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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: The 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 megacity Delhi, India (collected from 2015-2019) to address the 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; periodical variations in these emissions; whether the emissions are local, regional or trans-boundary). Integration of these problems are addressed using the statistical approaches potential source areas (PSA) [using hybrid modelling i.e., potential source contribution factor (PSCF)], conditional bivariate probability function (CBPF) and principal component analysis (PCA). Further, seasonal PSCF and CBPF indicates local sources (highly polluted residential, 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, Afganistan and Bay of Bengal) air parcel pattern also contribute to the aerosol loading at the site. Moreover, 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 activites in the local and regional regions of mega-city Delhi results in a polluted atmospheric, exposure to such atmosphere has a great impact on the human health and climate [1]. Addition to regional and local source's contribution, meteorological parameters like wind speed and wind direction plays dynamic role in pollutants distribution [2-3]. Mass concentration at the downwind regions (low local emissions) are effected by the long range transported aerosols [1]. Important studies has been focused on the carbonaceous particles (organic carbon OC, elemental carbon EC and water soluble organic carbon (WSOC)) such particle disturb the atmospheric chemistry resulting poor air quality [4]. Organic carbon contains large number of volatile compounds where EC is defined by non-volatile compounds further EC shows a strong light absorbing species [5]. Because of the light weight EC have tendency to travel long range so EC can be consider as a metric over the receptor sites

[6-7]. Biomass burning and incomplete in automobiles leads to the EC. While OC generates from sources like gasoline and diesels. Whereas, WSOC may be classified into hydrophilic (moderately) and hydrophilic (strong) portion [8]. Intrested one understand the particulate matter pathways, sources, optical, physical and chemical properties by analyzing the concentration, composition size at the receptor sites [9-10].

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

2. Methodology

2.1. Observation Site

Measurements of PM_{10} were carried out for five year (2015-2019) at the rooftop of CSIR-National Physical Laboratory, Delhi (28˚38′N, 77°10′E) at 10m (AGL). This observation site reflects an urban background including walled traffic roads and junctions points, agriculture and residential sector 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 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 central pollution control board (CPCB), India, was accepted for sampling throughout. Filters were properly dessicated and stored (at - 20˚ C) and weighted before and after the sampling so to get the mass of collected PM_{10}. Gravimetrical method (using microbalance: M/s. Sartorius, reolution: ± 10 μg) was applied to calculate the concentration of PM_{10}. The samples and their concentrations were further carried 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 a 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 detail about the analytical procedure for OC, EC has been mentioned in [13]. For WSOC analysis TOC-LCPH/CPN, M/s. Shimadzu, Japan Total Organic Carbon Analyzer was runned. Operational calibration was done following standard protocol. Instrumentation details can be found in [14-15]. Assembly of WD-XRF (wave lenghth dispersive X-Ray Fluorescence spectrometer) is used for the analysis of elements in PM_{10}. This setup is supposed to quantify elements ranging (B - U). For intensity error blank filters were also analysed. 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 small dimension which still have the information of large data set [17-18-19]. Extraction is done by forming new orthogonal variables as principle components thus to get the similarity patter between observations and variables [19]. Steps are followed by standardization then orthogonal transformation with Varimax rotation.

2.5. Conditional bivariate probability function (CBPF)

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

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

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

2.6. Trajectory Analysis

Considering the influences of transported pollutants, 5-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 5h 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 defination,

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

As $P'(t+\Delta t)$ is the first guess position of the particle.

$P(t+\Delta t)$ 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. Analysing 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

$$PSCF_{ij} = m_{ij} / n_{ij} \quad (3)$$

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° x 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 annaul (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 [23-18-24-14]. Likewise, EC (6.7 ± 5.2 μgm^{-3}) with range (0.9 – 35.6 μgm^{-3}), OC (25.3 ± 14.6 μgm^{-3}) with rang (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.

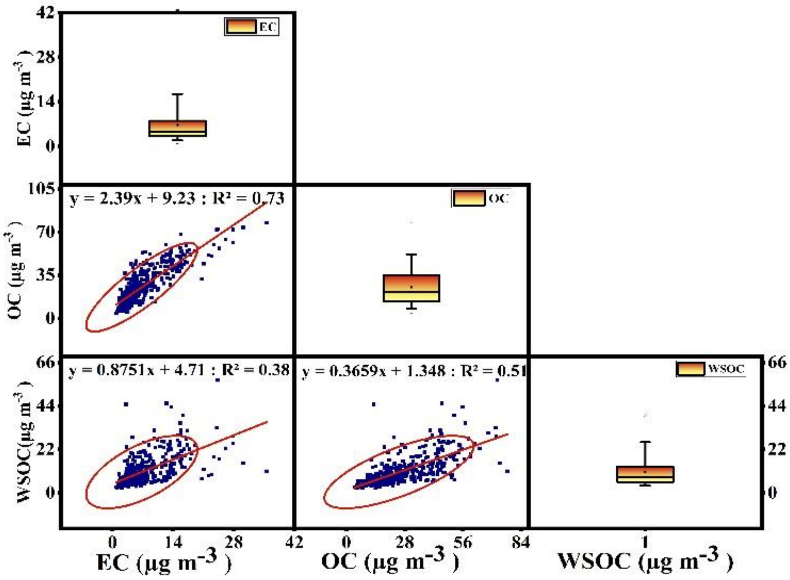


Figure 1. Annual scatter plot of EC *vs* OC, OC *vs* WSOC and the box plots of EC, OC, WSOC

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

Carbonaceous species	Concentration	Range
PM_{10} (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 parametes 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 to classify different possible sources or factors. Implications of the this tool result in five factors (Table 2). With an extracted variance of 18.59% factor-1 was higly 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 emssion 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); 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 defines the ws, annual PM_{10} concentration values > 75th percentile of total observations were attributed 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 place and traffic junction in the north-west and north-east direction including small scale industries in north-west direction.

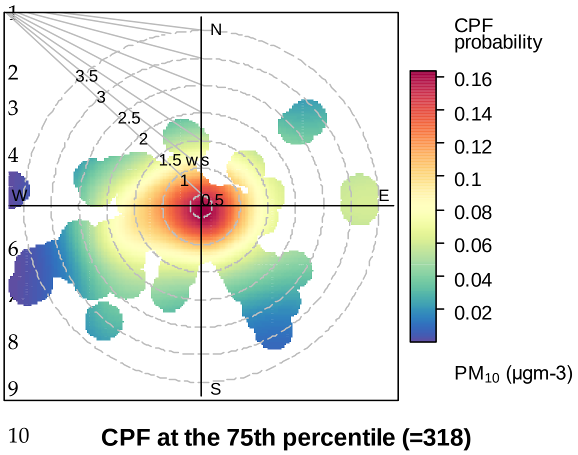
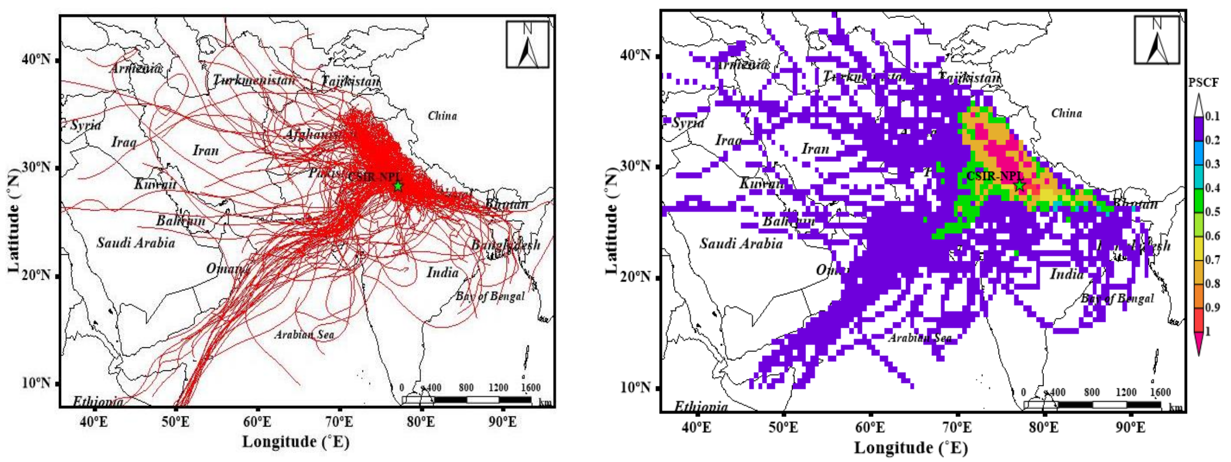


Figure 2. Conditional bivariate probability function (CBPF) plot for the observation site

3.4. Trajectory and potential source contribution factor (PSCF)

Figure 3a. reflects the airparcel pathways to the receptor site. The airparcel 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-boundry Pakistan, Afganistan, Arabian sea, Bay of Bengal, Bangladesh etc.,. Furthermore, PSCF profile reflects the source contribution from local, regional and trans-boundry respectively. Beside local contribution, regional contribution dominates the trans-boundary during the dry season (post-monsoon and winter) because of the increasing incineration activities (e.g., crop residual burning) in the region Punjab and Haryana.



(**a**) (**b**)

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, Afganistan, 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 RB and SKS; Data collection and analysis were performed by RB and MR; the first draft was written by RB and SKS. Data interpretation was carried out by RB, SKS, and TKM. All the authors read and approved the final manuscript.

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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