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Assessment of particulate matter variation during 2011–2015 over a tropical station Agra, India

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HIGHLIGHTS

- Daily average and seasonal variations of RSPM and SPM were analysed over a period of 2011-2015 at Agra.
- Back Trajectory analysis was done to analyse the aerosol pathways associated with air mass transport.
- Wavelet analysis was employed to assess the periodicity in the time series data.
- Role of transported pollution at study site Agra has been analysed seasonally.

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ABSTRACT

Air quality over Agra is deteriorating and causing a serious threat to people residing in the city as well as to World heritage site- Tajmahal. In the present study, daily average concentrations of Suspended Particulate Matter (SPM) and Respirable Suspended Particulate Matter (RSPM) were analysed over a period of 2011–2015 at four stations in Agra city, namely: Taj, Itmad-Ud-Daula, Rambagh and Nunhai. The concentrations are above threshold values when compared to specify standards for a healthy environment (by India, US, WHO, EU and China - Class I and Class II) for all the seasons except monsoon and the values are highest in the month of November and lowest in the month of August and September. Variation of RSPM and SPM were found to be positively correlated with each other with values of 0.76 (Taj), 0.72 (Itmad-Ud-Daula), 0.69 (Rambagh), 0.77 (Nunhai). The study illustrates that the levels of SPM and RSPM are not showing any decreasing trend over Agra even after closing of industries and taking other precautions inside the city by Government of India. The study clearly identifies that local control of pollution sources are not enough and pollution is being transported from nearby regions to keep the daily pollution value higher than threshold. Source regions of transported pollutants over Agra have been analysed by using Weighted Potential Source Contribution Function (WPSCF) for both SPM and RSPM. Wavelet analysis of monthly averaged values of RSPM and SPM data sets has shown the existence of semi-annual and annual periodicity over the study region.

1. Introduction

The presence of foreign particles, gases and other pollutants in the air in such concentration which is harmful to living entities and environment is termed as air pollution (WHO Online Ref.; Schmale et al., 2014). Constituents like Nitrogen dioxide, Ozone, Sulphur dioxide, Respirable Suspended Particulate Matter (RSPM) and Suspended Particulate Matter (SPM) are considered to be as main atmospheric pollutants and play an important role in affecting the human health, global environment and climate change (Schmale et al., 2014). Globally the proportions of these constituents are increasing rapidly due to increased vehicular pollution, urbanization, population, and industrialisation (Karagulian et al., 2015). Size and chemical composition of particulate matter (PM) have strong influence on the human health and environment, altering radiation budget by absorption and scattering (Ram and Sarin, 2012, Meena et al., 2014; Pipal et al., 2014c, Chitranshi et al., 2015).

Agra, heritage city of India is situated in the state Uttar Pradesh of India. It is the place where one of the seven wonders of world – Taj Mahal is situated. Assessment of air quality and aerosol load over Agra has always been an issue of prime concern by researchers

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and policy makers and it has been observed that values of aerosol loading is mostly higher than threshold values (e.g. Central Pollution Control Board (CPCB) report 2006; Safai et al., 2008; Kulshrestha et al., 2009; Singh et al., 2010; Dey et al., 2012; Pipal et al., 2014a; Meena et al., 2014; Chitranshi et al., 2014; Saini et al., 2014). It has been found that this increased PM concentration is affecting the monument adversely (Hicks and Kumari, 1987; Bergin et al., 2015). To provide a cleaner environment to public and to protect Taj Mahal, Government of India has taken strict measures in the past decade. Major actions being taken up for cleaner environment involves (i) usage of natural gas for the production processes, (ii) Continuous monitoring of existing industries which use coal or coke near the city boundary and no permission to new industries which use coal and coke inside the city area, (iii) Shut down of all the brick industries within a radius of 20 km of Agra city, (iv) Fitting scrubbers to diesel driven vehicles and use of low sulphur content diesel, (v) Restriction on usage of cars within a radius of 2 km from the monument to protect Taj Mahal, (vi) Use of battery driven/electric cars for the tourists (CPCB report, 2006).

The present study aims at assessment of SPM and RSPM concentration in Agra over a period of 2011–2015, and focused on finding an answer to the question: "Whether the measures implemented by Govt. of India on a local scale are improving air quality over Agra city?" We have analysed temporal (diurnal and seasonal) variation of RSPM and SPM concentration over Agra at four stations: Taj, Itmad-ud-Daula, Rambagh and Nunhai, over a period of 2011–2015, collected by Central Pollution Control Board (CPCB), Govt. of India. National Oceanic and Atmospheric Administration (NOAA) Hybrid Single Particle Lagrangian Integrated Trajectories (HYSPLIT) model (Draxler and Hess, 1998), which has been proven to provide a good model for back trajectory analysis (e.g. Fleming et al., 2012; Cheng et al., 2013; Chandra et al., 2014; Pipal et al., 2014c; Yadav et al., 2014) is employed in the present study to estimate long-range aerosol transport pathways to Agra. Source regions of transported pollutants over Agra have been analysed by using Weighted Potential Source Contribution Function (WPSCF) for both SPM and RSPM. Dispersion runs for cases during study period over the study regions are performed for understanding the dispersion patterns advecting over site. Use of wavelet analysis in the present study explores if there is any periodicity existing in the variation of RSPM and SPM over the study region.

2. Study area and data description

Agra (27.12° N, 78.17° E, 169 m above MSL), well known for one of the wonders of the world: Tajmahal, is situated in the state of Uttar Pradesh, India. Agra is a tropical, semiarid region with hot scorching summers and cold winters. The year has been divided into four seasons in the present study as pre-monsoon (March to May), monsoon (June to September), post-monsoon (October to November) and winter (December to February). The city is densely populated with the population density of 433 persons/km^2 (Census, 2011). River Yamuna is flowing through Agra city and Taj Mahal is situated on the west bank of Yamuna (Pipal et al., 2014a). Agra is surrounded by the Thar Desert from west to north-west (spatial distance ~ 300 km), and major cities including New Delhi in the north-west (spatial distance ~176 km), Firozabad in the east (spatial distance ~ 35 km), Mathura (spatial distance ~ 50 km), Hathras (spatial distance ~ 45 km), and Etah (spatial distance ~ 72 km) in the north-westerly to north-easterly direction.

CPCB has four monitoring stations in the city, namely: Taj (27.175° N, 78.0422° E), Itmad-Ud-Daula (27.1929° N, 78.031° E), Rambagh (27.2054° N, 78.0383° E) and Nunhai (27.2023° N, 77.78031° E). The location of Agra and zoomed on to four sites is shown in Fig. 1. Nunhai and Rambagh are highly industrialized areas, contributing to a remarkable growth in the concentration of pollutants. Various activities that contribute to air pollution over Agra other than vehicular emissions includes emissions from rubber processing, glass, and ferrous casting industries, biomass burning (Meena et al., 2014, Dayal et al., 2010).

The daily average concentration of RSPM (also can be called as $PM_{2.5}$) and SPM (also can be called as PM_{10}) at four sites: Taj, Itmad-Ud-Daula, Rambagh and Nunhai, collected by CPCB over a period of 2011–2015 are used in the present study. Wind data (3 hourly) from National Climate Data Centre (NCDC) (http://www.ncdc.noaa.gov/) has been used for analyzing wind roses to get a picture of wind speed and direction variation over Agra during period of study.

Back Trajectory and dispersion analysis has been performed using National Centres for Environment Prediction (NCEP)/National Centre for Atmospheric Research (NCAR) GDAS (Global Data Assimilation System) 1^{O} data and HYSPLIT 4 model June 2015 release (svn:761) (Stein et al., 2015; Draxler, 1999; Draxler and Hess, 1997). GDAS 0.5° data is missing vertical velocity and it has been found out recently by Su et al. (2015) that GDAS 1° data is better in computing back trajectories compare to GDAS 0.5° data because of this reason. Hence we have used GDAS 1° data only in present study. Therefore our model configuration for trajectory runs can be summarized as follows: Model version: HYSPLIT 4 model June 2015 release (svn:761), with Input data: GDAS 1^{O} data, Run time: −48 h for each 6 h interval, Model domain: not fixed with boundaries. Trajectories are allowed to go the extent they can move in the global domain, Trajectories computation method: Isentropic trajectories, Model altitude for trajectory computation: 500 m above ground layer (AGL), and Model top for trajectory computations: 10000 m AGL.

For dispersion runs also, we have used GDAS 1° data. The model has been allowed to use a deposition of 0.1 cm/s with a release top of 50 m AGL and release bottom at 0 m AGL. We have assumed that a unit mass has been released for 1 h at 12 UTC of the day and 24 h backward dispersion has been computed with averaging period of 6 h with top of averaged layer at 100 m AGL. For dispersion case studies, we have run online version of HYSPLIT (Rolph, 2016).

3. Methodology

3.1. Weighted Potential Source Contribution Function (WPSCF)

Back trajectory analysis was done over Agra for the four seasons to analyse the aerosol pathways from different sources at an altitude of 500 m above the ground level using HYSPLIT 4 model (Draxler and Hess, 1998). 48 h air mass isentropic back trajectories at Agra at an interval of 6 h (i.e. 00, 06, 12 and 18 UTC of every day) have been computed to analyse the long-term transport of aerosols from different regions. There are various methods for analysing back trajectory statistics, popular ones including- Concentration Weighted Trajectory (CWT) and Potential Source Contribution Function (PSCF). Both are trajectory statistical methods and it has been found by ensemble average correlation coefficient study that PSCF calculations are better than CWT (Kabashnikov et al., 2011). It is also found that both PSCF and CWT analysis are good in identifying the major source regions but performing different in identifying the moderate sources (Xin et al., 2016). The PSCF value for a given grid cell can be calculated by endpoints of trajectories in that grid cell. PSCF can be defined as (Han et al., 2007, Xu and Akhtar, 2010):

$$\mathrm{PSCF}(i,j) = (a_{i,j}/b_{i,j}) \quad (1)$$

$b_{i,j}$ and $a_{i,j}$ are the total number of back trajectory endpoints falling

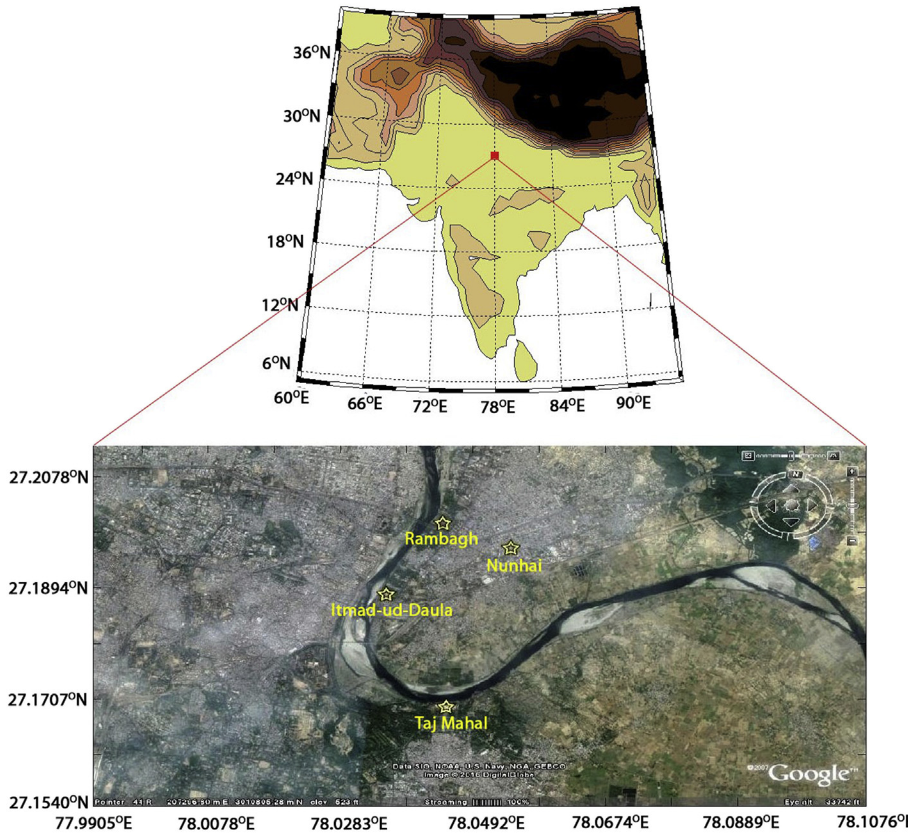

Fig. 1. Geolocation of Agra with monitoring stations.

into grid cell (i,j), during all days, and in days when receptor concentrations were higher than specified criterion value respectively. An empirical weight function can be used to reduce the influence of $b_{i,j}$ on the PSCF values (Zeng and Hopke, 1989; Xu and Akhtar, 2010):

$$W_{i,j} = \begin{cases} 1.0 & b_{i,j} > 2 \times \text{Average} \\ 0.75 & \text{Average} < b_{i,j} \leq 2 \times \text{Average} \\ 0.5 & 1/2 \times \text{Average} < b_{i,j} \leq \text{Average} \\ 0.15 & 0 < b_{i,j} \leq 1/2 \times \text{Average} \end{cases} \quad (2)$$

where Average is the average number of trajectory endpoints in all grid cells.

Thus, the Weighted PSCF (WPSCF) can be defined as:

$$\text{WPSCF}(i,j) = (a_{i,j}/b_{i,j})W_{i,j} \quad (3)$$

The threshold values for SPM and RSPM to be used in WPSCF has been taken as 100 and 60 $\mu g/m^3$ as suggested by Indian standards threshold for a clean environment (CPCB report, 2015).

3.2. Periodicity test using wavelet analysis

Wavelet is a signal processing tool for determining dominant modes within a time series by decomposing the time series into time-frequency domain (Torrence and Compo, 1998). Wavelet scalogram is being used for determination of periodicity in time series data of pollutant concentrations and proven to detect the dominant periods (Zeri et al., 2016; Ramachandran et al., 2013). In the present study, periodicity test is performed on monthly averaged data sets using continuous wavelet transform approach, using Morlet mother wavelet, normalised by the standard deviation. Morlet wavelet, consisting of a plane wave modulated by Gaussian for a wavelet function $\psi_0(\eta)$ depending on non-dimensional time parameter η can be explained as:

$$\psi_0(\eta) = \pi^{-1/4} e^{i\omega_0\eta} e^{-\eta^2/2} \quad (4)$$

where ω_0 is non-dimensional frequency (Torrence and Compo, 1998). The wavelet power spectrum using Morlet wavelet for a Gaussian white noise normalised by standard deviation has been plotted for the present study.

4. Results and discussion

4.1. Daily average concentration variation of RSPM and SPM over Agra

The concentration of RSPM and SPM shows a significant daily average variation at all the four stations during the study period. The highest, lowest and mean concentrations of daily average RSPM were found to be 622 $\mu g/m^3$, 11 $\mu g/m^3$, 164.91 $\mu g/m^3$ (Taj), 697 $\mu g/$

Table 1
Particulate matter standards implemented by India, USA, EU, WHO and China (Class I and Class II). Values are in $\mu g/m^3$.

	Region	India	USA	EU	WHO	China Class I	China Class II
Daily	$PM_{2.5}$	60	35	–	25	35	75
	PM_{10}	100	150	50	50	50	150
Annual	$PM_{2.5}$	40	12	25	10	15	35
	PM_{10}	60	–	40	20	40	70

m^3, 16 μg/m^3, and 185.81 μg/m^3 (Itmad-ud-Daula), 621 μg/m^3, 21 μg/m^3, and 179.38 μg/m^3 (Rambagh) and 644 μg/m^3, 35 μg/m^3, and 245.08 μg/m^3 (Nunhai), respectively for the period of study. The highest, lowest and mean concentration of daily average SPM were found to be 1058 μg/m^3, 28 μg/m^3, and 298.15 μg/m^3 (Taj), 1134 μg/m^3, 16 μg/m^3, and 367.26 μg/m^3 (Itmad-ud-Daula), 904 μg/m^3, 52 μg/m^3, and 374.49 μg/m^3 (Rambagh) and 1217 μg/m^3, 42 μg/m^3, and 526.58 μg/m^3 (Nunhai), respectively during 2011–2015. The standards of PM permissible level adopted by India, USA, EU, WHO and China (Class I: Applies to national parks, conservation areas, and designated historical sites and Class II: Applies to residential, commercial, cultural, industrial, and heavily trafficked areas) are specified in Table 1 (Guerreiro et al., 2014; Cao et al., 2013; CPCB report, 2015). The plot of daily average concentration variation of RSPM and SPM is shown in Fig. 2a and b, which clearly indicates that the concentrations for most of the days in the annual cycle are higher than the specified permissible levels in Table 1. The concentration of RSPM (Fig. 2a) was highest in the months of November and December (post monsoon and winter season) whereas, concentrations were lowest in the months of August and September (monsoon season). July will have more rain than September in the area. Lower values in September compared to July (although the differences in values are not very large) may be due to two reasons: 1. Rainy conditions will wash out the pollutants. 2. On non rainy days, atmosphere is less cloudy in September (which is withdraw phase of monsoon over the region) compare to July (onset month of monsoon, having more rainfall), which allows vertical mixing with clear sky and advection of pollutants from nearby places is not that high which was observed in July. We believe that this is the reason why we have lower values in September compare to July over the study region.

Lowest concentrations during monsoon period can be attributed to rainfall and washing out of pollutants over the region. The higher concentrations of RSPM over Agra during November and December can be attributed to both natural and man-made activities such as vehicular emissions, coal and biomass and biofuel combustion, crop residual burning and dust transport from nearby dry regions along with stable conditions prevailing in the atmosphere (Pipal et al., 2014a).

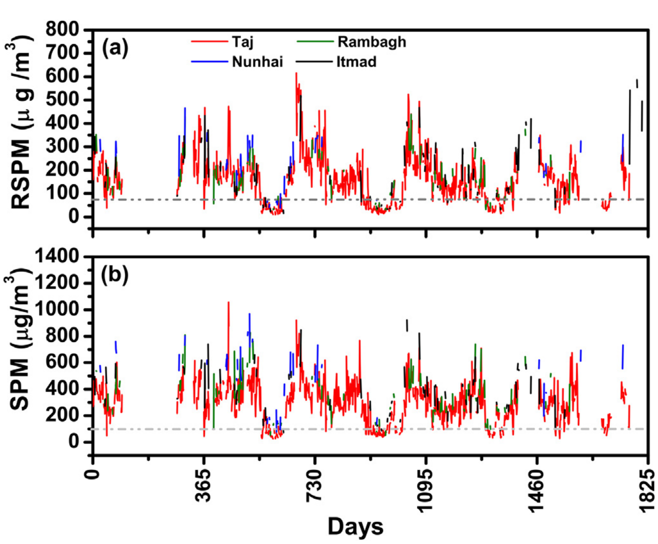

Fig. 2. Variation of daily average concentrations of (a) RSPM and (b) SPM at all four monitoring stations for the period 2011–2015. Gaps are due to data unavailability. The grey colour dash-dot line indicates the Particulate matter standards implemented by India. The x-axis represents years and 0–365 should be read as 2011, 365–730 is 2012 and so on.

The daily average variation of SPM is shown in Fig. 2b. The daily average values of concentrations for SPM are highest in the months of March (pre-monsoon) and November (post-monsoon) and lowest in the months of August and September (monsoon). Lowest SPM values are coinciding with RSPM lowest values during monsoonal rainfall. For SPM also, high values are observed in November. However, in the pre-monsoon month of March, SPM values are quite high and found to be comparable with values observed in months of November, December; which was not the case with RSPM. Higher values of SPM during March may be because of associated dust storms in and around the study region, which will increase the concentration of dust particles in the atmosphere considerably (Dipu et al., 2013).

Trend analysis (using Mann-Kendall test) for the time series of both SPM and RSPM shows that there is no trend existing in data. However, if we fit a linear trend line, it shows that for SPM at all the four sites slope values are (Taj: slope = −0.0747, Rambagh: slope = −0.0651, Itmad-ud-Daula: slope = −0.0939, Nunhai: slope = −0.0958) whereas for RSPM, it was observed that slope values for three stations (Taj: slope = −0.042, Rambagh: slope = −0.013, Nunhai: slope = −0.015) but positive for Itmad-ud-Daula (slope = +0.026).

The absence of trend (expected to be negative) shows that the measures taken up by the Government of India at a local scale (CPCB report, 2006) are not helpful to improve the air quality, as the values are still high and crossing threshold limits for most of the days. In the case of RSPM values at Itmad-ud-Daula, slope is positive (however not significant). Percentage of RSPM and SPM exceeding India and China Class II threshold values (as these two are highest in class) for daily average values at all four sites in Agra is shown in Table 2. It was observed that the percentage crossing threshold values for RSPM and SPM were high (>77% in any case) when compared to that of standards specified by CPCB NAAQS (India) and China (Class-II) which are highest in the category as specified in Table 1. The results are showing how severely air quality is affected at the region and health consequences faced by local residents due to these high alarming values throughout the year cannot be neglected.

4.2. Seasonal variation of RSPM and SPM concentration over Agra

Seasonal variation of both RSPM and SPM over Agra showed a considerable variation in all the seasons during the study period. Table 3 is showing seasonal values for RSPM and Table 4 for SPM during 2011–2015.

For RSPM, at Taj station, winter season values are highest followed by post-monsoon, pre-monsoon, and monsoon at all four stations. Only in the year 2012, post monsoon value (290.38 ± 144.83) is higher than winter values (216.07 ± 75.28). For Rambagh, Itmad and Nunhai, winter and post-monsoon values are higher and almost of the same order. Maximum values are either in winter or in post-monsoon for these sites. Pre-monsoon values are coming next in magnitude followed by monsoon values, which are

Table 2
Percentage of RSPM and SPM daily average concentrations exceeding standards implemented by India and China (Class II).

Site	RSPM		SPM	
	>60 μg/m^3	>75 μg/m^3	>100 μg/m^3	>150 μg/m^3
Taj	81.19	77.91	85.88	79.69
Itmad	87.03	82.42	94.31	87.53
Rambagh	86.75	83.12	96.79	86.72
Nunhai	93.52	88.74	99.65	95.47

Table 3
Seasonal concentrations of RSPM over Agra. Terms in bracket indicates standard deviations.

	Year	Pre-monsoon	Monsoon	Post-monsoon	Winter
Taj	2011	155.15 (±46.94)	–	198.43 (±72.92)	230.75 (±103.21)
	2012	197.53 (±80.66)	78.09 (±69.48)	290.38 (±144.83)	216.07 (±75.28)
	2013	159.63 (±39.78)	52.88 (±41.77)	214.81 (±125.99)	241.59 (±98.75)
	2014	129.28 (±40.22)	94.59 (±62.12)	154.04 (±58.26)	181.23 (±61.24)
	2015	136.95 (±62.18)	54.28 (±20.82)	195.95 (±62.41)	203.91 (±48.33)
	Average	***155.71 (±53.96)***	***69.96 (±48.55)***	***210.72 (±92.88)***	***214.71 (±77.36)***
Rambagh	2011	146.86 (±47.70)	–	221.67 (±81.96)	243.40 (±80.97)
	2012	180.13 (±50.03)	93.95 (±74.21)	329.21 (±173.63)	206.24 (±81.36)
	2013	200.57 (±51.74)	76.24 (±39.48)	259.37 (±120.89)	221.18 (±88.16)
	2014	173.67 (±77.49)	114.67 (±68.66)	262.82 (±94.54)	196.61 (±74.58)
	2015	171.32 (±80.37)	72.20 (±32.89)	161.56 (±43.77)	214.79 (±44.48)
	Average	***174.51 (± 61.47)***	***89.26 (± 53.81)***	***246.92 (± 102.96)***	***216.44 (± 73.91)***
Itmad	2011	162.42 (±44.56)	–	222.17 (±87.94)	265.37 (±95.01)
	2012	172.35 (±95.80)	80.67 (±59.24)	291.70 (±143.40)	234.43 (±80.70)
	2013	199.20 (±47.86)	60.79 (±35.86)	255.54 (±145.72)	260.52 (±111.61)
	2014	160.21 (±68.21)	115.07 (±71.04)	288.00 (±112.24)	243.57 (±99.23)
	2015	141.95 (±61.46)	63.08 (±15.62)	370.35 (±202.79)	284.13 (±106.68)
	Average	***167.22 (± 63.57)***	***79.91 (± 45.44)***	***285.55 (± 138.41)***	***256.60 (± 98.64)***
Nunhai	2011	203.81 (±64.09)	–	264.67 (±132.55)	320.29 (±114.04)
	2012	247.86 (±90.63)	116.62 (±82.21)	368.16 (±147.42)	286.45 (±72.06)
	2013	278.13 (±74.15)	110.25 (±77.31)	–	284.16 (±108.50)
	2014	–	–	412.00 (±70.57)	358.87 (±132.37)
	2015	206.00 (±79.85)	74.50 (±20.49)	246.78 (±59.02)	257.67 (±74.95)
	Average	***233.95 (± 77.18)***	***100.46 (± 60.01)***	***322.90 (± 102.39)***	***301.48 (± 100.38)***

lowest in the range. The high values during post monsoon and winter season may be attributed to the locally emitted and transported pollutants and stable conditions persisting over the area during that period. There may be existence of temperature inversion layer over the region, which will further help in enhancing the pollutants and making high concentrations over the study area. Lower depth of mixed layer, low temperatures, and less wind speed helps the pollutants to stagnate and shows a significant increase in the concentration during winters over the region (Tiwari et al., 2015). As mentioned earlier, burning of biomass, coal, and local anthropogenic activities also contribute to high concentrations of PM over Agra in winter and post-monsoon season (Pipal et al., 2014c).

However, in the case of SPM, post monsoon season values are highest attributing to aerosol load due to crop residual burning, biomass burning, and incirnation. Winter and pre-monsoon values are almost of the same magnitude and little lower in magnitude to that of winter. It is noticed that the values are high in comparison to RSPM in all the seasons. The high load in the pre-monsoon season may be attributed to transported aerosols from dust storms of the Thar Desert and the Middle East winds. During monsoon, the load was observed to be lowest as the pollutants are washed out because of heavy rainfall (Kulshrestha et al., 2009).

The average pollutant variation of PM over Agra from previous studies in comparison to present study findings are shown in Table 5. All the previous studies show that mean mass concentration of particulate matter is high at Agra over decades. The values are on the higher side when compared with most of the previous

Table 4
Seasonal concentrations of SPM over Agra. Terms in bracket indicates standard deviations.

	Year	Pre-monsoon	Monsoon	Post-monsoon	Winter
Taj	2011	336.26 (±101.76)	–	379.30 (±83.72)	370.58 (±118.38)
	2012	409.64 (±140.47)	190.20 (±174.60)	473.84 (±166.85)	338.41 (±98.74)
	2013	364.55 (±102.44)	125.16 (±83.12)	335.72 (±176.23)	350.41 (±123.09)
	2014	274.16 (±92.79)	216.79 (±145.25)	283.17 (±97.07)	256.27 (±84.77)
	2015	285.95 (±149.64)	133.48 (±48.76)	328.95 (±87.49)	306.33 (±74.51)
	Average	***334.11 (± 117.42)***	***166.41 (± 112.93)***	***360.19 (± 122.27)***	***324.40 (± 99.90)***
Rambagh	2011	369.20 (±101.55)	–	507.13 (±118.06)	426.69 (±120.94)
	2012	501.50 (±117.63)	295.72 (±228.76)	568.78 (±184.42)	415.76 (±140.35)
	2013	460.96 (±144.68)	183.97 (±98.74)	400.92 (±165.41)	351.23 (±118.97)
	2014	352.29 (±135.85)	286.92 (±161.31)	489.36 (±146.54)	312.96 (±119.36)
	2015	350.74 (±168.60)	203.90 (±116.67)	393.44 (±105.33)	362.11 (±85.35)
	Average	***406.94 (± 133.66)***	***242.63 (± 151.37)***	***471.93 (± 143.95)***	***373.75 (± 116.99)***
Itmad	2011	471.00 (±94.48)	–	522.33 (±168.08)	471.07 (±146.19)
	2012	496.30 (±186.15)	272.47 (±196.18)	588.05 (±196.72)	429.43 (±121.37)
	2013	416.16 (±100.25)	177.08 (±105.96)	509.29 (±247.83)	427.32 (±174.62)
	2014	354.35 (±132.09)	258.37 (±157.72)	464.27 (±141.27)	370.39 (±104.71)
	2015	303.35 (±173.20)	157.67 (±52.92)	406.64 (±102.36)	349.50 (±65.32)
	Average	***408.23 (± 137.23)***	***216.40 (± 128.19)***	***498.12 (± 171.25)***	***409.54 (± 122.44)***
Nunhai	2011	604.91 (±120.27)	–	605.50 (±148.79)	641.45 (±199.86)
	2012	645.31 (±176.82)	339.80 (±231.21)	767.32 (±207.91)	594.40 (±162.86)
	2013	592.40 (±151.61)	261.50 (±154.01)	–	539.89 (±193.61)
	2014	–	–	671.33 (±139.37)	597.50 (±192.97)
	2015	466.48 (±181.05)	263.52 (±89.04)	535.00 (±102.56)	469.09 (±130.10)
	Average	***577.27 (± 157.44)***	***287.60 (± 158.08)***	***644.79 (± 149.66)***	***568.47 (±175.88)***

Table 5
Mean mass concentrations of PM over Agra observed during different studies.

S. No	Period of study	Author	Studied parameter	Mean mass concentration (μg/m^3)
1	April 1991–June 1992	Kulshrestha et al., 1994	SPM	368.5
2	Dec 2004	Safai et al., 2008	TSP	Foggy/Hazy days - 475 ± 101, Clear sky days - 338 ± 38
3	Dec 2005–Dec 2006	Rajput and Lakhani, 2010	TSP	TSPM- 198–690, PAH- 15–392 ng m^{-3}
4	May 2006–Mar 2008	Kulshrestha et al., 2009	$PM_{2.5}$ and PM_{10}	Urban- PM_{10}-154.2, $PM_{2.5}$-104.9 Rural- PM_{10}-148.4, $PM_{2.5}$-91.1
5	May 2006–Sept 2006 May 2007–Sept 2007	Satsangi et al., 2011	PM_{10} and $PM_{2.5}$	SJC- PM_{10}-121.8 ± 28.3, $PM_{2.5}$-73.5 ± 14.6 DB- PM_{10}-68.7 ± 24.8, $PM_{2.5}$-28.6 ± 14.6 BS- PM_{10}-54.9 ± 20.3, $PM_{2.5}$-39.4 ± 16.3
6	Jan–Dec 2007	Singh et al., 2010	PM_{10}	PM_{10}-160
7	Mar 2007–Feb 2008	Singh and Sharma, 2012	PM_{10}	PM_{10}-155.47 ± 77.97
8	May 2010–April 2011	Pachauri et al., 2013	Carbonaceous $PM_{2.5}$	$PM_{2.5}$-79.7 ± 40.5
9	2010–2011	Pipal et al., 2014a	$PM_{2.5}$, PM_{10}	Roadside- $PM_{2.5}$-97.2, PM_{10}-242.6 Semirural- $PM_{2.5}$-230.5, PM_{10}-121.2
10	2010–2012	Chitranshi et al., 2014	PM_{10}	PM_{10}-72.98 - 141.75± (39.70–85.24)
11	May–Aug 2011	Meena et al., 2014	Carbonaceous $PM_{2.5}$	$PM_{2.5}$-55.3 ± 17.4
12	Dec 2011–Feb 2012	Pipal et al., 2014b	Carbonaceous $PM_{2.5}$	$PM_{2.5}$-183 ± 114
13	Nov 2011–June 2012	Bergin et al., 2015	$PM_{2.5}$,TSP	TSP-135 ± 55, $PM_{2.5}$-60 ± 39
14	Jan 2011–Dec 2015	Present Study	SPM, RSPM	Taj: SPM- 298 ± 155, RSPM- 165 ± 103 Rambagh: SPM- 367 ± 170, RSPM- 179 ± 100 Itmad: SPM- 374 ± 186, RSPM- 186 ± 113 Nunhai: SPM- 527 ± 225, RSPM- 245 ± 126

studies but of the same magnitudes of order as observed by Pipal et al. (2014b); Rajput and Lakhani (2010); Safai et al. (2008). We believe that change of monitoring sites locations at different parts inside the city may be prime reason for this discrepancy in the values along with use of different instrumentation and different methods for calculating the values of RSPM and SPM.

4.3. Correlation between SPM and RSPM variation

Correlations between the RSPM and SPM were analysed to estimate the impact of emissions and various atmospheric phenomena over aerosol accumulation and dispersion with size dependency over Agra (Yadav et al., 2014). Scatter plots were generated between daily average concentrations of RSPM and SPM at Agra during years 2011–2015 (Fig. 3) to investigate the correlation. The variation of RSPM and SPM show a good agreement in all the seasons at four sites. It is evident from the scatter plots that RSPM and SPM are positively correlated at Taj ($r^2 = 0.76$), Itmad-ud-Daula ($r^2 = 0.72$), Rambagh ($r^2 = 0.69$), and Nunhai ($r^2 = 0.77$). Fitting 1:1 line shows that SPM values are on the higher side for all sites during the study period.

4.4. Wind roses for the study area

Wind roses have been plotted using NCDC 3 hourly datasets over Agra during period of study to get a picture of wind speed and

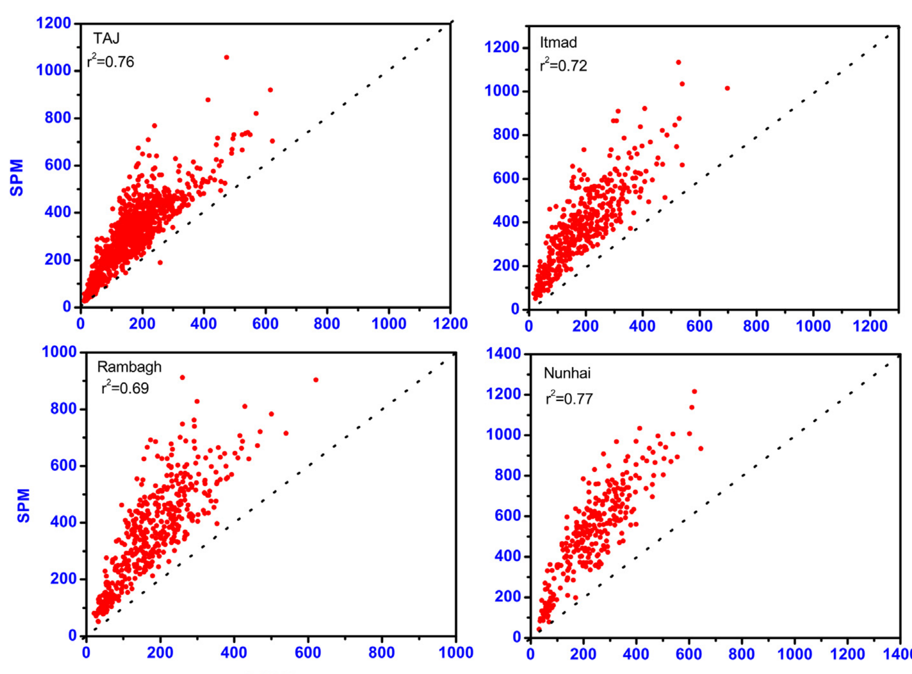

Fig. 3. Correlation between daily average values of SPM and RSPM at all four monitoring stations for the period 2011–2015.

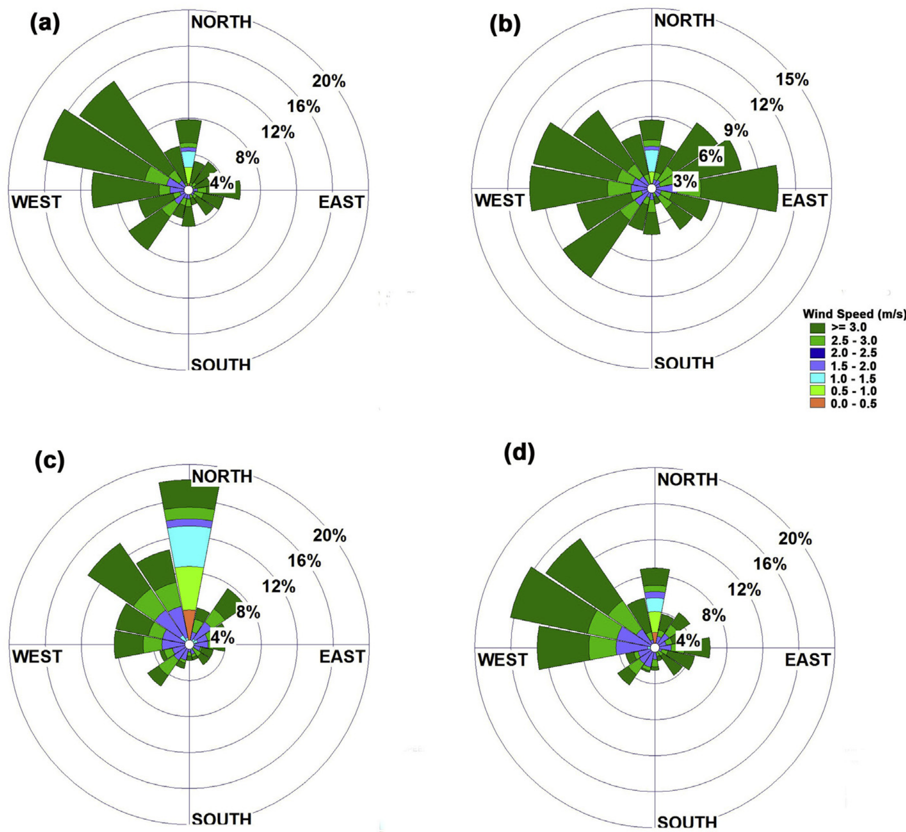

Fig. 4. Wind rose diagrams for Agra for (a) Pre-monsoon, (b) Monsoon, (c) Post-monsoon, and (d) Winter season.

direction variation (Fig. 4). During pre-monsoon season (Fig. 4a), it has been observed that predominant wind direction is north west with wind speeds mainly in the rangers of $\geq$3 ms^{-1}. There is also a westerly and northerly component of wind and even smaller components in north east and south east directions, respectively. In the monsoon season (Fig. 4b), this north westerly component continues to be present over the site. Along with this north west component, strong westerly, easterly and south westerly components are also observed over the site. Wind speed values were on higher side $\geq$3 ms^{-1} in this season as well.

During post-monsoon (Fig. 4c), we can see clearly changing of wind direction over the site. Predominant winds are northerly now with variable wind speeds and components of north westerly winds were also observed. Other directions winds are negligible compare to north to north west sector in post-monsoon season. Situation in winter season (Fig. 4d) is similar to pre-monsoon season (Fig. 4a) with a difference in wind speed patterns over the study site. It is clear from the wind rose diagrams that wind from north west direction is always approaching the Agra. Noticeable thing is that Delhi is situated in this direction (spatial distance ~176 km), and this wind pattern is indicating that Delhi's pollution might be transported to Agra during all seasons. To further investigate this fact, we have to investigate WPSCF results, discussed in next section.

4.5. Back trajectory and WPSCF analysis

Seasonal back trajectory analysis was done over Agra to analyse the aerosol pathways from different sources at an altitude of 500 m above the ground level using HYSPLIT 4 model with use of GDAS 1° data (Draxler and Hess, 1998). We have calculated 48 h air mass isentropic back trajectories at Agra at an interval of 6 h (i.e. 00, 06, 12 and 18 UTC of every day) which emphasizes the transport of aerosols from different regions (not shown here). WPSCF plots for 0.5° resolution has been generated for both RSPM and SPM by using threshold values of 60 μg/m^3 and 100 μg/m^3 as suggested by Indian standards threshold for a clean environment (Table 1). WPSCF plot for RSPM has been shown in Fig. 5 and for SPM has been shown in Fig. 6. Both RSPM and SPM potential origin sources are showing similarities as expected. The weighted potential source regions for values < 0.1 are shaded in light violet colour in both the figures.

For the WPSCF of RSPM (Fig. 5), pre-monsoon (Fig. 5a), post-monsoon (Fig. 5c) and winter (Fig. 5d) are showing potential source regions from north-westerly directions and extent is crossing Delhi in all three seasons. In monsoon season (Fig. 5b), however, we can clearly see the wind reversal at the site and transportation is from south westerly direction over the site. During pre-monsoon and winter season, the aerosols are being transported with winds from Thar Desert (Rajasthan) (Fig. 5a). Due to this reason the aerosol concentration is high during March–May. In monsoon season, Agra is covered by the marine air from the Arabian Sea and Bay of Bengal (Fig. 5b) as well as rainfall during monsoon washes out pollution in the atmosphere, which may be attributed to lower concentrations. In post monsoon season (Fig. 5c); the prevailing winds from local sources as well as from Indo-Gangetic plain (IGP) play a prominent role in elevating the pollutant concentration over Agra. Biomass burning, coal, and fossil fuel combustion contribute to the high aerosol load during post-monsoon season (Pachauri et al., 2013). During winter (Fig. 5d), air mass transportation to Agra is not very different to post monsoon season, but additionally, air masses from Thar desert

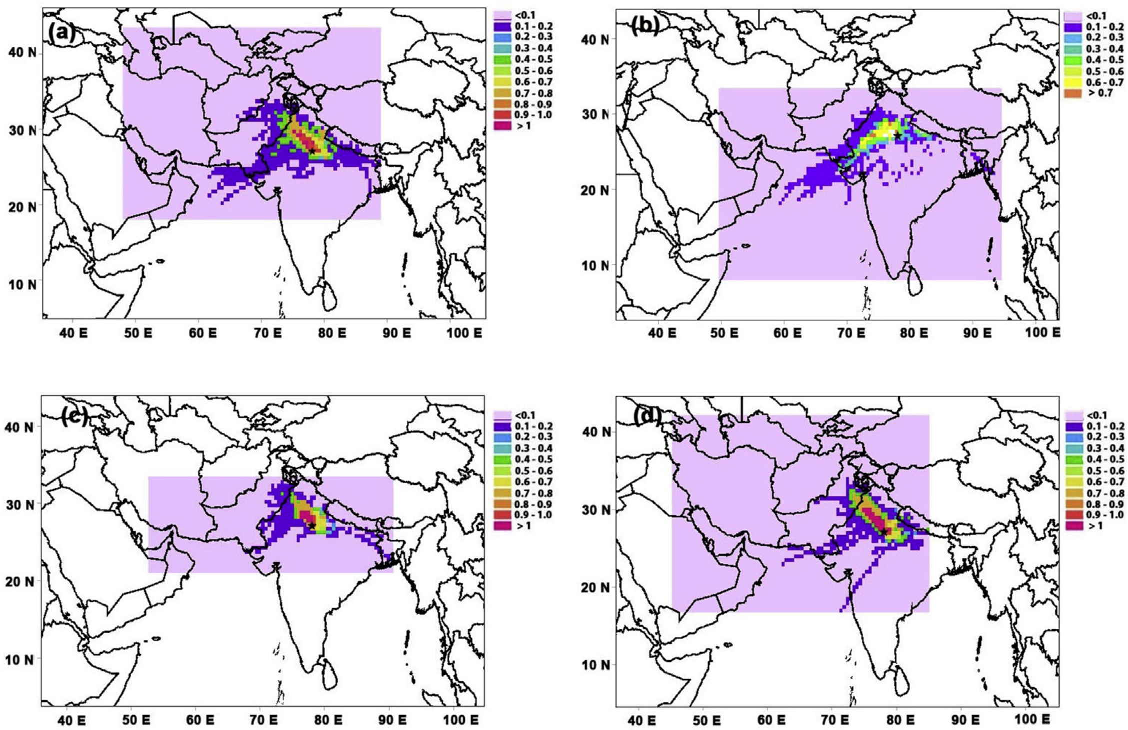

Fig. 5. Maps of weighted Potential Source Contribution Function (WPSCF) values for RSPM for (a) Pre-monsoon, (b) Monsoon, (c) Post-monsoon and (d) winter season using 500 m AGL, 48-h isentropic back trajectories.

(parts of Rajasthan) also arrive at Agra depicting a significant growth in the pollutants concentration.

WPSCF of SPM (Fig. 6) during different seasons is showing similarity with that of WPSCF for RSPM (Fig. 5). Higher SPM values are observed during pre-monsoon season (Fig. 6a) which verifies our hypothesis of dust transport from the aforesaid regions to Agra

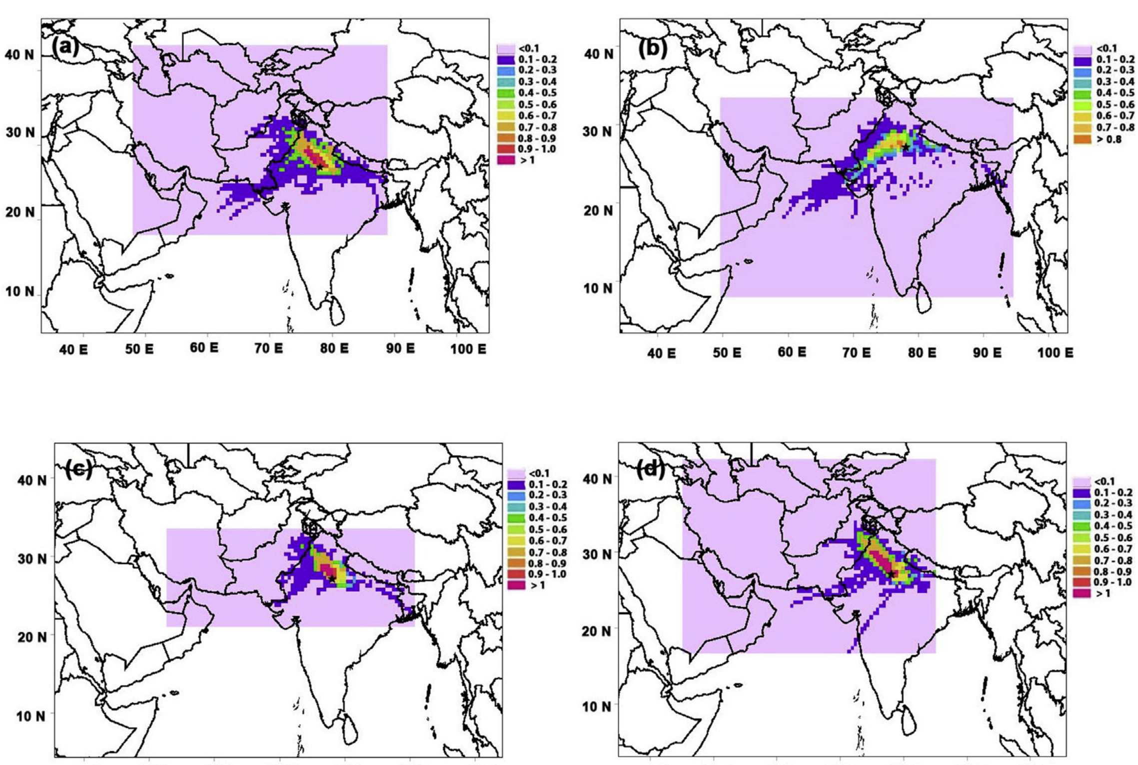

Fig. 6. Maps of weighted Potential Source Contribution Function (WPSCF) values for SPM for (a) Pre-monsoon, (b) Monsoon, (c) Post-monsoon and (d) winter season using 500 m AGL, 48-h isentropic back trajectories.

during these months. SPM values are showing high range in WPSCF plot for monsoon season (Fig. 6b) compare to RSPM and this can be understood as transport of coarser particles from the central and western parts of India in comparison to fine particles.

The results are clearly showing that potential sources for the contribution of RSPM and SPM are not local in nature and most of the pollutants are being transported from the north-westerly direction. Metropolitan city Delhi is in north-west direction to Agra (at a spatial distance of ~176 km) and results indicate that highly polluted air from Delhi is being transported to Agra in all the seasons. The high levels of RSPM and SPM at study area are also indicating that all the practices to decrease locally generated pollution over the area are not helpful in maintaining low levels of pollution as expected.

4.6. Dispersion analysis using HYSPLIT

To provide a conclusive statement on pollution dispersion towards the city of Agra, we used HYSPLIT dispersion modeling for showing examples of spatial distribution of pollution dispersion during the different seasons (Monsoon, pre-monsoon, post-monsoon, and winter). We choose case studies (selected randomly) during the study period over Agra and focus on release of a unit mass dispersion considering particles for selected cases of pre-monsoon (28 April 2014), monsoon (12 August 2014), post-monsoon (20 October 2014), and winter (16 January 2015). Backward dispersion has been computed for 24 h period with averaging period of 6 h each using online version of HYSPLIT (Rolph, 2016). Results are shown in Fig. 7 for pre-monsoon and monsoon and in Fig. 8 for post-monsoon and winter. Agra station is shown by a star symbol in the figures and 4 concentric circles of 100 km each are marked in all the figures.

If we see the dispersion trend for pre-monsoon (Fig. 7a–d), the 6 h averaging in Fig. 7a is clearly showing plume is arriving from north-west direction. Going back in time for 12 h, 18 h and 24 h (Fig. 7b–d) respectively, it is confirmed that direction of plume dispersion is from north-west. After 12 h averaging point in Fig. 7c and d the plume is crossing the second concentric circle which is 200 km and that means the pollution plume is crossing Delhi around 12–18 h back. Before 18 h back, plume is advecting even further northwest to the site as seen in Fig. 7d but the concentration was less as expected. During monsoon, however, the plume is dispersing from west to north westerly direction (Fig 7e–g). Interesting thing to note here is that even after 18 h averaging backward in time, the concentrations were high compare to pre-monsoon season, which may be attributed to cloudy conditions and advecting of pollutants in horizontal direction with less mixing in upward directions over the area. These results are matching with the results of RSPM and SPM PSCF analysis shown in Figs. 5 and 6, respectively.

In case of post-monsoon and winter also (Fig. 8), the results are showing that a 24 h backward dispersion is coming from north-west direction. In the case of post-monsoon (Fig. 8a–d) shows that plume dispersed has less horizontal extent, whereas during winter season (Fig. 9a–d) the concentrations are higher up to 24 h averaging. The results are indicating that higher concentrations are advecting in winter season as observed in PSCF analysis. The results are showing that direction is north-west in case of pre-monsoon, post-monsoon, and winter, whereas plume is advecting from west-northwest in monsoon season. All four seasons, the plume is advecting passing the Delhi and nearby region before reaching to Agra, which were also observed in PSCF analysis (Figs. 5 and 6) and wind roses (Fig. 4) over the site.

4.7. Periodicity test using wavelet approach

Above findings further motivated us to crosscheck the spatial variation of periodicity in the SPM and RSPM catalogue (2011–2015). The wavelet power spectrum using Morlet wavelet for a Gaussian white noise normalised by standard deviation has been shown in Fig. 9. We noticed there is a dominance of semi-annual and annual periodicity in both SPM and RSPM at Taj site (Fig. 9a–d). Annual periodicity is observed for the Itmad-ud-Daula site in both SPM and RSPM (Fig. 9b and e), compare to semi-annual periodicity which is only observable for RSPM. Similarly, at Rambagh site also, both semi-annual and annual periodicities are clearly observable for SPM (Fig. 9c), while RSPM has the only semi-annual periodicity (Fig. 9f). We have not performed analysis at

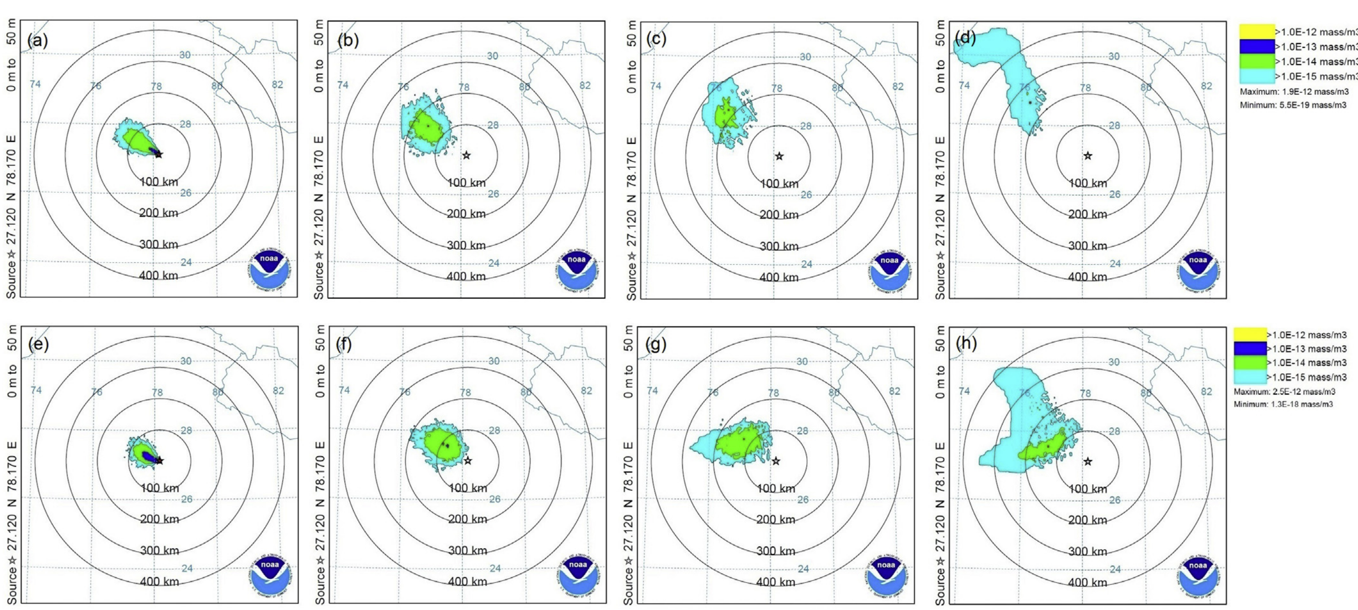

Fig. 7. Dispersion maps using HYSPLIT for Pre-monsoon (28 April 2014) shown in Figures (a) - (d), and Monsoon (12 August 2014) shown in figures (e) - (h), starting at 12 UTC of day averaged for 06 h back ((a) and (e)), 12 h back ((b) and (f)), 18 h back ((c) and (g)), and 24 h back ((d) and (h)) in time. Symbol star denotes the Agra site location.

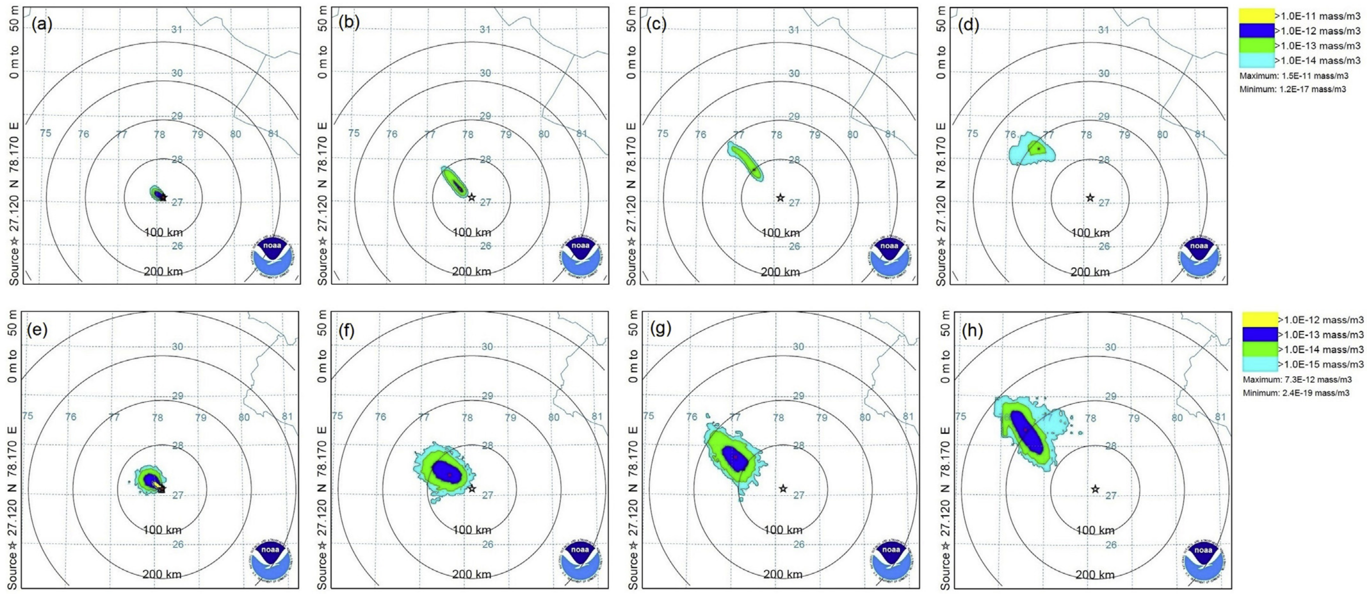

Fig. 8. Dispersion maps using HYSPLIT for Post-monsoon (20 October 2014) shown in Figures (a) - (d), and Winter (16 January 2015) shown in figures (e) - (h), starting at 12 UTC of day averaged for 06 h back ((a) and (e)), 12 h back ((b) and (f)), 18 h back ((c) and (g)), and 24 h back ((d) and (h)) in time. Symbol star denotes the Agra site location.

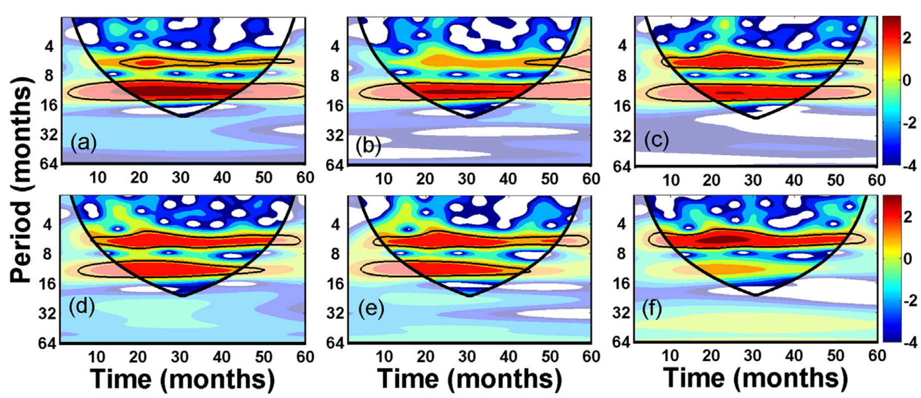

Fig. 9. Wavelet power spectrums for Taj [(a) SPM and (d) RSPM], Itmad-ud-Daula [(b) SPM and (e) RSPM] and Rambagh [(c) SPM and (f) RSPM] using, Morlet mother wavelet. Black contour represents 95% confidence level for white noise.

Nunhai as data is too sparse and not suitable for monthly averages. However as the sites are not very far from each other in geographical locations, we can expect the existence of semi-annual and annual periodicity in SPM and RSPM variations over the region.

5. Conclusions

We have analysed the daily average concentration variations of RSPM and SPM over Agra for the period 2011–2015. Four stations Taj, Itmad-ud-Daula, Rambagh and Nunhai were considered to observe the daily average variation, seasonal variation of RSPM and SPM. The analysis depicted that the average annual concentrations of RSPM (164.9–245.08 μg/m^3) and SPM (298.15–526.58 μg/m^3) for the four sites were found to be very high when compared to the standards specified by Pollution Control Board of India. Higher values for daily average concentrations of RSPM have been observed in post monsoon and winter season, whereas for SPM higher values are persisting in pre monsoon and post monsoon season. Winter season is also accounting the high values of SPM, however, less than post monsoon values. Monsoon has been identified as the season with lowest values of RSPM and SPM at the study region.

There is no trend existing in the time series of SPM and RSPM. The daily average values are still high at all stations and crossing threshold limits for the study period. The results are showing that even after taking measures to reduce pollution at the site by Government of India, high values are persisting over time.

WPSCF analysis has shown that the aerosol transport is mainly due to the winds from urbanized areas and cities in north-westerly direction throughout the year. Some transported pollution is also arriving at the site from Thar Desert of Rajasthan during pre-monsoon season, from the Arabian Sea and the Bay of Bengal in monsoon season, from IGP, adjacent regions and cities during post-monsoon and winter season. The present study is clearly indicating that it is not only locally generated pollution but transported pollution mostly from the nearby industrialized and urbanized areas which are critically affecting the air quality of Agra city. This may be the reason why even after implementing controlling measures by Govt. of India for a cleaner environment in Agra city, air quality over the region is still deteriorating. This fact has been also re-established by dispersion analysis by HYSPLIT for cases chosen for different seasons over the study area. The time series data of

SPM and RSPM shows the existence of annual and semi-annual periodicity for the study period. Present study discovers that the threat to Taj Mahal and residents of Agra still exists because of high concentration levels of pollutants and mitigation measures must be improved not only at local scales but also at nearby regions. However, in order to reach a conclusion, further data collection and analysis at various places in the vicinity to Agra is required along with theoretical and experimental understandings of pollution release and source apportionment studies over the region.

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