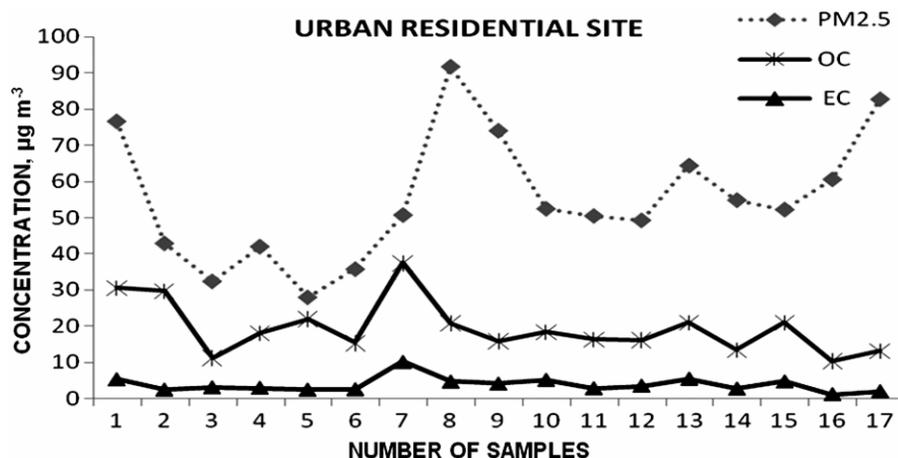


Fig. 2 — Variation of PM<sub>2.5</sub>, OC and EC concentrations at urban residential site

Siming District has higher concentration of OC than Agra, while Seoul (Korea) and XZ, Zhuhai shows lower concentration of OC. EC concentration at the urban residential (Agra) is equal to the concentration of EC at Ahmadabad and Siming District while lower than BU, Hong Kong; XZ, Zhuhai and Seoul, Korea. Variability in the OC and EC concentrations at different urban residential sites observed is reported in Table 1. This variation may be attributed to their sources which may be either primary or secondary or both. At BU, Hong Kong and Seoul, Korea, the major source of carbonaceous aerosols is motor vehicle exhaust which is a primary source. In Siming District, OC concentrations were not only affected by primary sources like exhaust of local pollutants but also by meteorological factors while at Ahmedabad, the contributing factor is primary as well as secondary sources, that is biomass burning. Hence, it may be concluded that the difference in values at the sites could be due to the variation in primary and secondary sources of carbonaceous aerosol at different sites.

### 3.1 Relationship between OC and EC

The origin of carbonaceous aerosol can be estimated on the basis of the relationship between OC and EC<sup>32,33</sup> as shown by the regression between the OC and EC concentrations in Fig. 3. At urban residential site, relatively strong OC-EC correlations were observed. Atmospheric elemental carbon is from primary anthropogenic sources and is not formed by reactions involving gaseous hydrocarbon precursors in the atmosphere. Organic carbon may be emitted as primary particles directly from sources, but secondary

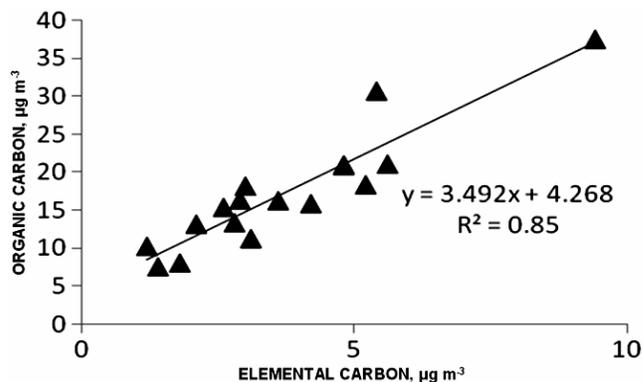


Fig. 3 — Regression between OC and EC at urban residential site

organics can also be formed in the atmosphere from the low vapour pressure products of atmospheric chemical reactions. Regression equation between OC and EC is obtained and shown in Fig. 3. The high coefficient of determination ( $r^2$ ) of OC and EC ( $r^2 > 0.85$ ) implies similar emission sources (i.e. vehicle exhaust) contributing to ambient carbonaceous particles. High OC-EC correlations ( $r = 0.92$ ) indicate either common sources or sources of similar strength.

### 3.2 OC/EC ratios

Carbonaceous aerosol represents a mixture of various emission sources (EC and primary OC) and secondary OC formed by atmospheric reaction processes. The ratio of OC to EC concentrations (OC/EC) can, therefore, be used to study the emission and transformation characteristics. Typical emission sources include diesel and gasoline powered vehicular exhaust (OC/EC = 1.0–4.2), wood

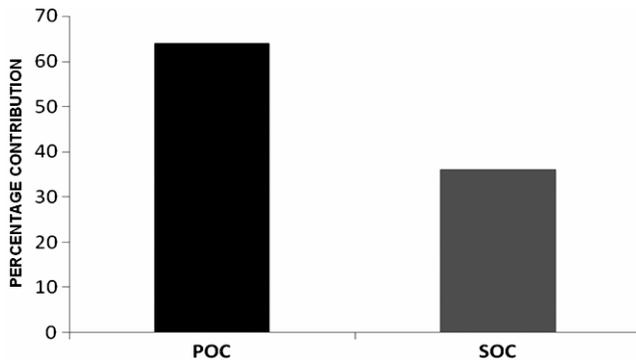


Fig. 4 — Comparison of POC and SOC at urban residential site

combustion (16.8–40.0), residential coal smoke (2.5–10.5), kitchen emissions (32.9–81.6) and biomass burning (7.7), etc.<sup>34-36</sup>

The average OC/EC ratio at the sampling site is between 3.5 and 11.9. An overall average of  $5.4 \pm 1.6$  is observed at urban residential site. Studies have related the OC/EC ratio to secondary organic particle formation<sup>30,37,38</sup>. A primary OC/EC ratio of 2.2 is assumed to indicate the general direction of increasing secondary organic particle concentrations<sup>31,37</sup>. However, the average OC/EC ratio obtained in this study for  $PM_{2.5}$  is greater than 2. This suggests that in addition to vehicular exhaust emission, there is formation of secondary organic carbon because of the gas/particle conversion of gaseous hydrocarbon precursors.

### 3.3 Estimate for secondary organic carbon concentration

Minimum OC/EC ratio method is used to estimate the secondary organic carbon ( $OC_{sec}$  or SOC) formation<sup>32,39</sup> using following equation:

$$OC_{primary} = (OC/EC)_{min} * EC$$

where,  $OC_{primary}$ , is the primary OC or POC; and  $(OC/EC)_{min}$ , the minimum ratio observed. It has been suggested that particle samples with minimum OC/EC ratios contain almost exclusively primary carbonaceous compounds<sup>40</sup>. Therefore, the concentration of secondary organic carbon (SOC) can be calculated from the equation:

$$OC_{secondary} = OC_{total} - OC_{primary}$$

where,  $OC_{secondary}$ , is the secondary OC (SOC);  $OC_{total}$ , the total OC (TOC); and  $(OC/EC)_{min}$ , the minimum ratio observed. The  $(OC/EC)_{min}$  of 3.5 have been observed for this site and used in the calculation<sup>39</sup>.

At this site, POC is  $12.9 \pm 8.4 \mu g m^{-3}$ , while SOC is  $7.2 \pm 6.3 \mu g m^{-3}$ . At the urban residential site, POC and

SOC accounted for 64.2% and 35.8% of TOC as shown in Fig. 4. The percentage of estimated SOC at urban residential site is in agreement with percentages in urban areas found in other studies<sup>32,39,40</sup>.

## 4 Conclusions

Carbonaceous aerosols are analyzed at urban residential site Khandari, Agra. During the study period, the 24-h  $PM_{2.5}$  concentration ranged  $55.3 \pm 17.4 \mu g m^{-3}$ . OC concentrations in  $PM_{2.5}$  ranged 7.6 - 37.5  $\mu g m^{-3}$  with an average of  $18.2 \pm 6.4 \mu g m^{-3}$ , while EC concentrations ranged 1.2 - 9.4 at the urban residential site with an average of  $3.2 \pm 1.3 \mu g m^{-3}$ . OC/EC ratio at the site is between 3.5 and 11.9. At this site, although both primary sources and secondary sources contribute to total carbonaceous aerosol, primary sources dominate and are more than 1.8 times secondary sources.

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