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Exceedances and trends of particulate matter (PM$_{2.5}$) in five Indian megacities

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HIGHLIGHTS

• Monthly mean PM$_{2.5}$ levels have declined over Indian megacities during 2014–2019.
• Pervasive anthropogenic activities with meteorology govern peak diurnal PM$_{2.5}$ values.
• Winter months are the most polluted across five megacities and monsoon is cleanest except for Chennai.
• PM$_{2.5}$ levels during monsoon months are 65%–85% lower than the winter months.
• The number of days exceeding the national air quality standards is in decline.

GRAPHICAL ABSTRACT

ABSTRACT

Fine particulate matter (PM$_{2.5}$) is the leading environmental risk factor that requires regular monitoring and analysis for effective air quality management. This work presents the variability, trend, and exceedance analysis of PM$_{2.5}$ measured at US Embassy and Consulate in five Indian megacities (Chennai, Kolkata, Hyderabad, Mumbai, and New Delhi) for six years (2014–2019). Among all cities, Delhi is found to be the most polluted city followed by Kolkata, Mumbai, Hyderabad, and Chennai. The trend analysis for six years for five megacities suggests a statistically significant decreasing trend ranging from 1.5 to 4.19 μg/m$^3$ (2%–8%) per year. Distinct diurnal, seasonal, and monthly variations are observed in the five cities due to the different site locations and local meteorology. All cities show the highest and lowest concentrations in the winter and monsoon months respectively except for Chennai which observed the lowest levels in April. All the cities consistently show morning peaks (~08:00–10:00 h) and the lowest level in late afternoon hours (~15:00–16:00 h). We found that the PM$_{2.5}$ levels in the cities exceed WHO standards and Indian NAAQS for 50% and 33% of days in a year except for Chennai. Delhi is found to have more than 200 days of exceedances in a year and experiences an average 15 number of episodes per year when the level exceeds the Indian NAAQS. The trends in the exceedance with a varying threshold (20–380 μg/m$^3$) suggest that not only is the annual mean PM$_{2.5}$ decreasing in Delhi but also the number of exceedances is decreasing. This decrease can be attributed to the recent policies and regulations implemented in Delhi and other cities for the abatement of air pollution. However, stricter compliance of the National Clean Air Program (NCAP) policies can further accelerate the reduction of the pollution levels.

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1. Introduction

Air quality in megacities is a major concern for human health where a large portion of the population lives, and the pollution levels often
exceed the limit values (Kumar et al., 2015; Zheng et al., 2017). Among all pollutants, PM$_{2.5}$ (particles less than 2.5 μm in diameter) poses a greater risk as it can penetrate deep into the human body (Xing et al., 2016). It has been estimated that exposure to outdoor PM$_{2.5}$ is the fifth leading risk factor worldwide and the third leading risk factor in India (GBD 2015 Risk Factors Collaborators, 2016). Globally, exposure to PM$_{2.5}$ accounts for 4.2 million deaths and over 100 million disability-adjusted life-years in 2015 (GBD 2015 Risk Factors Collaborators, 2016). A growing number of epidemiological evidence of acute and chronic impacts of PM$_{2.5}$ on human health, besides its role in perturbing weather and climate (Fuzzi et al., 2015), has led the scientific community to monitor levels of PM widely across urban, suburban and rural regions of different countries in the last decade. However, inter-comparison of these results is not always possible either because of the difference in sampling or monitoring instrument or due to different sampling duration. This requires a sampling network that works on one principle with large spatial coverage.

In recent times, United States Environment Protection Agency (USEPA) has come up with PM$_{2.5}$ monitoring at the U.S. Embassy and Consulates (USEC) in various countries using Federal Equivalent Method (FEM) approved instrument Beta Attenuation Monitor (BAM-MetOne 1020) and is providing hourly measurements of PM$_{2.5}$ in 27 countries. This USEC data has been used for the study of PM$_{2.5}$ levels in the urban environment for different purposesviz., to study the trend and characteristics of PM$_{2.5}$ (Chen et al., 2020; Fontes et al., 2017; Seekankh et al., 2018; Liang et al., 2016; Battenier et al., 2016; San Martini et al., 2015), to compare with other data and model evaluation (Jiang et al., 2015; Li, 2020; Matthias et al., 2017; Mukherjee and Toohey, 2016; Shimadera et al., 2016; Uno et al., 2014; Wang et al., 2018), and to estimate the health impacts (Han et al., 2020; Lowsen and Conway, 2016; Luong et al., 2020; Nhung et al., 2020; Tian et al., 2020; Wang et al., 2020; You et al., 2016; Zhang et al., 2020). While most of the studies have been carried out in China, few studies have been carried out for other countries including Vietnam (Hien et al., 2019; Luong et al., 2020), Japan (Shimadera et al., 2016) Indonesia (Kusuma et al., 2019), Mongolia (Hill et al., 2017), Bangladesh (Auvee and Bashar, 2019), and Singapore (Liu and Salvo, 2018).

In India, monitoring of PM$_{2.5}$ started in 2013 at the US embassy in national capital New Delhi, and gradually, the network has spread to four more megacities of India viz., Kolkata, Mumbai, Hyderabad, and Chennai at their respective consulates. This data set provides a unique opportunity to study the PM$_{2.5}$ trend in five Indian megacities which are facing PM$_{2.5}$ pollution problems (WHO, 2018). An advantage of using USEC AQ data is that all AQ sites use the same type of monitoring instruments (FEM BAM-1020 manufactured by MetOne Instruments) (MetOne, 2016) and data is processed using a common quality control protocol defined by USEPA (Ray and Vaughn, 2013). This allows for a fair comparison across the sites. Moreover, USEC AQ sites show lower concentration as compared to nearby AQ sites by a local agency (Bhardwaj and Pruthi, 2019). This suggests that the USEC AQ sites are in relatively cleaner areas, therefore unaffected by unregulated and highly localized sources. Additionally, the real-time, as well as archived PM$_{2.5}$ concentration data, are provided to the user community for further analysis to improve the understanding of the variability and trends in PM$_{2.5}$ across different cities.

USEC AQ data for India has been mostly utilized for PM$_{2.5}$ variability and trend analysis (Bhardwaj and Pruthi, 2019; Chen et al., 2020; Dhammapala, 2019; Pommier et al., 2018; Seekankh et al., 2018; Yang et al., 2018). Also, the data has been utilized for comparison with the PM$_{2.5}$ data collected by Central Pollution Control Board (CPCB) in New Delhi (Bhardwaj and Pruthi, 2019), for utilization as ambient values for indoor air quality study in Mumbai (Lueker et al., 2020), and verification of simulated results (Mahesh et al., 2019; Seekankh et al., 2017). Analysis of hourly PM$_{2.5}$ carried out for nearly four years by two different studies at all Indian USEC sites from Jan-2013 to Oct-2016 (Seekankh et al., 2018) and four sites excluding Kolkata from Mar-2015 to Dec-2018 (Chen et al., 2020) shows Delhi being the most polluted city. Both the studies report winter to be the most polluted season for all the cities because of increased household emissions (Chowdhury et al., 2019; Guttikunda and Calori, 2013) and lower boundary layer (Sathyanaidh et al., 2017). The lowest concentration was reported in the Indian summer monsoon season due to the wet scavenging of the particles by rain (Tiwari et al., 2013b). A well-developed morning and evening peaks were observed for Chennai and Hyderabad whereas for other cities, evening peak was shown to last longer. A study by Gurjar et al. (2016) found increasing trend in PM over Delhi and no trend over Kolkata and Mumbai for a period of eight years (2005–2012). No notable trend in PM$_{10}$ concentration for 12 years (2004–2015) was reported in a similar study by Pant et al. (2019). In another study by Yang et al. (2018) during 2014–2015, no comparable decline in PM was found in Indian cities. So far, we have not come across any study that has suggested a significant declining trend in air quality in Indian cities despite the measures taken by the local authorities.

Previous studies have covered the spatial variation of PM$_{2.5}$ in terms of diurnal and seasonal trends, yet it has not been detailed for individual cities. Also, the annual trend observed for the cities is either not discussed or when discussed remains inconclusive. Additionally, none of the earlier studies have conducted a detailed exceedance and trend analysis. To address these gaps, we use USEC PM$_{2.5}$ data measured at all USEC sites in India for six years (1-January-2014 to 31-December-2019) to study the variability and trend. In addition, we perform the exceedance and episode analysis for pollution from 2014 to 2019 with respect to the Indian national ambient air quality standards (NAAQS, CPCB, 2009) and World Health Organization standards (WHO, 2005). The results of this study will help to address the effectiveness of the recent initiatives to improve the air quality and to meet the national air quality standards. The results of this work will support the National Clean Air Program (NCAP, MoECC, 2019), which aims to reduce particulates by 20–30% by 2024, keeping 2017 as the base year.

2. Data and methodology

2.1. US Embassy and consulate data

Hourly PM$_{2.5}$ concentrations measured at five locations, US Embassy New Delhi and US consulates in Kolkata, Mumbai, Hyderabad, and Chennai (Fig. 1) have been obtained for six years (1 Jan 2014 to 31 December 2019). The data from 1 March 2015 to 31 December 2019 are available from the AirNow website (https://www.airnow.gov). The record priors to 1 March 2015 has been taken from the consulate website (https://in.usembassy.gov/embassy-consulates/new-delhi/air-quality-data/).

2.1.1. Site description

The geographical location of all five sites along with the local potential emission sources are tabulated in Supplementary Table 1 and are shown in Fig. 1. New Delhi, the national capital of India with more than 16 million population, is in the northern part of India. Kolkata, with a population of over 14 million people, is in the north-eastern part of India. The city is located around 100 km away from the Bay of Bengal. Mumbai, with a population of over 18 million people is located on the west coast (Arabian Sea) and Chennai with more than 8 million population is located on the south-east coast (Bay of Bengal). Hyderabad, an inland city located in the south-central part of India, has a population of over 7 million people. Delhi and Kolkata are also in the Indo Gangetic plains (IGP), which is one of the most polluted regions of India (Ojha et al., 2020).

2.1.2. Climatology of five cities

Delhi has a semi-arid climate with extreme dryness, hot summer, cold and foggy winters, and moderate rainfall during monsoon. The
climate of Mumbai is characterized by hot summers and humidity throughout the year and heavy monsoon rainfall. Kolkata has a humid wet and dry tropical climate with hot and humid summers, moderate winter. Hyderabad has a tropical wet and dry climate with hot and dry summers, humid in other seasons, and moderate winters. Chennai, being close to the equator and near the coast, has humid summers and dampness throughout the year. Chennai receives maximum rainfall during the North-East monsoon season (October to December) (Rajeevan et al., 2012) and other cities receive maximum rainfall during the South-West summer monsoon season (June–September) (Pattanaik and Rajeevan, 2010). The detailed climatology of each city is available at the Indian Meteorological Department (IMD) Climatology of Smart Cities, IMD (2020).

2.1.3. Potential local sources

While meteorology plays an important role in controlling the air quality, the local emission sources mainly household and traffic emissions (Singh et al., 2018a) impact the local air quality. In order to know the potential local sources, the open street maps of the 2 km × 2 km area surrounding the embassy/consulates (red circle) are shown in Fig. 1a–e. Chennai site (Fig. 1a) is influenced by traffic emissions as it is close to a major road intersection. Kolkata and Hyderabad (Fig. 1b, c) sites are surrounded by residential areas where household emissions can be a potential source. Moreover, Hyderabad is also surrounded by major roads on three sides. Mumbai site (Fig. 1d) is surrounded by a road network and mixed sources. However, there is a big public parking lot nearby Mumbai site that can have an impact on the PM$_{2.5}$ levels. The New Delhi site (Fig. 1e) is in the well-managed area having embassies of other countries. The site is close to open roads with moderate traffic.

2.1.4. Monitoring instrument and data quality control

The PM$_{2.5}$ concentrations across all the sites are monitored in real-time using FEM BAM-1020, having a standard range of 0–1000 μg m$^{-3}$, resolution of ±0.1 μg m$^{-3}$ and 24 h average lower detection limit less than 1.0 μg m$^{-3}$, and the data is processed using a common quality control protocol defined by USEPA (Ray and Vaughn, 2013). However, we found that there were still negative values with valid flag and outliers (sudden spikes) present in the data set. Therefore, we have further processed a quality control check to remove the outliers. Any data point which is more than three local scaled median absolute deviations (MAD) from the local median of the data within a running window of 6 h has been considered as an outlier. As the PM$_{2.5}$ data does not have a normal distribution (Supplementary Fig. 1), it would be the best choice to use MAD over standard deviation as a measure of local spread. This method removes 2.1–3.6% of data that had been marked as valid data (Supplementary Table 1 and Supplementary Fig. 2).

2.1.5. Meteorological data

The Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) hourly surface reanalysis meteorological products from 1 January 2014 to 31 December 2019 has been obtained from NASA’s Global Modeling and Assimilation Office (GMAO). These products are available at a horizontal resolution of 0.5° × 0.625° (https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/). Details of the MERRA-2 products and evaluation has been reported by Gelaro et al. (2017), Randles et al. (2017) and Buchard et al. (2017). The hourly meteorological data (wind, temperature, precipitation, planetary boundary layer height-PBLH) have been extracted from the corresponding grids of the latitude and the longitude of the five megacities (Supplementary Table 2). The MERRA-2 wind speed and temperature have been validated against the
surface meteorological observations are taken from the NOAA’s National Climatic Data Center (NCDC) online portal (https://www7.ncdc.noaa.gov/CDO/cdo).

2.2. Method

The air quality data has been analyzed from 2014 to 2019. We have used Seasonal Trend decomposition procedure based on LOESS (LOcally wEighted Scatterplot Smoothing) smoothing (STL) (Cleveland et al., 1990) to estimate the trend in PM$_{2.5}$ as adopted by Bigi and Ghermandi (2016) to study the trend in PM$_{2.5}$ in the Po Valley, Italy. STL is a widely used filtering procedure for decomposing time series into three components: trend, seasonal, and remainder or residual. The decomposition is based on a sequence of smoothing procedures using a locally weighted regression known as LOESS (Cleveland et al., 1990). The LOESS smoother is based on fitting a weighted polynomial regression for a given time of observation, where weights decrease with distance from the nearest neighbor. Time-series of monthly mean PM$_{2.5}$ concentration at all five megacities were decomposed in trend, seasonal, and remainder components using STL procedure (Cleveland et al., 1990). As the PM$_{2.5}$ data is not normally distributed (Supplementary Fig. 4), time-series data were log-transformed before STL decomposition to attain normally distributed residuals and to control heteroscedasticity. Time-series data were back-transformed from logarithmic decomposed data to analyse the trend. A significant slope in the monthly trend component was calculated using Generalized least squares (GLS) regression (Brockwell and Davis, 2002) for each site within a 95% confidence interval (CI) with a significance level (alpha = 0.05). GLS is used to estimate the linear relation between time and observation, where weights decrease with distance from the nearest neighbor.

Exceedance analysis has been performed and the number of threshold exceedances has been calculated by keeping the threshold of daily mean PM$_{2.5}$ equal to 60 μg/m$^3$ and 25 μg/m$^3$ as per NAAQS (CPCB, 2009) and WHO (WHO, 2005) standards respectively. The linear trend (Singh et al., 2018b) in the annual exceedances in six years has been calculated for each site within 90% confidence interval (Cl) with a significance level (alpha = 0.1) because of the small sample size (Labovitz, 1968). We have also calculated the number of pollution episodes and the length of each episode. An episode has been considered when the daily mean PM$_{2.5}$ has exceeded the threshold continuously for three or more days.

3. Results and discussion

3.1. Diurnal, seasonal and monthly variations in PM$_{2.5}$

The variation in the PM$_{2.5}$ levels for a location is an interplay of emissions, geography, and meteorological conditions (Alimisis et al., 2018; Ganguly et al., 2019; Nair et al., 2007). Previously, the diurnal and seasonal variations have been reported for five cities for the period of fewer than four years 2013–2016 by Sreekanth et al. (2018) and for four cities excluding Kolkata for four years (2015–2018) by Chen et al. (2020). However, these studies did not discuss the trend in the PM$_{2.5}$. Various pollution mitigation schemes along with the public awareness programs (NCAP, MoEFCC, 2019) have been implemented in India since the availability of the USEC data. Therefore, we analyzed the data for a longer period of six years for all available USEC sites in India to study the trend, exceedance and variations in details.

The diurnal variation of PM$_{2.5}$ for Chennai, Kolkata, Hyderabad, Mumbai, and New Delhi is presented in Fig. 2 across the four different seasons. The diurnal mean PM$_{2.5}$ values shown in the figure are averaged over the six years (2014–2019). The comparison of the annually averaged diurnal variation across all the cities is shown in Fig. 2(f). A detailed plot of the diurnal variation with the median and 5th and 95th percentile (hourly concentration across six years) is shown in Supplementary Fig. 3. The maximum values are attained in the winter season because the baseline PM$_{2.5}$ values remain high as a result of stagnant atmospheric conditions (Sreekanth et al., 2007; Tiwari et al., 2013b; Tyagi et al., 2017) and increased emissions (Guo et al., 2017, 2019; Schnell et al., 2018). Delhi exhibits the highest PM$_{2.5}$ concentration for all the seasons. For all the cities, monsoon months (JJA) show the lowest PM$_{2.5}$ levels owing to the wet scavenging and washout by rain during the South-West monsoon, except for Chennai, where most of the rainfall occurs in North-East monsoon season.

Fig. 2. Diurnal variation of PM$_{2.5}$ in different seasons for (a) Chennai, (b) Kolkata, (c) Hyderabad, (d) Mumbai, (e) New Delhi, and (f) Annual diurnal variation across all cities. Scales are different.
All the cities consistently show morning peaks around (-08:00–10:00 h). A shift of up to two hours in the morning peak hours is due to the season and the geographical location. For a city, the winter peak appears at a later hour of the day than the summer peak hour because of late sunrise and onset of human activities in winter. Among all, the cities located in the eastern side of India (Kolkata, Chennai, and Hyderabad) show a peak around an hour earlier than the cities located further west (Delhi and Mumbai) because of the early sunrise and human activities in the eastern cities. The cities Hyderabad and Chennai show a sharp peak during morning traffic hours, whereas the same is not true for New Delhi, Mumbai, and Kolkata. The higher levels of PM$_{2.5}$ during nighttime leads to a smaller peak during morning traffic peak hours. A similar study carried out by Chen et al. (2020) has attributed this to the higher population of Mumbai and New Delhi. This may not be the sole reason as the population of Hyderabad and Chennai are also large enough to enhance the nighttime emissions. The traffic sources in the vicinity of the monitoring station (Fig. 1) along with the local meteorology may be responsible for the sharp peak during the morning hours. The morning peak is attributed to the morning fumigation effect after the sunrise (Stull, 2012; Nair et al., 2009), along with morning traffic and household emissions (Tiwari et al., 2013b) trapped within the evolving shallow boundary layer. The diurnal peak of PM$_{2.5}$ occurs in the morning hours for all the cities except for Kolkata where the peak PM$_{2.5}$ is found at midnight. For Kolkata, a similar variation in BC has been reported by Talukdar et al. (2015) who have also shown the highest peak for BC during midnight rather than morning traffic peak time. This could be due to the late evening household emissions. Moreover, higher wind prevalence in the city of sea-breeze during the early morning to afternoon as compared to the other periods of the day (Gururaja et al., 2019) can also explain the lower concentration in the day time as compared to the night in Kolkata.

The lowest level of PM$_{2.5}$ during a day for all the cities is found in late afternoon hours (~15:00–16:00 h) due to higher PBLH (Supplementary Fig. 4) (Sathyanadh et al., 2017) allowing for vertical mixing and dilution of the surface pollutants. In later times, the increased traffic and household emissions along with the decrease in PBLH lead to a rise in PM$_{2.5}$ levels. One distinct feature to notice is that Chennai night-time concentrations are lower than the daytime concentrations whereas Kolkata, Mumbai, and New Delhi have consistently elevated PM$_{2.5}$ levels throughout the nighttime. Chennai site is located in the vicinity of traffic sources (Fig. 1) and the night-time minima in Chennai corresponds to the lowest traffic emissions after midnight (Srimuruganandam and Shiva Nagendra, 2010). This suggests that the night-time minima in Chennai are possibly due to the lower night-time local emissions and efficient dispersion as compared to other cities. The diurnal variations for each city associated with each season are shown in Supplementary Fig. 3. The black lines in each of the subplots show mean PM$_{2.5}$ whereas the other colored lines in each of the subplots show median PM$_{2.5}$ values. The shaded region shows the range of PM$_{2.5}$ values with the lower and upper boundaries correspondingly defined by the 5th percentile and 95th percentile of the hourly concentrations across six years.

Fig. 3 shows the variations in monthly mean, median, and 5th percentile and 95th percentile of the hourly concentrations across six years for the five cities. All cities show the highest and lowest concentrations in the winter and monsoon months, respectively. The winter maxima are associated with increased emissions and lower PBLH (Tiwari et al., 2013b). The PM$_{2.5}$ levels start to decrease as summer begins because of the increased PBLH as a result of strong surface heating. The surface heating starts from Southern India, which also favors the onset of sea-breeze, makes Chennai cleanest in April. In later months, the levels converge to a similar level across all cities in monsoon because of the wet scavenging of the particles. For Delhi, August month is found to be the cleanest month because of the wet deposition of the pollutants and increased soil moisture which leads to less dust resuspension (Singh et al., 2020a). The PM$_{2.5}$ levels in Delhi reach to maximum in November due to the increased biomass burning (Beig et al., 2020; Ravindra et al., 2019a) around Delhi. During this period, the downwind transport of pollutants from the agricultural bio-mass burning region of Punjab and Haryana adds extra particulates either directly or through a gas to particle conversion (Wang et al., 2005; Sharma et al., 2014) leading to pollution episodes (Kanawade et al., 2020).

We also calculated the ratio of the highest to the lowest monthly mean PM$_{2.5}$ concentration for a city to know the extent of the variability.

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**Fig. 3.** Monthly variation of PM$_{2.5}$ for (a) Chennai, (b) Kolkata, (c) Hyderabad, (d) Mumbai, (e) New Delhi, and (f) Annual monthly variation across all cities. Mean (black line) and median (color line) are shown as the line plot, whereas the shaded region shows 5th percentile and 95th percentile of the hourly concentrations across six years for the five cities.
within a year. As the highest pollution levels are found in winter and lowest in monsoon, it can also be considered as the most polluted to the cleanest ratio. This ratio is found to be high for Kolkata (6.98) and Delhi (6.82) followed by Mumbai (4.9), Hyderabad (3.18), and Chennai (2.98). This suggests that for Kolkata and Delhi, the winter months PM$_{2.5}$ levels can be 7 times higher than the monsoon levels.

### 3.2. Trends in PM$_{2.5}$ at five cities

Here we utilize the USEC PM$_{2.5}$ data to calculate the annual trend with the monthly mean PM$_{2.5}$ across all five Indian cities for six years. Monthly mean PM$_{2.5}$ time series at all five locations were decomposed in trend, seasonal and remainder components using STL procedure and the slope in the trend component was calculated using GLS. STL decomposition of the monthly mean PM$_{2.5}$ along with GLS fitted models for the five cities are shown in Fig. 4. The equation shown in the figure depicts the GLS linear regression slope with 95% CI and the same has been shown in Table 1. As can be seen from Fig. 4, all the cities show a significant decline (negative) in trend ranging from 1.5 to 4.19 μg/m$^3$ per year. The highest decline in trend of 4.19 ± 1.12 μg/m$^3$ per year was found for New Delhi whereas Chennai exhibit a declining trend of 2.61 ± 0.52 μg/m$^3$ per year (−7.59 ± 1.51%/year) which is highest in terms of the percentage decline.

We have also checked whether the trend in the PM$_{2.5}$ is affected by trend in the meteorological parameters such as wind speed, PBLH, and precipitation during the six years. These meteorological parameters are obtained from MERRA-2 reanalysis and have been validated (Supplementary Table 2) against the surface observations at the airports. The trend in the meteorological parameters has been calculated in the same way as it was done for PM$_{2.5}$. The calculated trend in wind speed, temperature, PBLH, and annual precipitation is shown in Supplementary Fig. 5. It is found that wind speed, temperature, PBLH and precipitation do not exhibit a significant change during the study period. Therefore, this analysis confirms that the reduction in PM$_{2.5}$ is not due the meteorology but due to the reduction in emissions. Various pollution mitigation schemes along with the public awareness programs (NCAP, MoEFCC, 2019), could have led to the reduction of PM$_{2.5}$ levels in Delhi.

### 3.3. Analysis of PM$_{2.5}$ exceedances and episodes

Air quality standards, guidelines, objectives, targets, and limit values are defined by the local authorities to control air pollution. The levels

![Fig. 4. STL decomposition and GLS regression of monthly mean PM2.5 for the five Indian megacities over a period of six years (2014–2019). The black line shows the STL trend component and the red line shows GLS fitted slope representing the overall trend in PM2.5.](image-url)
below the standard or limit value are considered to be acceptable in terms of the known effects on human health and the environment. An exceedance is defined when the concentration of a pollutant exceeds the air quality standards or limit values. The air quality standards have been defined by various agencies across the globe through air quality directives, acts, or guidelines. The EU's air quality directives (2008/50/EC), US-EPA's clean air act, India's Air (Prevention and Control of Pollution) Act and WHO Air Quality Guidelines set key air pollutant concentrations thresholds and national ambient air quality standards (NAAQS) that should not be exceeded in a given period. The limiting values have been defined over daily and annual scales. The limit values for annual mean PM2.5 are set at 25 μg/m3 by EU (EEA, 2019), 35 μg/m3 by the US (USEPA, 2015), 25 μg/m3 by the UK (UK DEFRA, 2010), 10 μg/m3 by WHO (WHO, 2005), and 40 μg/m3 by Indian NAAQS (CPCB, 2009). The daily mean (24 h average) ambient PM2.5 standards defined by the US, WHO, and India are 35, 25, 60 μg/m3 respectively. EU and UK have not defined daily limits for PM2.5.

In this section, we have analyzed the USEC PM2.5 data to study the number of threshold exceedances of daily mean PM2.5 as per the WHO (25 μg/m3) and Indian-NAAQS (60 μg/m3) values for six years. India is among the list of countries that experience an increase in high-pollution events during the last decade (Pommier et al., 2018). The persistently high levels of PM2.5 for three or more than three days defines an episode of high PM2.5 levels. We have also calculated the number of pollution episodes and the length of each episode. The average number of percentage exceedances, number of episodes, and duration of the PM2.5 pollution episodes calculated for each city are shown in Fig. 5. The number of exceedances for each year is shown in Supplementary Fig. 6.

As can be seen from the yearly plots (Supplementary Fig. 6), all the five cities exceed the WHO standards for more than 50% of days for all the years except for Chennai during 2015, 2018, 2019. Delhi represents the highest number of exceedance days as compared to the other four cities. The exceedance days are relatively higher for all inland cities and lower for coastal cities Chennai and Mumbai. The Indian NAAQS exceedances are also found to be highest for Delhi where more than

| City      | Trend (μg/m³ per year) | Trend (%/year) | P-value |
|-----------|------------------------|----------------|---------|
| Chennai   | −2.61 ± 0.52           | −7.59 ± 1.51   | 0       |
| Kolkata   | −1.81 ± 0.82           | −2.24 ± 1.02   | 0       |
| Hyderabad | −1.30 ± 0.71           | −2.83 ± 1.35   | 0.0001  |
| Mumbai    | −1.79 ± 0.96           | −3.16 ± 1.69   | 0.0004  |
| New Delhi | −4.19 ± 1.12           | −3.71 ± 0.99   | 0       |

Fig. 5. Variability of PM2.5 episodes and their related statistics that exceeded the PM2.5 values corresponding to WHO standards (25 μg/m³) and Indian-NAAQS (60 μg/m³) during the 2014–2019 period for the five Indian megacities.
200 days exceeds the limit values of 60 μg/m³. Chennai experienced the lowest number of exceedances. The number of exceedances is found to be around 100–150 days for other cities. Previously, Gordon et al. (2018) have also mentioned that for more than half of the National Ambient air quality Monitoring Program (NAMP) stations of India measured PM_{2.5} and PM_{10} values that routinely exceeded Interim Target-1 levels (75 and 150 μg/m³ for daily and 35 and 70 μg/m³ annually, respectively). A simulation study using WRF-Chem for India (Bran and Srivastava, 2017) for the year 2008 found PM_{2.5} mass concentration 2–3 times higher than Indian NAAQS and WHO standards. Moreover, remote sensing based study by Dey et al. (2012) has shown that 51% of the Indian population is exposed to the levels that exceed the WHO annual air quality threshold of 35 μg/m³.

Fig. 5 describes the statistics of the occurrence of such episodes in terms of exceedance of days in comparison with WHO and Indian NAAQS. Higher the number of days in a pollution episode, the higher is the chances of chronic exposure for the people. The plot in Fig. 5 (a) shows the percentage of days for the respective cities which exceeded the threshold of WHO and Indian standards. The number of exceedances is highest for Delhi and lowest for Chennai. Fig. 5b shows the number of episodes for a city, whereas Fig. 5c, d describe mean and median duration of episodes respectively. Chennai and New Delhi show the contrasting result in terms of the highest and lowest number of episodes respectively, that exceeded the WHO standards. Sometimes, the number of episodes can be misleading as shorter episodes can increase in the number as it is seen in Chennai. The number of episodes must be combined with the duration of the episode. It can be noticed that Chennai has shorter pollution episodes while Delhi has longer episodes. Combining both the number and duration of episodes, New Delhi still shows the highest number of days under these pollution episodes that exceed the WHO standards.

In terms of Indian NAAQS, Delhi experiences on an average 15 number of episodes having a median, mean, and maximum duration of 7, 14, and 66 days respectively. The mean duration of the episode for Chennai, Kolkata, Hyderabad and Mumbai is equal to 6, 18, 11, 11 respectively. The maximum duration (Fig. 5–e) of a single episode for Chennai, Kolkata, Hyderabad, Mumbai and Delhi is 43, 78, 89, 67, 114 days respectively in terms of WHO exceedances and 10, 60, 31, 35, 66 days in terms of Indian NAAQS. It is to be noticed that New Delhi experienced a single pollution episode that lasted for more than three months as per WHO standards and more than two months as per Indian NAAQS.

3.4. Trends in exceedances

Environmental agencies around the globe define different threshold values and target values, therefore, we have calculated the number of exceedances and trends for different thresholds values from 20 μg/m³ to 380 μg/m³. Fig. 6 shows the number of exceedances (magenta) and the trend in the number of exceedances per year (blue) for varying threshold values. The shaded blues region shows a 90% CI. The dashed red line shows the threshold values as per the WHO guidelines and the red dash-dot red line shows the threshold as per Indian NAAQS guidelines. It is found that the number of exceedances decreases as the threshold is increased for all the sites. Kolkata, Hyderabad, and Mumbai do not show...
any significant trend for all threshold values. Chennai shows a significant negative trend in the number of exceedances for the threshold values up to 35 μg/m³ and above this threshold, there is no significant trend is noticed. It is found that New Delhi exhibit a negative trend in the number of exceedances, however, the trend is significant up to around 200 μg/m³. We found earlier a significant decreasing trend in PM_{2.5} in Delhi. This analysis confirms that not only PM_{2.5} is decreasing in Delhi but also the number of exceedances is decreasing.

3.5. Chemical composition and source signature

The chemical composition of PM_{2.5} offers vital information on the contributions of specific sources and help to understand aerosol properties and processes. PM_{2.5} chemical components have been found to vary considerably among different sites across the globe (Snider et al., 2016). Global population-weighted PM_{2.5} concentrations were dominated by particulate organic mass, secondary, mineral dust as well as secondary inorganic aerosols such as sulfates, nitrates and ammonium (Philip et al., 2014). In addition to the observed trend of PM_{2.5}, it is also important to know the variability and trend in the chemical composition. The relation between PM_{2.5} exposure and associated health effects is linked with physical and chemical characteristics of the PM_{2.5} and therefore requires attention along with its sources for better management of urban air pollution (Braziewicz et al., 2004; Srimuruganandam and Nagendra, 2011). However, the unavailability of long-term chemical composition records restricts the detailed analysis of the possible sources. Moreover, one can conduct modeling analysis of PM_{2.5} composition but it is considerably challenging because of the combination of uncertainties in the magnitude and spatial and temporal allocation of primary PM_{2.5} emissions and our limited understanding of the chemical production pathways for secondary constituents (Marbur et al., 2008; Appel et al., 2008). As the continuous records of the chemical composition of PM_{2.5} are not available, we gather this information from existing literature to discuss the chemical composition of PM_{2.5} as well as the probable emission source. To date, several studies involving chemical constituent analysis and related source apportionment have been carried out for the national capital Delhi, whereas for the other four megacities (Kolkata, Mumbai, Hyderabad, and Chennai) these kinds of studies are limited. Depending on the prevalent sources, these studies vary widely in their reported chemical constituents. Most commonly reported chemical constituents studied are metals, carbonaceous fractions (EC, OC), cations (Na\(^{+}\), K\(^{+}\), Mg\(^{2+}\), Ca\(^{2+}\), NH\(_{4}\)\(^{+}\)), and anions (SO\(_{4}\)\(^{2-}\), NO\(_{3}\)\(^{-}\), Cl\(^{-}\)).

For the national capital Delhi, the PM_{2.5} is reported to be mainly constituted of EC, OC, Na\(^{+}\), K\(^{+}\), Mg\(^{2+}\), Ca\(^{2+}\), NH\(_{4}\)\(^{+}\), and anions (SO\(_{4}\)\(^{2-}\), NO\(_{3}\)\(^{-}\), Cl\(^{-}\)), some trace metals and heavy metals (Pant et al., 2015; Saxena et al., 2017; Sharma and Mandal, 2017; Jain et al., 2020). While NO\(_{3}\)\(^{-}\) and NH\(_{4}\)\(^{+}\) were observed to be highest during winter followed by post monsoon>summer>monsoon (Jain et al., 2020; Kota et al., 2018; Sharma and Mandal, 2017; Pant et al., 2015), SO\(_{4}\)\(^{2-}\) was reported to be most abundant during summer followed by monsoon>post monsoon>winter (Jain et al., 2020; Pant et al., 2015). Secondary NO\(_{3}\) is thermally unstable at higher temperatures whereas at low temperatures during winter its formation is favorable (Cesari et al., 2018). Higher photochemical activities during the summer season and high humid conditions during monsoon favors the formation of secondary SO\(_{4}\)\(^{2-}\) (Jain et al., 2020; Goel et al., 2018; Pant et al., 2015). While Na\(^{+}\) is observed highest during monsoon owing to its sea origin (Jain et al., 2017; Saxena et al., 2017), Cl\(^{-}\) is also significantly linked with wood combustion, open waste burning, coal combustion and industries (Rai et al., 2020; Ali et al., 2019; Pant et al., 2015), and therefore is observed highest during winter (Saxena et al., 2017; Sharma and Mandal, 2017; Jain et al., 2017; Sharma et al., 2016) along with the biomass burning marker ion K\(^{+}\) (Sudheer et al., 2014; Tiwari et al., 2013a). Organic components of PM_{2.5} like levoglucosan has been linked with biomass burning in winter (Pant et al., 2015). PAHs, linked with biomass burning and road transport, showed the highest concentration during winter, followed by post monsoon>summer>monsoon (Gadi et al., 2019; Singh et al., 2011). EC and OC which are emitted from vehicular emissions and biomass burning (Ram and Sarin, 2011; Sharma et al., 2016) are reported to be higher during winter than summer (Jain et al., 2017; Sharma and Mandal, 2017; Tiwari et al., 2014). Elemental contribution analysis showed the higher contribution of road dust and soil (marked with higher Si value) during summer whereas for the winter season, the contribution of biomass burning was high (marked with higher K value) (Sharma and Mandal, 2017; Saxena et al., 2017; Jain et al., 2020; Pant et al., 2015).

For Kolkata, roughly 50% of the PM_{2.5} mass was reported to be constituted of ions (Na\(^{+}\), K\(^{+}\), Mg\(^{2+}\), Ca\(^{2+}\), NH\(_{4}\)\(^{+}\), SO\(_{4}\)\(^{2-}\), NO\(_{3}\)\(^{-}\), Cl\(^{-}\)) and carbonaceous particles (EC, OC) (Chatterjee et al., 2012). Higher concentrations of NH\(_{4}\), SO\(_{4}\), and NO\(_{3}\) were observed during winter than summer, however, the SO\(_{4}\)\(^{2-}\) oxidation ratio, which is an indicator of secondary SO\(_{4}\)\(^{2-}\) formation, was found to be highest during summer months. Kolkata showed the highest Na\(^{+}\) during monsoon, whereas higher Cl\(^{-}\) was observed during dry seasons (Chatterjee et al., 2012) and were linked with biomass and coal burning. EC and OC for Kolkata as well showed the highest levels during winter, followed by summer and lowest in monsoon (Chatterjee et al., 2012; Talukdar et al., 2015). For Mumbai, the major chemical constituents observed were ions (Na\(^{+}\), K\(^{+}\), Ca\(^{2+}\), NH\(_{4}\), SO\(_{4}\), NO\(_{3}\), Cl\(^{-}\)) and carbonaceous particles (EC, OC) and some trace and heavy metals. While Secondary ions (NH\(_{4}\), SO\(_{4}\), and NO\(_{3}\)), EC and OC were observed higher either during winter or post-monsoon season owing to inland contribution. Non-sea salt sources were of anthropogenic origin (Joseph et al., 2016). Elemental analysis showed a significant contribution of the sea during monsoon, and soil dust during the summer season (Police et al., 2018). For Hyderabad, the studied chemical constituents were EC and OC (Ali et al., 2016). While both EC and OC showed the highest winter concentration than during summer, followed by the monsoon, the concentration variation during winter to summer transition for the two carbonaceous fractions is quite different. EC showed a significant decrease during the transition of winter to summer, the same was not true for OC as secondary organic carbon formation and biomass burning added to the total OC levels during summer. Elemental analysis showed the importance of sources like resuspended dust and vehicular emission for the city (Gummneni et al., 2011). For Chennai, the reported chemical constituents of PM_{2.5} were ions (Na\(^{+}\), K\(^{+}\), Ca\(^{2+}\), Mg\(^{2+}\), NH\(_{4}\)\(^{+}\), SO\(_{4}\)\(^{2-}\), NO\(_{3}\)\(^{-}\), Cl\(^{-}\), F\(^{-}\)) and some trace and heavy metals (Jose et al., 2019; Srimuruganandam and Nagendra, 2011). The ionic content of PM_{2.5} showed the dominance of SO\(_{4}\)\(^{2-}\) and NH\(_{4}\)\(^{+}\) during winter followed by monsoon>summer (Srimuruganandam and Nagendra, 2011). Marine aerosols showed a significant contribution for the coastal city (Jose et al., 2019).

We also performed the five days backward trajectories analysis using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT version 4) model (Stein et al., 2015) to characterize the source region of air masses coming to the megacities. Five days backward trajectories arriving at an elevation of 500 m at USEC sites were computed at an interval of six hours for the period 2016–2019. Then, cluster analysis was carried out to finally come up with five major trajectories for four different seasons. The seasonal back-trajectory plots (Fig. 7) show broadly consistent air mass movement patterns during winter and monsoon seasons and scattered directions during summer and post-monsoon season. Delhi received the majority of air masses from North-west during winter (33%), summer (68%), and post-monsoon season (55%). The majority of the air mass coming from the North-west to Delhi represent the downwind transport of pollutants from the agricultural bio-mass burning region of Punjab and Haryana (Singh et al., 2020; Beig et al., 2020). Moreover, the wind trajectories coming from the eastern part of IGP can add extra particulates either directly or through a gas to particle conversion (Wang et al., 2005; Sharma et al., 2014). Air masses coming from the transboundary region, extending to Pakistan, Afghanistan, Iran, Gulf countries, and further west to the
Mediterranean region, also contribute significantly to Delhi. During monsoon, Delhi receives the air mass from the Arabian sea as well as from the east part of IGP. For Mumbai, most of the air masses originate from the nearby Arabian sea during monsoon, post-monsoon, and summer season. The contribution from the central Indian region during winter (55%) and post-monsoon (47%) are significant. Mumbai also gets transboundary air pollutants during winter and summer from countries located in the Middle-East region. For Kolkata, the IGP region is the major source of air masses during winter (81%) and post-monsoon (39%), and during the summer air masses originated from the land (IGP and central India) as well as from the Bay of Bengal. Moreover, during monsoon season the trajectory follows south-west monsoon wind pattern therefore the air masses originate from the Bay of Bengal as well as from the Arabia sea. For south Indian cities Hyderabad and Chennai, most of the air masses originate from adjacent seas during all the seasons. Hyderabad received 67% of air masses via sea route originating from the Bay of Bengal and the eastern part of IGP during the winter (34%). During summer, 81% of the air masses are from sea regions (35% from the Bay of Bengal and 46% from Arabian sea traveling via land west of Hyderabad). During monsoon, all the air masses originated from the Arabian sea. For Chennai, winter season follows the north-east monsoon wind pattern and gets all the air masses from the Bay of Bengal with majority origination from the IGP region. During summer and post-monsoon, Chennai receives majority air masses from Bay of Bengal (~40%) and the Arabian sea (~38%) via the sources located in the west of Chennai. All the air masses during the monsoon originated from the Arabian sea. While the air masses originate from various directions during the different seasons based on the synoptic weather conditions, the contribution of local sources remains significant (Guttikunda et al., 2019) for the cities. Although the discussion of seasonal variation of chemical constituents, source apportionment and trajectory analysis give an idea of probable sources and regions, a detailed quantitative analysis of source region attribution (Rai et al., 2020) for different seasons can provide better clarity on contributing sources and regions and can further help to understand the observed reduction in PM$_{2.5}$ in the megacities.

### 3.6. Possible linkage of the observed trend with the recent mitigation measures

Major sources of primary PM$_{2.5}$ in India are emissions from the household, power sector, industries, transport, open burning (crop and waste) and dust (Conibear et al., 2018; Guo et al., 2017; Venkataraman et al., 2018). Although the household emissions are dominant across India (Apte and Pant, 2019), vehicular exhaust and dust resuspension (Singh et al., 2020a) remain the dominant local source in Indian cities (Guttikunda et al., 2014, 2019). Other urban sources include construction dust, industrial exhaust, and domestic cooking and heating (Guttikunda et al., 2019). Most of the sources of PM$_{2.5}$ in urban areas are local, however non-local contribution can be significant (Guttikunda et al., 2019;
Guo et al., 2017). For eg.in Delhi, local sources contribute ~70% of total PM$_{2.5}$, but the non-local sources contribute over 30% especially in winter (Guo et al., 2017). The emissions neighboring rural areas, contribute to the urban pollution in India (Guttikunda et al., 2019; Ravindra et al., 2019a, 2019b). Rural households in India rely on kerosene for light in the absence of electricity, and on wood, dung, and other solid fuels for cooking and heating (Chowdhury et al., 2019; Ravindra et al., 2019c). Use of these fuels emit particles, gaseous pollutants, and volatile organic compounds, and therefore are a significant source of secondary particulate matter in both rural and urban areas (Pervez et al., 2019; Rooney et al., 2019). In addition to household and traffic emissions, open waste burning is also a significant contributor to the total PM$_{2.5}$ in Indian cities (Guttikunda et al., 2019; Kumari et al., 2019). The open waste burning is prevalent during winter and over the urban areas with low socio-economic status (Nagpure et al., 2015). Other sources that determine the urban PM$_{2.5}$ levels include industries, thermal power plant, brick production, and use of diesel generator sets, however, the influence of these sources is highly variable (Guttikunda et al., 2019).

Future emissions scenario studies conducted over India predict an increase in PM$_{2.5}$ (Venkataraman et al., 2018; Pommiere et al., 2018), however studies (Chowdhury et al., 2019; Purohit et al., 2019; Venkataraman et al., 2018; Conibeer et al., 2018; Bhanarkar et al., 2018) have shown that significant reduction in PM$_{2.5}$ is achievable by implementation of strict measures to reduce the PM$_{2.5}$ emissions. It has been shown by Chowdhury et al. (2019) that a transition from bad fuel to clean fuel in the household has the potential to significantly reduce the PM$_{2.5}$ levels at the national level. However, at the urban or city level, where cleaner fuel is used, reduction in vehicular emissions (exhaust and non-exhaust) can bring down the PM$_{2.5}$ levels as observed during the COVID lockdown in Indian cities (Kumar et al., 2020; Singh et al., 2020c) and significantly reduce the traffic exposure (Singh et al., 2020b) in urban areas.

We propose that the reduction in PM$_{2.5}$ levels across the cities is due to the recent measures taken to reduce the ambient pollution levels in India. The major recent initiatives that might have helped in the reduction include the launch of the National Air Quality Index (AQI) for public awareness, the formation of Environment pollution (prevention and control) authority, implementation of a Graded Response Action Plan (GRAP) and Comprehensive Action Plan (CAP) for prevention, control and mitigation of air pollution in Delhi and NCR. National Clean Air Program (NCAP) was launched in January 2019 with comprehensive mitigation actions along with augmentation of the air quality monitoring network across the country. A tentative national target of 20–30% reduction of PM$_{2.5}$ and PM$_{10}$ concentration by 2024 was proposed under NCAP taking 2017 as a reference year. Besides, several steps have been taken for creating awareness among the general population (PIB, 2019).

As solid fuels are the biggest contributor to the household emissions across India, a scheme (Pradhan Mantri Ujjwala Yojana, PMUY, pmuy.gov.in) was launched in 2016 to ensure cleaner fuel (Liquefied petroleum gas, LPG) for all households. As the PM$_{2.5}$ emission from LPG is lower than that of solid fuel (Deepthi et al., 2019), the implementation of PMUY across India would have reduced PM$_{2.5}$ levels mainly at the regional level (Chowdhury et al., 2019). However, people’s attitudes towards fuel usage may lessen the expected reduction in emission linked with this switch to cleaner fuel usage as solid fuels are much cheaper and easily available (Ravindra et al., 2019c). A massive rural electrification scheme (Deen Dayal Upadhyay Grameen Jyoti Yojana, DDUGY, ddugy.gov.in) should have also contributed to the reduction in PM$_{2.5}$ by reducing the emissions from kerosene lighting. However, frequent power cuts might minimize the impact of the benefit achieved through electrification (Hou and Urpelainen, 2020). In order to mitigate the air pollution from waste burning, five waste management rules on solid waste, hazardous waste, plastic waste, biomedical waste, and e-waste have been revised and implemented in 2016 (Solid Waste Management Rules, 2016). Although waste management is a major challenge (Kumar et al., 2017), a major step to improve the door-to-door waste collection and disposal as a part of Swachh Bharat Mission (swachhbharatmission.gov.in; Ghosh, 2016) in urban areas could have resulted in the improvement in air quality.

For the reduction of traffic exhaust emissions, the emission standard of the fleet was improved to BS-IV from April 2017 and BS-VI was scheduled from April 2020. The old fleet scrappage program was launched, and electric vehicles are being promoted. Apart from this, shifting to alternate cleaner fuels like LNG, ethanol blending in petrol are some of the steps taken for the cleaner transport sector. Moreover, the use of a modern public transport system was promoted in recent years to reduce traffic emissions. While these measures reduce the exhaust emissions, the maximum reduction is expected in road dust resuspension emission (Singh et al., 2020a, 2020b) by regular road dust cleaning by mechanized vacuum dust cleaners (Goyal et al., 2019; Gulia et al., 2018). Other measures include dust control from the building and road construction activities. The stringent measures to limit the crop residue burning. (Lohan et al., 2018; Ravindra et al., 2019b; Bhuvaneswari et al., 2019) and revised standards for coal-based thermal power plants by 2017 (Guttikunda and Jawahar, 2018) could have potentially helped in the reduction of PM$_{2.5}$ in the cities. Our proposition is supported by the study of Purohit et al. (2019) who have reported that the air pollution control measures implemented until June 2018, should deliver an overall decline of ambient PM$_{2.5}$ levels of about 14% by 2030 and stabilize the pollution levels despite economic growth. However, a detailed analysis is required to link the recent control measures with the observed reduction in the PM$_{2.5}$ in the five megacities.

4. Conclusions and discussion

This study reports a detailed analysis of the variabilities and trends in the PM$_{2.5}$ concentration measured at the US embassy and consulates in the five megacities (Chennai, Kolkata, Hyderabad, Mumbai, and New Delhi) in India during the period of six years (2014–2019).

Distinct diurnal, monthly and seasonal variations are observed in the five cities due to the different site locations and local meteorology. However, all cities show the highest and lowest concentrations in the winter and monsoon months, respectively. The winter maxima are associated with increased emissions and lower PBLH. Monsoon months are found to be the cleanest month because of wet deposition and increased soil moisture which leads to less dust re-suspension. The concentration during monsoon is found to be around 15% of the winter levels for Kolkata and Delhi, whereas for other cities is found to be around 20–33%. Delhi exhibits the highest PM$_{2.5}$ concentration in all the seasons. For all the cities, monsoon months show the lowest PM$_{2.5}$ levels due to the wet scavenging during the South-West monsoon, except for Chennai, where most of the rainfall occurs in North-East monsoon season during October–December. One of the distinct features in Delhi is that the PM$_{2.5}$ levels are found maximum in November due to the increased biomass burning around Delhi. Moreover, the firecrackers emissions during Diwali, which is celebrated around this period, further adds to the total pollution in Delhi leading to severe pollution episodes for few days (Ganguly et al., 2019). For Chennai, the lowest level is found in April instead of monsoon because of less rainfall during South-West monsoon and onset of sea-breeze. All the cities consistently show morning peaks around (~08:00 – 10:00 h) due to the fumigation effect and morning peak traffic and household emissions trapped within the evolving shallow boundary layer. A shift of up to two hours in the morning peak hours is due to the season and the geographical location. The lowest level of PM$_{2.5}$ for all the cities is found in late afternoon hours (~15:00 – 16:00 h). However, Chennai also shows minimum value after midnight (~01:00 h) when the levels become equal to the day-time low.

A statistical trend analysis based on STL decomposition of the monthly mean PM$_{2.5}$ concentration has been performed. We find a statistically significant decreasing (negative) trend (within 95% confidence interval) for all the cities during the study period. The calculated decreasing (negative) trend is equal to 2.61 ± 0.52 μg/m$^3$/year for
Chennai, 1.81 ± 0.82 μg/m³/year for Kolkata, 1.50 ± 0.71 μg/m³/year for Hyderabad, 1.79 ± 0.96 μg/m³/year for Mumbai and 4.19 ± 1.12 μg/m³/year for Delhi. While Delhi shows the highest decline per year, Chennai has been found to exhibit the fastest percent decline per year. Analysis of MERRA-2 meteorological parameters suggests no significant change in the annual mean wind speed, temperature, PBLH, and precipitation in the past six years. Despite that, PM2.5 has been found to exhibit a declining trend.

We have also reported the number of threshold exceedances of daily mean PM2.5 as per the WHO (25 μg/m³) and Indian NAAQS (60 μg/m³). In addition, the number of pollution episodes and length of each episode (levels above limit values continuously for three or more days) has been reported. We found that the PM2.5 levels in the cities exceed WHO standards for more than 50% of days in a year with a few exceptional years in Chennai. The exceedances in terms of Indian NAAQS are found to be highest for Delhi with more than 200 days of exceedances. Chennai is found to have the lowest number of exceedances. The number of exceedances is found to be around 100–150 days for other cities. Delhi experiences an average 15 number of episodes per year in terms of Indian NAAQS, with a median, mean, and maximum duration of 7, 14, and 66 days respectively. The mean duration of the episode for Chennai, Kolkata, Hyderabad, and Mumbai is equal to 6, 18, 11, 11 respectively. The maximum duration of a single episode for Chennai, Kolkata, Hyderabad, and Delhi is 43, 78, 89, 67, 114 days respectively in terms of WHO exceedances and 10, 60, 31, 35, 66 days in terms of Indian NAAQS. We found that New Delhi experienced a single pollution episode that lasted for more than three months as per WHO standards and more than two months as per Indian NAAQS.

We have calculated the number of exceedances and trends in the exceedances for different threshold values from 20 μg/m³ to 380 μg/m³. Kolkata, Hyderabad, and Mumbai do not show any significant trend for all threshold values; however, Chennai shows a significant negative trend in the number of exceedances for the threshold values up to 35 μg/m³. New Delhi exhibits a negative trend up to around 200 μg/m³. This suggests that not only the annual mean PM2.5 is decreasing in Delhi but also the number of exceedances is decreasing.

So far, we have not come across any study that has suggested a significant declining trend in air quality in Indian cities despite the measures by the local authorities. This is the first study, to the best of our knowledge, that has reported statistically significant decreasing trends of PM2.5 in Indian megacities. This decrease can be attributed to the recent policies and regulations (NCAP, MoEFFC, 2019) implemented in Delhi and other cities for the abatement of air pollution. The implementation of source sector-specific measures related to vehicular emissions, road dust resuspension and other fugitive emissions, cleaner fuel, biomass/municipal solid waste (MSW) burning, industrial pollution, construction, and demolition activities, etc., were the major steps towards the mitigation of air pollution. The mitigation measures implemented until June 2018 were expected to deliver an overall decline of ambient PM2.5 despite economic growth (Purohit et al., 2019). While a reduction in PM2.5 is found in Delhi, it continues to be the most polluted city among five megacities. With the annual rate of reduction observed here, it may take another two decades for the pollution levels to come within Indian NAAQS levels. Therefore, stricter compliance of the NCAP policies can further accelerate the reduction of the pollution levels to reduce the health impacts across all India. The results presented in this study will support the National Clean Air Program of India and help to evaluate the pollution reduction in Indian megacities due to decreased activities post COVID-19 lockdown in India (Singh et al., 2020c).

Declaration of competing interest

The author(s) declare(s) that there is no conflict of interest regarding the publication of this article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.141461.

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CRediT authorship contribution statement

Vikas Singh: Conceptualization, Formal analysis, Investigation, Writing - original draft, Writing - review & editing. Shweta Singh: Writing - original draft, Investigation, Formal analysis, Writing - review & editing. Akash Biswal: Visualization, Investigation, Formal analysis, Writing - review & editing.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.141461.

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