Atmospheric Environment 204 (2019) 142–150

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Tracking ambient $PM_{2.5}$ build-up in Delhi national capital region during the dry season over 15 years using a high-resolution (1 km) satellite aerosol dataset

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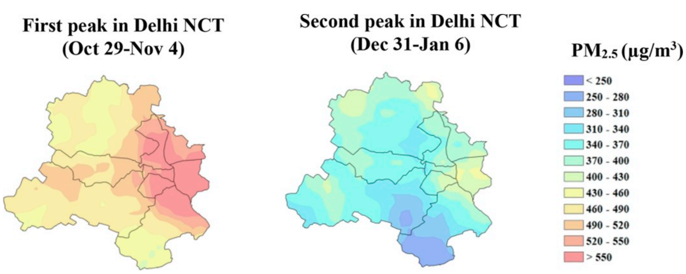
GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
$PM_{2.5}$
Delhi
Episodes
MAIAC AOD
Global Human Settlement
Sub Soil Water Act 2009

ABSTRACT

The Delhi National Capital Region (NCR) is among the most polluted areas in the world. We tracked ambient $PM_{2.5}$ (fine particulate matter) build-up in Delhi NCR during the dry season (October–June) by analysing 15 years (2001-02 to 2015-16) of high-resolution (1 km) satellite data that has been bias corrected using coincident in-situ data. Ambient $PM_{2.5}$ concentrations are 1.25 times lower in the upwind regions compared to the downwind areas of Delhi NCR, with the difference being attributed to outflow from the Delhi National Capital Territory (NCT). We identify two major peak pollution episodes – the first occurs from the end of October to early November, while the second occurs toward the end of December through early January. Mean ambient $PM_{2.5}$ concentrations remain > 300 μg/m^3 (five times the Indian 24-h national standard) for several weeks around the two peak pollution episodes. The first peak is attributed to pollution transport from upwind areas affected by open biomass burning, coupled with stable atmospheric conditions, while the second is attributed to enhanced local emissions and perhaps secondary aerosol formation under favorable meteorological conditions. The implementation of the "Sub-soil Water Preservation Act" in 2009 in Punjab (a state upwind of Delhi NCT) reduced the time between paddy and wheat cultivation seasons; as a result, open biomass burning increased, resulting in a 9% increase in weekly $PM_{2.5}$ concentration over Delhi NCT since 2009 during the first pollution episode. To the best of our knowledge, this is the first time MODIS MAIAC 1 km satellite data are used to generate pollution statistics over India. Our results demonstrate the potential of high-resolution satellite data in tracking pollution build-up at an urban scale and may help in air quality management.

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https://doi.org/10.1016/j.atmosenv.2019.02.029
Received 6 July 2018; Received in revised form 11 December 2018; Accepted 17 February 2019
Available online 26 February 2019

1. Introduction

Exposure to ambient particulate matter less than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) has been causally associated with chronic obstructive pulmonary disease (COPD), ischemic heart disease (IHD), stroke, lung cancer, diabetes, pregnancy-related complications, and low birth weight among other outcomes (Anenberg et al., 2010; Krewski et al., 2009; Pedersen et al., 2013; Pope et al., 2002; Slama et al., 2007; Smith et al., 2014; Sun et al., 2013; Weinmayr et al., 2015). Premature deaths in India due to ambient $PM_{2.5}$ exposures have been rising since 1990, with the most recent estimate for India (2016) at approximately 1.0 million premature deaths per year (https://vizhub.healthdata.org/gbd-compare/india).

The National Capital Territory (NCT) of Delhi is recognized as one of the most polluted regions in the world (Guttikunda and Calori, 2013; Tiwari et al., 2012; Pan et al., 2016). Delhi NCT houses about 16.7 million people; the larger Delhi NCR, which includes several districts of Haryana and Uttar Pradesh, is home to about 46 million people. Several studies have estimated that annual $PM_{2.5}$ concentrations in Delhi NCT exceed India's annual National Ambient Air Quality Standard (annual NAAQS) of 40 μg/m^3 (that is 4 times higher than the WHO annual air quality guideline) by more than 200% (Chowdhury et al., 2017; Chowdhury and Dey, 2016; Dey et al., 2012; Pal et al., 2018; Tiwari et al., 2013, 2012). Saraswat et al. (2013) estimated the mean annual $PM_{2.5}$ concentration over Delhi to be 133 μg/m^3 using a land-use regression model. Apte et al. (2011) found that in-road $PM_{.5}$ exposure in Delhi NCT exceeded the 24-hr NAAQS (~2.5 times higher than the WHO 24-hr air quality guideline) of 60 μg/m^3 by 1.5 times. Goel et al. (2015) examined the $PM_{2.5}$ concentration for multiple transport microenvironments in Delhi and found that on-road $PM_{2.5}$ concentration exceeded ambient measurements for all modes of transports. A recent report (Sharma and Dixit, 2016) estimated the winter $PM_{2.5}$ concentration over several locations in Delhi to spike eight times above the daily NAAQS standard, primarily attributed to secondary aerosol particle formation (NO_3^- + SO_4^{-2} + NH_4^+ and secondary organics), followed by combustion related infusion of black carbon and elemental carbon particles. Goel and Guttikunda (2015) estimated that vehicular emission of particulate matter increased steadily from the 1990s to early 2000s, after which it decreased significantly until 2012. Pant et al. (2017) carried out personal exposure monitoring on a small sample in Delhi and concluded that auto-rickshaws are the most polluted microenvironments.

Over the last two decades, multiple policies have been implemented to reduce air pollution in Delhi NCT, including shutting down 1328 factories responsible for hazardous emissions, revamping the public transport system by introducing the Delhi Metro, conversion of fleet transport vehicles to compressed natural gas (CNG), and reduction of sulphur content in diesel (Chowdhury et al., 2017; Goel and Pant, 2016; Narain and Krupnick, 2007). In 2015, the National Green Tribunal Act banned the use of diesel vehicles older than 15 years within the city. In 2016, an odd-even traffic intervention (which allowed odd and even numbered cars classified by the last digit of the number plates to alternately use roads on odd and even days respectively) was implemented twice for 15 days, each from January 1st and April 15th, without noticeable benefit (Chowdhury et al., 2017). On the contrary, very few policies have been undertaken to tackle pollution in the equally polluted and larger Delhi NCR, which consists of districts from surrounding states of Haryana (Gurugram, Faridabad, Jhajjar, Panipat, Rohtak, Rewari and Sonipat) and Uttar Pradesh (Bulandsahar, Meerut, Ghaziabad, Baghpat and Gautam Budh Nagar) alongside NCT.

Despite the implementation of these policies in Delhi NCT, ambient $PM_{2.5}$ concentrations in NCT and NCR remain almost three times higher than annual NAAQS. Multiple factors explain the lack of success of these policies to curb ambient air pollution, the primary being the geographic location of the Delhi NCT and NCR and the prevailing meteorological conditions. Long-range transport of pollution emitted from open biomass burning in upwind rural regions during the crop burning season, dust transport during the summer, and transport of pollution emitted from brick kilns throughout the year (Cusworth et al., 2018) add to local sources such as transport, construction, diesel generators, power plants, industries, residential use, etc.

Air pollution episodes can be tracked using a range of high-quality, in-situ measurement techniques, including gravimetric sampling, TEOM (tapered element oscillating microbalance) instruments, and/or BAMs (beta attenuation monitors). $PM_{2.5}$ monitoring in Delhi began in 2008; the network has since expanded to 35 + monitors in the NCT. We note that the number of ground-based monitors outside of Delhi NCT but within Delhi NCR (which encompasses both urban and rural areas) is relatively few till date. Hence it is not known whether the pollution build-up during the dry period (end of September to the end of June) is a more recent phenomenon and whether its magnitude has intensified in recent years. Similarly, available in-situ data are geographically sparse and insufficient to understand the spatial heterogeneity across the Delhi NCR.

In this paper, we report $PM_{2.5}$ build-up over Delhi NCR in the dry season for the period of September 2001 to June 2016 using 1 km resolution satellite-derived aerosol data. We identify the peak episodes of ambient $PM_{2.5}$ concentrations over NCT and their changes over time. We further present the 15 year s average dry-period climatological $PM_{2.5}$ concentration map of Delhi NCR at 1 km resolution, examine changes in ambient $PM_{2.5}$ exposure at a weekly scale during the dry period, and identify rural to urban differences in pollution episodes.

2. Data and methodology

2.1. Satellite-based ground-level PM2.5 modelling

We use a new aerosol optical depth (AOD) dataset that was generated at a high resolution (1 km × 1 km) using the Multi-Angle Implementation of Atmospheric Correction (MAIAC) algorithm (Lyapustin et al., 2011, 2018) applied to the MODIS (Moderate Resolution Imaging Spectroradiometer) data. The MAIAC AOD is retrieved as a combined Aqua and Terra product. Due to explicit surface characterization, MAIAC provides global AOD retrievals both over dark and bright surfaces and features improved cloud detection, general lack of urban bias and a significant increase in spatial coverage as compared to dark-target (Levy et al., 2013) and deep blue (Hsu et al., 2013) algorithms, particularly in partly cloudy conditions (Chudnovsky et al., 2013b). A few studies have already used the AOD generated through MAIAC algorithm to estimate concentration to $PM_{2.5}$ at high spatial resolution (Chudnovsky et al., 2013a,b; Di et al., 2016; Just et al., 2015; Kloog et al., 2014, 2015; Lee et al., 2016).

We estimate $PM_{2.5}$ from MAIAC-AOD by multiplying spatially and temporally varying conversion factors at 0.1° × 0.1° spatial resolution obtained from GEOS-Chem chemical transport model. A detailed description of deriving the conversion factors is provided elsewhere (van Donkelaar et al., 2014, 2010a). We downscale the conversion factors to 1 km resolution using spline interpolation. Inferred $PM_{2.5}$ is first evaluated against coincident ground-based $PM_{2.5}$ measurements (that are publicly available) from NCT operated by Central Pollution Control Board (CPCB) and Delhi Pollution Control Committee (DPCC). Consistent data, available from 12 sites from 2013 onwards, are used for the calibration. We temporally match $PM_{2.5}$ (averaged over 4 h, 10:00 to 14:00 to cover Terra and Aqua overpass) retrieved by the ground-based monitoring stations with the MODIS MAIAC retrieved $PM_{2.5}$ within 500 m radii of the respective ground-based monitoring site. Comparisons are made between ~1400 pairs of coincident ground and satellite-based estimates. We adopt a percentile-based bias correction method following our previous studies (Chowdhury et al., 2017; Pande et al., 2018) where we calibrate satellite retrieved $PM_{2.5}$ at every 2nd percentile with respect to the ground-based $PM_{2.5}$. We observe a low bias in inferred $PM_{2.5,}$ as we had in our previous studies with the

MODIS 3 km (MOD04_3k) aerosol product (Chowdhury et al., 2017) and the MISR V22 aerosol product (Dey et al., 2012). The bias ($\Delta PM_{2.5}$ = in-situ $PM_{2.5}$ – MAIAC $PM_{2.5}$) is found to have a linear relation with in-situ $PM_{2.5}$. We correct this low bias of inferred $PM_{2.5}$ using regression. The bias-corrected MAIAC $PM_{2.5}$ validates well (R^2 = 0.88, significant at 95% CI and a slope of 0.9) with remaining coincident in-situ $PM_{2.5}$ data (Fig. 1). The errors in the regression coefficients translate to an overall ~16% uncertainty in the inferred $PM_{2.5}$ relative to in-situ $PM_{2.5}$. We use the bias-corrected MAIAC $PM_{2.5}$ data further to examine ambient $PM_{2.5}$ ground-level concentrations in the Delhi NCR during 2001–2016.

Since our focus is to track pollution build-up during the dry season (typically from the end of September when the south-west monsoon ceases over Delhi NCR until the end of June of the subsequent year when the south-west monsoon reaches Delhi NCR), we first average all valid 1 km^2 $PM_{2.5}$ that are bias-corrected within the NCR for each day and present the analysis at weekly scale by then averaging over 7 days to ensure enough samples (e.g. see Fig. S1 in supporting information [SI] that depicts the total number of valid retrievals over Delhi NCT in a week) for robust statistics. In our analysis, week 1 represents Sep 24–30 every year and week 39 represents the 3rd week of June, just before the conventional dates of monsoon onset over the NCR.

2.2. Meteorological analysis

Meteorological conditions (e.g. wind direction and speed, mixing layer depth, precipitation etc.) play an important role in modulating $PM_{2.5}$ concentrations over any region at varying time-scales, i.e. from diurnal to seasonal, annual, and decadal. (Chelani, 2013; Upadhyay et al., 2018). We use ERA Interim data at 0.125° (~12.5 km) spatial resolution for analyzing mixing layer depth (MLD), wind speed and wind direction at the weekly temporal resolution for our study period. Precipitation during the dry season is only < 9% of the precipitation received during the monsoon season in Delhi.

Fig. S2 (see SI) shows the map of India with our study area of the Delhi NCR highlighted in light blue and the smaller Delhi NCT highlighted in dark blue along with the mean wind direction and speed during the dry season. We recognize that the Delhi NCR, which engulfs Delhi NCT, is also heavily polluted and demands similar mitigation policies as planned and implemented over the Delhi NCT to curb pollution.

2.3. Fire count data as a proxy of open biomass burning in Punjab and Haryana

The states of Punjab and Haryana are known as the bread bowls of India. Approximately about 2.98 and 1.35 million hectares in Punjab and Haryana are used for the cultivation of rice in semiaquatic submerged land, according to the Ministry of Agriculture (GoI, 2016). Although Punjab is small compared to many Indian states, it is the third largest rice producer in India after West Bengal and Uttar Pradesh. These states are estimated to produce about 26.6 million tons of paddy straw annually out of which only ~20% is utilized elsewhere (as animal fodder, mulch over soil, or in industry in-situ and ex-situ incorporation (GoI, 2018a, 2016)). Given that these states have two cropping seasons with typically 10–15 days between rice harvesting and subsequent wheat sowing, the farmers resort to burning the paddy residues to prepare the field for wheat cultivation, which generates high levels of $PM_{2.5}$ every year. This occurs typically from late October until early November. The typical wind direction in these months transports pollution toward Delhi NCR and thus causes severe pollution episodes in this region. A recent study (Cusworth et al., 2018) attributes 78% of the observed $PM_{2.5}$ enhancement in the NCT during this time to open biomass burning. We analyze MODIS Fire Information for Resources Management System (FIRMS) data which captures fire activities at 1 km × 1 km spatial resolution. Only observations with high confidence (> 70%) are included in our analysis. We compute weekly aggregates of all fire occurrences within the magenta box marked in Fig. S2 that includes Punjab and Haryana at weekly scale to understand its variability vis-à-vis $PM_{2.5}$ variability in the NCR.

2.4. Global human settlement data for rural and urban classification

We identify each 1 km grid within Delhi NCR as rural/urban/no settlement based on the Global Human Settlement Layer (GHSL) settlement model (SMOD) data (Melchiorri et al., 2018). GHS-SMOD data engages a number of information layers (Dijkstra and Poelman, 2014) to classify each 1-km grid into 4 classes - high density urban, low density urban, rural and unpopulated/no settlement. For this study, we combine the low- and high-density urban clusters to represent urban areas within NCR and estimate ambient $PM_{2.5}$ concentration and track build-up of $PM_{2.5}$ during the dry period in the three classes - rural, urban and no settlement. Grids classified as no-settlement are termed as minimal-settlement.

3. Results

This section is divided into three parts. First, we discuss the general pattern of pollution build-up based on 16 years of satellite data. Next, we investigate the change in amplitude of the pollution build-up following a major policy implementation in the upwind state. Lastly, we discussed the pollution build-up in view of rural, urban and minimal settlement areas within Delhi NCR.

3.1. Pollution build up in the dry season

Fig. 2a depicts the 15-year average dry-period ambient $PM_{2.5}$ concentrations over Delhi NCR (with the boundary of the NCT marked in bold line) and its trend in Fig. 2b (the green contours depict the trend estimated with p-value < 0.1). Fig. 2a is overlain with a mean wind pattern for the time period of interest. To describe the gradient of $PM_{2.5}$ concentration across the NCR, we focus on two regions - A and B in the upwind and downwind flanks of the NCR respectively, depending on the dominant wind direction. We estimate the long term (15 years) dry season averaged $PM_{2.5}$ concentration over NCR to be 171 ± 23 μg/m^3. In the same time period, the long term $PM_{2.5}$ concentrations in the upwind box A and downwind box B are estimated to be

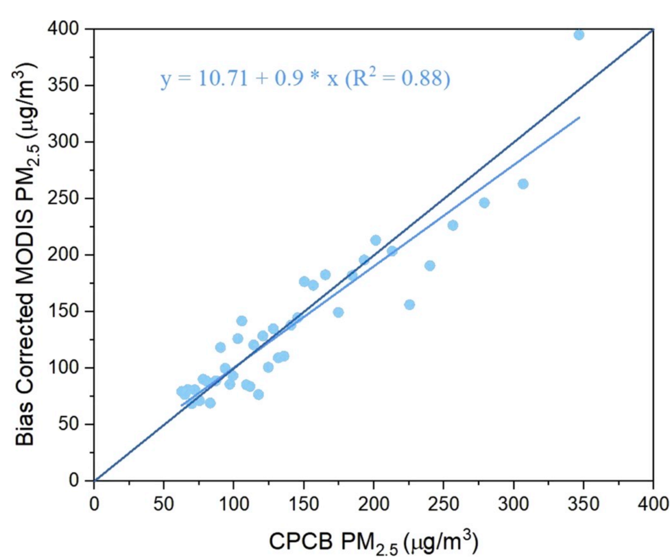

Fig. 1. Scatterplot between bias-corrected MAIAC-$PM_{2.5}$ (after calibration with ~1400 data points) and coincident remaining in-situ $PM_{2.5}$ measured at CPCB sites in Delhi. $PM_{2.5}$ measurements from ground-based sites for the year 2016 are randomly selected for validation.

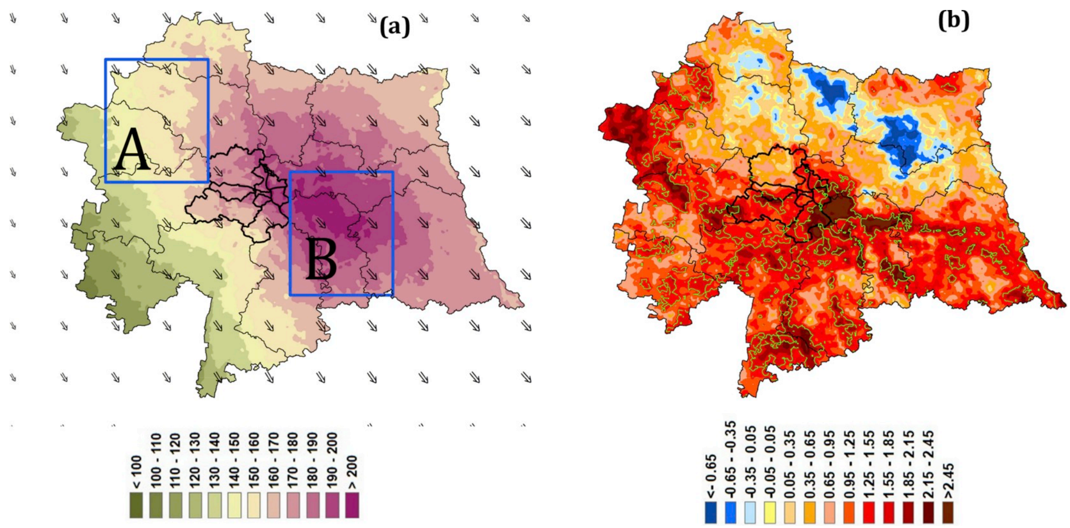

Fig. 2. (a) Mean ambient $PM_{2.5}$ ground-level concentrations (μg/m^3). The two blue boxes represent the upwind (A) and downwind (B) flanks of Delhi NCR and (b) trend (μg/m^3 per year) over the NCR during the dry season for the period 2001–2016. The green contours mark the trend estimated with a p-value < 0.1. Delhi NCT boundary is marked by a bold line. Surface wind is also marked in the left panel. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

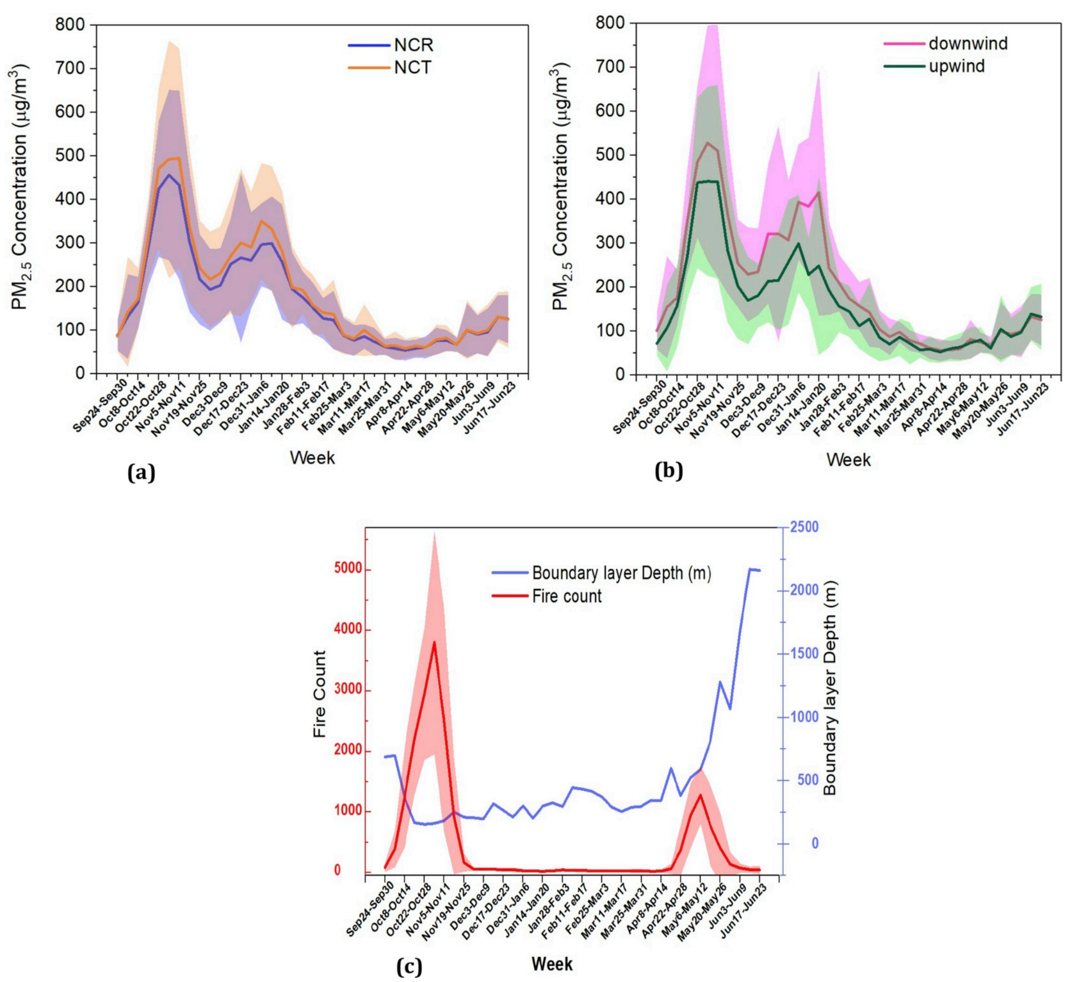

Fig. 3. (a) Weekly $PM_{2.5}$ concentration over Delhi NCR and NCT. The blue and orange lines depict mean (shaded areas represent ± 1σ, σ is standard deviation) $PM_{2.5}$ concentration; (b) Weekly $PM_{2.5}$ concentration over the upwind and downwind boxes marked as A and B in Fig. 2a, and (c) Weekly mean boundary layer depth over Delhi NCR (in blue) and the number of fire events in the upwind region (demarcated by magenta box in Fig. S2 in SI) in red with shades showing ± 1σ. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

161.2 ± 19 μg/m^3 and 202.1 ± 24.4 μg/m^3, which implies that the $PM_{2.5}$ concentration in the downwind region of Delhi NCR is 1.25 times the $PM_{2.5}$ concentration in the upwind region. This is also consistent with results from a recent report (ARAI and TERI, 2018). The higher concentration in the downwind region may be attributed to the 'megacity outflow' of pollution from Delhi NCT (marked in bold in Fig. 2a) which affects the downwind districts in Delhi NCR. Though this spatial gradient is persistent throughout the post-monsoon (October–November), winter (December–February) and summer (March – May) seasons (Fig. S3 in SI), its magnitude vary across seasons. Mean (± 1 standard deviation) ground-level $PM_{2.5}$ concentration (290 ± 54 μg/m^3) during the post-monsoon season is close to five times the daily NAAQS. Pollution level (210 ± 35 μg/m^3) remains more than 3 times higher than the daily NAAQS in the winter season. During the summer season, though $PM_{2.5}$ mean ground-level $PM_{2.5}$ concentrations decrease substantially over the NCR (90 ± 13 μg/m^3), they remain substantially higher than the daily NAAQS. We note that WHO 24-h guideline for $PM_{2.5}$ is much lower (25 μg/m^3) and therefore health risks would exist even if the Indian standard is met.

Fig. S4 depicts the inter-annual variability in dry season $PM_{2.5}$ concentration over Delhi NCR. $PM_{2.5}$ concentration increases steadily up to 2013–2014 dry season. In the next two dry seasons, it decreases. This post-2014 dip observed in the satellite data is in parity with the trend obtained from CPCB data (Fig. S5). Since the meteorology has not changed drastically after 2014 (Fig. S4 and S6), we speculate that the observed decrease could be a result of a reduction in local emissions. This can be confirmed only if emission inventory within Delhi NCR is updated for every year. The trend analysis computed from mean values of dry-season $PM_{2.5}$ concentration (Fig. 2b) reveals that $PM_{2.5}$ has been increasing at a rate of > 1.5 μg/m^3 per year (the green contours in Fig. 2b show values which are statistically significant at 90% CI, $p < 0.1$) in the NCT and southern parts of the greater NCR since 2001. Though the north-eastern flank of the NCR shows a decreasing trend, it is not significant (at 90% CI, $p < 0.1$).

Weekly time series of Delhi NCR and NCT $PM_{2.5}$ concentrations averaged over the 15-year period are shown in Fig. 3a, while Fig. 3b depicts $PM_{2.5}$ concentration in the upwind and downwind areas (represented by boxes A and B in Fig. 2a). The first week represents September 24–30 of each year. Supplementary Video 1 depicts the changes in weekly spatial distribution of $PM_{2.5}$ over Delhi NCR. Ambient $PM_{2.5}$ concentration over the entire NCR (including Delhi NCT, the upwind and downwind boxes) starts rising from October 15–21, coinciding with the increase in open biomass burning (red line in Fig. 3) in the upwind regions of Punjab and Haryana. Prevailing northwesterly winds (Fig. S2) transport the resulting pollution into the NCT that is trapped in the stable atmospheric condition indicated by low boundary layer depth (Fig. 3) and low wind speed (Supplementary Video 1). $PM_{2.5}$ concentration continues to rise over the next 3 weeks and reaches 456 ± 195 μg/m^3 in October 29-November 4 over Delhi NCR. In Delhi NCT, the peak $PM_{2.5}$ pollution episode extends by a week until November 5–11 when peak $PM_{2.5}$ reaches 494 ± 250 μg/m^3 before it starts decreasing. In the downwind area (Box B in Fig. 2a), $PM_{2.5}$ concentration peaks during October 29-November 4 when mean $PM_{2.5}$ concentration reaches 528 ± 257 μg/m^3. The corresponding peak $PM_{2.5}$ in the upwind area (Box A in Fig. 2a) during the same week is 17% lower (441 ± 214 μg/m^3). Overall it can be noticed that the magnitude of first peak $PM_{2.5}$ concentration is higher in Delhi NCT than in Delhi NCR and is higher over the downwind area than in the upwind area. Note that this estimate is a 15-year average and therefore such a massive rise in pollution level over a long period of time is expected to have substantial health impacts (both acute and chronic). Once the fire count starts decreasing after October 29-November 4 and wind strengthens, $PM_{2.5}$ concentrations fall. However, relatively shallow boundary layer keeps average pollution level above 200 μg/m^3 over Delhi NCR. $PM_{2.5}$ concentrations briefly decrease below 200 μg/m^3 for a couple of weeks in the upwind area.

Supplementary video related to this article can be found at https://doi.org/10.1016/j.atmosenv.2019.02.029.

$PM_{2.5}$ concentration again starts rising from December 17–23 and reaches a second peak (300 ± 130 μg/m^3) during January 7–13. $PM_{2.5}$ concentration starts decreasing after reaching a peak of 299 μg/m^3 during December 31-January 6 in the upwind area while in the downwind area the peak sustains for a couple of more weeks until January 14–20 when it reaches 415 μg/m^3 (67% higher than the upwind area for the same week). We attribute the second peak over the NCR to rise in local emissions from solid-waste and open biomass burning (for heating), enhancement in secondary aerosol formation (Nagar et al., 2017) and transport of pollution from adjoining states by converging winds (Fig. S7 in SI). $PM_{2.5}$ concentration in Delhi NCR thereafter drops and stabilizes in the range 50–100 μg/m^3 by March 25–30 as the boundary layer starts to expand (Fig. 3c) enhancing the rate of dispersion. $PM_{2.5}$ concentration increases again since May 20–26, which is attributed to enhanced dust transport from arid regions by north-westerlies (Das et al., 2013), but large boundary layer depth impedes ground-level $PM_{2.5}$ concentration from exceeding 120 μg/m^3. We observe a dwarf second peak in fire events on May 5–11 due to the burning of crop residues generated from wheat harvesting. However, unlike during the first fire count peak, pollution is not transported to the NCR this time due to changes in wind direction (Fig. S8), thus resulting in lower $PM_{2.5}$ concentration over Delhi NCR as compared to the first peak (October 29-November 4).

In-situ CPCB $PM_{2.5}$ data in Delhi NCT are averaged for 39 weeks during the 2015-16 and 2016-17 dry season. The weekly averaged in-situ $PM_{2.5}$ also depicts two major episodes that coincide with the peaks obtained from satellite data (Fig. S9 in SI). The difference in magnitude of the peaks obtained from the in-situ $PM_{2.5}$ and satellite $PM_{2.5}$ may be explained by the spatial resolution of the data that go into estimating the weekly average. While the in-situ $PM_{2.5}$ for each week is obtained by averaging measurements obtained from 12 sites, the weekly satellite-derived $PM_{2.5}$ is obtained by averaging ~9000 data points.

3.2. Is Diwali effect detectable at weekly scale?

Diwali, also known as the "festival of lights" is celebrated across India and is commemorated by extensive burning of fireworks. Though the exact dates of Diwali vary from year to year, it typically falls in between late October and early November. This is the same time period as the first pollution episode and when peak fire counts are identified (discussed in the previous sub-section, Fig. 3). Several studies in Delhi and across India have examined the effect of firework use during Diwali on air quality and health (Ghei and Sane, 2018; Lin, 2016; Mukherjee et al., 2018; Pandey et al., 2016). These studies have found that $PM_{2.5}$ increases significantly during and immediately after the Diwali event. To understand whether the impact of firework emission during Diwali on $PM_{2.5}$ build-up is detectable at a weekly scale, we estimate the (a) $PM_{2.5}$ concentration during the week encompassing the Diwali festival, termed as 'with Diwali days' and (b) $PM_{2.5}$ concentration of the same week by excluding the Diwali days ('Diwali' days are considered to be the day of 'Diwali', the preceding day and the succeeding day), termed as 'without Diwali days'.

The difference (5.6%) in the median value of weekly $PM_{2.5}$ 'without Diwali days' (374 μg/m^3) and 'with Diwali days' (353 μg/m^3) averaged over 15 years (Fig. 4) is statistically insignificant. We interpret that though Diwali (being a 1–3 day event) enhances $PM_{2.5}$ concentration over the NCT at an hourly to daily scale (e.g. Ghei and Sane, 2018; Lin, 2016; Mukherjee et al., 2018; Pandey et al., 2016), the impact is not detectable at weekly time scale.

3.3. Post 2009 enhancement in the first peak pollution episode

Fig. 5a and b depict inter-annual variability of weekly $PM_{2.5}$ and fire count over Delhi NCR. It can be seen that the peak period (3 weeks

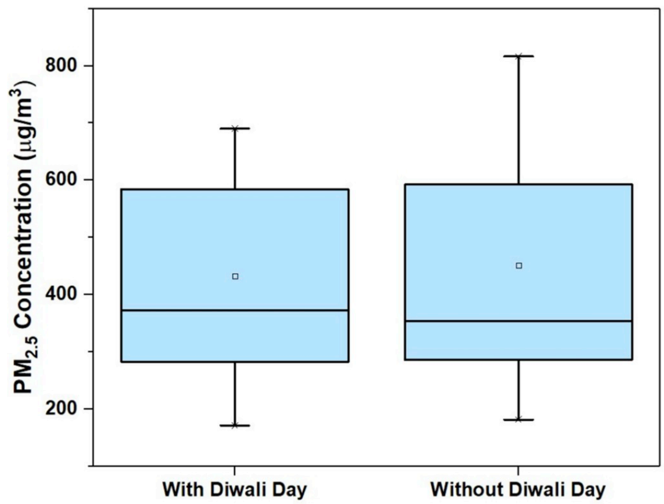

Fig. 4. Box plot (showing 5th to 95th percentile ranges) of ambient $PM_{2.5}$ concentrations during "Diwali" week with and without "Diwali" days.

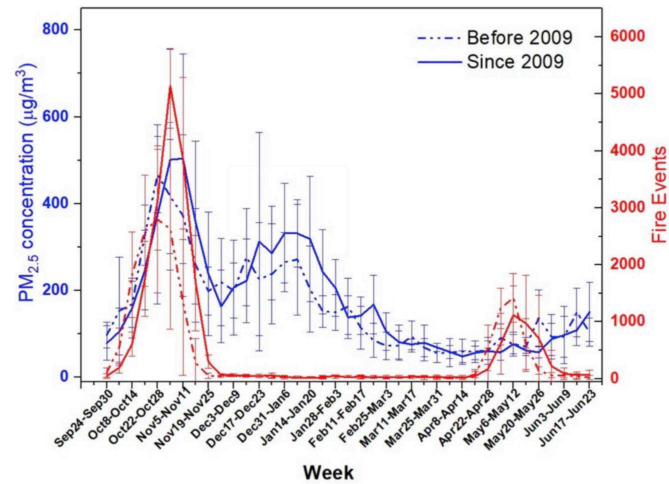

Fig. 6. Weekly $PM_{2.5}$ concentration (in blue) and fire events (in red) in the pre-2009 (dashed lines) and post-2009 era (solid lines) over NCR. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

following the week of October 15–21) fire events have become more intense over the years resulting in steeper and elongated peaks of $PM_{2.5}$ lasting for 2–3 weeks in Delhi NCR. A drop in fire events in 2015 is also reflected in comparatively low $PM_{2.5}$ during this peak episode.

In March 2009, the Government of Punjab drafted legislation entitled 'The Punjab Preservation of Sub Soil Water Act' based on the initiative of the Punjab State Farmers Commission in an attempt to preserve sub-soil water resources (Singh, 2009; Tripathi et al., 2016). This act restricts farmers from sowing rice paddies before May 10 and from transplanting paddies before Jun 10, shifting the rice transplantation season and delaying paddy harvesting, in turn forcing farmers to clear fields for wheat cultivation by burning paddy waste within a few days before sowing wheat. Having less time to manage large amounts of paddy stubble, the farmers resort to crop residue burning rather than other methods to process the residues, such as transporting them to mills or using them as fertilizer or mulch. This is reflected in the 83% increase (from ~2800 to ~5150 fire events) and a shift in peak fire counts from week October 22–28 to October 29-November 4 (Fig. 6). We note that the area under rice cultivation in Punjab did not change drastically after 2009 to drive the radical increase in post-2009 fire events (the area under rice cultivation increased from 2.8 thousand hectares in 2009 to 2.89 million hectares in 2015). As a result, mean ($\pm 1\sigma$) ambient $PM_{2.5}$ concentrations in the Delhi NCR during the first episode increased from $467 \pm 87\,\mu g/m^3$ (in October 22–28) before 2009 to 504 ± 364 (in Nov5–Nov11) $\mu g/m^3$ since 2009 (indicating 7.8% increase of $PM_{2.5}$ concentration and a 2 week shift in the peak).

The corresponding increase in peak $PM_{2.5}$ concentration in Delhi NCT is 8.3% and the peak shifts by 1 week. In the upwind area, the corresponding numbers are 5.3% and 2 weeks and 11% and 1 week in the downwind area respectively (see Fig. S10a-c). Though the enhancement in mean peak concentration may not be large, the persistence of the first peak pollution episode has extended in duration in all the domains (Delhi NCR, Delhi NCT, upwind downwind areas) post 2009.

3.4. The rural-urban divide in pollution build-up in Delhi NCR

Fig. 7 shows the spatial distribution of rural (21.5%), urban (23.3%) and minimal-settlement (remaining 55.2%) areas in Delhi NCR. The corresponding numbers for Delhi NCT are 8%, 86.5%, and 5.5% respectively. Table 1 enlists the mean $PM_{2.5}$ concentration in these different settlement classes for in Delhi NCT, NCR, upwind and downwind areas (marked by box A and B in Fig. 2a). Our results indicate that ambient $PM_{2.5}$ concentration in the urban grids in Delhi NCR is 5% and 7.7% higher than rural and minimal-settlement grids. Furthermore, the urban grids in Delhi NCT have 3.8% higher $PM_{2.5}$ concentrations relative to the urban grids in Delhi NCR. It should be noted that the urban grids in Delhi NCR include the urban grids in Delhi NCT. A larger gradient in $PM_{2.5}$ concentration can be noted between the upwind and the downwind areas. The urban and rural grids in the downwind area are estimated to have 24.1% and 25.7% higher $PM_{2.5}$ concentrations than in the similar settlement classes in the upwind area. The minimal-

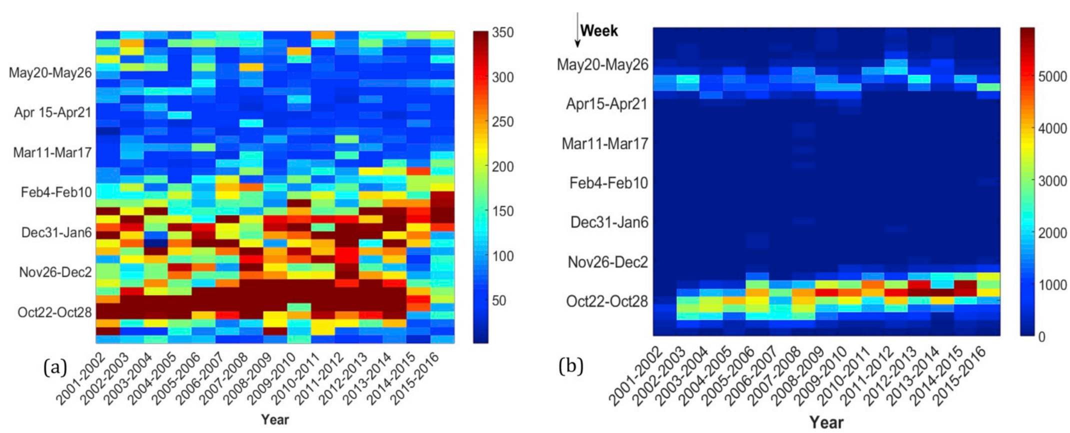

Fig. 5. Inter-annual variability in weekly (a) ambient $PM_{2.5}$ concentrations (in $\mu g/m^3$) in Delhi NCR and (b) fire count in the upwind regions.

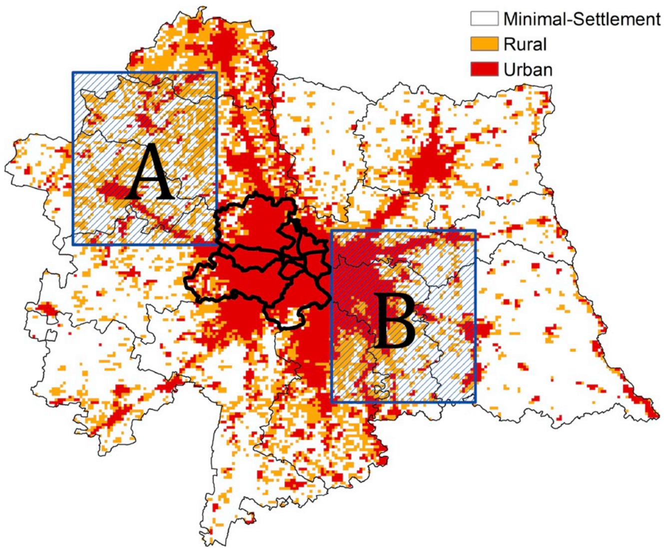

Fig. 7. Identification of 1 km grids in NCR as urban (dark green), rural (light green) and no settlement (white). The two boxes A, B depict the upwind box A and the downwind box B. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

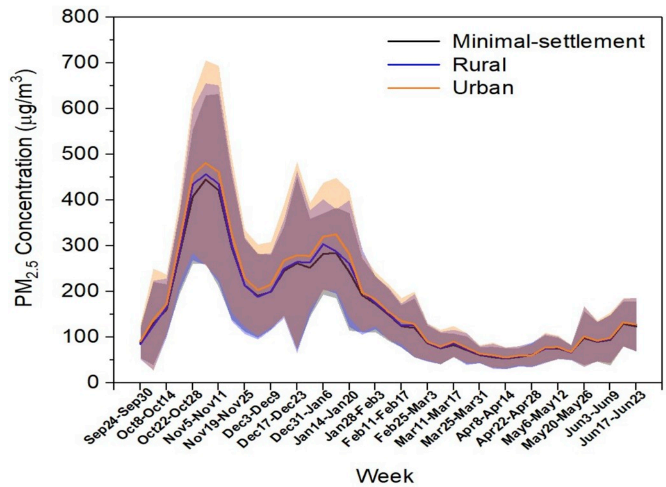

Fig. 8. Weekly $PM_{2.5}$ concentration over the urban (orange), rural (blue) and minimal-settlement (black) settlement classes of Delhi NCR. The solid lines depict mean (shade represents $\pm 1\sigma$) $PM_{2.5}$ concentration. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

settlement class in the downwind area has 25.1% and 13.5% higher $PM_{2.5}$ concentration than in the minimal-settlement classes in the upwind area and in the adjacent Delhi NCT. This further confirms that 'megacity outflow' by prevailing winds from Delhi NCT adds to the local emissions, resulting in higher $PM_{2.5}$ concentrations in the downwind area.

Weekly $PM_{2.5}$ build-up in the rural, urban and minimal-settlement areas in Delhi NCR is shown in Fig. 8. The first peak episode occurs in all three settlement classes at the same time (October 29-November 4) and indicates the strong influence of regional transport (from the area affected by open biomass burning as discussed earlier). Higher $PM_{2.5}$ concentration in the urban grids compared to the rural and minimal-settlement grids can be attributed to local emissions. However, we note that the urban-rural divide in $PM_{2.5}$ concentration in the NCR (or even in the NCT) is not large, as is commonly perceived. This implies a strong role of (1) meteorology and regional transport and (2) high emissions in rural areas of NCR perhaps from residential activities and brick kilns (Guttikunda and Goel, 2013), both of which modulate the build-up of $PM_{2.5}$ during the dry season.

4. Discussion and conclusions

In this study, we examine $PM_{2.5}$ build-up over Delhi NCT and the greater NCR during the dry period using 16 years (2001–2016) of new generation high resolution (1 km) satellite-based AOD data. Satellite-based AOD products are proven to be highly useful in monitoring pollution, especially in regions devoid of consistent ground-based measurements (van Donkelaar et al., 2010, 2014, 2016). We note that satellite-based estimates of $PM_{2.5}$ are conservative estimates because of two factors. In the presence of extremely high pollution cases, the atmosphere becomes too bright to retrieve aerosols and instead are misclassified as clouds. The same is true in the presence of fog, which is common in the winter season in NCR. Studies have observed that more particulate pollution enhances fog formation and leads to longer fog episodes (Mircea et al., 2002; Mohan and Payra, 2009). On the contrary, few studies have found that fog facilitates secondary aerosol formation (Chakraborty et al., 2016; Dall'Osto et al., 2009) and elevates $PM_{2.5}$ concentration. It is difficult to determine which process dominates. These issues merit further investigation by the satellite aerosol community in the future.

Our study indicates two peak pollution episodes in Delhi NCR. Climatologically, the first peak occurs in the week of October 29-November 4. We attribute this peak to transport of $PM_{2.5}$ from upwind crop residue burning sites under stable atmospheric condition. The second peak is slightly lower than the first peak and climatologically occurs in December 30-January 5. We associate this peak with a combination of enhancement in local emission, stable meteorological conditions, and trans-state pollution transport. Ambient $PM_{2.5}$ concentration is 9% higher in Delhi NCT than Delhi NCR. The megacity outflow contributes to the pollution level in the downwind area where $PM_{2.5}$ concentration is 24% higher in the dry season than in the upwind area. The duration of the peak episodes has extended in recent years.

Our results call for significant effort to design and implement policies within the entire NCR (including both urban and rural areas) to curb emissions to reduce $PM_{2.5}$ exposure. The first episode may be mitigated largely by controlling open biomass burning in the upwind states. The Government of India has recently laid out a multi-pronged strategy (GoI, 2018a) to address this issue, which includes incentives for in-situ and on field straw management techniques. To ensure proper implementation of this scheme, the government will need a strong enforcement mechanism and new awareness campaigns among farmers to educate them about other options for crop residues. The second episode can be curbed by reducing local emissions during this period. Recent efforts by the government include implementation of an odd-

Table 1
Mean ($\pm 1\sigma$) ambient $PM_{2.5}$ concentration ($\mu g/m^3$) in various settlement classes in our study region.

	Area-weighted $PM_{2.5}$	Rural $PM_{2.5}$	Urban $PM_{2.5}$	Minimal-settlement $PM_{2.5}$
NCR	171.5 ± 21.4	171.6 ± 21.1	180.2 ± 22.5	167.3 ± 21.1
NCT	186.9 ± 25	179.2 ± 24.3	187.1 ± 26	177.2 ± 22
Upwind area	161.2 ± 19	162.7 ± 17.2	162.4 ± 18.2	160.8 ± 18.6
Downwind area	202.1 ± 24.4	204.6 ± 24.5	201.5 ± 25.6	201.14 ± 24.3

even traffic intervention policy for few days within NCT. This, however, returned minimal dividends in terms of mitigating $PM_{2.5}$ levels (Chowdhury et al., 2017). This may be attributed to the remaining sources of $PM_{2.5}$ in NCR where similar mitigation policies were not enforced. Recently, the government of India has introduced the BS-VI fuel norms within the NCT which has potential to reduce tail-pipe SO_2 emission by 80% compared to older BS-IV norms and also reduce PAH emissions extensively (GoI, 2018b). Apart from concentrating on the transportation sector, the government is also extensively trying to introduce viable cleaner alternatives for industry and power sectors and provide clean energy for household uses, including heating and lighting.

All the policies implemented or planned by the government focus on reducing pollution within Delhi NCT. As demonstrated here, unless an inter-sectoral approach is taken to implement policy at a larger scale, it may not succeed. To truly improve air quality in Delhi NCR, programs must be taken to clean up numerous sectors throughout the broader Indo-Gangetic basin. As can be seen from our analyses, ~25% of Delhi NCR can be classified as rural and has high $PM_{2.5}$ concentration ($171.6 \pm 21.1\,\mu g/m^3$) comparable to urban $PM_{2.5}$ levels in Delhi NCR and NCT. Even in the minimal-settlement areas (that can be considered as background), the PM2.5 concentrations ($167.3 \pm 21.1\,\mu g/m^3$) is about 4 times higher than the annual NAAQS. Hence ambient $PM_{2.5}$ pollution in Delhi NCR is high over the entire domain and shall not be implicitly considered as an urban problem. Although Delhi NCT is well monitored with around 35+ ground-based monitoring stations, the larger Delhi NCR that includes heavily populated satellite cities like Faridabad, Ghaziabad, Noida, and Gurugram are scarcely monitored. Future monitoring in these areas is required to examine the efficacy of the Graded Response Action Plan formulated by the Govt. of India (http://cpcb.nic.in/uploads/GRAP_Notification.pdf).

Finally, we demonstrate the advantage of pollution monitoring using high-resolution satellite data for the development of air quality management plan at large scale, especially in India where adequate ground-based pollution monitoring network is currently lacking. Though the network is being expanded, it may take decades to reach adequate spatial and population-based coverage. In that way, satellite data can provide guidance in placing monitors by identifying local hot and cold spots.

Declaration

Authors declare no competing financial interest.

Acknowledgements

The work is partially supported by a grant from Department of Biotechnology under Indo-UK Program on 'Air Pollution and Human Health' (BT/IN/UK/APHH/41/KB/2016-17). SC and SD acknowledge the Fulbright-Nehru Doctoral Fellowship and Fulbright-Nehru Academic and Professional Excellence Fellowship, respectively. MODIS 1 km AOD data are archived at LP DAAC. We thank the anonymous reviewers for providing constructive comments that helped to improve the earlier version of the manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2019.02.029.

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