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# Source and Source Region of Carbonaceous Species and Trace Elements in $PM_{10}$ over Delhi, India †

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**Abstract:** This study investigated the carbonaceous species [elemental carbon (EC), organic carbon (OC), water soluble organic carbon (WSOC)] along with the trace elements (Al, S, Ti, Mn, Fe, Cu, Zn, As, Br, Pb, Cr, F, Cl, Na, K, Mg, Ca, P) in $PM_{10}$ over the megacity Delhi, India (collected from 2015–2019) to address certain significant scientific issues (i.e., what are the directionality or pathway of these emissions; what are the possible emission sources which are distressing the observation site; what are the periodical variations in these emissions; and whether the emissions are local, regional, or trans-boundary). Integration of these problems are addressed using various statistical approaches including potential source areas (PSA) [using hybrid modelling i.e., potential source contribution factor (PSCF)], the conditional bivariate probability function (CBPF), and principal component analysis (PCA). Furthermore, seasonal PSCF and CBPF indicate both local source (highly polluted residential areas, traffic congestions, and industrial emissions) and regional sources (Haryana, Punjab) dominancy during winter and post-monsoon seasons at the receptor site, whereas during summer and monsoon along with local source and the regional, trans-boundaries (Indo-Gangatic plane, Pakistan, Afghanistan, and Bay of Bengal) air parcel patterns also contribute to the aerosol loading at the sites. Moreover, the PCA approach framed four common sources [crustal/road dust (RD), industrial emission (IE), fossil fuel combustion + biomass burning (FCC+ BB), vehicular emission (VE)] with one mixed source over the sampling site of Delhi.

**Keywords:** $PM_{10}$; OC; EC; elements; PSCF; CBPF; PCA; Delhi



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## 1. Introduction

Enhancement of anthropogenic activities in the local and regional regions of the megacity Delhi has resulted in a polluted atmospheric; exposure to such an atmosphere has a great impact on the human health and climate [1]. In addition to regional and local sources' contribution, meteorological parameters such as wind speed and wind direction play a dynamic role in the distribution of pollutants [2,3]. Mass concentration at the downwind regions (low local emissions) are affected by long range transported aerosols [1]. Important studies have been focused on the carbonaceous particles (organic carbon (OC), elemental carbon (EC) and water-soluble organic carbon (WSOC)); such particles disturb the atmospheric chemistry resulting in poor air quality [4]. Organic carbon contains large number of volatile compounds wherein EC is defined by non-volatile compounds and further EC shows a strong light absorbing species [5]. Due to their light weight, ECs have a tendency to travel a long range. Thus, EC can be considered as a metric over receptor sites [6,7]. Biomass burning and incomplete in automobiles leads to ECs, while OC is generated from sources such as gasoline and diesel. On the other hand, WSOC may be classified into hydrophilic (moderately) and hydrophilic (strong) portions [8]. Interestingly,

one can understand the particulate matter pathways, sources, optical, physical and chemical properties by analyzing their concentration and composition size at receptor sites [9,10].

Delhi is one of the metropolitan megacities of Asia where urbanization, industrialization, and economic growth are very rapid. It is surrounded by the Indo-Gangetic plain (IGP) in the East, the Thar desert in the West, the Himalayas in the North, and the hot plains in the South region [11]. It is important to observe the atmosphere of such an urbanized city.

## 2. Methodology

### 2.1. Observation Site

Measurements of $PM_{10}$ were carried out for five years (2015–2019) at the rooftop of the CSIR-National Physical Laboratory, Delhi (28°38′ N, 77°10′ E) at 10 m (AGL). This observation site reflects an urban background including walled traffic roads, junction points, and agriculture and residential sectors with small scale industries in the north-west [11].

### 2.2. Sample Collection

$PM_{10}$ samples ($n$ = 452) were collected on pre-baked PallFlex tissue quartz filters using a respirable dust sampler (average flow rate 1.13 $m^3$ $min^{-1}$; Model: AAS 212 NL, Make: M/s. Ecotech, India) installed at the rooftop of CSIR-NPL, New Delhi from January 2015–December 2019. The sampler used in this study was periodically calibrated using National Standards [11]. The meteorological parameters were [such as wind speed (WS, accuracy: ±2%), wind direction (WD, accuracy: ±3°), temperature (T, accuracy: ±1°), and relative humidity (RH accuracy: ±2%)] also collected during the $PM_{10}$ sampling. National ambient air quality standards (NAAQS) protocol by the Central Pollution Control Board (CPCB), India, was accepted for sampling throughout. Filters were properly desiccated, stored (at −20 °C), and weighted before and after the sampling so to get the mass of collected $PM_{10}$. The gravimetrical method (using microbalance: M/s. Sartorius, resolution: ±10 µg) was applied to calculate the concentration of $PM_{10}$. The samples and their concentrations were further investigated for the study of organic carbon (OC), elemental carbon (EC), water soluble organic carbon (WSOC), trace elements, trajectories, potential source contributor factor (PSCF), and conditional bivariate probability function (CBPF).

### 2.3. Analysis (OC, EC, WSOC, Trace Metals)

$PM_{10}$ samples along with filter blank samples were punched to an area 0.536 $cm^2$ and carried out OC and EC analysis using thermal/optical carbon analyzer (DRI Model 2001A, Atmoslytic Inc., Calabasas, CA, USA) working on the principle of preferential oxidation (Improve-A protocol) [12]. The details concerning the analytical procedure for OC and EC have been mentioned in [13]. For WSOC analysis, the TOC-LCPH/CPN (M/s. Shimadzu, Kyoto, Japan) total organic carbon analyzer was used. Operational calibration was conducted following standard protocol. Instrumentation details can be found in [14,15]. Assembly of WD-XRF (wave length dispersive X-ray fluorescence spectrometer) was used for the analysis of elements in $PM_{10}$. This setup was supposed to quantify elements ranging from B to U. With respect to intensity, error blank filters were also analyzed. Details are available in [16].

### 2.4. Potential Component Analysis (PCA)

Statistical multivariate tools based on true eigen vector (PCA) was used for source apportionment of $PM_{10}$. PCA is a dimensionality- reduction statistical tool; it reduces the large data set dimension to a small dimension which still has the information of a large data set [17–19]. Extraction is conducted by forming new orthogonal variables as principal components, thereby achieving a similar pattern between observations and variables [19]. Steps are followed by standardization and then orthogonal transformation with Varimax rotation.

### *2.5. Conditional Bivariate Probability Function (CBPF)*

Including the meteorological parameter (ws and wd) along with the pollutants CBPF discriminate the sources and the directionality. Mathematically defined as

$$\text{CBPF} = (m_{\Delta\theta,\,\Delta u}/n_{\,\Delta\theta,\,\Delta u}) \text{ condition: } C \geq x \quad (1)$$

The numerator represents the number of samples in wind sector ($\Delta\theta$) with wind speed ($\Delta u$), and the denominator represents the total number of samples. C is measured concentrations and x is the threshold criterion.

### *2.6. Trajectory Analysis*

Considering the influences of transported pollutants, five-days isentropic backward trajectories arriving at study site, Delhi (28°38′ N, 77°10′ E) at 500 m above ground level (AGL) (including the winds in the lower boundary region and neglect surface frictions) were calculated every 5 h using HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model [20]. Backward trajectory represents the range of particulate matter. Further trajectory is the time integration of particle position vector in space as particles are assumed to follow the wind.

Mathematically definition,

$$\{P(t + \Delta t) = P(t) + 0.5\{V(P,t) + V(P't + \Delta t)\}\Delta t, \quad (2)$$

As P′(t + Δt) is the first guess position of the particle.
P(t + Δt) is the final position of the particle.
And V(P,t) is the velocity of the particle.

### *2.7. Potential Source Contribution Factor (PSCF)*

PSCF is a statistical approach used to measure the residence time of air parcels for a given geographical area. Depending on the geographical scale the entire geographic region covering the trajectories is divided into a series of grid cells. Analyzing the trajectory pathways PSCF identifies the source regions. Back trajectories from the receptor sites are represented by the segment endpoints. By defining the number of endpoints that fall in the *ij*th cell as $n_{ij}$ and the number of endpoints that corresponds to a $PM_{10}$ concentration above the criterion when arriving at receptor in the same grid cell as $m_{ij}$ [21].

Mathematically

$$\text{PSCF}_{ij} = m_{ij}/n_{ij} \quad (3)$$

One can interpret PSCF as conditional probability by defining the potential contributions of a grid cell. Arbitrary weighted function $W_{ij}$ is multiplied to the PSCF to scale down the uncertainty due to small $n_{ij}$. In the present study domain the grid size is 1.0° × 1.0° further the regions extends from 40° E to 90° E and 10° N to 40° N for all the study sites.

## 3. Result and Discussions

### *3.1. Concentration Profile*

Table 1 contents the annual statistical results of the $PM_{10}$ and the carbonaceous species. 5-year annual (2015–2019) average concentrations for $PM_{10}$ was observed to be 237 ± 104 μg $m^{-3}$ with ranging from 31–733 μg $m^{-3}$. This observed mass concentration exceeds by more than four times of the standard limit (annual: 60 μg $m^{-3}$) defined by NAAQS controlled by central pollution control board (CPCB), India. Analogous scientific results were reported in [16] i.e., 249 ± 103 μgm$^{-3}$, [13] i.e., 191 ± 45 μgm$^{-3}$, [11] i.e., 202 ± 74 μgm$^{-3}$, [22] i.e., 238 ± 106 μgm$^{-3}$, more like [14,18,23,24]. Likewise, EC (6.7 ± 5.2 μgm$^{-3}$) with range (0.9–35.6 μgm$^{-3}$), OC (25.3 ± 14.6 μgm$^{-3}$) with range (4.2–77.6 μgm$^{-3}$) and WSOC (10.6 ± 7.5 μgm$^{-3}$) with range (2.4–56.0 μgm$^{-3}$), respectively. Figure 1 showed a positive correlation between EC vs. OC ($R^2$ = 0.73) and OC vs. WSOC ($R^2$ = 0.51) signifying the same sources of origination (biomass burning and/or

vehicular emissions), moreover, annual OC/EC profile was 4.3 ± 1.6 (range: 1.3–12.6). Furthermore, the diagonal plots in Figure 1 represents the annual box plot (25~75%) for EC, OC and WSOC with the mean and median labels. Noted the seasonal variation of $PM_{10}$ mass concentration as post-monsoon > winter > summer > monsoon. Increasing incineration activities and low boundary layer during dry seasons leads to higher concentration.

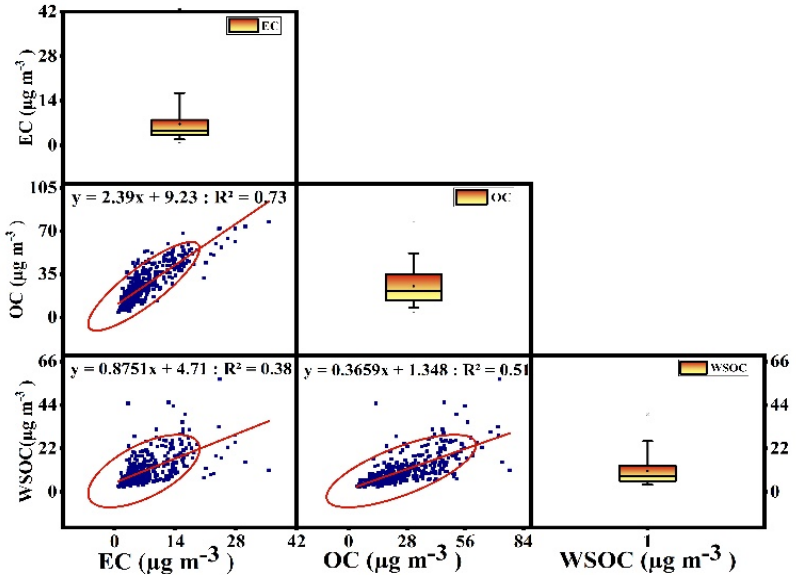


**Figure 1.** Annual scatter plot of EC vs. OC, OC vs. WSOC and the box plots of EC, OC, WSOC.

**Table 1.** Annual average concentrations (average ± SD) carbonaceous species of $PM_{10}$.

| Carbonaceous Species | Concentration | Range |
|---|---|---|
| PM10 (µg $m^{-3}$) | 237 ± 104 | 31–733 |
| EC (µg $m^{-3}$) | 6.7 ± 5.2 | 0.9–35.6 |
| OC (µg $m^{-3}$) | 25.3 ± 14.6 | 4.2–77.6 |
| WSOC (µg $m^{-3}$) | 10.6 ± 7.5 | 2.4–56.0 |
| OC/EC | 4.3 ±1.6 | 1.3–12.6 |

### *3.2. Source Apportionment*

Statistical extraction method PCA with rotation method (Varimax with Kaiser normalization) was applied to 21 chemical parameters of $PM_{10}$ (EC, OC, WSOC, Al, S, P, Mn, Ti, Br, Pb, Zn, Cr, Na, Ca, Fe, Mg, F, K, Cl, Cu, and As) so as to classify different possible sources or factors. Implications of this tool result in five factors (Table 2). With an extracted variance of 18.59% factor-1 was highly loaded with Al, Ti, Mn, P signifying crustal or road dust [16–25]. Pb, Zn, Cr, Na corresponds to the factor-2 loading to $PM_{10}$ with extracted variance 16.56%, suggesting Industrial emission origin [26]. Factor-3, with 15.59% extracted variance attributes to biomass burning plus fossil fuel combustion as there is significant loading of EC, OC, WSOC, K, S [27] with extracted variance of 15. 59%. Factor-4, extracted variance 13.56%, $PM_{10}$ is loaded with Ca, Fe, Mg, F, K attributing to a mix source i.e., biomass burning plus road dust. Factor-5, extracted variance 5.95%, Cu is the dominating load to $PM_{10}$ suggesting vehicular emission origin, brake linings during traffic cogestion emits Cu, thus a good traffic indicator [28].

**Table 2.** Potential component analysis (PCA) of $PM_{10}$ during study period.

| Species | Factor-1 | Factor-2 | Factor-3 | Factor-4 | Factor-5 |
|---|---|---|---|---|---|
| EC | - | 0.161 | 0.833 | 0.148 | - |
| OC | - | 0.154 | 0.876 | 0.118 | - |
| WSOC | - | 0.160 | 0.820 | - | - |
| Al | 0.893 | | 0.104 | 0.110 | 0.111 |
| S | 0.157 | | 0.554 | - | 0.205 |
| P | 0.927 | - | - | - | - |
| Mn | 0.897 | 0.109 | - | - | - |
| Ti | 0.798 | 0.373 | - | 0.160 | - |
| Br | 0.579 | 0.418 | 0.247 | | 0.105 |
| Pb | - | 0.854 | 0.159 | - | - |
| Zn | 0.129 | 0.846 | 0.155 | - | - |
| Cr | 0.436 | 0.823 | - | | |
| Na | - | 0.543 | 0.300 | 0.408 | 0.284 |
| Ca | 0.129 | 0.207 | 0.116 | 0.796 | 0.288 |
| Fe | 0.302 | | - | 0.764 | |
| Mg | - | | - | 0.653 | |
| F | - | 0.177 | 0.166 | 0.608 | |
| K | - | 0.340 | 0.549 | 0.558 | - |
| Cl | | 0.340 | 0.388 | 0.438 | - |
| Cu | 0.188 | - | 0.217 | | 0.805 |
| As | 0.208 | - | 0.263 | - | |
| % Variance | 18.59 | 16.56 | 15.59 | 13.56 | 5.95 |
| CV % | 18.59 | 35.15 | 50.74 | 64.30 | 70.25 |
| Sources | Crustal/RD | IE | BB + FFC | BB + RD | VE |

RD (road dust); IE (industrial emission); BB (biomass-burning); FFC (fossil fuel combustion); VE (vehicular emission); and CV (cumulative variance).

### *3.3. Conditional Bivariate Probability Function (CBPF)*

To stimulate the local source regions, CBPF was programmed in the present study. Figure 2 is the profile for CBPF (for 75% i.e., 318), where pollutant ($PM_{10}$) was computed along with the meteorological parameters (ws and wd). The radial pattern attributes the ws, annual $PM_{10}$ concentration values > 75th percentile of total observations to local regions with wind speed (0.5–1.5 m/s). The local region emissions could be from traffic, industrial emission and biomass burning, as the location is walled with the traffic junctions and residential area i.e., Patel-Nagar, Shadipur, Rajandar traffic junction in the north-west and north-east direction, including small scale industries in the north-west direction.

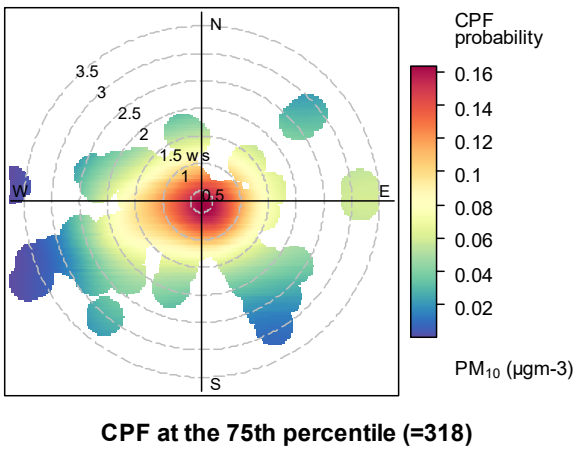


**Figure 2.** Conditional bivariate probability function (CBPF) plot for the observation site.

### *3.4. Trajectory and Potential Source Contribution Factor (PSCF)*

Figure 3a reflects the air parcel pathways to the receptor site. The air parcel followed the flow patter from regional region IGP in the north-east, Haryana, Punjab etc., in the

north-west, Gujarat, Rajasthan in the south-west, including the trans-boundary Pakistan, Afganistan, Arabian sea, Bay of Bengal, Bangladesh etc. Furthermore, PSCF profile reflects the source contribution from local, regional and trans-boundary, respectively. Beside local contribution, regional contribution dominates the trans-boundary during the dry season (post-monsoon and winter) due to the increasing incineration activities (e.g., crop residual burning) in the region Punjab and Haryana (Figure 3b).

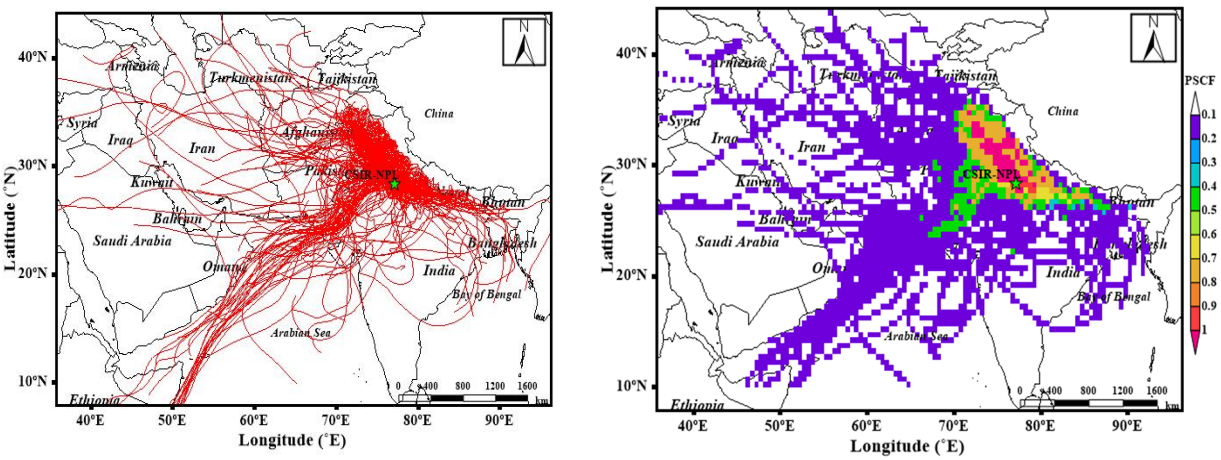


**Figure 3.** (**a**) Air parcels pathway (five days backward trajectories). (**b**) Potential source contribution factor (PSCF), for the observational site CSIR-NPL, New-Delhi.

## 4. Conclusions

Concluding with the high mean concentration of $PM_{10}$ (μg $m^{-3}$), PCA identified five possible sources (crustal/RD, BB + FCC, IE, VE, and mixed source), additionally CBPF identified the local regions contributing to the receptor site, whereas trajectory analysis and PSCF concluded the air parcel flow from IGP, Afghanistan, Pakistan, Arabian sea, Bangladesh, Haryana, Punjab etc., i.e., contribution from regional region along with the trans-boundary in addition to the local regions over the receptor site.

**Author Contributions:** Conceptualization by R.B. and S.K.S.; data collection and analysis were performed by R.B. and M.R.; the first draft was written by R.B. and S.K.S. data interpretation was carried out by R.B., S.K.S., and T.K.M. All authors have read and agreed to the published version of the manuscript.

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**Data Availability Statement:** The datasets developed during the current study are available from the corresponding author on reasonable request.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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