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# Long-term (2005–2012) measurements of near-surface air pollutants at an urban location in the Indo-Gangetic Basin

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Simultaneous long-term measurements of near-surface air pollutants at an urban station, New Delhi, were studied during 2005–2012 to understand their distribution on different temporal scales. The annual mean mass concentrations of nitrogen dioxide ($NO_2$), sulphur dioxide ($SO_2$), particulate matter less than 10 μm ($PM_{10}$) and suspended particulate matter (SPM) were found to be $62.0 \pm 27.6$, $12.5 \pm 8.2$, $253.7 \pm 134$ and $529.2 \pm 213.1$ μg/m$^3$, respectively. The 24-hr mean mass concentrations of $NO_2$, $PM_{10}$ and SPM were exceeded on ∼27%, 87% and 99% days that of total available measurement days to their respective National Ambient Air Quality Standard (NAAQS) level. However, it never exceeded for $SO_2$, which could be attributed to reduction of sulphur in diesel, use of cleaner fuels such as compressed natural gas, LPG, etc. The mean mass concentrations of measured air pollutants were found to be the highest during the winter/post-monsoon seasons, which are of concern for both climate and human health. The annual mean mass concentrations of $NO_2$, $PM_{10}$ and SPM showed an increasing trend while $SO_2$ appears to be decreasing since 2008. Air mass cluster analysis showed that north–northwest trajectories accounted for the highest mass concentrations of air pollutants (more prominent in the winter/post-monsoon season); however, the lowest were associated with the southeast trajectory cluster.

**Keywords.** Air quality; particulate matter; NAAQS; Indo-Gangetic Basin; back trajectory; urban environment.

## 1. Introduction

The ubiquitous presence of aerosols in the atmosphere affects Earth's energy budget, directly by scattering and absorbing solar radiation, and/or indirectly through cloud microphysics and thereby precipitation pattern (Ramanathan *et al.* 2001; Poschl 2005), and thus influences climate on a

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local as well as global scale (IPCC 2013). While natural aerosols are dominant and an integral part of the atmosphere on a global scale, the major non-natural aerosols are emitted by various anthropogenic sources like industrial processes, domestic fuel burning, transportation activities, etc. The chemically active aerosols such as oxides of nitrogen ($NO_x$), sulphur dioxide ($SO_2$), carbon monoxide (CO), hydrocarbon, hydrogen fluoride and particulate matter (PM) as well as other organic and inorganic compounds over South Asia have posed major environmental (i.e., air quality), climatic and health concerns in recent decades (Pope and Dockery 2006; Guttikunda and Calori 2013; Pachauri *et al.* 2013; Maji *et al.* 2015; Chowdhury and Dey 2016).

Many Indian metro cities, especially those in the Indo-Gangetic Basin (IGB), are amongst the most polluted cities in the world. This conglomeration of polluted cities in the IGB makes it one of the world's most populated and polluted river basin (Dey and Di Girolamo 2010). New Delhi is one of the urban mega-cities situated in the northwestern part of IGB. The severely degraded air quality of New Delhi has received much attention in recent years as it poses potential health hazards (Maji *et al.* 2015 and references therein). Recent studies have shown that the emission loads over New Delhi and its surroundings are still increasing rapidly. For example, Sahu *et al.* (2011, 2015) computed the emissions of various sources in New Delhi, e.g., PM, CO and $NO_x$ which showed the concentration of about 105, 703 and 255 kt/yr, respectively, for the year 2010. Mishra and Goyal (2015) have also carried out a quantitative assessment of emission of these pollutants, restricted to New Delhi urbanised area (referred to as National Capital Region) for the same year 2010. It was estimated that the emission of PM, CO and $NO_x$ was about 38, 235 and 133 kt/yr, respectively. According to these studies, the transport sector of New Delhi is defined as one of the major contributors to the total loading of air pollutants. Several studies aiming to address air quality of New Delhi in terms of PM emissions, impact of compressed natural gas (CNG), air toxicity and air quality index have been carried out in the recent past, but for a limited near-surface measurements (Goyal and Sidhartha 2003; Kathuria 2005; Kumar and Foster 2007; Bishoi *et al.* 2009; Tiwari *et al.* 2014). However, these studies could not delineate the annual variability of these criteria pollutants, which are indeed essential to chase the mitigation policy and manage current emissions of air pollutants. The present study aims to examine overall intra- and inter-annual behaviour of air pollutants at an urban station, New Delhi using long-term (2005–2012) near-surface measurements of $NO_2$, $SO_2$ and PMs alongside a few meteorological parameters. Furthermore, the present study delineates the relationship of measured air pollutants with the meteorological variables and the possible role of long-range transport to the air quality of the station.

## 2. Methods

### 2.1 *Description and meteorology of the site*

New Delhi (28.6°N, 77.2°E and ∼215 m above mean sea level) is located in the northwestern part of IGB, covering about 1500 $km^2$ and inhabited by about 17 million people (Census 2011). The present study was carried out at the Mayapuri site, situated in the central New Delhi region. The site represents a residential-cum-industrial area with some commercial activities. A population density map of New Delhi, indicating the location of the measurement site, Mayapuri is shown in figure 1. A very high to highest population density area surrounds the measurement site. New Delhi is bounded by the Himalayas in the north, plains of central India in the south and the Thar Desert in the west–southwest. Vehicular and industrial emissions are found to be the highest contributor to total air pollution in New Delhi (CPCB 2012). The contribution of vehicular emissions in New Delhi is estimated to be about 72% of the total air pollution loading (Goyal *et al.* 2006). About 7 million registered vehicles, two coal-fired power plants (Rajghat and Badarpur thermal power station), two gas-fired (Indraprastha gas turbine and Pragati power station) power plants and several small and medium scale industrial facilities are located in New Delhi (Mishra and Goyal 2015). Besides, dust emissions from road dust, construction, mining activities and windblown dust from barren lands also contribute significantly to the total PM loading over this region. Air quality of New Delhi gets worse particularly during the summer season, when this region receives natural dust aerosol from adjacent (e.g., Thar) and/or far-distance (Middle-east) deserts (Srivastava *et al.* 2011, 2014). This region also experiences worse air quality during the early winter season (November–December) due to burning of agricultural biomass residue for land clearing by

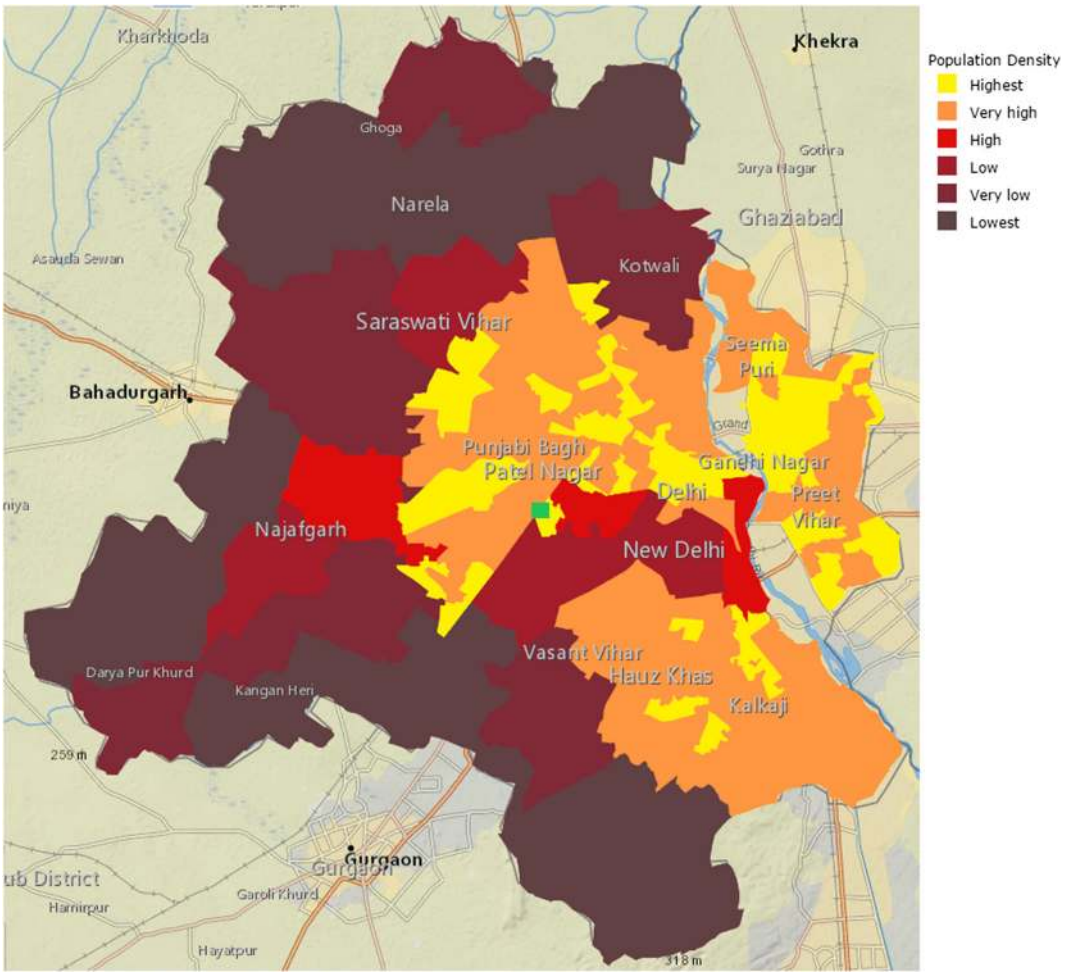


Figure 1. New Delhi region showing the location of the measurement site, Mayapuri (filled green square) and population density (filled colours). https://www.arcgis.com/home/webmap/viewer.html?webmap=9ba9043482e84b608659de3e1f7fc9ab.

farmers in northwest states (Haryana and Punjab) (Kaskaoutis *et al.* 2014; Sharma *et al.* 2017), leading to degraded air quality and visibility (Singh and Dey 2012; Tiwari *et al.* 2014).

New Delhi experiences a distinct annual weather pattern cycle with hot summer, humid monsoon and cold and dry winter (Srivastava *et al.* 2012). The northern part of India, especially the IGB, also experiences frequent foggy conditions during the winter season as a result of the fair-weather condition and shallow boundary layer height. Owing to this, air pollutants cannot be dispersed or mix with free troposphere resulting in higher loading at the surface which causes poor visibility. The climate of New Delhi is of typical monsoon-influenced humid subtropical climate and semi-arid, with high to extreme variation between summer and winter temperatures and precipitation patterns. Figure 2 shows the monthly mean near-surface meteorological parameters at New Delhi, obtained from the India Meteorological Department (IMD) over the period 2005–2012. Summer starts in the middle of March and the temperature climbs to greater than 45°C at times from April to June. Monsoon usually arrives around 29th June over this region and the winter weather conditions are marked by fog events and stable atmosphere with the temperature reaching about 2°C at times during December–January. Wind speed is found to be relatively higher during the summer and lower during post-monsoon and winter months. Wind direction shows large variability from east–southeast to west–northwest. The monthly mean visibility is observed to be lower during winter months owing to reduced boundary layer height and stable weather conditions. A slight dip in the visibility during summer months could be attributed to long-range transported dust from nearby or distant deserts (Pandithurai *et al.* 2008; Srivastava *et al.* 2011).

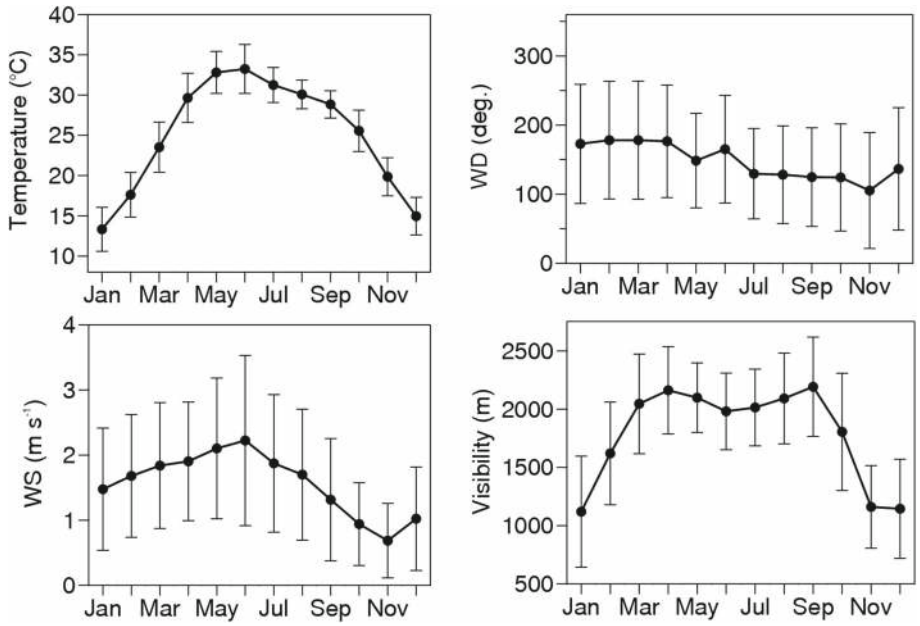


Figure 2. Monthly mean surface meteorological parameters; temperature, wind direction (WD), wind speed (WS) and visibility at New Delhi, averaged over the period from 2005 to 2012.

### 2.2 *Instrumentation and data analysis*

Data presented in this work was collected by Central Pollution Control Board (CPCB) at Mayapuri site in western New Delhi from 2005 to 2012. Several criteria pollutants are being monitored at this site, including $SO_2$, nitrogen dioxide ($NO_2$), respirable suspended particulate matter (RSPM) and suspended particulate matter (SPM). Measurements of these criteria pollutants are not available for latter half of the year 2006, for $SO_2$ during the year 2010 and for SPM for the year 2012 due to instrument malfunctioning. A total of 556 days of dataset was obtained over the entire study period.

RSPM is the particulates with aerodynamic diameter $<10\,\mu m$ size (hereafter referred to as $PM_{10}$). $PM_{10}$ is of great importance as the particles lower than this size can enter into the respiratory system, and therefore, they have received considerable attention in recent times owing to human health concern. $PM_{10}$ data was measured with a gravimetric method using a respirable dust sampler (RDS; Model: Envirotech APM 460). Air is drawn through a glass fibre filter of $20.3 \times 25.4$ cm size at a flow rate of 1.1 $m^3$/min. The mass of collected particle is determined by the difference in filter weights before and after sampling. The concentration of $PM_{10}$ in the chosen size range is calculated by dividing the weight gain of the filter by the volume of air sampled. The sampling was carried out every 8-hr round the clock. SPM is the particulates with a diameter less than $100\,\mu m$, which have a comparatively short lifetime in the atmosphere due to its mass. It is measured by using a high volume sampler (HVS; Model: Envirotech APM 415/430). Similar to $PM_{10}$, the SPM was sampled using a HVS at a flow rate of 1.1−1.5 $m^3$/min.

$SO_2$ is measured using the modified West and Gaeke method and UV-fluorescence method. In this method, $SO_2$ in ambient air was absorbed in a solution of 0.04 M sodium tetrachloromercurate at an average flow rate of 1 l/min. The detection range of the $SO_2$ concentration of $4-1050\,\mu g/m^3$ can be achieved; however, $SO_2$ in the range of $25-1050\,\mu g/m^3$ can only be measured under normal conditions. It is possible to measure concentration less than $25\,\mu g/m^3$ by sampling a larger volume of air, but only if, the absorber efficiency of the particular system is first determined and found to be satisfactory.

$NO_2$ is measured using the modified Jacobs and Hochheiser and chemiluminescence method (Purdue *et al.* 1972), where ambient $NO_2$ is collected by bubbling air through a solution of sodium hydroxide and sodium arsenite, considering $SO_2$ as a major interfering compound. In this technique, chemical reaction derived absorbance of the highly coloured azo dye is measured on a spectrophotometer at a wavelength of 540 nm to determine the $NO_2$ in the sample in the range of $9-750\,\mu g/m^3$. This method has an average bias of ∼3% over the range of $50-300\,\mu g/m^3$.

CPCB carries out quality assurance and quality control (QA/QC) programmes, which is an essential part of any monitoring system. The calibration of the instruments is done using top loading calibrator. In addition, service and repair of the instruments and evaluation of ambient air quality monitoring stations are also done by CPCB under the National Air Quality Monitoring Programme (NAMP). The details of the calibration of the instruments and QA/QC programmes are given elsewhere (CPCB 2012).

Here, we used 8 years (2005–2012) of near-surface measurements of criteria pollutants ($NO_2$, $SO_2$, $PM_{10}$ and SPM) from the Mayapuri site in the central part of New Delhi. We have performed the systematic analysis to examine intra- and inter-annual variability of the measured criteria pollutants. We further investigated the relationship between several weather parameters and criteria pollutants. The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) PC-version model was also used to identify different air mass trajectory clusters based on direction and source region.

## 3. Results and discussion

### 3.1 *Time-series analysis of air pollutants*

Figure 3 shows daily (with dots) and monthly (with solid line) mean time series of the measured near-surface criteria pollutants. The daily mass concentration of $NO_2$ was found to vary from 13.5 to 142.1 $\mu g/m^3$, with a mean value of 62.0 $\pm$ 27.6 $\mu g/m^3$ at New Delhi over the entire time period. For about 27% of data samples of $NO_2$ was found to exceed the corresponding National Ambient Air Quality Standard (NAAQS) level (80 $\mu g/m^3$). It is found that the $NO_2$ levels crossing the safe standard (i.e., NAAQS), particularly in the post-monsoon and winter periods. This could be attributed to the industrial (largely from power plants) along with the vehicular sources (George *et al.* 2013). Furthermore, the daily mass concentration of $SO_2$ was found to vary from 2 and 48.7 $\mu g/m^3$, with a mean value of 12.4$\pm$8.2 $\mu g/m^3$. Surprisingly, the daily mass concentration of $SO_2$ was found to be six times below its 24-hr NAAQS value (80 $\mu g/m^3$). This could be attributed to the use of low sulphur content fossil-fuel, use of fuel gas de-sulphurisation for industrial processes and/or implementation of CNG vehicles. Also, there has been a change in domestic fuel used from coal to LPG which may have contributed to reduction in ambient levels of $SO_2$. However, the former two factors are not widely regulated yet in India. Singh and Dey (2012) also showed the major effect of this policy intervention on the stabilisation of a rapid decrease in atmospheric visibility over New Delhi. More recently, Verma *et al.* (2015) also observed lower mass concentrations of $NO_2$ and $SO_2$ than NAAQS levels in Lucknow,

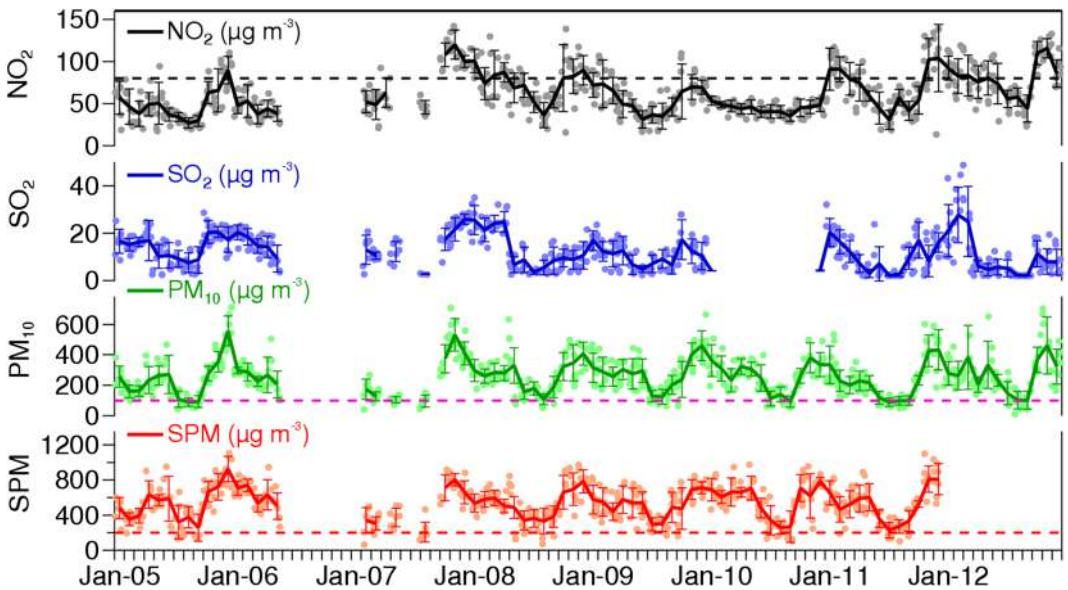


Figure 3. Daily (dots) and monthly (solid line) time series of mass concentrations of $NO_2$ (black), $SO_2$ (blue), $PM_{10}$ (green) and SPM (red) at Delhi during the entire study period. The dotted line indicates 24-hr averaged NAAQS level for $NO_2$ (80 $\mu g/m^3$), $SO_2$ (80 $\mu g/m^3$), $PM_{10}$ (100 $\mu g/m^3$) and SPM (200 $\mu g/m^3$). Error bars indicate standard deviation.

which is about 500 km to the southeast of New Delhi.

Daily mean $PM_{10}$ mass concentration was found to vary between 10.7 and 843 $\mu g/m^3$, with a mean value of $253.7 \pm 134\,\mu g/m^3$ over the entire time period. The $PM_{10}$ was found to be substantially higher than its 24-hr NAAQS value ($100\,\mu g/m^3$) for about 87% of the days of total observation days. SPM was found to vary from 64 to $1104\,\mu g/m^3$ with a mean value of $529.2 \pm 213.1\,\mu g/m^3$, which was higher than its prescribed 24-hr NAAQS value ($200\,\mu g/m^3$) for about 99% of days of the total observation days. It should be noted here that the standard used for SPM for comparison was available before November 2009; however, the new standards for various air pollutants (except SPM) were formed after November 2009 where concentration has been notified as one area class i.e., industrial, residential, rural and other areas (www.cpcb.nic.in). Recent findings indicate that large concentrations of PM in New Delhi have mainly resulted from construction, industrial processes and vehicular emission (Sahu *et al.* 2011; Verma *et al.* 2015). The resuspended dust, associated with the vehicular movement on the road, is also thought to be directly related to a number of vehicles, their weight and speed.

### 3.2 *Monthly and seasonal variation of air pollutants*

Figure 4 shows monthly mean box–whisker plot for mass concentrations of $NO_2$, $SO_2$, $PM_{10}$ and SPM. Box–whisker plot shows statistical variables such as mean, median and percentiles (10th, 25th, 75th and 90th). In general, all air pollutants showed more or less similar monthly mean variations with the highest values during the post-monsoon/winter season and the lowest during the monsoon season. The former is attributed to large rate of anthropogenic emission whereas the latter perhaps due to large wet removal of species. For particulate matter ($PM_{10}$ and SPM), a small secondary peak observed during March–June months is associated with the relative contribution from long-range transported dust aerosols from neighbouring and distant desert regions (Pandithurai *et al.* 2008; Srivastava *et al.* 2011, 2014). $NO_2$ mass concentration was the highest during the post-monsoon season (November, $88.1\,\mu g/m^3$) whereas $SO_2$ was the highest during the winter season (January, $19.7\,\mu g/m^3$). Both $PM_{10}$ and SPM mass concentrations were also high during the early winter (419.4 and $783.3\,\mu g/m^3$, respectively). The highest concentrations during the post-monsoon/winter season could be attributed to several factors such

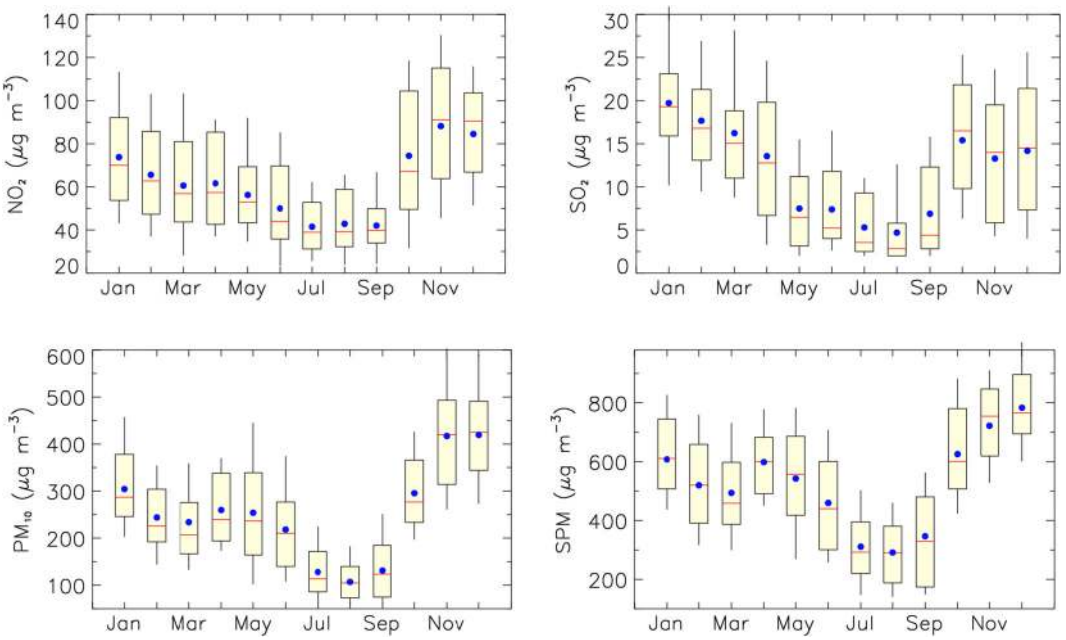


Figure 4. Monthly mean box–whisker plot for $NO_2$, $SO_2$, $PM_{10}$ and SPM over the entire study period. The horizontal solid red line indicates the median, filled blue dot indicates the mean, top and bottom of the box indicate the 75th and 25th percentile respectively, and top and bottom whiskers indicate the 90th and 10th percentiles, respectively.

as enhanced emission sources due to trash/biomass residue burning during this time of the year and stable weather conditions with calm wind and shallow boundary layer height (Tiwari *et al.* 2013a). A stable atmospheric condition restricts ventilation of near-surface air pollutants to free troposphere during the post-monsoon/winter season (Gaur *et al.* 2014). Furthermore, the burning of fire crackers during Deepawali acts as an additional abrupt source of fine PM over the course of a week or less to the atmosphere during post-monsoon season (Attri *et al.* 2001; Singh *et al.* 2003; Barman *et al.* 2009; Tiwari *et al.* 2012a; Singh *et al.* 2014). The major source of air pollutants during the post-monsoon season mainly includes crop residue burning for clearing agricultural fields for the next crop in surrounding states, including Punjab and Haryana (Hyvarinen *et al.* 2010; Kaskaoutis *et al.* 2014; Sharma *et al.* 2017). New Delhi region being in the downwind of these states receives higher loading of air pollutants. Awasthi *et al.* (2011) reported higher concentrations of PM $< 2.5\,\mu$m ($PM_{2.5}$, $69\,\mu g/m^3$) as compared to its NAAQS, which varied from 44 to $147\,\mu g/m^3$ at Patiala, located northwest of Delhi near the foothills of the Himalayas. They found that $PM_{2.5}$ increased substantially (∼78% higher than the background value) during the crop residue burning in the post-monsoon (October–November) season. Badarinath *et al.* (2006) and Kaskaoutis *et al.* (2014) also reported similar findings for the duration of the exhaustive burning of rice crop residue in IGB region during post-monsoon. A previous study done by Tiwari *et al.* (2013b) at New Delhi also showed an intra-seasonal variability of $PM_{2.5}$ mass concentration during 2007–2009. They observed the highest $PM_{2.5}$ concentrations during post-monsoon and winter periods (∼$150\,\mu g/m^3$), which was about 2–3 times higher as compared to the concentrations observed in summer (∼$70\,\mu g/m^3$) and monsoon (∼$45\,\mu g/m^3$). Pipal *et al.* (2014) have performed a comparative study between Delhi and Agra (which is urban location at about 200 km to the south of New Delhi) during the winter period of 2011–2012. Though the observed $PM_{2.5}$ concentrations were higher at both the stations, they were about 27% higher at New Delhi as compared to Agra. On the other hand, secondary organic aerosol, aerosol particles formed from the gas-to-particle and/or particle–particle transformation processes of organic compounds, accounts for a substantial fraction of tropospheric aerosols (Kroll and Seinfeld 2008). Previous studies also report a high frequency of secondary aerosol formation at New Delhi and other sites in India (Hyvarinen *et al.* 2010; Kanawade *et al.* 2014) which can contribute significantly to the total PM in the atmosphere.

Table 1. *Correlation coefficient among all measured air pollutants and meteorological parameters.*

| | $NO_2$ | $SO_2$ | $PM_{10}$ | SPM | Temp | WS | VIS |
|---|---|---|---|---|---|---|---|
| $NO_2$ | 1 | 0.45 | 0.68 | 0.55 | −0.52 | −0.25 | −0.54 |
| $SO_2$ | – | 1 | 0.41 | 0.41 | −0.54 | −0.13 | −0.32 |
| $PM_{10}$ | – | – | 1 | 0.91 | −0.55 | −0.41 | −0.64 |
| SPM | – | – | – | 1 | −0.52 | −0.44 | −0.58 |
| Temp | – | – | – | – | 1 | 0.48 | 0.82 |
| WS | – | – | – | – | – | 1 | 0.52 |
| VIS | – | – | – | – | – | – | 1 |

During monsoon, however, the lowest concentrations of all pollutants are linked to either rapid dispersion due to high wind speed or due to rain wash out processes (Tiwari *et al.* 2012b). Previous studies also linked the variation in seasonal concentration of air pollutants with meteorological parameters (e.g., wind direction, mixing layer) (Goyal and Sidhartha 2002; Tiwari *et al.* 2013b; Gaur *et al.* 2014). The criteria pollutants considered in this study showed reasonable negative correlation with temperature, wind speed as well as visibility (table 1), indicating that meteorology plays an important role in the dispersion and transformation of air pollutants in the atmosphere. The correlation coefficient of all the pollutants with temperature and visibility (except for $SO_2$) is about −0.5; however, it is relatively less negatively correlated (about −0.3) with the wind speed during the study period. Wallace and Kanaroglou (2009) reported that correlation between temperature and $PM_{2.5}$ increased by about 54% during the night and decreased by about 14% during the daytime. This is perhaps due to the low-level inversion and lower temperature. The local-to-regional scale weather pattern may also change the temporal and spatial variation of air pollution levels. Winds, one of the crucial parameters, play a key role in dispersion and rigorous mixing of pollutants within the atmosphere. Strong winds are supposed to flush the air pollutants, and low winds are supposed to allow pollution to stagnate within the region itself. Tiwari *et al.* (2013b) separated $PM_{2.5}$ mass concentrations in different wind speed conditions such as calm conditions, wind speed $\leq 5$ m/s and wind speed

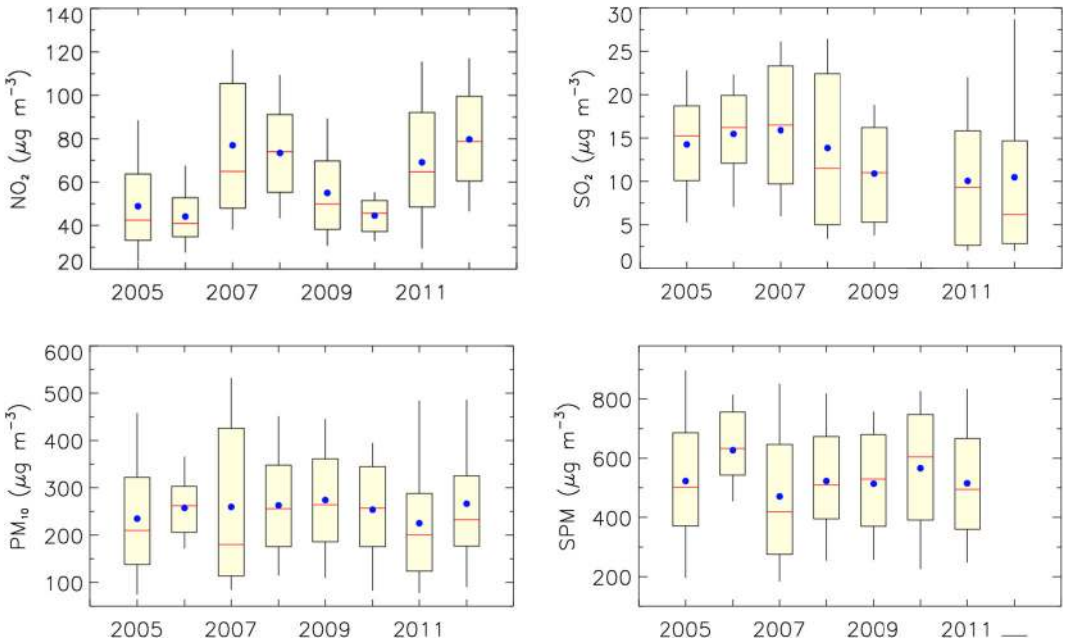


Figure 5. Yearly mean box–whisker plot for $NO_2$, $SO_2$, $PM_{10}$ and SPM over the entire study period. The horizontal solid red line indicates the median, filled blue dot indicates the mean, top and bottom of the box indicate the 75th and 25th percentile, respectively, and top and bottom whiskers indicate the 90th and 10th percentiles, respectively.

≥5 m/s. They observed that the concentrations of $PM_{2.5}$ lowered effectively with increasing wind speed indicating the dilution of pollutants through dispersion. Similar findings were also reported from other parts of world including Sharma *et al.* (2002) in Canada, Cao *et al.* (2009) in China, Ramsey *et al.* (2014) in Oklahoma City, Cheng and Lam (1998) in Hong Kong, Ruellan and Cachier (2001) in Paris and many others. Furthermore, modelling efforts have also been put forward to understand the influence of meteorological parameters on the dispersion of air pollutants (Kesarkar *et al.* 2007; Elshazly *et al.* 2012; Mishra and Goyal 2016).

### 3.3 *Inter-annual variability of air pollutants*

Figure 5 shows inter-annual variability of measured near-surface air pollutants with box–whisker plot, with statistical variables such as mean, median and percentiles (10th, 25th, 75th and 90th). The present observations show a rise in $NO_2$, RSPM and SPM levels whereas a decreasing trend is observed in $SO_2$. A decreasing trend in $SO_2$ may be attributed to various interventions that have taken place over the last decade such as shutting down of hazardous industries, reduction of sulphur content in petrol and diesel, use of cleaner fuel for transport (such as CNG) and domestic (such as LPG) purposes (Saxena *et al.* 2012; Gurjar *et al.* 2016). However, at the same time the number of vehicles has increased exponentially which is one of the major sources of $NO_2$ and other pollutants. A recent study based on ozone monitoring instrument retrieved tropospheric $NO_2$ column data showed that there is clearly an increasing trend in $NO_2$ concentration over Indian industrial sector, mainly reflecting the rapid expansion of the power sector as a result of fast population and economic growth (Krotkov *et al.* 2016). It could be that $NO_2$ emission from coal-fired power plants is not entirely regulated in India and the installation and operation rates of $NO_2$ emission control devices are also very low.

The ambient concentrations of $PM_{10}$ and SPM have been higher than NAAQS for more than a decade now and appear to show a gradually increasing trend. There are several anthropogenic sources of PM to the atmosphere such as industrial process, domestic coal burning, thermal power plants, vehicular emissions, biomass (crop residue, cow dung, etc.) burning and construction activities. The other natural sources may include road dust, wind-blown soil dust or desert dust. Previous studies estimated that industrial processes (including thermal power plants) contribute to about 80% of $PM_{10}$ emission in New Delhi whereas vehicular emissions contribute only about 15% (Reddy and Venkataraman 2002; Gurjar *et al.* 2004). Among vehicular emissions, diesel driven vehicles were found to be the major contributor to total PM,

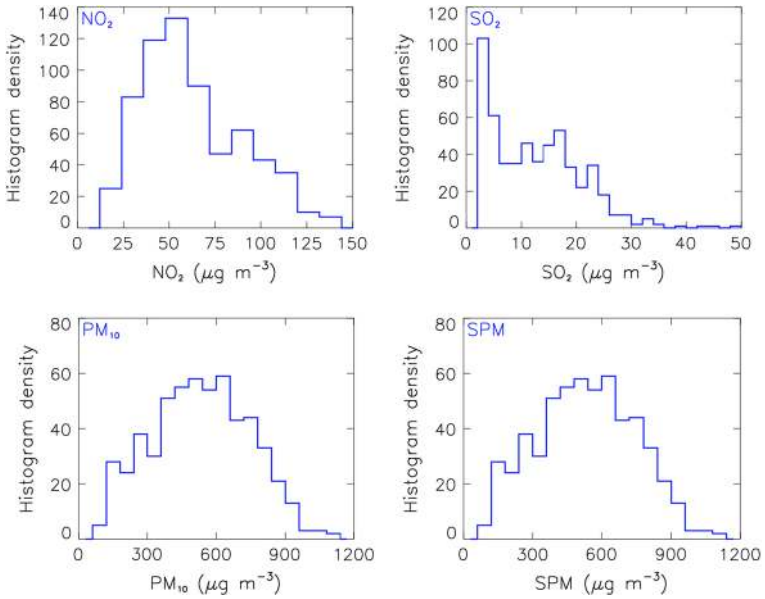


Figure 6. Histogram density plots for $NO_2$, $SO_2$, $PM_{10}$ and SPM at Delhi over the entire study period.

with emission ranging from 3% to 22% as per the auto fuel policy report (Mashelkar *et al.* 2002). In addition to these primary sources, burning of municipal solid waste has recently emerged as one of the major sources of PM, particularly in cites of developing nations (Guttikunda *et al.* 2014; Wiedinmyer *et al.* 2014). In the present study, information about total registered motor vehicles in Delhi was obtained from the Offices of State Transport Commissioners/UT Administrations (https://data.gov.in) during the study period from 2005 to 2012 and shown in figure S1 of the supplementary material. A gradual increase in vehicle number density over the station during the study period appears to be an important cause for the observed increased concentration of gases and PMs. Other less significant sources, but may be important include non-exhaust particles from electric vehicles (Timmers and Achten 2016) and corrosion of road pavements/vehicle components.

Figure 6 shows histogram density plots for daily mean mass concentrations of $NO_2$, $SO_2$, $PM_{10}$ and SPM over the entire study period. For $NO_2$, about 66% of data points lie between 25 and 75 $\mu g/m^3$, with only about 5% of data values less than 5 $\mu g/m^3$. For $SO_2$, about 38% of data values lie between 10 and 20 $\mu g/m^3$, with about 42% of data points less than 10 $\mu g/m^3$. For $PM_{10}$, about 38%, 48%, 11% and 2% data points lie in the range of 1–200, 200–400, 400–600 and 600−800 $\mu g/m^3$, respectively. For SPM, about 7%, 21%, 33% and 28% of data points lie in the range of 1–200, 200–400, 400–600 and 600−800 $\mu g/m^3$, respectively. The mass concentration of all the measured air pollutants, except $SO_2$, was observed to exceed from their respective prescribed NAAQS level. However, the range of mass concentration of the highest contribution of $NO_2$ is near its NAAQS level, and the range of highest contribution of $SO_2$ is below its NAAQS. On the other hand, the observed range of mass concentration of the highest contribution of $PM_{10}$ and SPM was found to be subsequently above their respective NAAQS levels.

### 3.4 *Trajectory cluster analysis*

We used PC-version HYSPLIT model (Draxler and Rolph 2010) to calculate 5-day air mass backward trajectories starting from New Delhi at 11:30 am local time at 500 m above ground level, which is being the lower mixed layer and to avoid low-level terrain. Gridded wind field data from the Global Data Assimilation System have been used with a spatial resolution of $1^\circ \times 1^\circ$ and a time resolution of 1 hr (Kanamitsu 1989). Figure 7 shows four different air mass trajectory clusters over the entire time period only for those days when measurements are available; namely 'north–northwest (N–NW)', 'west–southwest (W–SW)', 'southeast (SE)' and 'local slow moving (LSM)' air trajectories. LSM trajectories are defined as those are in general vicinity of the measurement site. Air mass backward trajectories are those encountered diverse

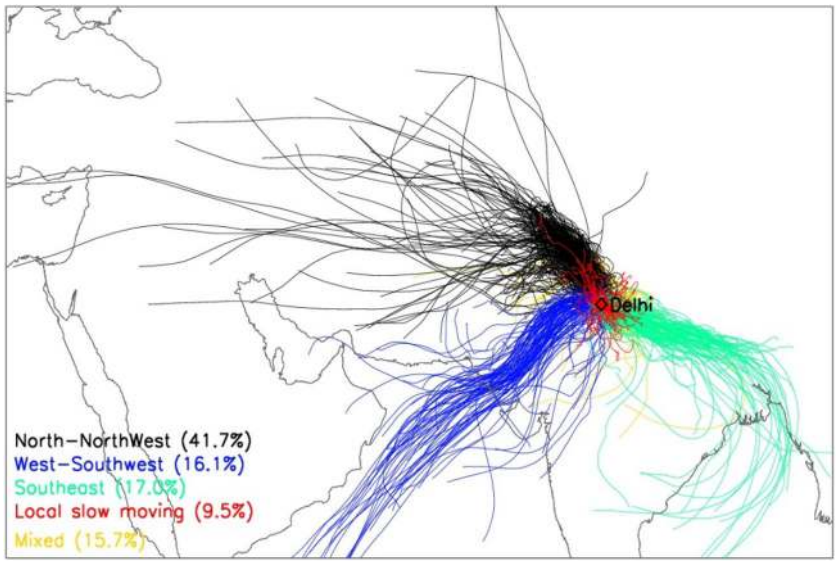


Figure 7. HYSPLIT calculated 5-day air mass backward trajectory clusters over entire time period for which measurements are available. Air mass back trajectory cluster obtained based on direction from which air mass is coming, and those could not be identified are indicated by 'mixed' cluster (yellow colour).

Table 2. *Calculated mean mass concentrations of criteria pollutants for observed trajectory clusters at Delhi.*

| | Trajectory occurrence (%) | $NO_2$ ($\mu g/m^3$) | $SO_2$ ($\mu g/m^3$) | $PM_{10}$ ($\mu g/m^3$) | SPM ($\mu g/m^3$) |
|---|---|---|---|---|---|
| N–NW | 41.7 | 71.9±29.6 | 15.4±8.6 | 314.2±122 | 608.8±190.2 |
| W–SW | 16.1 | 25.1±27.5 | 3.6±5.5 | 102.4±130.8 | 209.2±257.3 |
| SE | 17.0 | 23.3±26.6 | 2.92±5.1 | 69.1±92.6 | 141.4±200.1 |
| LSM | 9.5 | 56.9±40.8 | 9.8±8.1 | 241.8±188.8 | 438.2±336.1 |
| M | 15.7 | 50.5±31.2 | 10.7±8.8 | 198.2±128.5 | 408.0±268.2 |

N–NW: north–northwest; W–SW: west–southwest; SE: southeast; LSM: local slow moving; M: mixed.

source regions grouped as additional 'mixed' trajectory cluster. New Delhi received air mass from N–NW direction on about 42% days of total observation days, higher compared to any other directions. However, air mass trajectory cluster was contributed to be lower (9.5%) over the station for LSM. For each of these clusters, we have further calculated mean mass concentrations for air pollutants (table 2). The higher mass concentrations for all pollutants were associated with the trajectories from N–NW region, having the highest mean mass concentrations of $NO_2$ ($71.9 \pm 29.6\,\mu g/m^3$), $SO_2$ ($15.4 \pm 8.6\,\mu g/m^3$), $PM_{10}$ ($314.2 \pm 122\,\mu g/m^3$) and SPM ($608.8 \pm 190.2\,\mu g/m^3$). It is not surprising as the wind direction is mostly N–NW during the winter/post-monsoon season. To substantiate this we also analysed trajectory data on a seasonal basis during the entire study period and is shown in figure S2(a–d) of the supplementary material. The figure clearly shows that the highest contribution (∼60% of the total observation days) of air masses reaching over the station is from N–NW direction during the winter/post-monsoon seasons. This further suggests an important role of meteorology in the dispersion of air pollutants, as discussed in table 1. The lowest mean mass concentrations of air pollutants were observed to be associated with the SE trajectory cluster, which could be due to wet scavenging of air pollutants during the monsoon season.

## 4. Conclusions

Simultaneous measurements of various criteria pollutants such as $NO_2$, $SO_2$, $PM_{10}$ and SPM were carried out at an urban mega city, New Delhi in the western IGB during the period from January 2005 to December 2012. The present study provided long-term investigations of mass concentrations of

these air pollutants and their association with surface weather parameters and air mass trajectories over the station. The major findings of the present study are as follows:

- The mean mass concentrations of $NO_2$, $SO_2$, $PM_{10}$ and SPM at New Delhi were observed to be about $62 \pm 28$, $12 \pm 8$, $254 \pm 134$ and $529 \pm 213\,\mu g/m^3$, respectively, with significant intra- and inter-annual variability over the entire study period.
- Mean mass concentrations of $NO_2$, $PM_{10}$ and SPM were found to exceed by ∼27%, 87% and 99%, respectively, compared to their prescribed NAAQS levels. However, the mass concentration of $SO_2$ never exceeded its NAAQS.
- The measured criteria pollutants showed a reasonable negative correlation (about −0.5) with temperature and visibility; however, relatively less correlation was observed with the wind speed (about −0.3), suggesting that the meteorology plays an important role in dispersion and transformation of air pollutants.
- Air mass cluster analyses suggested that the maximum air mass contribution (∼42% of the total observation days) was from N–NW direction, which was more prominent during the winter/post-monsoon season (∼60% of the total observation days), and was associated with higher loading of all air pollutants at New Delhi urban site. However, lower mean mass concentrations of air pollutants were associated with SE trajectory cluster.

The present study provides insight into near-surface air pollutants at an urban mega-city New Delhi site and their relationship with surface weather parameters. Air quality has become a major challenge in recent times, particularly in developing nations, like India with increasing energy demand due to large population growth. Long-term exposure to bad air quality has potential threat not only to human health but also to the climate and thus our ecosystem. Thus, precautionary measures are needed to chase the mitigate policy and manage current emissions of air pollutants.

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