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Variability of outdoor fine particulate ($PM_{2.5}$) concentration in the Indian Subcontinent: A remote sensing approach

Sagnik Dey [a,*], Larry Di Girolamo [b], Aaron van Donkelaar [c], S.N. Tripathi [d,e], Tarun Gupta [d,e], Manju Mohan [a]

[a] Centre for Atmospheric Sciences, Indian Institute of Technology Delhi, Hauz Khas, New Delhi 110016, India
[b] Department of Atmospheric Sciences, University of Illinois at Urbana Champaign, 105 South Gregory St., Urbana, IL 61801, USA
[c] Department of Physics and Atmospheric Science, Dalhousie University, Halifax, Nova Scotia, Canada
[d] Department of Civil Engineering, Indian Institute of Technology Kanpur, UP 208016, India
[e] Centre for Environmental Science and Engineering, Indian Institute of Technology Kanpur, UP 208016, India

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ABSTRACT

The rapid increase of aerosols over the Indian Subcontinent over the last decade has the potential for severe health implications. However, the lack of a dense network to measure $PM_{2.5}$ (particles with aerodynamic diameter < 2.5 μm) hinders health risk assessments at regional scale. Here, we utilize Multiangle Imaging SpectroRadiometer (MISR)-retrieved columnar aerosol optical depths to estimate surface $PM_{2.5}$ based on recently published conversion factors that account for the composition and vertical distribution of aerosols. We examine the space–time variability of bias-corrected (utilizing coincident *in-situ* observations) $PM_{2.5}$ over the Indian Subcontinent for the period Mar 2000–Feb 2010. We show that 51% of the subcontinent's 1.4 billion people are exposed to pollution that exceed the World Health Organization's highest annual air quality threshold of 35 μg m^{-3}, while another 13% and 18% are exposed in the ranges 25–35 and 15–25 μg m^{-3} respectively. Of the remaining population who breathe clean air, only 25% live in urban areas. In many regions, the high-levels of pollution are persistent rather than episodic. $PM_{2.5}$ concentrations in the rural areas of the Indo-Gangetic Basin are higher than many urban centers in peninsular India. Five hotspots (where $PM_{2.5}$ increases by > 15 μg m^{-3} over the ten-year period) are identified, which cover parts of the eleven Indian states and Bangladesh affecting ~23% of the population. Our results highlight the urgent need to carry out local cohort studies at these hotspots to better understand the health impacts under local conditions.

1. Introduction

The acute and chronic health impacts from short and long-term exposures to particulate matter are well established in the literature (e.g. Balakrishnan et al., 2002a, 2002b; Cohen et al., 2005; Zanobetti & Schwartz, 2009; Pope et al., 2009; Pope, 2000; Pope et al., 2011a, 2011b; Anenberg et al., 2010, 2011; Brauer et al., 2012). Epidemiological studies examining these impacts rely on long-term ambient (both indoor and outdoor) measurements of particulate matter concentration. In the Indian Subcontinent, home to ~1.4 billion people, high aerosol loading (Dey & Di Girolamo, 2010) and its continuing rapid increase over the last decade (Dey & Di Girolamo, 2011) is a major concern for the potential impacts on health.

Annually, 400–550 thousand premature deaths have been attributed to indoor air pollution in India (Smith, 2000). The epidemiological studies focusing on outdoor air pollution in this region (e.g. Balakrishnan et al., 2011; Rajarathnam et al., 2011) mostly utilize PM_{10} (particles with aerodynamic diameter smaller than 10 μm) that is routinely monitored by the Central Pollution Control Board (CPCB) of India through a network of 342 sites under the National Air Quality Monitoring Programme. These data suggest that PM_{10} concentration has increased at some sites, while it also decreased at some sites in the last few years (CPCB, 2008). PM_{10} enters our respiratory tract, but $PM_{2.5}$ reaches the alveoli region; hence $PM_{2.5}$ is considered as a better exposure indicator than PM_{10} for health impact assessments (WHO, 2006). Recently, CPCB initiated monitoring of $PM_{2.5}$ in six major cities, while other efforts include air quality monitoring and forecasting during Commonwealth Games 2010 (e.g. Sahu et al., 2011). While this is an excellent start, the numbers of monitoring stations are too few for a complete and accurate assessment of regional health risks given the very high spatial and seasonal variability of aerosol loading (Dey & Di Girolamo, 2010). Moreover, most of the CPCB sites are concentrated in the urban areas, leaving the large rural population unchecked. Lack of integrated regional scale analysis of exposures to $PM_{2.5}$ and lack of comprehensive long-term $PM_{2.5}$ monitors in the Indian Subcontinent motivated us to carry out the present exposure analysis using satellite data.

Satellite data can be useful for examining global air quality in the absence of a robust database of *in-situ* $PM_{2.5}$ (hereafter denoted as $[PM_{2.5}]_I$) (e.g. van Donkelaar et al., 2010; Liu et al., 2009; Gupta et al., 2006). While there are numerous factors that contribute to the

* Corresponding author. Tel.: +91 11 2659 1315; fax: +91 11 2659 1386.
E-mail address: sagnik@cas.iitd.ac.in (S. Dey).

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uncertainties in using satellite data to retrieve $PM_{2.5}$ concentrations (e.g. Hoff & Christopher, 2009), these studies have found reasonable agreements with $[PM_{2.5}]_I$ measured at many locations across the globe — particularly when additional chemical and meteorological information from other sources was added to the analysis. Here we present a climatology of the spatial and temporal variability of $PM_{2.5}$ in the last decade (Mar 2000–Feb 2010) over the Indian Subcontinent by applying information from a global chemical transport model and $[PM_{2.5}]_I$ measurements to the MISR aerosol retrieval (hereafter denoted as $[PM_{2.5}]_{MISR}$). The health implications of the decadal exposure to $PM_{2.5}$ are discussed along with the potential sources of uncertainties at a regional scale based on the World Health Organization (WHO) air quality guidelines (WHO, 2006), as well as more recent epidemiological studies (Pope et al., 2009; Pope et al., 2011a, 2011b; Anenberg et al., 2011; Balakrishnan et al., 2011).

2. Satellite data analysis

The MISR Level 2 (Version 22) aerosol product includes columnar aerosol optical depth (AOD) at four wavelengths, segregated by size and shape of the particles and the single scattering albedo (Kahn et al., 2010). Global evaluations of MISR data quality have been reported in detail elsewhere (Kahn et al., 2009, 2010) and include detailed comparisons with the Aerosol Robotic Network (Holben et al., 1998). MISR–AOD is well correlated with AERONET (correlation coefficient, $R=0.83$) over the Indian Subcontinent, but is biased low and this bias (e.g. the bias is 30% for AOD$=0.5$) increases linearly with increasing AOD (Dey & Di Girolamo, 2010). The climatological distribution of MISR–AOD over the Indian Subcontinent has been successfully explained by emission factors, synoptic scale meteorology and topography (Dey & Di Girolamo, 2010; 2011).

Our region of interest (40°N–EQ and 65°–99°E longitude) is subdivided into a $0.5°\times0.5°$ grid, as this grid size provides a robust sample size beyond a grid at the original Level 2 17.6 km$\times$17.6 km resolution, while remaining small enough to capture the spatial variability in aerosol characteristics (Dey & Di Girolamo, 2010). Median AOD is derived for each $0.5°\times0.5°$ grid from all Level 2 pixels whose central coordinates fall within a single $0.5°\times0.5°$ grid for each day. The grids with zero population (i.e. over the oceans) have not been considered for $PM_{2.5}$ statistics shown in Figs. 3 and 4 because of our focus on exposure analysis.

We calculate annual mean $[PM_{2.5}]_{MISR}$ from daily estimates for the 10-year period using spatially varying monthly climatological conversion factors (η, the ratio of $PM_{2.5}$ to columnar AOD) calculated from the GEOS-Chem chemical transport model (v8-01-04; http://www.geos-chem.org) at MISR overpass times. Complete details on the derivation of η are provided in van Donkelaar et al. (2010). In brief, GEOS-Chem solves for the temporal and spatial evolution of aerosol and trace gases using meteorological data sets, emission inventories, and equations that represent the physics and chemistry of atmospheric composition. Daily η values for 2001–2006 were interpolated from the $2°\times2.5°$ resolution used for GEOS-Chem in van Donkelaar et al. (2010) to $0.5°\times0.5°$ resolution used here and averaged to a monthly climatology for application to MISR–AOD values. The simulated aerosol relative vertical structure accounts for the effect of elevated aerosol layers that are observed frequently during premonsoon to monsoon seasons (e.g. Lau & Kim, 2006; Jaidevi et al., 2012) and may potentially impact the AOD–$PM_{2.5}$ relationship. The simulated relative aerosol vertical structure showed good agreement in central Asia with CALIPSO (Cloud-Aerosol Lidar Infrared Pathfinder Satellite Observations) observations during June–December 2006. Uncertainties in η and in a combined MISR-MODIS AOD resulted in a global, population-weighted $[PM_{2.5}]_{MISR\text{-}MODIS}$ uncertainty (1σ) of 25% with a reasonable degree ($R>0.75$ at $2°\times2.5°$) of regional correlations with $[PM_{2.5}]_I$ measurements over many parts of the globe (van Donkelaar et al., 2010).

We compared $[PM_{2.5}]_{MISR}$ with available coincident $[PM_{2.5}]_I$ in India (Fig. 1) to quantify the bias over our study area. The present validation effort, as an extension of van Donkelaar et al. study (2010), was carried out to understand the applicability of η values derived for the period 2001–2006 from MODIS-MISR combined AOD to determine $PM_{2.5}$ from MISR–AOD for 10-year period. Two kinds of datasets were used. Daily mean $PM_{2.5}$ values were measured at Delhi during Dec 3, 2007–May 31, 2008 using a 15-channel aerosol spectrometer (Mohan & Payra, 2008) and at Kanpur during Nov 4, 2009–Feb 26, 2010 using a $PM_{2.5}$ sampler (Gupta et al., 2011). These in-situ measurements were taken on the residential academic campuses of IIT Delhi and IIT Kanpur, thus they may be considered to be more representative of regional pollution levels compared to sites in vicinity of local, intense sources.

$[PM_{2.5}]_{MISR}$ at 17.6 km$\times$17.6 km resolution (Level 2) surrounding these two measurement sites have been compared with $[PM_{2.5}]_I$ (Fig. 1a). $[PM_{2.5}]_{MISR}$ shows a statistically significant linear relation ($R=0.71$) with $[PM_{2.5}]_I$, but with a low bias. The low bias may have its roots in several places. In heavily polluted environments containing a large fraction of absorbing aerosols, MISR has a low bias in AOD of 20–30% (Kahn et al., 2009). This may partly account for the bias observed in Fig. 1a. Comparison of $[PM_{2.5}]_I$ with $PM_{2.5}$ derived from AERONET–AOD using the same η at Kanpur (Fig. 2a) reveals a higher slope (0.39) of the best fit line relative to Fig. 1a, which is consistent with the low bias of MISR–AOD relative to AERONET–AOD observed in previous studies (Dey & Di Girolamo, 2010; Kahn et al., 2009, 2010). The low bias of MISR–AOD over India was previously quantified by Dey and Di Girolamo (2010), where the bias increases with an increase in AOD. The model used in van Donkelaar et al. (2010), as with all global models used in aerosol studies to date, are known to produce a low bias in AOD over the Indian Subcontinent (e.g. Ganguly et al., 2009), which if not proportionally represented by a decrease in simulated $PM_{2.5}$ would impact the accuracy of η. Despite the good overall global agreement of satellite-derived $PM_{2.5}$ (slope$=0.86$; $R=0.83$), van Donkelaar et al. found a large underestimation in values over India (supplementary material in van Donkelaar et al., 2010), consistent with our findings. The reasons for the bias in AOD in these models are not well understood, but could result if the lowest vertical layer of the model is too coarse relative to the injection heights of emissions, vertical mixing is too fast, and/or emission inventories are too low. While model chemistry and dynamics do account for the diurnal cycle in deriving η, the model's emission inventory is not diurnal. A recent study (Rehman et al., 2011) suggests a strong multi-modal diurnal variability in aerosol concentration in a village near Kanpur, with a minimum at the time of the MISR mid-morning overpass time. This could also contribute to the low bias in $[PM_{2.5}]_{MISR}$ relative to $[PM_{2.5}]_I$. Lower correlation ($R=0.65$) between MISR–AOD and $[PM_{2.5}]_I$ (Fig. 2b) relative to the correlation between $[PM_{2.5}]_{MISR}$ and $[PM_{2.5}]_I$ shown in Fig. 1a ($R=0.71$) suggests that using η improves this analysis. Despite the low bias, the correlation shown in Fig. 1a is indicative that large scale spatial and temporal influences on the aerosol field are being captured in deriving $[PM_{2.5}]_{MISR}$.

We further compared monthly averaged $[PM_{2.5}]_I$ that were reported at Hyderabad (Latha & Badarinath, 2010) during Jan–Dec 2003, Anantpur (Balakrishnaiah et al., 2011) during May 2006–Apr 2007, Agra (Kulshrestha et al., 2009) during May 2006–Mar 2008, Kaikhali in Sunderban (Mukherjee et al., 2010) during Dec 2003–Nov 2006, Kanpur and Delhi (locations shown in Fig. 3a) with monthly mean $[PM_{2.5}]_{MISR}$ (Fig. 1b). Again, a good correlation exists between the two datasets, with a low bias in monthly-mean $[PM_{2.5}]_{MISR}$ relative to monthly-mean $[PM_{2.5}]_I$. Note that the low-bias in the monthly comparison is greater (slope of Fig. 1b is smaller than that of Fig. 1a) relative to the daily comparison. We suspect that the key reason for this difference in bias is that the MISR–AOD retrievals, hence $[PM_{2.5}]_{MISR}$, are only done when skies are clear; while monthly $[PM_{2.5}]_I$ includes both clear and cloudy days. Based on the meteorological controls of air pollution alone, there is no reason to expect $PM_{2.5}$ to be the same for clear

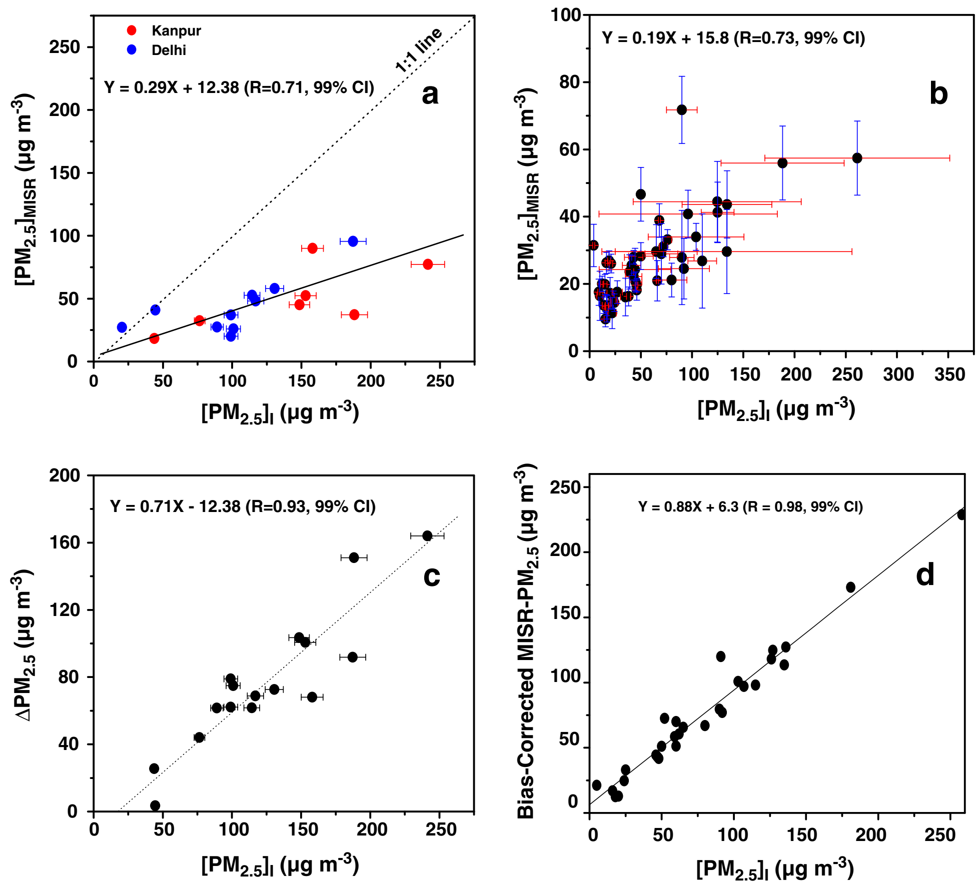

Fig. 1. A Scatter-plot between (a) daily $[PM_{2.5}]_{MISR}$ and $[PM_{2.5}]_I$ at Kanpur (red circles) and Delhi (blue circles), (b) monthly $[PM_{2.5}]_{MISR}$ and $[PM_{2.5}]_I$ at Delhi, Kanpur, Hyderabad, Anantpur, Agra and Sunderban, (c) bias in daily $[PM_{2.5}]_{MISR}$ and $[PM_{2.5}]_I$ and (d) bias-corrected $[PM_{2.5}]_{MISR}$ (using Eq. (1)) and $[PM_{2.5}]_I$. Both daily (representative of clear days) and monthly (in-situ measurements consider both clear and cloudy days, while MISR-retrievals cover only clear days) comparisons show statistically significant (at 99% confidence level, CI) linear relationships with a low bias at high-$PM_{2.5}$ condition. The error bars in 1a and 1c indicate the uncertainties of in-situ measurements, while they represent ±1 standard deviation (σ) in 1b around the mean values. The dotted line in 1a represents 1:1 line. The bias in $[PM_{2.5}]_{MISR}$ increases linearly with an increase in $[PM_{2.5}]_I$ (statistically significant at 99% CI). The relation has been used to produce the $\overline{PM_{2.5}}$ statistics displayed in Table 2 and used for exposure study (Figs. 3 and 4). Locations of the in-situ observations are shown in Fig. 3a. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

and cloudy days. In addition, the MISR and *in-situ* measurements likely have not sampled the same days within a specific month to calculate the monthly average which may lead to increased uncertainty.

We quantify the MISR-bias ($\Delta PM_{2.5} = [PM_{2.5}]_I - [PM_{2.5}]_{MISR}$) based on coincidently sampled daily values at Kanpur and Delhi, which linearly increases with increasing $[PM_{2.5}]_I$ (Fig. 1c) and can be represented by the relation ($R = 0.93$ and $p < 0.01$, significant at 99% CI)

$$\Delta PM_{2.5} = 0.71[PM_{2.5}]_I - 12.38 \tag{1}$$

We corrected this bias for $[PM_{2.5}]_{MISR} > 12.38$ µg m^{-3}, so as to not allow negative $PM_{2.5}$ values and use the bias-corrected $[PM_{2.5}]_{MISR}$ (hereafter referred to as $\overline{PM_{2.5}}$) for the exposure analysis and the statistics presented in Fig. 3 and Table 2. The error (±0.07) in the slope of the linear fit of the bias translates to an uncertainty of ±3% in estimated $\overline{PM_{2.5}}$, in addition to any uncertainty due to η and MISR–AOD. For example, an uncertainty of ±10% in η leads to an uncertainty of ±8% in the threshold value (12.38 µg m^{-3}) of bias correction in our estimates. We stress that in correcting for bias, there is an assumption that the bias remains the same over our entire study domain and over all seasons. The application of this correction, however, should allow quantitative interpretation of these $PM_{2.5}$ estimates by calibrating against bias in both η and MISR–AOD. The degree to which the assumption of a constant calibration holds will require further $[PM_{2.5}]_I$ measurements coincident with the MISR overpass from different locations characterized by different emission sources. The spatial and temporal analysis of aerosol properties presented in Dey and Di Girolamo (2010, 2011) can be used as a guide in choosing these locations. We have applied the bias correction to mean monthly $[PM_{2.5}]_{MISR}$ from various sites (shown in Fig. 1b) and compared with $[PM_{2.5}]_I$ (Fig. 1d) to examine this issue. The slope of the best-fit line increases from 0.19 (in Fig. 1b) to 0.88 (in Fig. 1d) with $R = 0.98$. This supports the fact that the bias correction significantly improves estimation of satellite-based $PM_{2.5}$ and strengthens our approach to establish bias-corrected $PM_{2.5}$ climatology assuming uniform bias over the subcontinent in absence of any further in-situ measurements. Until such measurements are made, bias correction using data presented in Fig. 1b may be used as a guide to the uncertainty caused by this assumption – assuming the sampling differences of this data set and MISR (as discussed above) are ignored. By ignoring this, an upper bound on this error is reached, since coincident sampling at these locations with MISR overpasses on clear days would lead to a smaller error. In doing so, the difference between the bias-corrected $\overline{PM_{2.5}}$ from Eq. (1) and that derived from Fig. 1b is ~8%. A better constraint on the uncertainty in $\overline{PM_{2.5}}$ from MISR using our approach can only be achieved with more long term *in-situ* data coincident with MISR overpasses, as well as further improvements to the GEOS-Chem model (including resolution) and associated assimilated data. Until then, we proceed with the present

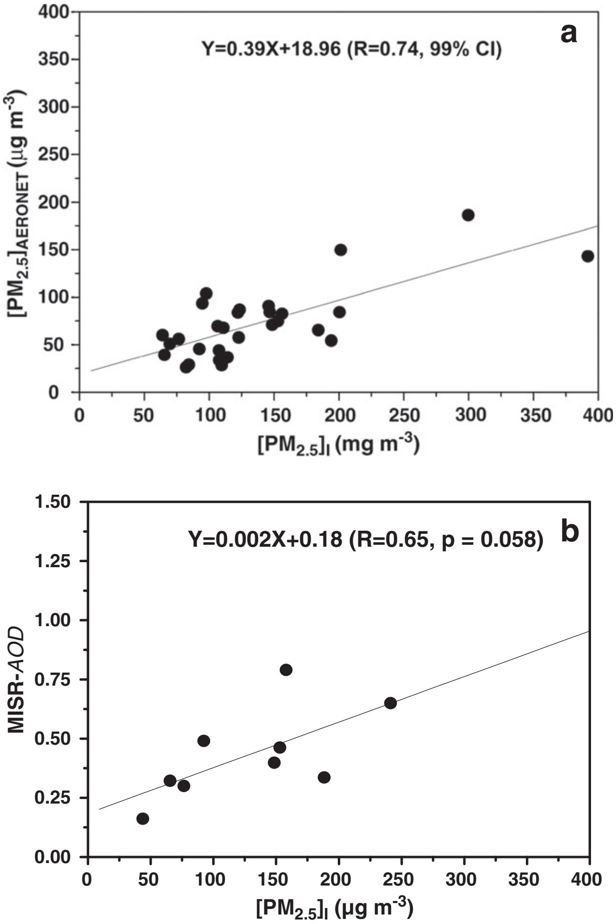

Fig. 2. Scatter plot between (a) daily $[PM_{2.5}]_{AERONET}$ and $[PM_{2.5}]_I$ and (b) MISR–AOD and $[PM_{2.5}]_I$ at Kanpur. $[PM_{2.5}]_{AERONET}$ has been derived by converting the AERONET-retrieved AOD using the same conversion factors (η) that were used to derive $[PM_{2.5}]_{MISR}$.

uncertainty analysis to study the spatio-temporal distributions of $PM_{2.5}$ in the last decade.

3. Spatio-temporal variability of $\overline{PM_{2.5}}$

Population data were collected from 'Socioeconomic data and application center' website (http://sedac.ciesin.columbia.edu/gpw/global.jsp#) for the year 2000 and 2010 at ~5 km×5 km resolution and have been projected onto our 0.5°×0.5° grid following Tobler et al. (1997). The urban extent data are used to distinguish between the 'urban' and 'semi-urban and rural' population within each 0.5°×0.5° grid. Population data of the year 2000 is considered for the exposure analysis because this is the population that has been exposed to decadal changes in pollution. The exposure of the population distribution to annual and daily $\overline{PM_{2.5}}$ is examined based on WHO air quality guidelines, which were guided by epidemiological studies prior to 2006, including Cohen et al. (2005), Dockery et al. (1993), Jerrett et al. (2005), Katsouyanni et al. (2001), Pope et al. (1995, 2002), and Samet et al. (2000).

The ten-year mean annual $\overline{PM_{2.5}}$ (Fig. 3a) exceeds WHO standard of 10 μg m^{-3} (see Table 1 for WHO standards and interim targets, IT) over 70% (49% of the inhabited area) of the subcontinent's area, of which the heavily populated Indo-Gangetic Basin (IGB) and the largely uninhabited Taklamakan Desert stand out with the largest values. 255, 181 and 720 million people (~18%, ~13% and ~51% of the total ~1.4 billion population respectively) are exposed to annual $\overline{PM_{2.5}}$ in the range WHO IT-2 to IT-3, IT-1 to IT-2 and >IT-1, respectively. Out of these 255, 181 and 720 million populations, ~24%, ~18% and ~21% respectively are urban populations. Only ~18% population in the subcontinent is breathing clean air, of which, 25% live in urban area. Our results suggest that not only the urban population, but a large semi-urban and rural population is also at health risk due to this enormous particulate concentration. It must be noted that the magnitude of such impacts on morbidity and mortality may differ from place to place depending on the seasonal variability of $PM_{2.5}$ composition, outdoor exposure time, and other demographic variables (Pope et al., 2011a). For example, larger fraction of dust particles in rural areas may have different impact on human health compared to urban areas dominated by combustion-generated particles. It is not possible to examine this issue here because of the lack of robust chemical composition data from MISR. Qualitatively, exposure at the $PM_{2.5}$ hotspots in Fig. 3a that are characterized by dominant anthropogenic particles as shown in Dey and Di Girolamo (2010) have higher health risks than any other places.

We further focus on the 46 large urban centers with population >1 million (Table 2), because the air quality may be severely affected due to the rapid migration of the population into urban centers for socio-economic reasons. Previously, India did not have any air quality standard for $PM_{2.5}$ and only recently in 2009, the Indian air quality standard for $PM_{2.5}$ was determined as 60 μg m^{-3} for 24-hr average and 40 μg m^{-3} for annual average (www.cpcb.nic.in/National_Ambient_Air_Quality_Standards.php). The annual average Indian $PM_{2.5}$ standard (Table 1) is close to WHO IT-3. Three key points emerge from the statistics summarized in Table 2:

(i) Mean annual $\overline{PM_{2.5}}$ exceeds WHO IT-1 at 29 urban centers, lies in between IT-1 and IT-2 at 10 centers and between IT-2 and IT-3 at 5 centers, whereas it is less than 15 μg m^{-3} at only two urban centers (Bangalore and Coimbatore). Delhi has the highest mean (±1σ, σ is the standard deviation representing the temporal variability) annual $\overline{PM_{2.5}}$ (148.4±67 μg m^{-3}) followed by Meerut (96.4±48 μg m^{-3}). $\overline{PM_{2.5}}$ is more than double of IT-1 at other metro areas in the IGB and Mumbai. $\overline{PM_{2.5}}$ concentrations exceed the annual Indian standard in 18 urban centers.

(ii) The inter-annual variability of $\overline{PM_{2.5}}$ in these centers is not uniform because of the large heterogeneity in seasonal cycles of pollutant emission (Dey & Di Girolamo, 2011).

(iii) Mean annual $\overline{PM_{2.5}}$ does not correlate with the urban centers' population, a proxy for anthropogenic activities (e.g. mean (±1σ) $\overline{PM_{2.5}}$ of 88.7±53 μg m^{-3} at Agra with a population of 1.3 million is more than six times of mean $\overline{PM_{2.5}}$ of 14.4±9 μg m^{-3} at Bangalore with a population of 5.7 million). Since there are no major policy or cultural differences in emission factors across India, the climatology in Table 2 points to the important roles of meteorology in redistributing the pollutants (Dey & Di Girolamo, 2010) based on the broader scale PM emissions (Streets et al., 2003; Venkataraman et al., 2005).

The episodic nature of such high annual $\overline{PM_{2.5}}$ has also been examined. Figs. 3b–d show the percent of days per year when daily $\overline{PM_{2.5}}$ exceeds the three WHO daily interim targets mentioned in Table 1. It is again of note that the daily exposure statistics represent percent of clear days in a year, because MISR aerosol retrievals are not possible on cloudy days (hence our results may vary with $[PM_{2.5}]_I$ statistics that use both clear and cloudy days). Daily $\overline{PM_{2.5}}$ exceeds IT-1 in 40–50% of days in the western to central IGB and Mumbai. The frequency increases to 60–75% for IT-2 and >75% for IT-3 in these regions. This clearly suggests that high annual $\overline{PM_{2.5}}$ is not episodic; rather the high pollution level persists throughout a substantial period of the year, particularly in the heavily populated and industrialized IGB and Mumbai regions (Table 2).

Next we calculate the rate of change of $\overline{PM_{2.5}}$ per month through linear trend analysis of deseasonalized $\overline{PM_{2.5}}$ over the 120 months period. The total changes of $\overline{PM_{2.5}}$ per 0.5°×0.5° grid ($\Delta\overline{PM_{2.5}}$) in

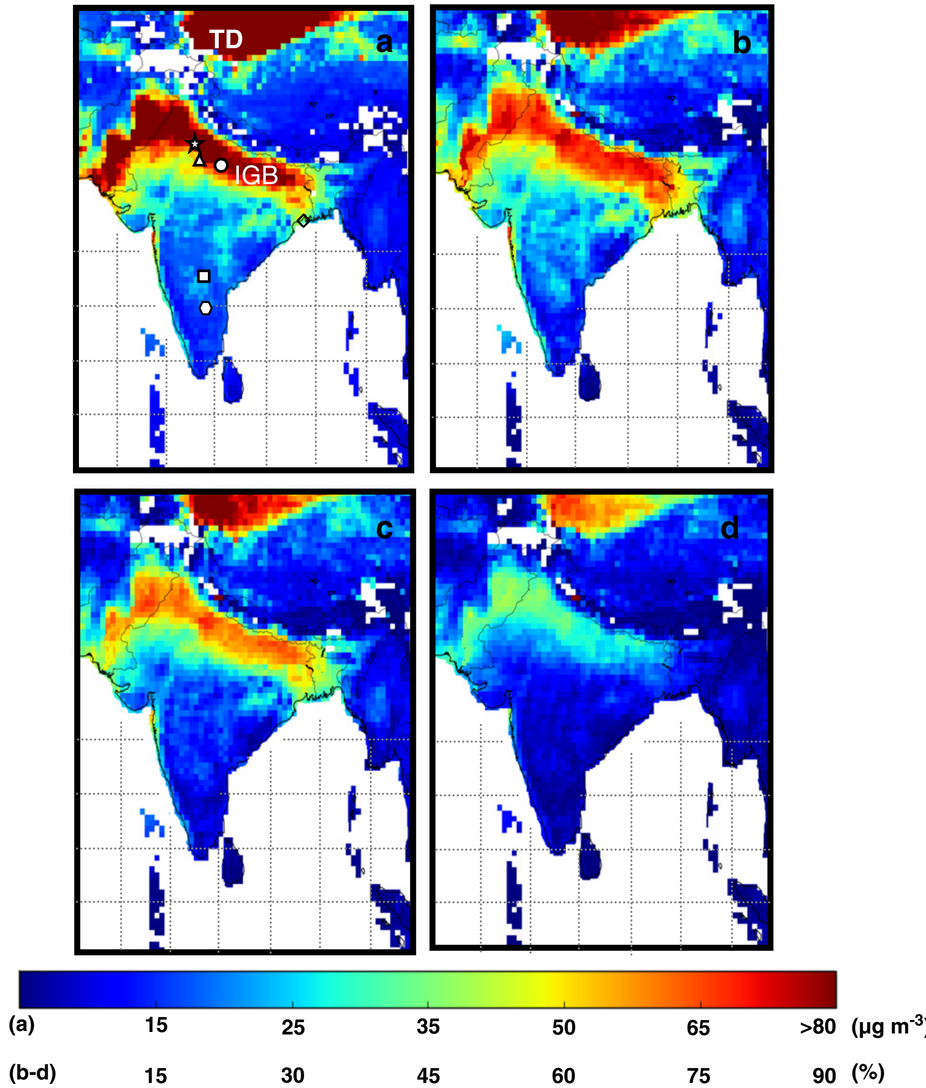

Fig. 3. Spatial distributions of (a) mean annual $\overline{PM_{2.5}}$ concentration (in μg m^{-3}) and percentage of clear days per year with mean daily $\overline{PM_{2.5}}$ exceeding (b) 37.5 μg m^{-3} (WHO IT-3), (c) 50 μg m^{-3} (WHO IT-2) and (d) 75 μg m^{-3} (WHO IT-1) during Mar 2000–Feb 2010 over the Indian Subcontinent. 'IGB' and 'TD' are acronyms of Indo-Gangetic Basin and Taklamakan Desert. 'White' regions represent 'water' or 'no data'. Note different scales for Figs. 3a and b-d. Locations of Delhi, Kanpur, Agra, Hyderabad, Anantpur and Sunderban are shown by 'star', 'circle', 'triangle', 'square', 'hexagon' and 'diamond' respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the last decade are then estimated (Fig. 4) by multiplying the rate of change of $\overline{PM_{2.5}}$ per month by 120 (i.e. total number of months). Most of the regions do not show statistically significant rate of increase ($p>0.05$, i.e. at 95% CI, which roughly corresponds to $\Delta\overline{PM_{2.5}}<15$ μg m^{-3}; although $\Delta\overline{PM_{2.5}}<15$ μg m^{-3} is also observed at few sites for significant rate of increase per month) or decrease of $\overline{PM_{2.5}}$ per month. The statistics have been compiled for all major urban areas with population > 1 million in the subcontinent (last column in Table 2), where bold numbers indicate $\Delta\overline{PM_{2.5}}$ that are estimated based on statistically significant rate of increase of $\overline{PM_{2.5}}$ per month (at 95% CI). The regions where $\overline{PM_{2.5}}$ has increased by more than 15 μg m^{-3} in the last decade are characterized as hotspots. The largest hotspot (marked as H1 in Fig. 4) stretches from Punjab and Haryana in the western IGB to eastern Uttar Pradesh. The second hotspot (H2 in Fig. 4) covers the rural areas of Bihar, West Bengal and northern Bangladesh. The third hotspot (H3) covers parts of Orissa and Chhattisgarh, while the fourth (H4) and fifth (H5) hotspots cover parts of Gujarat and Maharashtra, and Andhra Pradesh respectively. Nearly 23% of the total population in the subcontinent is living under these five hotspots.

Table 1
WHO and Indian standards for annual (1st row) and 24-hr (2nd row) average $PM_{2.5}$ concentration and the three interim targets of WHO (WHO, 2006). All $PM_{2.5}$ concentrations are in μg m^{-3} ('N/A' means 'not available').

	Annual 24-hr	Interim Target 3	Interim Target 2	Interim Target 1
WHO standard	10	15	25	35
	25	37.5	50	75
Indian standard	40	N/A	N/A	N/A
	60	N/A	N/A	N/A

It must be noted that the statistics has been presented here at $0.5° \times 0.5°$ spatial resolution, whereas significant sub-pixel variability in $PM_{2.5}$ likely exists in megacities like Delhi (Kumar et al., 2007). Such sub-pixel variability should be accounted for in more detailed analysis of health effects using a dense local network of $[PM_{2.5}]_I$. Assumption of climatological η over the years may induce a bias in estimated changes of $\overline{PM_{2.5}}$, because η may also change over the years in this region due to a change in emissions at the source that are not accounted for by our model and due to change in meteorology that influences aerosol vertical distribution. However, since the η values are considered to be constant over the period of study, $\overline{PM_{2.5}}$ trends

Table 2
Mean ($\pm 1\sigma$) monthly statistics of $\overline{PM_{2.5}}$ (in μg m^{-3}) over the urban centers (arranged in decreasing order of population) with population greater than 1 million based on the population data of the year 2000. Total changes in $\overline{PM_{2.5}}$ concentration in a 10 year period ($\overline{\Delta PM_{2.5}}$) are given in the last column, where the bold numbers indicate total changes based on statistically significant rate of increase per month at 95% CI. The names of the urban areas in India where mean annual $\overline{PM_{2.5}}$ exceeds the Indian annual standard (Table 1) are written in 'bold' font.

Sr. No.	Urban Center (Country)	Latitude (N) Longitude (E)	Population (million)	Annual $\overline{PM_{2.5}}$ (μg m^{-3})	# Clear days (in %) for daily $\overline{PM_{2.5}}$> WHO IT-3	# Clear days (in %) for daily $\overline{PM_{2.5}}$> WHO IT-2	# Clear days (in %) for daily $\overline{PM_{2.5}}$> WHO IT-1	$\overline{\Delta PM_{2.5}}$ in 10 years (in μg^{-3})
1	**Mumbai** (India)	18.9 72.8	17.66	78.8 ± 42	54.0	44.0	23.2	**24.6**
2	**Delhi** (India)	28.5 77.2	16.39	148.4 ± 67	80.0	70.6	42.1	**31.1**
3	**Kolkata** (India)	22.5 88.3	13.55	88.0 ± 45	63.8	54.8	17.2	23.0
4	Karachi (Pakistan)	24.8 67.0	9.33	76.6 ± 53	63.5	47	36.5	8.2
5	Dhaka (Bangladesh)	23.7 90.3	7.67	46.6 ± 28	57.9	48.8	13.1	4.1
6	Chennai (India)	13.1 80.3	6.42	21.3 ± 12	9.1	5.8	1.9	3.6
7	Lahore (Pakistan)	31.5 74.3	5.14	79.5 ± 42	71.8	63.1	42.2	11.5
8	Bangalore (India)	12.9 77.6	5.68	14.4 ± 9	9.8	5.3	2.3	0.01
9	Hyderabad (India)	17.4 78.4	5.53	22.9 ± 13	15.5	11.8	3.9	**14.1**
10	Ahmadabad (India)	23.0 72.5	4.69	36.5 ± 26	34.3	24.2	13.2	**16.2**
11	Pune (India)	18.4 73.8	3.81	19.4 ± 15	38.3	27.9	10.0	3.9
12	Chittagong (Bangladesh)	22.4 91.8	2.94	29.9 ± 24	50.7	39.8	8.1	7.9
13	**Kanpur** (India)	26.4 80.2	2.84	82.0 ± 45	77.9	68.4	33.1	**33.3**
14	**Surat** (India)	21.2 72.8	2.81	43.3 ± 27	41.0	27.6	14.2	4.6
15	**Lucknow** (India)	26.8 80.8	2.42	84.6 ± 47	77.7	70.1	35.2	**23.1**
16	**Jaipur** (India)	26.9 75.8	2.32	64.0 ± 42	49.9	38.0	25.6	8.5
17	Nagpur (India)	21.1 79.3	2.26	33.9 ± 20	29.5	14.6	5.2	**14.9**
18	Faisalabad (Pakistan)	31.2 73.2	2.00	84.1 ± 46	76.5	67.9	48.4	−2.3
19	**Patna** (India)	25.6 85.1	1.71	86.6 ± 48	78.1	71.7	33.1	**12.5**
20	Indore (India)	22.4 75.5	1.64	27.9 ± 14	25.4	17.1	8.8	**6.4**
21	Vadodara (India)	22.3 73.2	1.49	37.7 ± 31	36.7	21.5	11.1	9.0
22	Bhopal (India)	23.2 77.4	1.45	25.5 ± 15	34.6	23.9	10.9	3.4
23	Coimbatore (India)	11.0 76.9	1.44	10.7 ± 4	7.7	5.0	2.3	0.5
24	**Bhubaneswar** (India)	20.3 85.8	1.43	41.0 ± 29	34.7	22.8	8.1	**14.8**
25	Rawalpindi (Pakistan)	33.6 73.1	1.41	33.5 ± 22	42.2	33.4	23.4	0.3
26	**Ludhiana** (India)	30.9 75.8	1.39	95.3 ± 50	81.0	74.5	47.9	9.2
27	Kochi (India)	9.9 76.2	1.36	25.5 ± 19	32.7	21.1	11.7	**13.0**
28	Visakhapatnam (India)	17.7 83.2	1.33	22.2 ± 17	2.5	2.1	0.4	**12.1**
29	**Varanasi** (India)	25.2 82.9	1.32	71.1 ± 44	76.1	65.5	30.3	**16.1**
30	**Agra** (India)	27.2 78.1	1.32	88.7 ± 53	81.4	71.5	40.8	**21.5**
31	Multan (Pakistan)	30.2 71.4	1.19	91.4 ± 44	76.8	68.5	47.6	−3.3
32	Madurai (India)	9.8 78.1	1.19	20.2 ± 16	10.2	5.3	3.1	5.7
33	**Meerut** (India)	28.9 77.7	1.17	96.4 ± 48	75.9	67.7	38.5	4.1
34	Hyderabad (Pakistan)	25.4 68.4	1.17	94.6 ± 66	68.9	59.0	38.6	−1.3
35	Khulna (Bangladesh)	22.8 89.6	1.17	44.3 ± 29	59.4	47.4	12.8	11.8

Table 2 (*continued*)

Sr. No.	Urban Center (Country)	Latitude (N) Longitude (E)	Population (million)	Annual $\overline{PM_{2.5}}$ (μg m^{-3})	# Clear days (in %) for daily $\overline{PM_{2.5}}$> WHO IT-3	# Clear days (in %) for daily $\overline{PM_{2.5}}$> WHO IT-2	# Clear days (in %) for daily $\overline{PM_{2.5}}$> WHO IT-1	$\overline{\Delta PM_{2.5}}$ in 10 years (in μg^{-3})
36	Gujranwala (Pakistan)	32.1 74.2	1.13	72.1 ± 40	66.8	57.4	40.5	3.9
37	Narayanganj (Bangladesh)	23.6 90.5	1.13	42.6 ± 28	55.4	46.1	11.5	**10.7**
38	Jabalpur (India)	23.1 79.9	1.12	32.2 ± 20	36.5	24.6	12.7	**9.7**
39	**Jamshedpur** (India)	22.8 86.2	1.10	56.2 ± 39	56.6	40.2	15.3	**17.0**
40	**Asansol** (India)	23.7 86.9	1.09	75.2 ± 37	69.9	58.1	22.6	**13.6**
41	Nasik (India)	20.0 73.5	1.08	26.4 ± 21	29.5	21.1	10.7	**12.8**
42	**Dhanbad** (India)	23.8 86.4	1.06	68.6 ± 34	57.9	45.8	17.9	4.4
43	**Allahabad** (India)	25.4 81.5	1.05	74.9 ± 42	73.3	62.8	27.6	19.0
44	**Amritsar** (India)	31.6 74.9	1.01	73.5 ± 41	70.0	62.1	40.5	**22.6**
45	Vijaywada (India)	16.5 80.6	1.01	22.1 ± 15	18.7	5.7	3.3	**9.0**
46	Rajkot (India)	22.3 70.8	1.00	28.3 ± 22	41.4	29.9	18.9	3.0

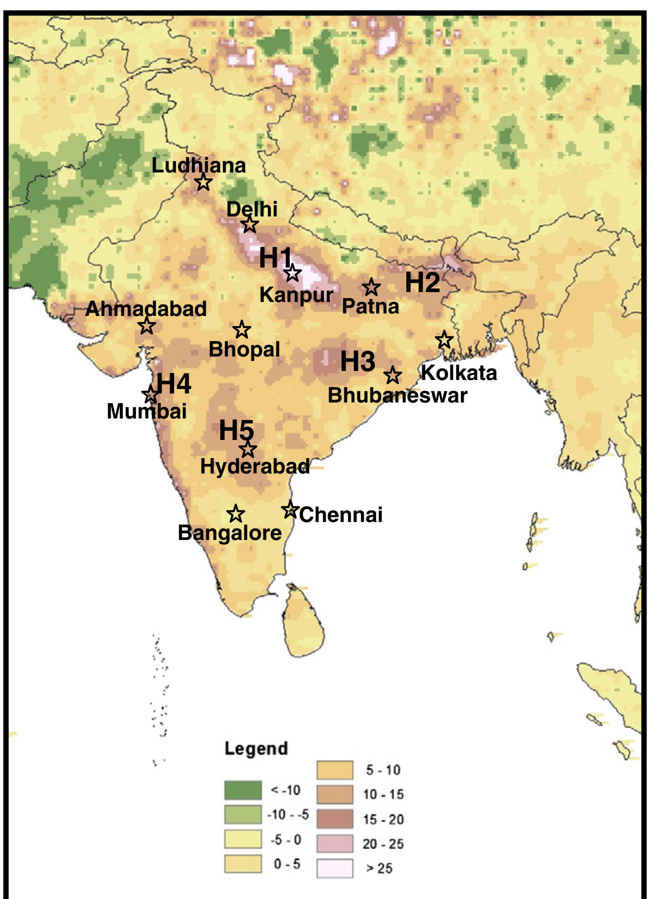

Fig. 4. Spatial distribution of total changes in $\overline{PM_{2.5}}$ concentration (in μg m^{-3}) during Mar 2000-Feb 2010 over the Indian subcontinent. Increase of $\overline{PM_{2.5}}$ by > 15 μg m^{-3} are characterized as hotspots. Five hotspots (marked as H1 to H5) are identified across India and Bangladesh. Locations of some of the large urban centers are also shown (by open star) for a better reference. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

are essentially controlled by MISR–AOD trends. Although trending analysis of AOD using satellite data must be approached with caution due to numerous potential sources of errors in aerosol retrievals, including changes in surface reflectance and in radiometric calibration, as discussed in Li et al. (2009), they have been accounted for here in using MISR following Dey and Di Girolamo (2011). Other recent studies (e.g. Porch et al., 2007; Streets et al., 2009; Zhang & Reid, 2010; Kaskaoutis et al., 2011) have also found increases in AOD at various regions across the subcontinent. Using data from multiple sources, Brauer et al. (2012) have also shown an increase in $PM_{2.5}$ during the period 1990–2005 over the Indian Subcontinent. Decreasing trend in surface-reaching solar radiation (Wild et al., 2005) and visibility (Wang et al., 2009) over this region further provide indirect evidences of increasing aerosol concentration. As pointed out by Dey and Di Girolamo (2011), H1, H3 and H5 are characterized by an increase in aerosols of mostly anthropogenic origin in the post-monsoon to winter seasons, while H2 and H4 are characterized by increase of aerosols from both anthropogenic and natural sources. In contrast, the large decrease (by more than 10 μg m^{-3}) in $\overline{PM_{2.5}}$ over southwestern Pakistan (Fig. 4) is attributed to a decrease in AOD, possibly due to an increase in vertical air motion observed in the region over the past decade (Dey & Di Girolamo, 2011).

4. Implications for human health impacts

Discussion is warranted about the uncertainties in interpreting the exposure analysis for health impacts. According to WHO guideline (WHO, 2006), long-term exposure to mean annual $PM_{2.5}$ > IT-1, > IT-2 and > IT-3 has 15%, 9% and 3% higher mortality risks, respectively, relative to long-term exposure to annual standard. The corresponding daily standards are 75 μg m^{-3}, 50 μg m^{-3} and 37.5 μg m^{-3} (Table 1). Daily $PM_{2.5}$ exceeding these three standards for ≥ 3 days in a year has 5%, 2.5% and 1.2% higher short-term mortality risks relative to exposure to daily $PM_{2.5}$ < 25 μg m^{-3}. These interim targets have been determined based on the epidemiological studies done in developed countries, where $PM_{2.5}$ concentration greater than 35 μg m^{-3} was considered to be very high (e.g. Jerrett et al., 2005; Pope et al., 2002). $PM_{2.5}$ concentration is twice as large as that of IT-1 at many places in the subcontinent throughout a substantial fraction of the year. The mortality risks from exposure to this high persistent $PM_{2.5}$ needs to be better quantified.

Recently, Pope et al. (2011a) suggested that a 10-year exposure carries only a slightly larger (~2%) relative mortality risk compared to an annual exposure based on evidence of a non-linearity between risk and time scale of the exposure. However, the exposure-response relation is highly variable depending on various socio-economic and demographic factors *viz.* indoor vs. outdoor exposure duration, lifestyle, nutrition level, smoking habit etc. (e.g. Brauer et al., 2012; Pope et al., 2011a, 2011b; Pope & Dockery, 2006; Cohen et al., 2005; Mohan et al., 2010). For example, Balakrishnan et al. (2011) has not found any strong evidence of non-linearity in the exposure-mortality relation in Chennai city in India based on the analysis of PM_{10}. Another study conducted in Delhi (Rajarathnam et al., 2011) has found evidence of flattening of mortality-exposure curve at $PM_{10} > 400\ \mu g\ m^{-3}$. Based on the analysis of health data (mortality from cardiopulmonary causes in adults, lung cancer and acute respiratory infections in children) from major urban areas in the world, Cohen et al. (2005) concluded that risk of death linearly increases with an increase in annual $PM_{2.5}$ in the range 7.5–50 $\mu g\ m^{-3}$ and the risk does not increase significantly in response to further increase in $PM_{2.5}$ beyond 50 $\mu g\ m^{-3}$. Such thresholds in $PM_{2.5}$ are not well-defined for India, particularly at the hotspots shown here where high $PM_{2.5}$ concentration is persistent throughout the year.

Personal exposure time may be longer in view of the fact that our $\overline{PM_{2.5}}$ estimates are representative of outdoor conditions only and representative of broad (0.5°×0.5°) areas. Estimated burden of diseases for cardiopulmonary and lung cancer mortality due to outdoor $PM_{2.5}$ is quite high in the subcontinent, but the uncertainties may be as high as 50% due to uncertainties in exposure-response relations and varying concentration thresholds (Anenberg et al., 2010). In the future, stricter Indian standard may be adopted based on epidemiological studies. Improved assessment of the uncertainties in estimating health impacts of outdoor $PM_{2.5}$ in the subcontinent due to all these factors and justification of the above standard are only possible with detailed cohort studies within the hotspots identified in this study. The exposure-response relation at this persistently high ambient $PM_{2.5}$ concentration needs to be re-examined to better quantify the health effects of this enormous pollution level. The robust long-term $PM_{2.5}$ database presented here may guide future studies in that direction.

5. Summary and conclusions

We present ten-year statistics of the spatial patterns of outdoor $PM_{2.5}$ in the Indian Subcontinent for the first time based on satellite data. Our primary focus is on the exposure analysis in view of the WHO air quality guidelines. The present study utilizes the methodology discussed in van Donkelaar et al.'s (2010) and Brauer et al.'s (2012) study, but reports many additional contributions. Our focus is on the Indian Subcontinent, where a synoptic view of space–time variability of $\overline{PM_{2.5}}$ is lacking. We have carried out detailed comparison with *in-situ* data to quantify the bias in the remote-sensing based methodology at regional scale, which suggests the initial space-based $\overline{PM_{2.5}}$ estimates may be regionally underestimated and that a more detailed analysis of local AOD–$PM_{2.5}$ relationships over India is needed. The statistics of space–time distribution of $\overline{PM_{2.5}}$ has been generated for the full decade. Moreover, the persistency of the pollution level has been examined in view of the WHO interim targets. The changes in $\overline{PM_{2.5}}$ during the last decade have also been estimated. The major conclusions of the present study are as follows:

1. We show that nearly 1.1 billion people (~82% of the total population) from the eight developing countries in the subcontinent are breathing air with $PM_{2.5}$ concentration in excess of the WHO-air quality guidelines (of 10 $\mu g\ m^{-3}$) for a substantial period of the year. This has significantly worsened over the past decade.
2. $PM_{2.5}$ concentration in the populated rural areas of the IGB is larger than many urban centers in peninsular India. Mean annual $\overline{PM_{2.5}}$ is persistently greater than 50 $\mu g\ m^{-3}$ in the IGB and Mumbai metropolitan area.
3. Five hotspots covering parts of eleven Indian states and parts of Bangladesh are identified, where $PM_{2.5}$ has increased by >15 $\mu g\ m^{-3}$ (interim target 3 of WHO) over the past decade. 23% population of the subcontinent is at risk due to exposure to this enormous rise of pollution at these five hotspots.

Our results demonstrate first regional scale synoptic view of outdoor air particulate pollution over the Indian Subcontinent for an entire decade. The background concentration and rise in magnitude of aerosol loading over the past decade are alarming. We call for more focused cohort studies at local scales using *in situ* observations. In the Indian Subcontinent, emphasis so far has been on indoor air pollution (e.g. Smith, 2000; Balakrishnan et al., 2002a, 2002b; Hu & Balakrishnan, 2005). The effect of outdoor particulate matter concentration on mortality has only been examined in few urban centers (e.g. Balakrishnan et al., 2011; Rajarathnam et al., 2011), and has only been based on PM_{10} concentration. Future expansion of ground-based monitoring networks to monitor $PM_{2.5}$ should consider the hotspots identified in this study. In addition, future satellite aerosol retrievals should be done at scales much smaller than that of MISR's 17.6 km retrieval. This will enable us to carry out better scale-matching with $[PM_{2.5}]_I$ in the context of highly variable aerosol concentration typically found within urban settings and map the variability (e.g., Kumar et al., 2007). We recommend further actions to improve the risk assessments of health impacts of the large pollution over the Indian Subcontinent, namely:

(i) Generate a national health database and carry out cohort studies at both urban and rural hotspots.
(ii) Establish spatially-varying exposure-response relationships as function of the social and demographic variables appropriate for the Indian subcontinent.
(iii) Examine the composition of $PM_{2.5}$ at these hotspots, because varying levels of toxicity in $PM_{2.5}$ may have different exposure-mortality and exposure-morbidity relations.

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