Insights into the Regional Transport and Local Formation of Secondary Organic Aerosol in Delhi, India

Himadri Sekhar Bhowmik¹, Sachchida Nand Tripathi²*, Ravi Sahu¹, Ashutosh Kumar Shukla³, Vipul Lalchandani¹, Shamitaksha Talukdar¹, Nidhi Tripathi³, Lokesh Sahu³

¹ Department of Civil Engineering, Indian Institute of Technology Kanpur, Kanpur, India
² Department of Civil Engineering and Centre for Environmental Science and Engineering, Indian Institute of Technology Kanpur, Kanpur, India
³ Space and Atmospheric Sciences Division, Physical Research Laboratory, Ahmedabad, India

ABSTRACT

The organic carbon (OC) and elemental carbon (EC) content were measured from PM$_{2.5}$ quartz filter substrates collected over 24 hours in Faridabad, a heavily polluted city in Delhi national capital region (NCR), during January–February 2018. In this study, the secondary fraction of OC (SOC) was calculated using the “EC-tracer method”, and subsequently, secondary organic aerosol (SOA) was calculated. Two typical episodes were observed where SOA was elevated for multiple days. These occurrences were investigated for aged SOA formation through multi-day regional transport and locally generated SOA using ozone (O$_3$), carbon monoxide (CO), nitric oxide (NO), and volatile organic compounds (VOCs) as supplementary data. Higher OC/EC ratios during these episodes could be due to a strong primary source, and this possibility cannot be ruled out. However, similar sulfate dynamics with SOA consolidated the multi-day regional transport of SOA during the elevated SOA episodes. The moderate resolution imaging spectroradiometer (MODIS) fire count, planetary boundary layer height (PBLH) data, temperature, and RH were examined to strengthen the hypothesis that the elevated SOA days were due to multi-day regional transport of SOA and not locally formed. The ratios of volatile organic compounds (VOC) were analyzed to understand the origins of VOCs and also to explore the regional transformation and local formation of SOA. The toluene/benzene (T/B) and isoprene/benzene (I/B) ratios were observed during the multi-day elevated SOA episodes to distinguish the regional and local sources of VOCs. Concentration weighted trajectory (CWT) plots also confirmed our hypothesis. This study suggests that the higher SOC fraction during those episodes was most likely due to regional or aged SOA formation. Also, multi-day regional transport contributes to the higher and elevated SOA episodes during January–February 2018.

Keywords: OC, EC, Local SOA, Regional SOA

1 INTRODUCTION

Carbonaceous aerosol constitutes a significant fraction (10–70%) of PM$_{2.5}$ in the urban atmosphere (Na et al., 2004; Martinez et al., 2012; Chen et al., 2019; Bhowmik et al., 2020) which alters the earth radiation balance, reduces visibility, and causes health issues followed by premature death (IPCC, 2001; U.S. EPA, 2009). Carbonaceous materials in particulate matter (PM) can be divided into two main components, organic carbon (OC) and elemental carbon (EC). OC can be either emitted directly in the particulate phase (primary) or can be formed in the atmosphere from gas-to-particle conversion process (secondary) of the atmospheric low and semi-volatile organic compounds (SVOC) and/or from the absorption of the reactive organic gases onto the particle surface (Turpin et al., 2000). EC is mainly primary and emitted during incomplete combustion (Chow et al., 2004).
The composition of secondary organic aerosol (SOA) is very complex and changes according to the precursor gases. SOA contributes a significant fraction (50–90%) of the organic aerosol (OA) in urban areas (Feng et al., 2013; Bhowmik et al., 2020). The direct quantification of the secondary fraction of OC (SOC) is difficult to achieve due to the complexity of its production and formation pathways. However, SOA concentration has been quantified by some indirect methods using carbon isotope composition (López-Veneroni, 2009), chemical transport models (Zeng and Wang, 2011), receptor models, and the EC tracer method (Strader et al., 1999; Cabada et al., 2004; Bhowmik et al., 2020) in which SOC has been estimated by subtracting the primary contributions from measured OC (Schauer and Cass, 2000).

Previous studies have shown that Indo Gangetic Plain (IGP) is a polluted region with high concentration of carbonaceous aerosol (Rengarajan et al., 2007; Bhowmik et al., 2020; Nagar et al., 2017). Especially during winter, the IGP faces massive air pollution episodes along with extreme haze and smog formation and associated health issues (Bhowmik et al., 2020). However, the contributions of primary and secondary OC are largely unknown in IGP. Our previous paper Bhowmik et al. (2020) showed that IGP experiences high SOA production (~80–138 µg m⁻³) during post-monsoon (October–November) and winter (December–February) and is 30–60% of OA as estimated using the “EC tracer method”. This higher SOA contribution could be due to strong primary emissions and/or multi-day SOA formation and regional transport (Polidori et al., 2006). Concentration dynamics, multiple meteorological effects, and atmospheric stagnation events can be simultaneously used for better understanding the local and regional origin of SOA.

To the best of our knowledge, there are very few studies focusing on SOA in IGP, and no such research is focused on investigating local and multi-day regional transport of SOA. The development of appropriate control policies for ambient PM requires better understanding of the contribution and origin of SOA as it constitutes a significant fraction of PM. This paper presents a comprehensive investigation of the local and multi-day regional origin of SOA in Faridabad, India, an urban area in IGP and one of the most polluted urban areas (annual avg PM2.5 ~140 µg m⁻³) in the world (World Economic Forum). We have used detailed data of OC, EC, and O₃, while CO, NOₓ, NO, SO₂, and VOC data along with MODIS fire maps and HYSPLIT air mass back trajectory as supplementary.

2 METHODS

2.1 Site Description

Faridabad, an urban area in Haryana and a part of the National Capital Region (NCR) of Delhi, India, is among the top 10 most polluted urban areas globally (World Economic Forum). With rapid increase in numbers of small and large-scale industries, construction of residential colonies and regional transport of large-scale biomass burning emissions from upwind part of Haryana, Faridabad faces heavy air pollution episodes especially during post-monsoon (October–November) and winter (December–February). PM2.5 samples were collected with a high-volume sampler (1.13 m³ min⁻¹) placed on the second floor of the C block building (~7 m above the ground level) at Manav Rachna International Institute of Research and Studies (MRIIRS), Faridabad (28.45°N, 77.28°E ~278 m above MSL) (Fig. 1) during January–February 2018. A proton transfer reaction-time of flight-mass spectrometer (PTR ToF-MS) and NO, CO, SO₂, and O₃ gas analyzers were also deployed at the same building with their inlets kept nearby high volume sampler. The sampling site was an institute campus and very close to heavy trafficked road (~50 m away).

2.2 Sampling Details

Using a high volume sampler (TISCH, USA), 52 daily PM2.5 samples, including 4 blanks were collected on 8 × 12 inches Whatman quartz filter substrates from 4th January 2018 to 28th February 2018 during winter. Blank filter papers were placed in the sampler without running the pump to collect the field blanks. The sampled filters were then transferred to the Centre for Environmental Science and Engineering (CESE), Indian Institute of Technology (IIT) Kanpur, where they were stored at~20°C in the deep freezer prior to the analysis. The samples were analyzed using an EC-OC carbon analyzer for EC and OC. Hourly gas data (SO₂, O₃, CO, NO) and high-resolution (hourly) VOC data were obtained using gas analyzers and a PTR-ToF-MS, respectively. Details about the instruments can be found in the supplementary material.
Fig. 1. Sampling location with different emission sources. The red border line shows border of the National Capital Territory Delhi.

2.3 Secondary Organic Aerosol (SOA) Estimation

The ambient OC measured by OC-EC analyzer is defined as total OC (OC$_{\text{total}}$), and can be divided into primary (OC$_{\text{pri}}$ or POC) and secondary OC (OC$_{\text{sec}}$ or SOC) as described in Eq. (1):

$$OC_{\text{total}} = OC_{\text{pri}} + OC_{\text{sec}}$$  (1)

where, OC$_{\text{pri}}$ is largely emitted by combustion sources and a small or negligible fraction from non-combustion sources (e.g., biogenic and dust resuspension) as well. The non-combustion fraction of POC is the intercept in the EC and OC scatter plot (Strader et al., 1999). Hence, POC can be defined as in Eq. (2):

$$OC_{\text{pri}} = OC_{\text{comb}} + OC_{\text{non-comb}}$$  (2)

Considering primary OC/EC ratio is largely attributed to combustion sources in the urban areas, OC$_{\text{comb}}$ can be defined as in Eq. (3):

$$OC_{\text{comb}} = (OC/EC)_{\text{pri}} \times EC$$  (3)
where, EC is the ambient EC measured by OC-EC analyzer. Finally, assuming the primary OC/EC ratio remains constant, primary (OC$_{pri}$ or POC) and secondary OC (OC$_{sec}$ or SOC) can be defined as in Eqs. (4) and (5):

\[
OC_{pri} = (OC/EC)_{pri} \times EC + OC_{non-comb} \tag{4}
\]

\[
OC_{sec} = OC_{total} - OC_{pri} \tag{5}
\]

This indirect approach of measuring SOC is termed as “EC-tracer method” (Cabada et al., 2004). Estimation of SOC by “EC-tracer method” highly relies on (OC/EC)$_{pri}$ ratio. There are quite a few theories to determine the (OC/EC)$_{pri}$ ratio discussed in our previous study (Bhowmik et al., 2020). In Bhowmik et al. (2020), we calculated SOC incorporating Pio et al. (2011) approach. The (OC/EC)$_{pri}$ ratio was estimated using the lowest (OC/EC) ratios from the linear regression on a portion (~15%) of the whole data set. The calculated SOC from “EC-tracer method” was multiplied by a constant (OM/OC) to get SOA mass concentration. Though a value of 1.2–2.1 has been reported for urban areas in the available literature (Turpin and Lim, 2001; Rengarajan et al., 2007), in Bhowmik et al. (2020), a value of ~2 was used from the average OM/OC ratio of 2.0 ± 0.19 (Lalchandani et al., 2021) which was derived from the aerodyne high-resolution time of flight aerosol mass spectrometer (HR-ToF-AMS) deployed at the same site during January–March, 2018. A detailed discussion on the other approaches to calculate (OC/EC)$_{pri}$ and the calculation of SOC and SOA can be found in our previous study (Bhowmik et al., 2020).

3 RESULTS AND DISCUSSIONS

During January to February 2018, two episodes were noticed with multi-day elevated SOA and OC without any increase in the primary tracers-NO and EC and are denoted as Episodes A and B in Fig. 2. Episode A occurred during 22nd to 26th January, 2018 and Episode B from 22nd to 24th February, 2018. During the entire sampling period, SOA formation closely tracked the measured O$_3$ concentration except on Episodes A and B. During Episodes A and B, SOA remained elevated for a period of 5 and 3 days, respectively. In contrast, during that period, O$_3$ concentrations were typically lower than the other days (Fig. 2). The hourly gas data were averaged over 24 hours as per the sampling schedule i.e., from 6.30 am (Local Time) morning to next day 6.30 am morning. The low O$_3$ concentrations during Episodes A and B were insufficient to provide ample oxidant concentration to form SOA locally, suggesting either air mass transport from upwind or downward mixing from aloft. On 11th February 2018, O$_3$ concentration was as high as ~31 ppb. On previous days, O$_3$ decreased mostly due to NO scavenging, and this might be downmixing of O$_3$ from the aloft rather than photochemical formation (Polidori et al., 2006).

![Fig. 2. SOA, SO$_4^{2-}$ and ozone concentrations with 24 hours averaged PBLHs during January–February 2018.](image-url)
Polidori et al. (2006) also suggested that when SOA remains elevated over several days, it is most likely that it has been transported regionally or might have transformed through multi-day transport. When transport distances are large and photochemical reactions are possible, regional SOA formation is predominantly vertical transport from aloft. Corsmeier et al. (1997) showed that O3 could be elevated in the lower atmosphere by vertical transport from aloft and would cause the elevation of SOA in the lower atmosphere by taking down the SOA from the upper atmosphere to the lower atmosphere up to ground level. O3 from aloft can also react with NOx and form NO3 radicals. Isoprene reacts with NO3 radical, which leads to the formation of SOA (Claeys et al., 2004; Dommen et al., 2006). Moreover, the vapor pressure of the secondary species increases with an increase in temperature, which drives a higher percentage of total mass into the gas phase and less fraction into the aerosol phase (Gray et al., 1986). Thus, the lower nighttime temperature (4°C–10°C) during January–February favored the partitioning of oxidant products into the particle phase which contributed to an increase in SOA concentration.

On 30th January and 11th February 2018, the difference between OC and EC is lesser (p value < 0.05). Interestingly, during Episodes A and B, the variation in EC remained more or less the same as observed in the previous days while OC increased dramatically (Fig. 3). During Episodes A and B, the average SOC was ~70% and ~62% of OCtotal, respectively, whereas, on 30th January and 11th February 2018, when the EC was least the fraction was ~35% and ~37%, respectively. The SOC at a given space and time can be influenced by freshly formed SOA by photochemical reactions locally or aged SOA accumulated on previous days or transported regionally or a combination of both (Lim and Turpin, 2002). Pollutants can be transported vast distances from upwind locations or can be downmixed from aloft under favorable photochemical activity. Transport of vertical flux from aloft containing aged carbon mass fraction contributes to ground-level SOC concentrations (Lim and Turpin, 2002). Hence it is most likely that the additional OC is SOC. The higher OC/EC ratio (~9–10) during Episodes A and B suggest either regional secondary formation or could be due to a strong primary source of high OC/EC ratio. OC/EC ratios can be as high as ~10–16 for open biomass burning (Bhowmik et al., 2020; Andreae and Merlet, 2001; Ram et al., 2010; Sandradewi et al., 2008). Our previous study (Bhowmik et al., 2020) also showed that the pollution in NCR was influenced by mass-scale biomass burning from adjoining upwind states of Punjab and Haryana during winter and post-monsoon in 2018. The latter possibility cannot be ruled out since the aged or regionally transported air mass has a higher OC/EC ratio than the local emissions (Lim and Turpin, 2002). Considering that NO and CO can be used as local and regional combustion tracers (Polidori et al., 2006), it has been observed that during periods A and B, OC drastically increased (p value < 0.05) without any major increase in the primary tracers EC and NO, whereas CO increased (p value < 0.05) during the considered periods (Fig. 4). Interestingly, on 30th January, CO and NO decreased and on 11th February, both increased simultaneously.

As CO has a high lifetime in the atmosphere, it can act as a primary as well as a regional tracer. The increase in CO during Episodes A and B without any significant increase in NO could be due to the regional transport of CO. Whereas, the simultaneous decrease and increase in CO and NO
on 30th January and 11th February were possibly due to the similar or common origin of sources (primary combustion). NASA’s MODIS (Moderate Resolution Imaging Spectroradiometer) fire maps have been observed during these periods to determine whether CO is transformed regionally or formed locally. The OC/EC ratio along with O₃, CO, and NO dynamics on 30th January and 11th February, suggested that SOA could have been produced locally. From FIRMS (Fire Information for Resource Management System) fire count maps, it was observed that on 22nd January (Episode A) and 24th February (Episode B), the fire counts were less (p value < 0.05) as compared to 30th January (see supplementary Fig. S2). Hence, with the least count of open burning, it was impossible to produce more primary CO on 22nd January and 24th February than that on 30th January. On 11th February, although less fire count was observed, the sampling site coincided with one hotspot, which could be the possible reason for locally formed CO. Thus, during Episodes A and B, the contribution from regional CO was more likely. The increase in peaks of CO during Episodes A and B supported the regional origin of the additional OC.

Sulphates can be rapidly formed locally from SO₂ via aqueous-phase reactions with transition metal ions, H₂O₂ or NOₓ (Harris et al., 2013) and progressively via gas-phase reactions with OH radicals (Atkinson and Arey, 2003). The average RH during Episodes A and B were 39.7% and 40.2% RH, which are below the deliquescence RH (DRH) of inorganic salts (e.g., DRH for (NH₄)₂SO₄ is 80% RH at 25°C) (Lim et al., 2020). This suggested that the elevated sulphate during Episodes A and B was more regional than local. The high OC/EC ratios during these two episodes could have occurred due to the influence of strong primary sources. Hence, the possibility that SOA was freshly formed by photochemical reactions locally in that period could not be completely ruled out. However, the SOA and SO₄²⁻ dynamics supported the regional secondary formation (Fig. 2).

SOA could be elevated during Episodes A and B, due to lower planetary boundary layer height (PBLH). SOA variation with PBLH was observed to rule out the effect of meteorological parameters on elevated SOA periods i.e., A and B. The PBLH data were retrieved from the Global Data Assimilation System (GDAS) (ftp://arftp.arlhq.noaa.gov/pub/archives/gdas1) files with a 1° × 1° resolution on every three-hour interval. Three hourly PBLH data were averaged to the filter sampling period of 24 hours. The 24 hours averaged PBLHs remained more or less the same (800–900 m) throughout January (Fig. 2) and hence elevated SOA in Episode A should not be associated with the PBLH. During 16th to 20th February, the average PBLH was ~1300 m and SOA was as low as 15 µg m⁻³. During Episode B, the 24 hours averaged PBLHs were too high (1400–2200 m) to accumulate locally formed SOA. SOA formation also depends on temperature (T) (Bowman and Karamalegos, 2002) and relative humidity (RH) (Seinfeld et al., 2001). The average temperature and RH during the sampling period were 14.89 ± 1.68°C and 34.95 ± 4.84% respectively (see Fig. S1 in the supplementary material). It was observed that SOA yield is significantly lower under dry conditions and at lower temperatures (Mentel et al., 2008). Hence the multi-day elevation in SOA during Episodes A and B could not possibly be local in the less RH (39.7% and 40.2% RH during Episodes A and B) and lower temperature (13.83 and 17.52°C during Episodes A and B) and it was more likely to be transported from upwind or aloft.

**Fig. 4.** Daily-averaged CO and NO variations during the sampling period in Faridabad.
The regional formation of SOA very much depends on a complex way on its precursors and oxidants. VOCs are the precursors of SOA. The oxidizing agents (OH radical, O3, and NO3) in the atmosphere oxidize both biogenic and anthropogenic VOCs. The SVOCs and nonvolatile products nucleate to form new particles or condense onto particulate matters (Kleindienst et al., 2007). The diagnostic ratio of toluene/benzene (T/B) is typically used to infer the VOC sources. Benzene serves as an efficient tracer of anthropogenic emissions as its lifetime in the atmosphere is long enough to preserve the identity of VOC sources during their transport. A low T/B (< 2) ratio indicates that the pollutants have a strong influence of vehicular emissions (Yurdakul et al., 2013; Elbir et al., 2007; Oğuz et al., 2003), whereas T/B ratio greater than 2 infers sources other than vehicular emissions such as regional sources and T/B > 10 or higher indicates strong influence of industrial facilities nearby. To support the hypothesis that during Episodes A and D, elevated SOA are regional SOA, T/B ratio during these periods was observed. The hourly VOC data were averaged to 24 hours. During Episodes A and D, T/B ratios were more than 2, indicating a strong influence from regional sources. During Episodes A and B, T/B ratios were more than 2, indicating a strong influence from regional sources. On 30th January and 11th February, T/B ratios were lesser (~1.48 and 1.2), indicating pollution from the local vehicular emissions (Fig. 5).

Isoprene is mainly generated from biogenic sources and some anthropogenic sources. Isoprene/Benzene (I/B) ratio showed the opposite trend to T/B ratio during January–February 2018 in Faridabad. During Episodes A and B, I/B ratio was lesser than 1, which indicated less influenced by local emissions. On 30th January and 11th February, I/B ratio increased to ~1.5 indicating a strong impact from local anthropogenic emissions. Though the ratios (T/B and I/B) during Episodes A and B suggested local or regional influence, the ratios were very borderline. Hence, further investigation is required.

Back trajectories can be used to show the origin and transport path of a particular pollutant of polluted air mass at a given location over a specified time period. Back trajectory analysis was done using the PC-version of HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory, v4.1) (Draxler et al., 2020). The calculated trajectories were weighted with the calculated SOA concentration from the EC-tracer method using the Concentration Weighted Trajectory (CWT) model to track the air parcels causing change in their concentrations at the receptor site in an Igor-based user interface – ZeFir (Petit et al., 2017). The CWTs were calculated using the Eq. (6) specified in (Hsu et al., 2003) as-

\[
CWT_{ij} = \frac{1}{N} \sum_{k=1}^{N} C_k T_{jk}
\]

where CWT\(_{ij}\) is the average weighted concentration in \(i\) latitude and \(j\) longitude in \(\mu \text{g m}^{-3}\). \(C_k\) is the measured specie concentration of the arriving trajectory \(k\), \(N\) is the total number of trajectories,
and $T_{ijk}$ is the residence time of the trajectory $k$ within the $ij$th cell. Two CWT plots were made for Episodes A and B (Fig. 6).

The CWT plots for Episodes A and B (Figs. 6(a) and 6(b)) clearly shows the regional transport of SOA from north-west direction.

4 CONCLUSIONS

During January–February 2018, two pollution Episodes (A and B) were observed which were hypothesized as multi-day regional transport of SOA or aged SOA. Other two events on 30th January and 11th February observed in this study were taken as references for locally-formed SOA or freshly-formed SOA. Episodes A and B were characterized by multi-day elevated SOA and OC without any increase in NO and EC. During these episodes, $O_3$ concentrations were lower than past days and hence, were insufficient to oxidize SOA locally. The average SOC during Episodes A and B was > 50% of measured OC whereas, on 30th January and 11th February, the fraction was < 40%. The additional OC without any increment in the primary tracers (EC & NO) was most likely the SOC. Interestingly, the increment in CO during Episodes A and B created ambiguity in our hypothesis. While CO can act as a regional tracer parallel with primary tracer, NASA’s MODIS fire maps have been observed during these episodes. It was observed using FIRMS that the increase in CO during Episodes A and B was possibly due to transport of regional air masses rather than getting emitted locally. Although the possibility of higher OC/EC ratios due to strong primary source during Episodes A and B cannot be completely ruled out, the close track of $SO_4^{2-}$ with SOA during these episodes complements the regional transformation of SOA. The effect of meteorology was ruled out by observing the PBLHs, RH and temperature during the episodes. As VOCs are the precursors of SOA, the source locations of the VOCs were observed to support the SOA origin indirectly. The higher (> 2) T/B ratios and lesser (< 1) I/B ratios during Episodes A and B indicated strong influence of regional sources. In contrast, the lesser T/B ratio and the higher I/B ratio on 30th January and 11th February indicated the predominant local origin of the sources. Moreover, the CWT plots clearly showed the regional origin of SOA during Episodes A and B. All these findings suggest that SOA could be a substantial contributor to OC and could be produced locally (fresh) or transported regionally (aged). The elevated SOA during Episodes A and B was most likely due to multi-day regional transport whereas, on 30th January and 11th February SOA was fresh or locally formed. However, the complex day and nighttime chemistry as well as the contribution of cloud processing in $SO_4^{2-}$ formation could further lead to regional or local SOA formation.
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DISCLAIMER

The authors declare that they have no conflict of interest.

SUPPLEMENTARY MATERIAL

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