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Local characteristics of and exposure to fine particulate matter ($PM_{2.5}$) in four indian megacities

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ABSTRACT

Public health in India is gravely threatened by severe $PM_{2.5}$ exposure. This study presents an analysis of long-term $PM_{2.5}$ exposure in four Indian megacities (Delhi, Chennai, Hyderabad and Mumbai) based on in-situ observations during 2015–2018, and quantifies the health risks of short-term exposure during Diwali Fest (usually lasting for ~5 days in October or November and celebrating with lots of fireworks) in Delhi for the first time. The population-weighted annual-mean $PM_{2.5}$ across the four cities was 72 μg/m^3, ~3.5 times the global level of 20 μg/m^3 and 1.8 times the annual criterion defined in the Indian National Ambient Air Quality Standards (NAAQS). Delhi suffers the worst air quality among the four cities, with citizens exposed to 'severely polluted' air for 10% of the time and to unhealthy conditions for 70% of the time. Across the four cities, long-term $PM_{2.5}$ exposure caused about 28,000 (95% confidence interval: 17,200–39,400) premature mortality and 670,000 (428,900–935,200) years of life lost each year. During Diwali Fest in Delhi, average $PM_{2.5}$ increased by ~75% and hourly concentrations reached 1676 μg/m^3. These high pollutant levels led to an additional 20 (13–25) daily premature mortality in Delhi, an increase of 56% compared to the average over October–November. Distinct seasonal and diurnal variations in $PM_{2.5}$ were found in all cities. $PM_{2.5}$ mass concentrations peak during the morning rush hour in all cities. This indicates local traffic could be an important source of $PM_{2.5}$, the control of which would be essential to improve air quality. We report an interesting seasonal variation in the diurnal pattern of $PM_{2.5}$ concentrations, which suggests a 1–2 h shift in the morning rush hour from 8 a.m. in pre-monsoon/summer to 9–10 a.m. in winter. The difference between $PM_{2.5}$ concentrations on weekdays and weekend, namely weekend effect, is negligible in Delhi and Hyderabad, but noticeable in Mumbai and Chennai where ~10% higher $PM_{2.5}$ concentrations were observed in morning rush hour on weekdays. These local characteristics provide essential information for air quality modelling studies and are critical for tailoring the design of effective mitigation strategies for each city.

1. Introduction

Exposure to fine particulate matter (particles with an aerodynamic diameter less than 2.5 μm, $PM_{2.5}$) can pose a major threat to human health (Chowdhury and Dey, 2016; Gao et al., 2017, 2018a; Huang et al., 2018; Pope et al., 2009; Wang et al., 2017). As a rapidly developing country with an expanding population, India is suffering severe $PM_{2.5}$ pollution, with nine cities among the top ten most polluted cities in the world as reported by the World Health Organization (WHO, 2016). Exposure to high levels of $PM_{2.5}$ causes ~1 million premature mortality per year across India (Conibear et al., 2018a). In order to tackle this $PM_{2.5}$ pollution, the Central Pollution Control Board (CPCB) of India set revised National Ambient Air Quality Standards (NAAQS) in 2009 that included $PM_{2.5}$ regulations (CPCB, 2009). Some mitigation policies have been implemented in major Indian cities (Chowdhury et al., 2017; Sharma and Dixit, 2016), but limited improvement in air

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quality (~10% reduction in $PM_{2.5}$) has been seen (Chowdhury et al., 2017). $PM_{2.5}$ pollution is expected to further deteriorate in the coming decades (Chowdhury et al., 2018; Conibear et al., 2018b), due to rapid ongoing urbanization. This surface pollution over India also has important global implications through effective transport by the Asian summer monsoon to the upper troposphere and lower stratosphere, where pollutants can be re-distributed on a global scale and thus affect global climate forcing and air quality (Lelieveld et al., 2018; Liu et al., 2015; Yu et al., 2017).

Previous studies estimated health risks in India of exposure to $PM_{2.5}$ based on model analysis or satellite retrieves and mainly focused on long-term exposure (e.g., Chowdhury and Dey, 2016; Conibear et al., 2018a, b; Gao et al., 2018b; Lelieveld et al., 2015; van Donkelaar et al., 2015). In addition, intensive emissions and unfavourable meteorological condition for dispersion can significantly increase $PM_{2.5}$ and lead to hazardous short-term exposure with high health risks (Atkinson et al., 2014; Héroux et al., 2015). In-situ observations at high temporal resolution are valuable for more firmly grounded estimates of health risks. Furthermore, characterizing the seasonal and diurnal variations of urban $PM_{2.5}$ concentrations and their relationships to meteorology is the key to understanding the drivers of air pollution and devising effective mitigation strategies in Indian megacities (Schnell et al., 2018). Long-term in-situ monitoring studies are critical for a better understanding of these factors. However, only a few studies providing long-term observations of $PM_{2.5}$ have been undertaken, and most of these have focused on Delhi only (Sahu and Kota, 2017; Sharma et al., 2018). Information on local characteristics such as the diurnal variation in pollutant emissions is also critical for modelling studies. This information is scarce in India and models typically use a constant diurnal profile of emissions (e.g., Mohan and Gupta, 2018) or standard profiles from American or European cities to represent conditions in India (e.g., Marrapu et al., 2014). Long-term observations of the diurnal variation of pollutants would provide essential information for improving model performance.

This study presents a comprehensive summary of the seasonal and diurnal variation of urban $PM_{2.5}$ in four Indian megacities (Delhi, Chennai, Hyderabad and Mumbai), based on ground observations from 2015 to 2018. This analysis reveals the observation-based patterns of human activity and local temporal characteristics of emissions in each city, and hence provides valuable input for modelling studies. In addition, for the first time, we report the influences of weekend effect on the diurnal variations and quantify the health risks of short-term exposure during Diwali Fest. Finally, the cumulative exposure of urban residents to $PM_{2.5}$ and the corresponding health burdens are estimated for each city. The results of this study are valuable for the designation and implementation of mitigation policies on a city level aimed at improving air quality to meet the Indian NAAQS standards.

2. Materials and methods

2.1. Data

Datasets of pollutants measured between 1 March 2015 and 31 December 2018 are analysed in this study. An overview of the data is given in Table S1. Hourly $PM_{2.5}$ observations in Delhi, Chennai, Mumbai and Hyderabad (Fig. S1) are rountinely made at U.S. Embassy and consulates using a beta attenuation monitor (San Martini et al., 2015). These records are available from the AirNow website (https://www.airnow.gov/). The instruments are maintained and calibrated following the regulations of the U.S. Environmental Protection Agency (EPA, 2009, 2015). $PM_{2.5}$ observations from the U.S. Embassy are widely used in previous studies in India (Wang and Chen, 2019) and China (Lv et al., 2015, 2017; San Martini et al., 2015), and have been shown to be of good quality and in good agreement with other observations (Jiang et al., 2015; Mukherjee and Toohey, 2016).

We use hourly meteorological observations at the airport in each city (VIDP-Delhi, VOMM-Chennai, VABB-Mumbar and VOHY-Hyderabad). The flat topography surrounding these airports suggests that the observations are broadly representative of the dominant meteorological conditions in these cities. Historical records are archived by the National Oceanic and Atmospheric Administration, and are available from the National Climatic Data Center (https://www.ncdc.noaa.gov/data-access/). The height of the planetary boundary layer (PBL) is obtained from the European Center for Medium-Range Weather Forecasts (ECMWF) ERA-interim reanalysis at a 3-h interval and $0.125^{\circ} \times 0.125^{\circ}$ spatial resolution (https://www.ecmwf.int/).

2.2. Method

We estimate the long-term health impacts from exposure to ambient $PM_{2.5}$ concentrations, as these account for the majority of the health effects through capturing both acute and chronic responses. Following our previous works (Conibear et al., 2018a, b), we use integrated exposure-response (IER) functions (Burnett Richard et al., 2014), updated for the Global Burden of Disease GBD2016 (GBD, 2016) to estimate the relative risk (RR) of premature mortality due to exposure to $PM_{2.5}$ concentrations. There are IER functions with age-specific modifiers for chronic obstructive pulmonary disease (COPD), lower respiratory infection (LRI), ischaemic heart disease (IHD), cerebrovascular disease (CEV), and lung cancer (LC). We use the parameter distributions from the GBD2016 for 1000 simulations to derive the mean IER with 95% uncertainty intervals. The IER functions have uniform theoretical minimum risk exposure levels for $PM_{2.5}$ between 2.4-5.9 $\mu g/m^3$.

We use multi-year average annual-mean $PM_{2.5}$ concentrations from measurements made at U.S. diplomatic missions in Delhi (110 $\mu g/m^3$), Chennai (33 $\mu g/m^3$), Hyderabad (56 $\mu g/m^3$), and Mumbai (60 $\mu g/m^3$). Baseline mortality data are taken from the GBD2016 for India (GBD, 2018). Population size was taken from the lastest Indian Census data for 2011. Population age composition was taken from the GBD2016 population estimates for 2015 for India (GBD, 2017a).

Annual premature mortality (M) for each age and disease were estimated as a function of population (P), baseline mortality rates (I), and the attributable fraction (AF) for a specific relative risk (RR) (Equation (1)). The disease burden from LRI, IHD, CEV, COPD, and LC was estimated between 0 and 95 years upwards in 5 year groupings.

$$M = P \times I \times AF, \qquad AF = \frac{RR - 1}{RR} \tag{1}$$

Annual years of life lost (YLL) for each age and disease were estimated as a function of premature mortality and age-specific life expectancy (LE) from the standard reference life table from the GBD2016 (Equation (2)) (GBD, 2017b).

$$YLL = M \times LE \tag{2}$$

We estimate the short-term health impacts during Diwali Fest in Delhi from exposure to ambient $PM_{2.5}$ concentrations as all-cause premature mortality. The short-term health impacts are accounted for within the long-term health impacts, and are used to indicate the variation in the daily burden from acute responses (Héroux et al., 2015). We use the summary risk estimates (γ) from Atkinson et al. (2014) of 1.04% (0.52–1.56) per 10 $\mu g/m^3$ change in daily mean $PM_{2.5}$ concentrations (C_d), with respect to a reference $PM_{2.5}$ concentration (C_r) of 0 $\mu g/m^3$. We assume no upper concentration cutoff. India-specific risk functions for ambient $PM_{2.5}$ exposure do not currently exist, however, the use of the summary risk estimate of 1.04% is conservative when compared with the summary risk estimate of 1.2% from Levy et al. (2012) and 1.23% from WHO (2013). Baseline mortality data are taken from the GBD2016 for India for all ages for both genders combined (GBD, 2018). We convert these annual rates to daily rates (I_d) by dividing by 365.25, consistent with previous work due to the lack of daily data (West et al., 2007). We use first-three-day of Diwali Fest (320 $\mu g/m^3$) and October–November two-month (183 $\mu g/m^3$) averaged daily-mean $PM_{2.5}$

concentrations during 2015–2018 from the U.S. Embassy measurements for Delhi.

$$RR_d = 1 + [\gamma \times (C_d - C_r) \times 0.1] \quad (3)$$

$$M_d = P \times I_d \times \frac{RR_d - 1}{RR_d} \quad (4)$$

We use a linear exposure-response function with no cap on daily relative risk (RR_d), similar to a previous work (van Donkelaar et al., 2011), estimating daily relative risks following Equation (3). Daily premature mortality (M_d) is then estimated using Equation (4).

Using a logarithmic exposure-response function as in previous work (Crippa et al., 2016), our estimates of short-term premature mortality are about 10% larger than with a linear exposure-response function. To be conservative, we use the linear exposure-response function in this study.

3. Results

3.1. Overview of $PM_{2.5}$ in four megacities

The locations of Delhi, Chennai, Hyderabad and Mumbai are shown in Fig. 1, together with annual mean surface concentrations of $PM_{2.5}$ of anthropogenic origin over India in 2015 (van Donkelaar et al., 2011, 2015). Fig. 2 shows a calendar-view of daily average $PM_{2.5}$ concentrations in the four cities during 2015–2018, and monthly statistics are shown in Fig. S1. There is no clear inter-annual trend in $PM_{2.5}$ observed in these cities during 2015–2018. The Indian NAAQS classifies six different levels of air quality based on daily 24-h averaged $PM_{2.5}$ concentrations (Fig. 2). The two cleanest air quality levels, 'good' and 'satisfactory', are defined as healthy, and the others ($PM_{2.5} > 60\ \mu g/m^3$) are defined as unhealthy (CPCB, 2014). Delhi suffers the worst air quality among these cities, and the air quality levels are categorized as 'poor', 'very poor' or 'severe' for ~50% of the time. These hazy days mostly occur during October–February. The air quality in Chennai and Hyderabad is much better than Delhi, with few 'poor' air-quality days; and 'healthy' days counted up to 50% of the time in Hyderabad and most of the time in Chennai. Mumbai has better air quality than Delhi. This may be due to its coastal climate, where surface $PM_{2.5}$ is often diluted by clean air from the ocean. However, Mumbai still experiences about four months per year with air quality of 'poor' standard or worse. The Diwali Fest and New Year festivals make the air quality substantially worse in Delhi, as shown by the 'severe' days at the beginning of November and January (Fig. 2a). This suggests that the fireworks during the festivals contribute to an increase of $PM_{2.5}$ loading in Delhi significantly. However, there is no clear festival effect observed in the other three cities. It is unclear why no festival effect is observed in these other cities, although it may reflect lower firework use and more favourable meteorological conditions for dispersion in coastal cities.

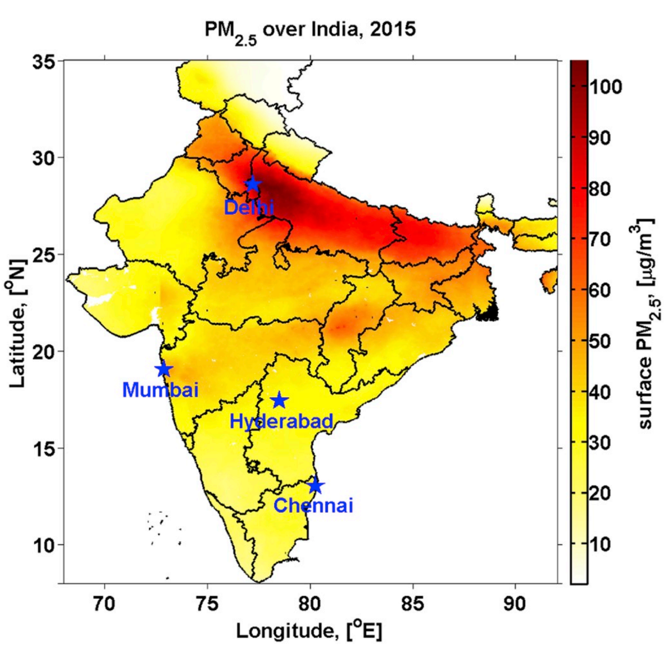

Fig. 1. Map of Delhi, Chennai, Hyderabad and Mumbai. Surface annual (2015) average of $PM_{2.5}$ is retrieved from satellite observations with sea-salt and dust excluded and at a relative humidity of 35% (van Donkelaar et al., 2015).

All cities suffered severe episodes of poor air quality, with maximum hourly $PM_{2.5}$ concentrations of 1676 $\mu g/m^3$, 1334 $\mu g/m^3$, 1107 $\mu g/m^3$ and 758 $\mu g/m^3$ in Delhi, Chennai, Hyderabad and Mumbai, respectively. In Delhi, the maximum hourly $PM_{2.5}$, observed during the Diwali Fest nights in 2016 and 2018, is ~70% higher than the highest level recorded in Beijing (980 $\mu g/m^3$), China (San Martini et al., 2015; Wang et al., 2014; Zheng et al., 2015). This strongly suggests that control of fireworks during the Diwali Fest would efficiently mitigate short-term $PM_{2.5}$ exposure in Delhi. This is also implied by a previous study (Singh et al., 2010), where a significant increase in particle loading by a factor of 2–6 compared with the period before and after Diwali Fest was found in Delhi during 2002–2007. Extreme episodes in other cities were observed at night-time (10 p.m.-2 a.m.) from the end of October to the beginning of December. The shallow planetary boundary layer (PBL) at night and intensive crop burning in this season are the likely reasons for these extremely high concentrations (Tiwari et al., 2013). Fig. S2 shows that there is a clear decrease in the frequency of high $PM_{2.5}$ concentrations in all cities as the PBL height increases. We also observe an anti-correlation between wind speed and $PM_{2.5}$ loading. With the same PBL height, $PM_{2.5}$ loading generally decreases as wind speed increases, and $PM_{2.5}$ is generally less than 100 $\mu g/m^3$ when the wind speed is greater than 4 m/s in all cities (Fig. S2). This is because the higher PBL and larger wind speed dilute the surface $PM_{2.5}$ (Chen et al., 2009; Mohan and Gupta, 2018).

In order to investigate the possible source regions of $PM_{2.5}$ for each city, we analyse the relationship between $PM_{2.5}$ concentration and wind direction (Fig. 3). Delhi is influenced by easterly and westerly/northwesterly winds, with high $PM_{2.5}$ concentrations (>150 $\mu g/m^3$) from both directions. The westerly and northwesterly winds have the highest frequency (~33%) and are associated with the most polluted episodes in Delhi. About 30% of the time $PM_{2.5}$ concentration in Delhi are higher than 150 $\mu g/m^3$, ~50% of which is associated with a westerly or northwestly wind. This indicates that crop biomass burning and desert dust could be major sources of $PM_{2.5}$ in Delhi. Punjab and Haryana are located to the northwest of Delhi, and are major sources of particles and gaseous precursors from crop burning during October–November (Cusworth et al., 2018; Jethva et al., 2018; Rastogi et al., 2014), when the worst air quality is observed in Delhi. Furthermore, previous modelling studies show significant increases (>50%) in aerosol loading when the westerly and northwesterly wind transports dust from the Thar Desert to Delhi during April–June (Kumar et al., 2014a, 2014b). In Hyderabad, another inland city, the easterly/westerly wind pattern is also dominant. The easterly wind brings a substantial amount of $PM_{2.5}$ to Hyderabad, but the conditions are better than in Delhi, with limited episodes of $PM_{2.5}$ concentration higher than 150 $\mu g/m^3$. Chennai and Mumbai are coastal cities with a prevailing onshore wind for 70–80% of the time which brings relatively clean marine air masses. The $PM_{2.5}$ concentrations are generally lower than 75 $\mu g/m^3$ when an onshore wind is present. The offshore wind brings pollutants from inland regions to the cities, but this occurs much less frequently (20–30%). These results indicate that there is a strong interaction between meteorology and $PM_{2.5}$ pollution, and strong local characteristics are found in each city. Detailed investigation of these local characteristics would be helpful in tailoring an effective mitigation policy for each city.

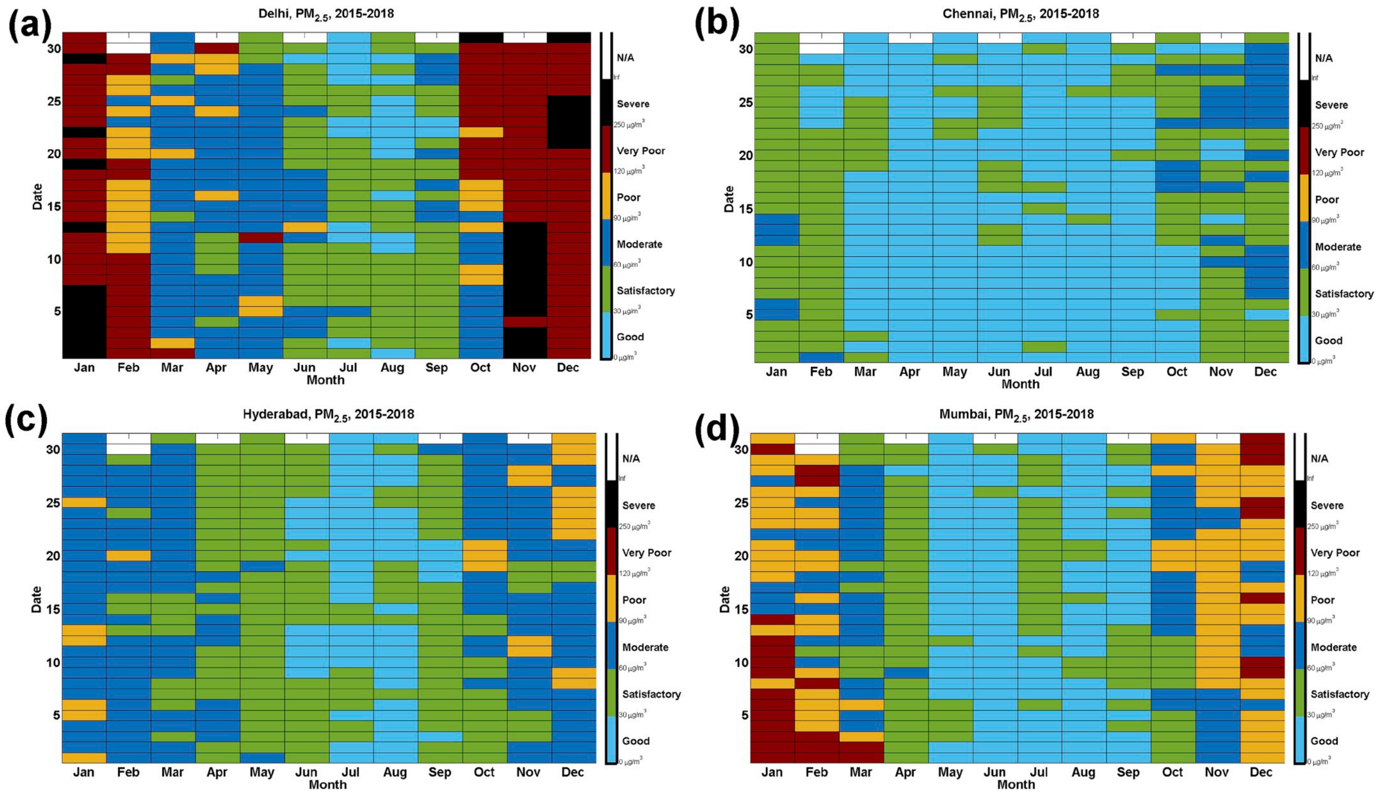

Fig. 2. Calendar-view of daily $PM_{2.5}$ air quality levels averaged over 2015–2018. (**a**) Delhi, (**b**) Chennai, (**c**) Hyderabad, and (**d**) Mumbai. The air quality levels are categorized following the Indian national air quality index definitions (https://app.cpcbccr.com/AQI_India).

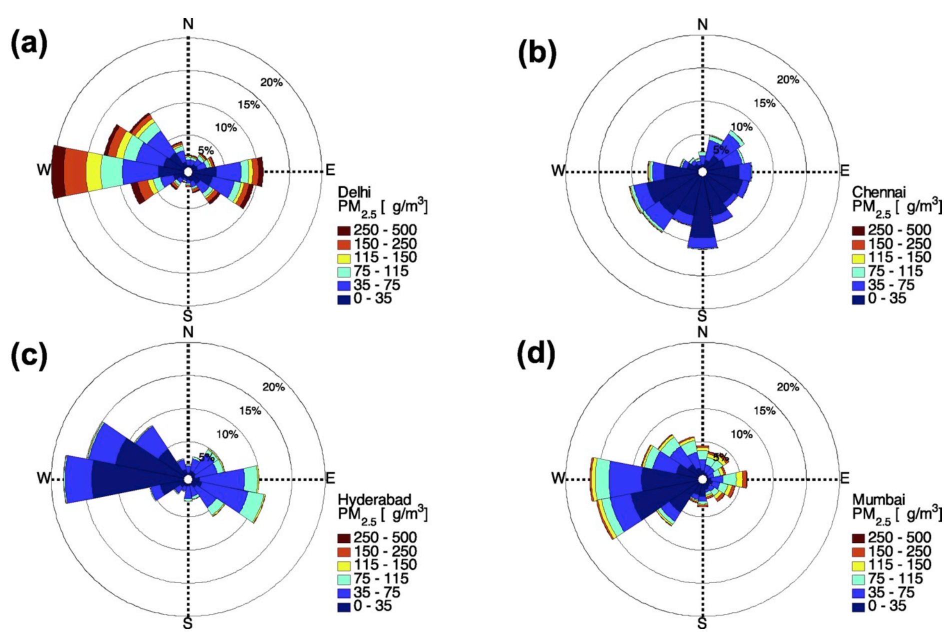

Fig. 3. Frequency distributions of $PM_{2.5}$ concentration as a function of wind direction. (**a**) Delhi, (**b**) Chennai, (**c**) Hyderabad, and (**d**) Mumbai.

3.2. Seasonal and diurnal patterns of $PM_{2.5}$

A distinct seasonal variation in the diurnal patterns is found, and this has different characteristics in each city (Fig. 4). Generally, the climate in India is characterised by four seasons: pre-monsoon/summer (March–May), monsoon (June–August), post-monsoon (September–November) and winter (December–February). Notable inter-seasonal changes in meteorology lead to significant differences in $PM_{2.5}$ loading. Benefitting from the cleansing effect of precipitation in the monsoon season (Ghosh et al., 2015), the hourly $PM_{2.5}$ is generally less than 50 $\mu g/m^3$ in the inland cities (Delhi and Hyderabad) and less than 30 $\mu g/m^3$ in the coastal cities (Chennai and Mumbai). Apart from cleansing by precipitation, frequent deep convection during summer monsoon in India can lift air pollutants near the surface to free troposphere or even upper troposphere, as reported by previous modelling and observational studies (Fadnavis et al., 2011; Kumar et al., 2015;

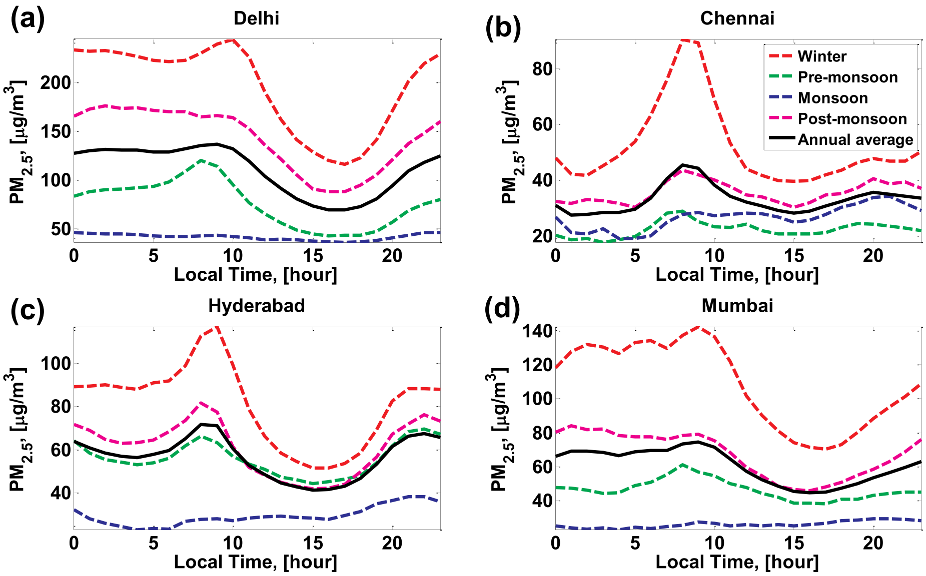

Fig. 4. Average diurnal variation of $PM_{2.5}$ concentrations for each season. (**a**) Delhi, (**b**) Chennai, (**c**) Hyderabad, and (**d**) Mumbai. The statistical values for each city in each season, including average, median, 75% percentile, 25% percentile, 95% percentile and 5% percentile, are given in Figs. S4–S8.

Lelieveld et al., 2018). This transport process dilutes air pollutants near the surface and could be one of the reasons that surface $PM_{2.5}$ concentration is the lowest during the monsoon season. Future works, with aircraft observations and modelling, are needed to quantify the relative importance of wash out and vertical transport in reducing concentrations of surface pollutants. Chennai benefits from prevailing onshore winds, with low $PM_{2.5}$ loadings in both the pre-monsoon and monsoon seasons (<30 μg/m^3). As a result of unfavourable meteorological conditions for dispersion and an increase in emissions from heating (Guttikunda and Calori, 2013; Guttikunda and Gurjar, 2012), winter is the most polluted season in all cities. The slow wind speeds and shallow PBL (Fig. S2) can trap $PM_{2.5}$ in the surface layer and increase its concentration (Hu et al., 2019; Zheng et al., 2015). The post-monsoon is the second most polluted season, with $PM_{2.5}$ higher than the annual averages. This inter-seasonal variation is consistent with the observations during 2013–2016 (Sreekanth et al., 2018) despite the rapid increase of anthropogenic emissions in India over the past decade (Li et al., 2017), indicating the importance of meteorology on the seasonal variation.

A clear diurnal pattern is found in all cities during winter, post-monsoon and pre-monsoon seasons (Fig. 4). However, no clear diurnal pattern is found during the monsoon season due to the influence of precipitation. The minimum $PM_{2.5}$ concentration during a day is generally found at 3–4 p.m. local time, possibly resulting from the dilution effect of the fully developed PBL in the afternoon (Fig. S3). $PM_{2.5}$ concentrations peak during the morning rush hour in all cities, the peaks approach 280 μg/m^3 (Delhi), 90 μg/m^3 (Chennai), 115 μg/m^3 (Hyderabad) and 140 μg/m^3 (Mumbai) in winter, respectively. It is interesting that the morning rush hour consistently shifts 1–2 h later from around 8 a.m. (pre-monsoon) to 10 a.m. (winter) in Delhi and Mumbai, and to 9 a.m. (winter) in Chennai and Hyderabad. A remarkably strong $PM_{2.5}$ peak is found during morning rush hour in Chennai and Hyderabad, with hourly $PM_{2.5}$ increased by ~50% and ~30% in 2 h, respectively. However, only a slight increase in $PM_{2.5}$ concentration is observed in Delhi and Mumbai, with an increase of ~10% in winter. These characteristics of $PM_{2.5}$ variation during morning rush hour may be related to the size of the population of each city. According to the latest census of India, there are around 4.6 and 7.0 million citizens in Chennai and Hyderabad, respectively; but more than 10 million citizens in Delhi and Mumbai (India Office of the Registrar General and Census Commissioner, 2011). Our results suggest that there is much greater human activity and emissions during the night in these two larger megacities leading to higher night-time $PM_{2.5}$ concentration but less variation during the morning. The morning rush hour lasts longer until 10 a.m. in winter in these megacities, in contrast to 9 a.m. in Chennai and Hyderabad. This is possibly because the busy traffic, also alarger city size would prevent a smooth commute and lead to longer commuting times (Alam and Ahmed, 2013; Srinivas, 2018). In addition, traffic is a major local source of $PM_{2.5}$ (~45%) in Delhi (Sahu et al., 2011). These results suggest that developing a more convenient and efficient public transport system and encouraging the usage could be a key to mitigate $PM_{2.5}$ pollution, especially in the biggest cities. More work on source apportionment is needed for each city to inform better targeted mitigation strategies.

3.3. Weekend effect in four cities

We report the influence of a weekend effect on the diurnal patterns of $PM_{2.5}$ in these cities, as shown in Fig. 5. No noticeable weekend effect is found in Delhi and Hyderabad. This is similar to Beijing and Chengdu in China (San Martini et al., 2015), with the diurnal patterns of $PM_{2.5}$ similar during weekdays and at the weekend. However, a notable weekend effect can be found in Chennai and Mumbai. The difference in the diurnal pattern of $PM_{2.5}$ between weekday and weekend is greatest before 10 a.m. A stronger morning rush hour is found in Chennai and Mumbai on weekdays, with ~10% higher $PM_{2.5}$ than at the weekend. This indicates that the decrease of traffic emissions in Mumbai and Chennai during weekend is probably the reason of weekend effect, and control of traffic emissions could be an efficient measure for improving air quality. In Chennai, $PM_{2.5}$ concentrations are about 5 μg/m^3 higher during night (12-5 a.m.) at the weekend than on weekdays; in contrast, $PM_{2.5}$ concentration is about 5 μg/m^3 lower at the weekend in Mumbai. These different weekend effects possibly indicate different life styles and $PM_{2.5}$ sources in each city. Further modelling and emission flux studies are needed to better understand the sources of $PM_{2.5}$ in each city.

3.4. Exposure to $PM_{2.5}$ and health impacts

We use these long-term in-situ observations to estimate the exposure of the population to $PM_{2.5}$ in Delhi, Chennai, Hyderabad and Mumbai. The annual averaged $PM_{2.5}$ loading in these cities is 110 μg/m^3, 33 μg/m^3, 56 μg/m^3 and 60 μg/m^3, respectively. The population-weighted annual mean $PM_{2.5}$ loading is 72 μg/m^3 across the four cities, which is

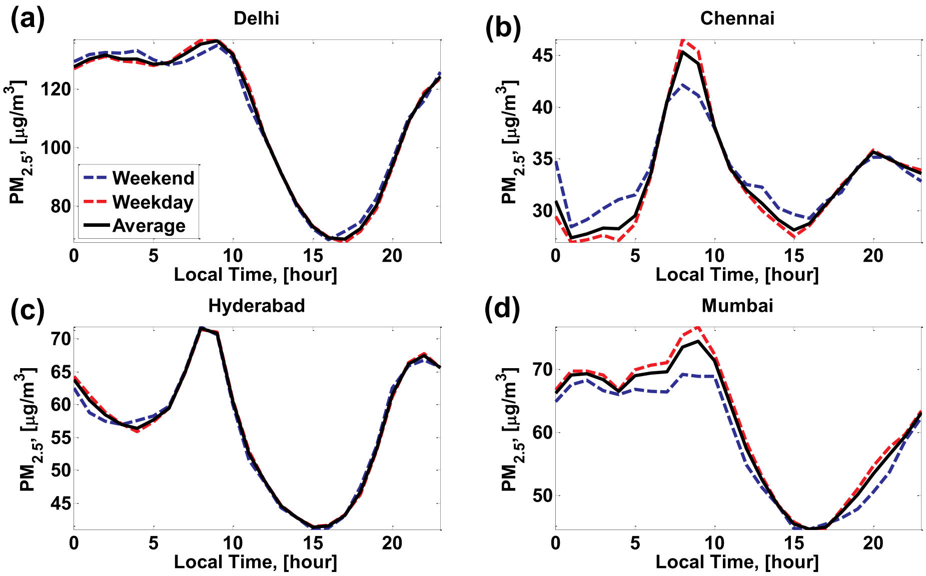

Fig. 5. Average diurnal variation of $PM_{2.5}$ concentrations on weekdays and at the weekend. (**a**) Delhi, (**b**) Chennai, (**c**) Hyderabad, and (**d**) Mumbai.

about 3.5 times higher than the global population-weighted value (20 μg/m^3, van Donkelaar et al., 2010) and ~22% higher than average Chinese city-level value (Zhang and Cao, 2015). The annual averaged $PM_{2.5}$ loading in Delhi is much higher than all Chinese major cities in the last five years (Wang et al., 2019). Fig. 6 shows the time integrated exposure, which indicates the proportion of time that a citizen is exposed to $PM_{2.5}$ concentrations over a given level over the four years measurement period. Citizens are exposed to unhealthy air quality ($PM_{2.5} > 60$ μg/m^3) for about 70% (Delhi), 15% (Chennai), 50% (Hyderabad) and 45% (Mumbai) of the time. The air quality is especially unhealthy in Delhi where citizens are exposed to 'severe' $PM_{2.5}$ pollution (>250 μg/m^3) for about 10% of the time. It is noteworthy that citizens of all four cities are exposed to air quality exceeding the 10 μg/m^3 WHO guideline nearly 100% of the time. $PM_{2.5}$ in all the cities except Chennai severely exceeds the revised Indian NAAQS standards of an annual average of 40 μg/m^3.

These continuous in-situ measurements give us an opportunity to make a robust assessment of long-term health impacts on a city scale in India (Fig. 7). We estimate that long-term ambient $PM_{2.5}$ exposure causes 10,200 (95% confidence interval: 6800–14,300), 2800 (1500–4100), 5200 (3100–7400), and 9500 (5800–13,600) premature mortality each year in Delhi, Chennai, Hyderabad, and Mumbai, respectively. Our premature mortality estimate for Delhi is reasonably

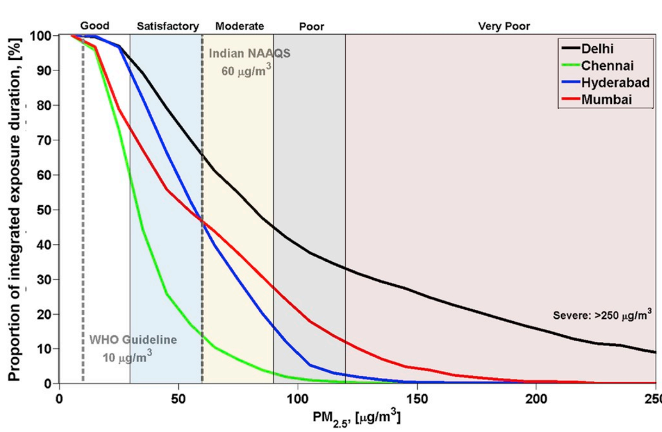

Fig. 6. Proportion of integrated exposure duration to $PM_{2.5}$ pollution at different levels in four cities.

agreed (~10% negative bias) with a previous estimate from the GBD2016 (GBD, 2016). We estimate that about 248,000 (168,000–340,700), 66,000 (37,400–96,800), 125,000 (78,300–176,100), and 230,000 (145,200–321,700) years of life are lost each year in Delhi, Chennai, Hyderabad, and Mumbai, respectively. The annual mortality rate per 100,000 population, which is independent of population size, is 93 (62–130), 60 (33–89), 74 (45–106), 76 (46–108) in Delhi, Chennai, Hyderabad, and Mumbai, respectively. Cardiovascular disease dominates the disease burden, with ischaemic heart disease (IHD) contributing ~40% and cerebrovascular disease (CEV) contributing ~30% in each city.

We estimate the health risks of short-term exposure during the New Year and Diwali Fest in Delhi and provide quantitative evidence to support control of fireworks. The fireworks during New Year enhance the $PM_{2.5}$ pollution in Delhi to some extent. The averaged $PM_{2.5}$ concentration during 1–3 January (276 μg/m^3) was about 20% higher than the monthly average of January (227 μg/m^3). This makes the daily premature mortality in Delhi slightly increase from January average of 43 (24–59) person per day to 50 (28–67) person per day during the New Year. The fireworks during Diwali Fest contribute substantially to the extremely high hourly concentration of $PM_{2.5}$ in Delhi (up to 1676 μg/m^3), leading to hazardous short-term exposure. Crop burning in Punjab and Haryana makes a large contribution to $PM_{2.5}$ loading in Delhi during October–November (Cusworth et al., 2018; Jethva et al., 2018), while fireworks in Diwali Fest can greatly worsen $PM_{2.5}$ pollution over the period of a few days (Singh et al., 2010). We find that the $PM_{2.5}$ concentration during Diwali Fest (including the festival start day and the following two days) is 75% higher (~320 μg/m^3) than the two-month average (~183 μg/m^3 in October–November) in Delhi over this four-year period. We estimate the short-term health impacts from ambient $PM_{2.5}$ concentrations during Diwali Fest at 56 (32–75) premature mortality per day in Delhi. This is an additional 20 (13–25) daily premature mortality, an increase of 56% compared with the October–November average of 36 (19–50) daily premature mortality. This highlights the importance of reducing firework emissions during Diwali Fest to improve public health.

3.5. Spatial representativeness and uncertainty

In order to analyse the spatial representativeness of observations in U.S. diplomatic missions in each city and the corresponding uncertainty, we extract surface $PM_{2.5}$ concentrations from a global high spatial

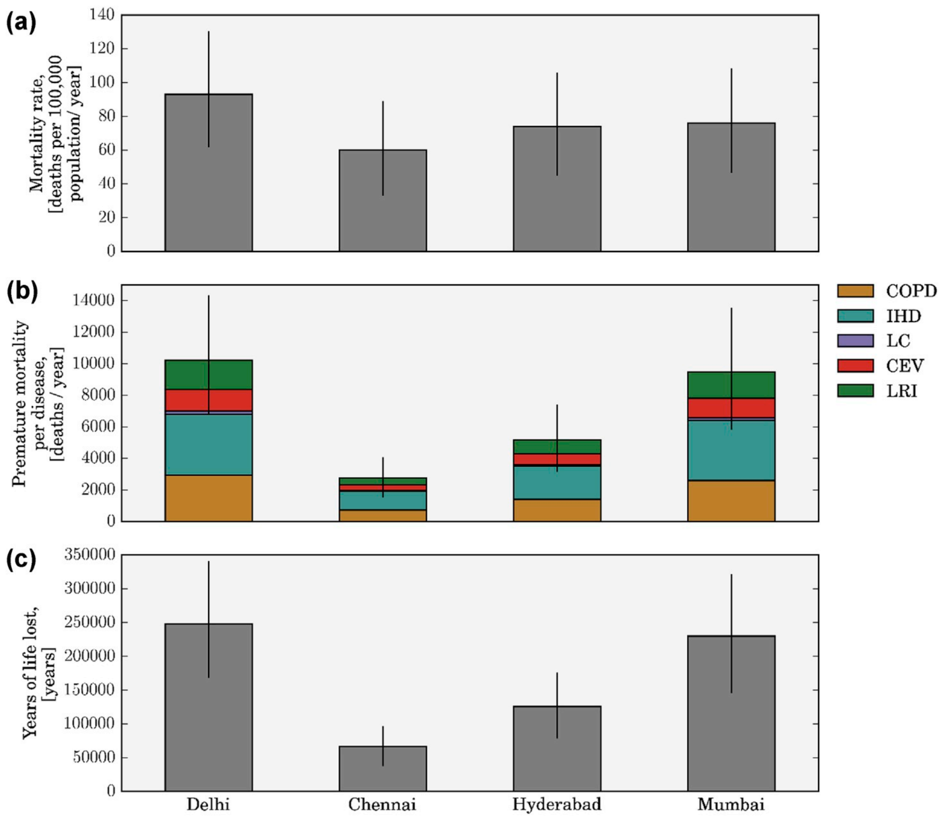

Fig. 7. Annual city-specific disease burden from long-term ambient $PM_{2.5}$ exposure. (**a**) Mortality rate per 100,000 population. (**b**) Premature mortality per disease of chronic obstructive pulmonary disease (COPD), lower respiratory infection (LRI), ischaemic heart disease (IHD), cerebrovascular disease (CEV), and lung cancer (LC). (**c**) Years of life lost.

resolution satellite-retrieved dataset (van Donkelaar et al., 2015, http://fizz.phys.dal.ca/~atmos/martin/?page_id=140). The extracted dataset includes the annual averaged (2015–2016) $PM_{2.5}$ concentration at locations of U.S. diplomatic missions and their surrounding regions within a distance of 20–100 km. This satellite-retrieved dataset is of high horizontal-resolution of 0.01 deg. × 0.01 deg. (lat-lon, about 1 km × 1 km). The retrieved data has been validated and widely adopted for global health effect analysis in previous studies (van Donkelaar et al., 2010, 2015). The standard deviation and ratios of $PM_{2.5}$ concentrations between U.S. diplomatic missions' locations and averages of surrounding regions are given in Fig. 8.

As shown in Fig. 8, the uncertainty in Chennai and Hyderabad is negligible, with difference between U.S. diplomatic missions and surrounding regions less than 5%, and the standard deviations increase

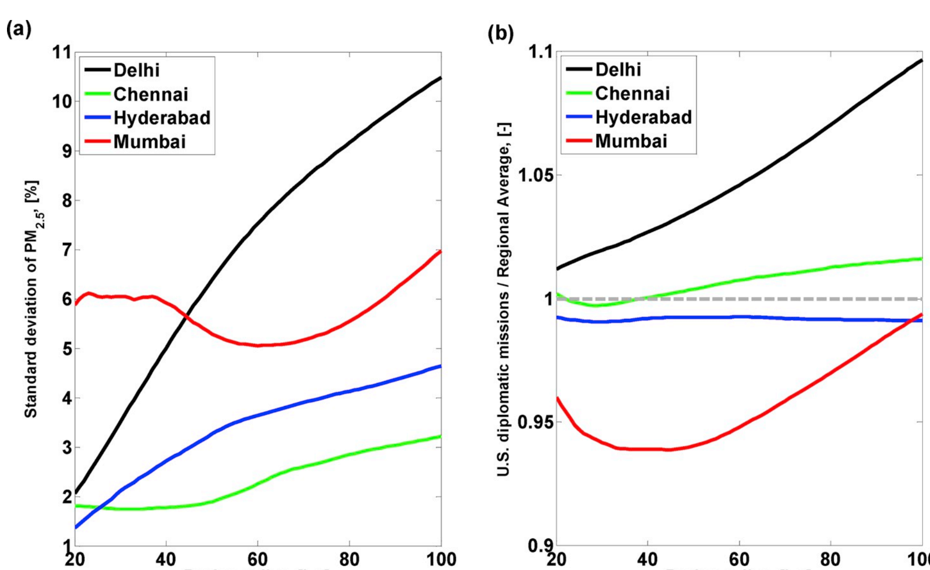

Fig. 8. Spatial representativeness of U.S. diplomatic mission observations in each city. (a) Standard deviation of PM2.5 mass concentrations in surrounding region as a function of region radius. (b) The ratio between U.S. diplomatic mission observation and regional average as a function of region radius.

slowly with the increase of distance from U.S. diplomatic missions but always less than 5%. This indicates a relatively homogeneous spatial distribution of $PM_{2.5}$ concentrations in Chennai and Hyderabad. In Mumbai, the standard deviation varies between 5 and 7%, with the minimum at a distance of ~60 km. This may be due to the influence of nearby large cities, such as Pune which is about 100 km away from Mumbai. The difference between U.S. diplomatic mission in Mumbai and the surrounding regional average is less than 6% in general, with the maximum underestimation of ~6% when the distance is about 40 km. This indicates that the observations of U.S. diplomatic mission in Mumbai well represent the nearby region, at least the region within 100 km. The representativeness of observations in the U.S. Embassy of Delhi decreases as the distance increases. The U.S. Embassy's observations may overestimate the $PM_{2.5}$ concentrations in Delhi compared with the regional average, but this overestimation is less than 5% and with standard deviations less than 6% when the distance (or region radius) is less than 60 km. However, the overestimation increases to ~10% with a standard deviation of ~10% when the distance is 100 km. This indicates a good representativeness of U.S. Embassy's observation for Delhi and its surrounding region within 60 km, but may overestimate the $PM_{2.5}$ concentration and the corresponding human exposure by ~10% if using U.S. Embassy's observations to estimate the $PM_{2.5}$ human exposure in a larger region of Delhi, such as with a radius of 100 km. This could be due to the higher urbanization level of Delhi, leading to a higher pollution level in/near the city center.

4. Conclusions and discussion

This study has estimated the health risks of long-term exposure to $PM_{2.5}$ based on in-situ observations in four Indian megacities (Delhi, Hyderabad, Chennai and Mumbai) during 2015–2018, and quantified the health risks of short-term exposure during Diwali Fest in Delhi for the first time. We also summarized the local characteristics of seasonal and diurnal variations of $PM_{2.5}$, and report the influence of a weekend effect on diurnal patterns. The results from this study are valuable for modelling studies and helpful in tailoring city-specific mitigation strategies.

Generally, substantial inter-seasonal variations in $PM_{2.5}$ are observed in the four cities, with the highest concentration during winter and the lowest during the monsoon season, when intensive wet scavenging lowers pollutant concentrations (Naja et al., 2014; Ojha et al., 2012). Winter is the most polluted season as a consequence of the shallow PBL and increased emissions from heating (Guttikunda and Calori, 2013; Guttikunda and Gurjar, 2012). Solid fuel burning is a common form of household heating in winter over India (Dumka et al., 2019; Jagadish and Dwivedi, 2018). To increase the efficiency of energy use and reduce $PM_{2.5}$ emissions in cities, we would suggest reduction in use of solid fuels (e.g., replace wood and coal with liquid petroleum gas or compressed natural gas) and implementation of central/electric heating systems with heating centres located in non-upwind regions (e.g., north or south of Delhi). The megacities of Delhi and Mumbai show a weak morning rush hour effect, but there is a strong one in Hyderabad and Chennai. For the first time, we report an interesting and consistent shift of about 2 h in the timing of the morning rush hour from pre-monsoon/summer (8 a.m.) to winter (9–10 a.m.), and analyse the influence of a weekend effect on the diurnal patterns of $PM_{2.5}$ in Indian megacities. The coastal cities of Chennai and Mumbai show a clear difference in morning $PM_{2.5}$ concentrations between weekdays and the weekend, but no noticeable difference was observed in the inland cities of Delhi and Hyderabad. These results indicate traffic emissions could be important sources of $PM_{2.5}$ and highlight the distinct local characteristics of human activity in each city, which is critical information for modelling studies. The four cities show significant differences in wind patterns and transport of $PM_{2.5}$, suggesting that different control strategies are needed for each city that take into account its local emission characteristics and meteorological conditions.

In this study, we report the high health risks of exposure to $PM_{2.5}$pollution in Indian cities and highlight hazardous short-term exposure during Diwali Fest in Delhi. Across the four cities, long-term exposure to $PM_{2.5}$ causes about 28,000 (95% confidence interval: 17,200–39,400) premature mortality and 670,000 (428,900–935,200) years of life lost each year. Fireworks during the Diwali Fest lead to severe air pollution in Delhi, and this is responsible for 56 (32–75) premature mortality per day, a 56% increase over the monthly average. More effective control policies are urgently required to mitigate the health burden and achieve sustainable development. Previous studies have shown that the dominant emission sources contributing to the disease burden from ambient $PM_{2.5}$ exposure are land transport in Delhi, residential solid fuel burning in Chennai and Hyderabad, and industrial coal burning in Mumbai (Conibear et al., 2018a). The disease burden is likely to increase substantially in future due to population ageing and growth, which enhance the susceptibility to disease, unless stringent emission control policies are implemented (Conibear et al., 2018b).

We have estimated the $PM_{2.5}$ exposure in the four cities with continuous observations, but it is noteworthy that some other Indian cities experience more severe air pollution (WHO, 2016). Continuous, widespread pollutant measurements across India would provide more complete information on regional pollutant characteristics and overall pollutant levels. More detailed measurements of the physicochemical properties of $PM_{2.5}$ in major cities, e.g., their composition and size distribution, would permit better characterisation of urban sources, and provide the information needed to design appropriate mitigation strategies.

Author contributions

Y. C. and O. W. conceived the study. Y. C. performed the analysis and interpreted the results with input from all co-authors. L. C. helped with the health effect assessment. The manuscript was written with input from all co-authors.

Additional information

The authors declare no competing financial interest.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Hourly measurements of $PM_{2.5}$ made at U.S. diplomatic missions in India are available through the AirNow platform maintained by the U.S. Department of State and the U.S. Environmental Protection Agency at https://www.airnow.gov/. Meteorological variables are available through the Integrated Surface Database—Surface Data Hourly Global data product maintained by the U.S. National Oceanic and Atmospheric Administration—National Climatic Data Center at https://www.ncdc.noaa.gov/. Y. W. would like to thank the China Scholarship Council for support through a PhD scholarship. Y. C. and O. W. would like to thank the NERC for funding (NE/P01531X/1 and NE/N006976/1). R. L. would like to thank the National Natural Science Foundation of China (grant no. 41305114). L. C. would like to thank the N8 consortium and EPSRC (grant EP/K000225/1). The paper is based on interpretation of scientific results and in no way reflect the viewpoint of the funding agencies.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.

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