Environmental Chemistry Letters (2018) 16:1009–1016
https://doi.org/10.1007/s10311-018-0706-y

ORIGINAL PAPER

Air pollutant levels are 12 times higher than guidelines in Varanasi, India. Sources and transfer

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Received: 16 October 2017 / Accepted: 15 January 2018 / Published online: 25 January 2018

Abstract

Air quality in an urban atmosphere is regulated by both local and distant emission sources. For air quality management in urban areas, identification of sources and their relationships with local meteorology and air pollutants are essential. The critical condition of air quality in Indo-Gangetic plain is well known, but lack of data on both local and distant emission sources limits the scope of improving air quality in this region. Concentrations of particulate matter of size lower than 10 µm (PM_{10}) were assessed in the highly urbanized Varanasi city situated in middle Indo-Gangetic plain of India from 2014 to 2017, to identify the distant air pollution sources based on trajectory statistical models and local sources by conditional bivariate probability function. Modifying effects of meteorology and air pollutants on PM_{10} were also explored. Mean PM_{10} concentration for the study period was 244.8 ± 135.8 µg m^{-3}, which was 12 times higher than the WHO annual guideline. Several distinct sources of traffic as the major source of PM_{10} were identified in the city. Trajectory statistical models like cluster analysis, potential source contribution function and concentration-weighted trajectory showed significant contributions from north-west and eastern directions in the transport of polluted air masses to the city. Dew point, wind speed, temperature and ventilation coefficient are the major factors in PM_{10} formation and dispersion.

Keywords Particulate matter · PM_{10} · Indo-Gangetic plain · Trajectory statistical models · Source · Urban

Introduction

Particulate matter (PM) is one of the leading health risk factors apart from its negative effects on plants, ecosystems and climate (Mukherjee and Agrawal 2017; Von Schneidemesser et al. 2015). Most of the urban areas are facing severe air quality problems due to PM (WHO 2016). PM in an urban environment depends upon local and long-distance sources, prevailing meteorology, land use type, the presence of other air pollutants and industrial emissions (Mukherjee and Agrawal 2017). Air quality guidelines, strict emission control and sustainable development measures have reduced PM levels, but lack of source identification and unavailability of long-term measurement data have narrowed the scope of air quality improvements in the developing countries. Indo-Gangetic plain of India is one of the heavily polluted regions of the world (Mukherjee and Agrawal 2017; Sharma et al. 2016). Research publications have pointed out the problem of PM pollution due to anthropogenic emissions and climatic factors, but studies related to long-term measurement data considering both local and distant sources and their relationships with prevailing meteorology and other air pollutants are still limited.

PM is emitted along with other gaseous pollutants, and therefore knowledge of other emission sources along with local sources is important in identifying pollution pattern in an urban environment. Source-receptor modelling is one of the most convenient ways to assess local air quality in relation to both short and distant sources based on local wind sector and back trajectory analysis. Based on the above facts, a 4-year continuous monitoring data was assessed to identify PM_{10} (particulate matter ≤ 10 µm) status, exceedances, local and distant sources, role of meteorology and other air pollutants in modifying PM levels in urban areas of Varanasi city located in middle Indo-Gangetic plain in India, utilizing source-receptor modelling approaches (bivariate conditional probability function for local sources and trajectory

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statistical models, i.e. potential source contribution function and concentration-weighted trajectories for distant sources).

Materials and methods

Study site

For analysis of PM_{10} status and sources, a highly urbanized city of Varanasi was selected. The city is located at 25.3176°N latitude, 82.9739°E longitude and 80.71 m above mean sea level in middle Indo-Gangetic plain in the north Indian state of Uttar Pradesh in India. Varanasi is ranked 29th in urban agglomeration ranking in India with an urbanization rate of 31.16% in 2011 (Census of India 2011). PM monitoring station in Varanasi is located at 25.3505°N latitude and 82.9783°E longitude in the northern sector of the city which is surrounded by heavy commercial and residential areas. The monitoring station is also surrounded by many small roads and two major national highways, 28 and 31 in both south-west and eastern directions from the monitoring site (Fig. 1).

Data collection and analysis

Air quality (PM_{10}, $PM_{2.5}$ [particulate matter ≤ 2.5 µm], SO_2, NO_2, CO and O_3) and meteorological data (wind speed, wind direction, temperature and relative humidity) for the period from July 2014 to May 2017 were downloaded from Central Pollution Control Board public database (cpcb.nic.in). For assessment of the relationship between air quality and other meteorological variables (dew point, rainfall and sunshine hours), data were collected from woundweather.com Web site for Varanasi city and from Banaras Hindu University. Planetary boundary layer height and ventilation coefficient were calculated from global data assimilation system meteorological data. Before the data analysis, data were scrutinized for outliers, normality and inconsistency by frequency analysis.

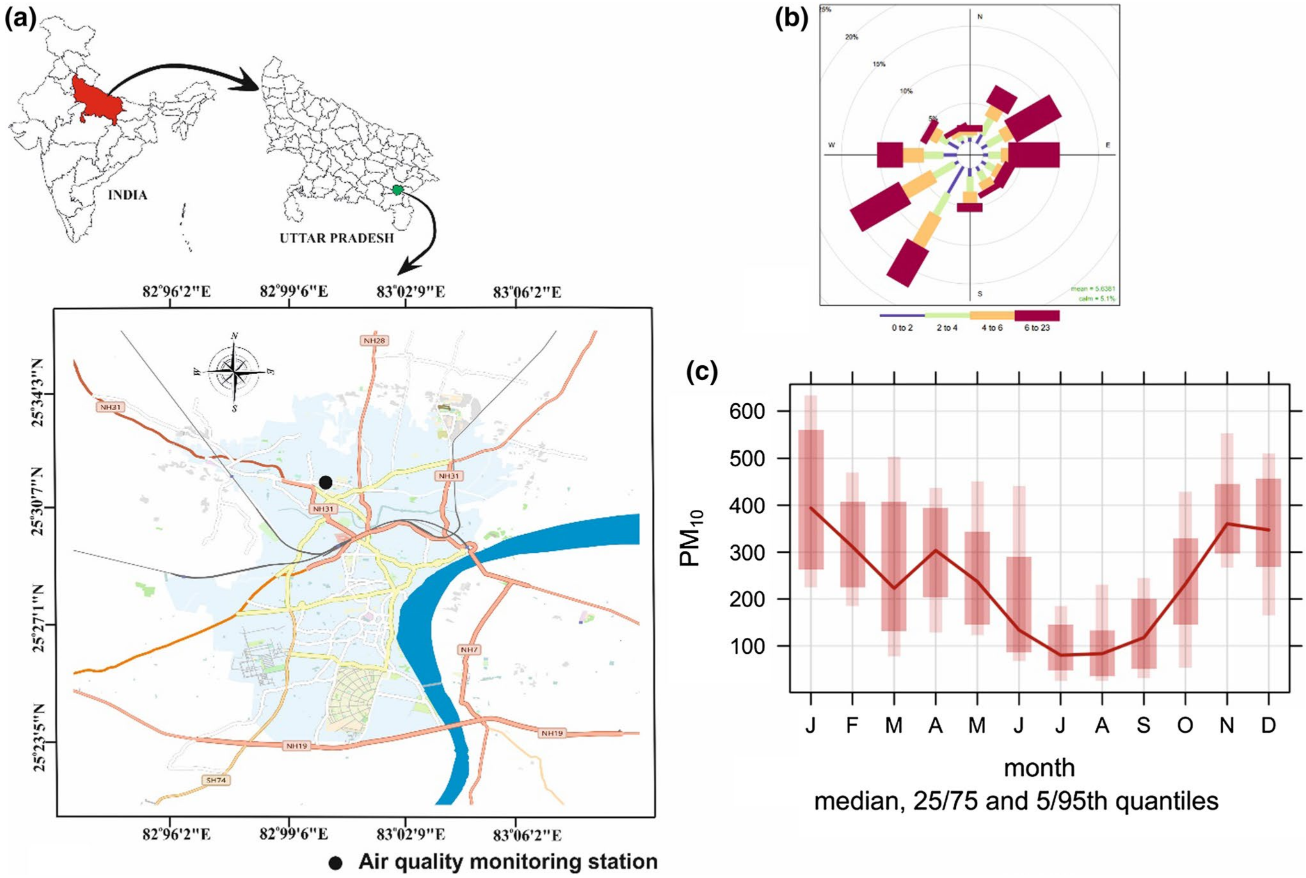


Fig. 1 **a** Map of study area showing Varanasi city and location of air quality monitoring station in the city. **b** Windrose diagram for the study period indicating south-west as major wind direction at high wind speed. **c** Box-and-whisker plot showing monthly variations in PM_{10} (µg m^{-3}) concentrations in Varanasi city during 2014–2017 with higher concentrations in December and January and minimum in July

Source apportionment

Conditional bivariate probability function

Source apportionment of PM_{10} for local and short-range sources was performed using conditional bivariate probability function, which assesses the probability of a wind direction and speed with specific pollutant levels. Pollutant concentration data are divided into cells defined by different ranges of wind direction and speed and calculated by following formula:

$$\mathrm{CBPF}_{\Delta\theta,\Delta U} = (m_{\Delta\theta,\Delta U}/c \geq x)/n_{\Delta\theta,\Delta U}$$

where CBPF represents conditional bivariate probability function, $m_{\Delta\theta,\Delta U}$ represents the number of samples in the wind sector $\Delta\theta$ with wind speed interval ΔU having concentration (c) greater than a threshold value x, $n_{\Delta\theta,\Delta U}$ is the total number of samples in that wind direction–speed interval. More details about the method are described in Uria-Tellaetxe and Carslaw (2014). Conditional bivariate probability function was performed through openair R package (Carslaw and Ropkins 2012) in r statistical software.

Trajectory statistical models

Backward trajectories were run for 96-h duration for the measurement period from July 2014 to May 2017 each day at intervals of 6 h employing the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model 4 (Draxler and Hess 2004) with the global data assimilation system meteorological data as input having a one-degree horizontal resolution at an elevation of 500 m above ground level. Cluster analysis was performed using PC version of HYSPLIT based on spatial variance computation between each endpoint of trajectory.

Potential source contribution function

Potential source contribution function is a receptor model which associates meteorological data with air pollution monitoring data to create probability values for determining areas of potential source contribution. If a source region located at (i, j coordinates) and an air parcel is passing through that location, it is probably above a percentile value of 75 or 90 that pollutant from the source location can be collected and transported via the trajectory to the receptor site and calculated as

$$\mathrm{PSCF}_{ij} = \frac{m_{ij}}{n_{ij}}$$

where PSCF represents potential source contribution function, n_{ij} is the total count of trajectory endpoints in the *ij*th cell and m_{ij} is the count of trajectory endpoints in the *ij*th cell associated with concentrations above the defined threshold.

Concentration-weighted trajectory

This is similar to potential source contribution function except it accounts for a residence time of trajectories instead of presence and absence of trajectories and calculated as

$$\bar{C}_{ij} = \frac{1}{\tau_{ijk}} \cdot \sum_{k=1}^{n} C_k \cdot \tau_k$$

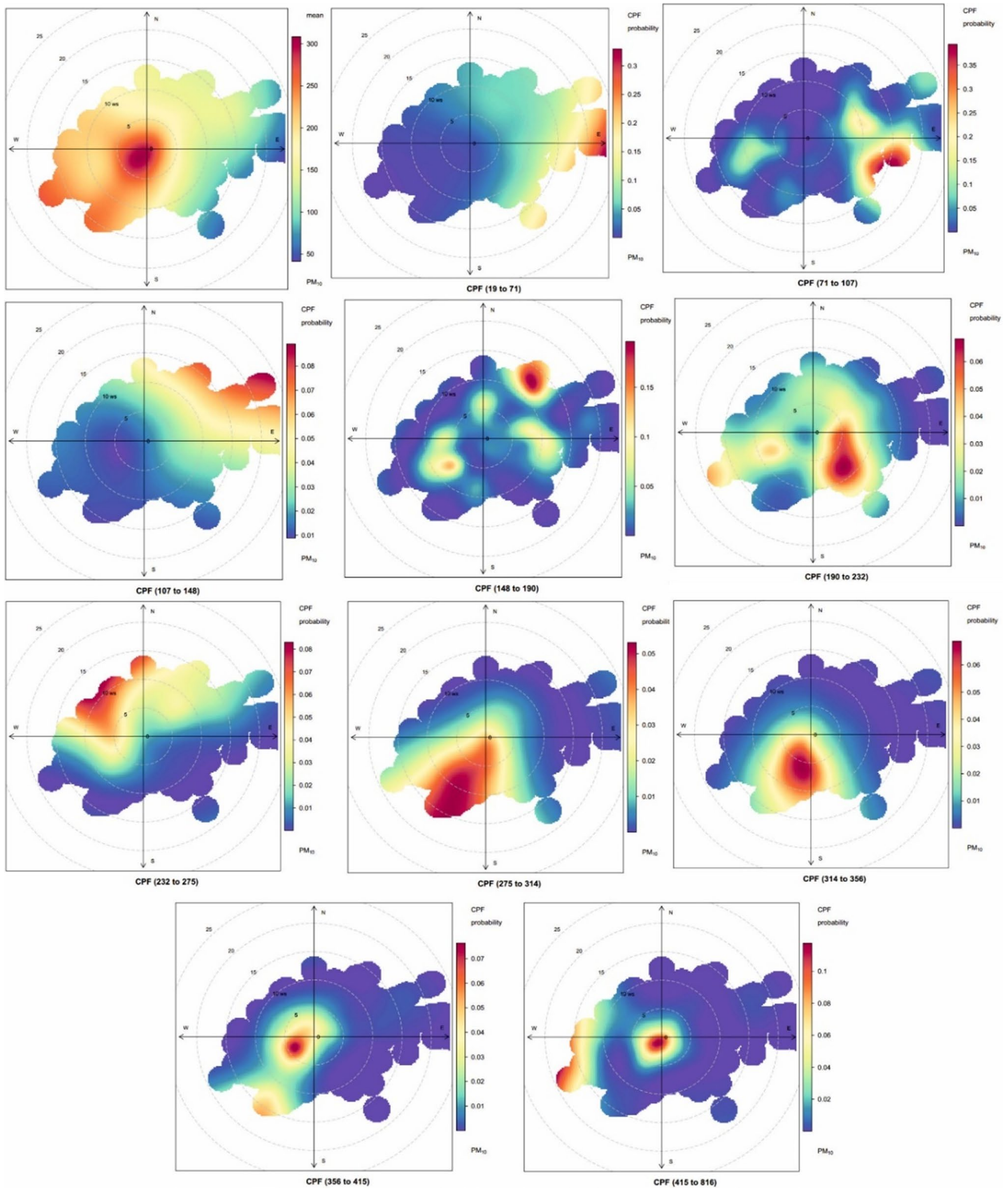
where $\bar{C}$ represents concentration-weighted trajectory, C_k is the concentration associated with trajectory endpoints in the ijth cell and τ_{ijk} is the resident time of trajectory endpoints in the *ij*th cell. Source trajectory analysis was performed through ZeFir v.3.40 (Petit et al. 2017) in Igor pro (wave metrics) v6.3 software. Trend in PM_{10} for 2014–2017 was analysed by Mann–Kendall test, and slope of the trend was estimated by Sen's slope (a nonparametric linear regression model). Hierarchical multiple regression was performed by SPSS software (SPSS Inc. version 23.0, IBM Corp, Armonk, NY).

Results and discussion

PM status and exceedances

Out of the total monitoring period of 1066 days (July 2014–May 2017), data of 823 days (77.20%) were assessed for PM_{10} status and exceedances. The mean PM_{10} concentration for said period was 244.8 (± 135.8) μg m^{-3} with values ranging between 19.28 and 816.1 μg m^{-3}. Mean concentration was, respectively, 4.081 and 12.24 times higher than the annual standard of Central Pollution Control Board, India (CPCB 2009), and World Health Organization (WHO 2005).

PM_{10} concentration varied significantly with the season ($p < .05$), but variations were non-significant for years. Mean PM_{10} concentration was maximum in the winter season (354.4 ± 92.85 μg m^{-3}), which were 4.539, 2.095 and 1.725 times higher than monsoon, post-monsoon and summer seasons. Most of this seasonal variation is attributed to planetary boundary layer height and other meteorological variables discussed in the later section of this paper. For the entire analysis period, PM_{10} level exceeded the Central Pollution Control Board annual and 24-h standard for ~ 93 and 82% days, whereas WHO 24-h standard was exceeded by ~ 96% of days. Exceedance of annual Central Pollution Control Board standard was maximum during winter (100%), followed by summer (99.50%), post-monsoon (94.04%) and was least in monsoon season (78.81%). Higher PM_{10} concentrations and exceedances are not unusual for this region, as previous reports by Mukherjee and Agrawal (2017) and Sharma et al. (2016) have also observed higher PM_{10} concentrations and exceedances in Indo-Gangetic plain.



CPF (19 to 71)

CPF (71 to 107)

CPF (107 to 148)

CPF (148 to 190)

CPF (190 to 232)

CPF (232 to 275)

CPF (275 to 314)

CPF (314 to 356)

CPF (356 to 415)

CPF (415 to 816)

Fig. 2 Conditional bivariate probability function plots for different concentration intervals of PM_{10} at the air quality monitoring site of Varanasi city for the period 2014–2017. Red to light green colour displays probable source areas with high probability in different directions from centre monitoring site with corresponding values in scale bar. *CPF*—conditional probability function

Varanasi is ranked 30th among Indian cities based on PM_{10} levels of global cities (WHO 2016).

After monsoon, PM_{10} concentration started to increase and showed peak concentration around January and then gradually declined from February to March and again showed elevation in April then followed a steep decline from May to July and remained constant up to August. In post-monsoon season, a sudden elevation was also recorded during months of September to November (Fig. 1c). Among the days of weeks, not much variations were recorded, but maximum value was recorded on Saturday and least on Sunday.

Time series analysis of PM_{10} based on Mann–Kendall test and Sen's slope showed a non-significant increase of 31.58 µg m^{-3} during the studied period. When time series trend was considered for different wind directions, a non-significant increasing trend was observed for most of the wind directions with a maximum increase from south-west direction (highly commercial region of the city). A significant positive increment of 64.66 µg m^{-3} was also recorded from south-east direction that may be due to increase in construction activity in this part of the city in last few years.

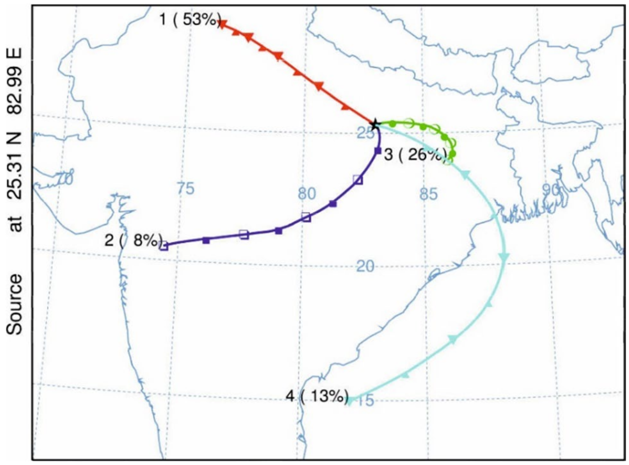


Fig. 3 Cluster analysis of backward air trajectories representing 96-h back trajectory clusters arriving at Varanasi city at 500 m above sea level during the period 2014–2017. The coloured lines represent the mean trajectory of each cluster, and values in parenthesis denote the percentage occurrence of backward air trajectories of each cluster. Cluster 1 located north-west of Varanasi city is the major trajectory pathway

Local PM_{10} source apportionment

The polar plot for entire data set showed two major source areas contributing maximally to the receptor site. The first sources are located in south-west direction under stable atmospheric condition (low wind speed) representing emission from local traffic, and the second sources under high wind speed are also located towards south-west direction representing emissions from the city centre (Fig. 2). When conditional bivariate probability function was run for different concentration intervals, several sources were detected. At lower interval (0–20% of data), diffused sources (lower probability values) are found from easterly direction at higher wind speed suggesting emission from nearby national highways, 28 and 31, whereas sources at moderate wind speed were located at south-east directions which intersect another busy road in the city with heavy commercial activity.

At the interval of 20–40%, two distinct sources were identified, one north-east, 4 km from the receptor site representing emission from traffic due to national highway 28 and another source from south-west direction where the industrial area of the city is located. At higher intervals (50–60%), sources were located towards north-west direction from receptor site; at the maximum interval (70–100%), all the PM sources were diffused and probably represent traffic areas at low wind speed mostly towards south-west direction from receptor site (Fig. 2). It is clear from the study that local sources were mostly due to traffic and road dust from major roads in the city and also from local combustion activities, whereas contribution from industries is minor as the industrial area is mostly related to storage activity with minor air pollutant emissions.

Distant source assessment of PM_{10} by trajectory statistical models

Cluster analysis of 96-h back trajectory data revealed 4 major air mass transport pattern reaching to Varanasi city. Four major clusters were further checked for their statistical significance based on Kruskal–Wallis test and were found significant at $p < .05$. Cluster 1 was the major cluster representing 53% of the total frequency which typically followed a longer flow pattern from north-west direction, suggesting its fast-moving nature. Cluster 2 (8%) showed pathway from the central region of India, originating from south-west direction with long transport pattern. Cluster 3 (26%) showed short transport patterns, indicating slow-moving air masses from eastern direction. Cluster 4 (13%) originated from the Bay of Bengal and followed a long curve pattern from south-east direction before reaching to the receptor site (Fig. 3).

Potential source contribution function and concentration-weighted trajectory found that at high probability, polluted air masses to Varanasi city originated from north-west, east and south-east directions (Fig. 4). In the summer season, higher concentrations were contributed from north-west, north-east and eastern directions. Long-range air masses were also detected from adjoining regions of Pakistan and Afghanistan, with major dense sources located within urban areas in upper Indo-Gangetic plain. In monsoon season,

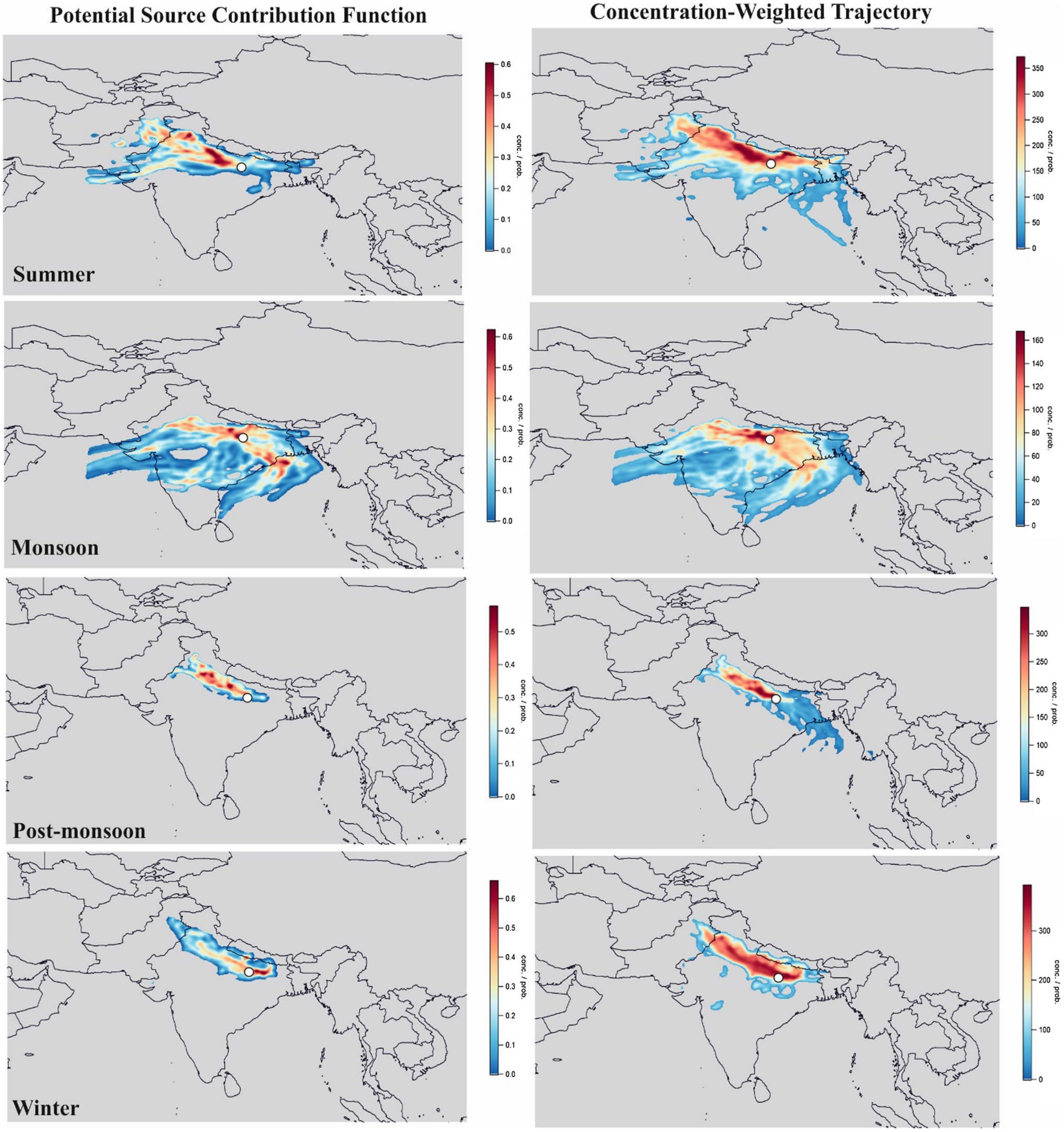


Fig. 4 Potential source contribution function and concentration-weighted trajectory maps for PM_{10} arriving at 500-m altitude at Varanasi city in different seasons. The Varanasi city is marked in circle, and values are displayed in colour scale with dark reddish brown colour indicating greater source potential and contribution of PM_{10} to monitoring site. (Color figure online)

major air masses bringing polluted air to receptor site were from north-west, south-east and north-east directions. Major areas in these regions are Delhi (capital of India), states of Haryana and Punjab, areas adjoining Varanasi district in north-west directions. The major source areas in east and south-east directions are from the states of Chhattisgarh, Bihar and Orissa and the Bay of Bengal. Post-monsoon season showed the unidirectional movement of polluted air masses predominantly from north-west direction. During winter season, higher PM was mainly originated from upper Indo-Gangetic plain and eastern region of Pakistan in north-west direction. PM_{10} was also contributed by Bihar and Chhattisgarh states due to the cycling of air masses in these areas at low planetary boundary layer height.

Most of the regions from where air masses travelled are identified amongst the most polluted regions of the world (WHO 2016). Several cities located on the air mass pathways are highly urbanized and industrial. In post-monsoon and winter seasons, entire north-west region is a huge source of biomass burning emissions from states of Punjab, Haryana and Uttar Pradesh, which bring higher PM mass to Varanasi. In summer season, a significant portion of PM is also added due to Thar Desert bringing a huge amount of desert dust to middle Indo-Gangetic plain (Mukherjee and Agrawal 2017). We also found sources from eastern direction which were mostly emitted through intensive burning activity and from mining operations in these areas.

Based on the data of cluster analysis, we calculated the ratio of local and distant sources contributions to PM_{10}. The ratio varied from 0.09 to 0.59 during the entire study period, suggesting a significant contribution of distant sources to local PM_{10}. The contribution was maximum during winter (39%) and was least during monsoon (9.4%). During the study period, we found mean $PM_{2.5}/PM_{10}$ ratio of 0.47 indicating a significant proportion of $PM_{2.5}$ in PM_{10}. The ratio varies with different emission sources, and a lower ratio is a marker of natural sources (windblown or road dust), while higher value of the ratio indicates towards anthropogenic sources (vehicular emissions, biomass burning and industrial emissions) because $PM_{2.5}$ emissions are normally higher from anthropogenic sources.

Role of meteorology and gaseous air pollutants

Hierarchical multiple regression was performed on normalized data of both air pollutants and meteorological variables by the stepwise approach to assess explained variability in PM_{10} (dependent variable) by independent variables (meteorological factors and air pollutants). Adjusted R^2 for model 1 which includes average dew point, wind speed, ventilation coefficient, minimum humidity, maximum and minimum daily temperature and planetary boundary layer height explained 68.5% variability in PM_{10} data due to meteorology, whereas model 2 representing air pollutants (CO, NO_2 and SO_2) explained 7.3% variability. This result suggests that PM_{10} dispersion or formation is regulated mostly through meteorological variables. Among the meteorological factors, PM_{10} showed maximum association with dew point followed by wind speed, temperature and ventilation coefficient. Dew point and temperature affect PM formation and removal, whereas wind speed and ventilation coefficient regulate PM dispersion.

Conclusion

PM_{10} concentrations in the urban area of Varanasi city were found to be critical with respect to national and international standards. Time series study showed a positive non-significant trend in PM_{10} level. Traffic and road dust were identified as the major factors for the local increases in PM_{10}. Trajectory statistical models like cluster analysis, potential source contribution function and concentration-weighted trajectory identified the significant contributions of PM_{10} from north-west and eastern directions through the transport of polluted air masses to Varanasi city. It is clear from our study that PM_{10} concentrations and variations are mostly regulated by local and distant emission sources and meteorological factors. Reduction in traffic emission and identification of other minor sources in local level and strict legislation at national level to reduce crop burning and industrial emissions in Indo-Gangetic plain are required to prevent excessive PM pollution in the region.

Acknowledgements Authors are thankful to Head, Department of Botany, Banaras Hindu University, and Coordinators, Center of Advanced Study in Botany and DST-FIST for providing necessary laboratory and central instrumental facilities for the research work. Department of Science and Technology, India, is also acknowledged for providing financial support in the form of an Inspire fellowship (IF120768) to Arideep Mukherjee. The authors are also grateful to anonymous reviewers and chief editor for their valuable suggestions which helped in improving the quality of the manuscript.

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