Local Characteristics of and Exposure to Fine Particulate Matter (PM$_{2.5}$) in Four Indian Megacities

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Highlights:

- PM$_{2.5}$ increased by 75% during Diwali in Delhi, causing 20 extra daily mortality

- A weekend effect is found in Mumbai and Chennai but not in Delhi and Hyderabad

- Distinct differences in diurnal pattern of PM$_{2.5}$ in different seasons and cities

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 hours 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

46 air quality modelling studies and are critical for tailoring the design of effective mitigation

47 strategies for each city.

48 **Keywords:** PM$_{2.5}$; Health effect; Diwali festival effect; Weekend effect; Long-term; Short-

49 term

50

51 **1. Introduction**

52   Exposure to fine particulate matter (particles with an aerodynamic diameter less than 2.5

53 µm, PM$_{2.5}$) can pose a major threat to human health (Chowdhury and Dey, 2016; Gao et al.,

54 2018a; Gao et al., 2017; Huang et al., 2018; Pope et al., 2009; Wang et al., 2017). As a rapidly

55 developing country with an expanding population, India is suffering severe PM$_{2.5}$ pollution,

56 with nine cities among the top ten most polluted cities in the world as reported by the World

57 Health Organization (WHO, 2016). Exposure to high levels of PM$_{2.5}$ causes ~1 million

58 premature mortality per year across India (Conibear et al., 2018a). In order to tackle this PM$_{2.5}$

59 pollution, the Central Pollution Control Board (CPCB) of India set revised National Ambient

60 Air Quality Standards (NAAQS) in 2009 that included PM$_{2.5}$ regulations (CPCB, 2009). Some

61 mitigation policies have been implemented in major Indian cities (Chowdhury et al., 2017;

62 Sharma and Dixit, 2016), but limited improvement in air quality (~10% reduction in PM$_{2.5}$) has

63 been seen (Chowdhury et al., 2017). PM$_{2.5}$ pollution is expected to further deteriorate in the

64 coming decades (Chowdhury et al., 2018; Conibear et al., 2018b), due to rapid ongoing

65 urbanization. This surface pollution over India also has important global implications through

66 effective transport by the Asian summer monsoon to the upper troposphere and lower

67 stratosphere, where pollutants can be re-distributed on a global scale and thus affect global

68 climate forcing and air quality (Lelieveld et al., 2018; Liu et al., 2015; Yu et al., 2017).

69   Previous studies estimated health risks in India of exposure to $PM_{2.5}$ based on model

70   analysis or satellite retrieves and mainly focused on long-term exposure (e.g., Chowdhury and

71   Dey, 2016; Conibear et al., 2018a, b; Gao et al., 2018b; Lelieveld et al., 2015; van Donkelaar

72   et al., 2015). In addition, intensive emissions and unfavourable meteorological condition for

73   dispersion can significantly increase $PM_{2.5}$ and lead to hazardous short-term exposure with high

74   health risks (Atkinson et al., 2014; Héroux et al., 2015). In-situ observations at high temporal

75   resolution are valuable for more firmly grounded estimates of health risks. Furthermore,

76   characterizing the seasonal and diurnal variations of urban $PM_{2.5}$ concentrations and their

77   relationships to meteorology is the key to understanding the drivers of air pollution and

78   devising effective mitigation strategies in Indian megacities (Schnell et al., 2018). Long-term

79   in-situ monitoring studies are critical for a better understanding of these factors. However, only

80   a few studies providing long-term observations of $PM_{2.5}$ have been undertaken, and most of

81   these have focused on Delhi only (Sahu and Kota, 2017; Sharma et al., 2018). Information on

82   local characteristics such as the diurnal variation in pollutant emissions is also critical for

83   modelling studies. This information is scarce in India and models typically use a constant

84   diurnal profile of emissions (e.g., Mohan and Gupta, 2018) or standard profiles from American

85   or European cities to represent conditions in India (e.g., Marrapu et al., 2014). Long-term

86   observations of the diurnal variation of pollutants would provide essential information for

87   improving model performance.

88   This study presents a comprehensive summary of the seasonal and diurnal variation of

89   urban $PM_{2.5}$ in four Indian megacities (Delhi, Chennai, Hyderabad and Mumbai), based on

90   ground observations from 2015 to 2018. This analysis reveals the observation-based patterns

91   of human activity and local temporal characteristics of emissions in each city, and hence

92   provides valuable input for modelling studies. In addition, for the first time, we report the

93   influences of weekend effect on the diurnal variations and quantify the health risks of short-

94 term exposure during Diwali Fest. Finally, the cumulative exposure of urban residents to $PM_{2.5}$

95 and the corresponding health burdens are estimated for each city. The results of this study are

96 valuable for the designation and implementation of mitigation policies on a city level aimed at

97 improving air quality to meet the Indian NAAQS standards.

98

99 **2. Materials and Methods**

100 **2.1 Data**

101   Datasets of pollutants measured between 1 March 2015 and 31 December 2018 are

102 analysed in this study. An overview of the data is given in Table S1. Hourly $PM_{2.5}$ observations

103 in Delhi, Chennai, Mumbai and Hyderabad (Fig. S1) are routinely made at U.S. Embassy and

104 consulates using a beta attenuation monitor (San Martini et al., 2015). These records are

105 available from the AirNow website (https://www.airnow.gov/). The instruments are maintained

106 and calibrated following the regulations of the U.S. Environmental Protection Agency (EPA,

107 2009, 2015). $PM_{2.5}$ observations from the U.S. Embassy are widely used in previous studies in

108 India (Wang and Chen, 2019) and China (Lv et al., 2017; Lv et al., 2015; San Martini et al.,

109 2015), and have been shown to be of good quality and in good agreement with other

110 observations (Jiang et al., 2015; Mukherjee and Toohey, 2016).

111   We use hourly meteorological observations at the airport in each city (VIDP-Delhi,

112 VOMM-Chennai, VABB-Mumbar and VOHY-Hyderabad). The flat topography surrounding

113 these airports suggests that the observations are broadly representative of the dominant

114 meteorological conditions in these cities. Historical records are archived by the National

115 Oceanic and Atmospheric Administration, and are available from the National Climatic Data

116 Center (https://www.ncdc.noaa.gov/data-access/). The height of the planetary boundary

117 layer (PBL) is obtained from the European Centre for Medium-Range Weather Forecasts

118   (ECMWF) ERA-interim reanalysis at a 3-hour interval and $0.125° \times 0.125°$ spatial resolution

119   (https://www.ecmwf.int/).

120   **2.2 Method**

121   We estimate the long-term health impacts from exposure to ambient $PM_{2.5}$ concentrations,

122   as these account for the majority of the health effects through capturing both acute and chronic

123   responses. Following our previous works (Conibear et al., 2018a, b), we use integrated

124   exposure-response (IER) functions (Burnett Richard et al., 2014), updated for the Global

125   Burden of Disease GBD2016 (GBD, 2016) to estimate the relative risk (RR) of premature

126   mortality due to exposure to $PM_{2.5}$ concentrations. There are IER functions with age-specific

127   modifiers for chronic obstructive pulmonary disease (COPD), lower respiratory infection (LRI),

128   ischaemic heart disease (IHD), cerebrovascular disease (CEV), and lung cancer (LC). We use

129   the parameter distributions from the GBD2016 for 1,000 simulations to derive the mean IER

130   with 95% uncertainty intervals. The IER functions have uniform theoretical minimum risk

131   exposure levels for $PM_{2.5}$ between 2.4–5.9 $\mu g/m^3$.

132   We use multi-year average annual-mean $PM_{2.5}$ concentrations from measurements made

133   at U.S. diplomatic missions in Delhi (110 $\mu g/m^3$), Chennai (33 $\mu g/m^3$), Hyderabad (56 $\mu g/m^3$),

134   and Mumbai (60 $\mu g/m^3$). Baseline mortality data are taken from the GBD2016 for India (GBD,

135   2018). Population size was taken from the lastest Indian Census data for 2011. Population age

136   composition was taken from the GBD2016 population estimates for 2015 for India (GBD,

137   2017a).

138   Annual premature mortality (M) for each age and disease were estimated as a function of

139   population (P), baseline mortality rates (I), and the attributable fraction (AF) for a specific

140   relative risk (RR) (Equation 1). The disease burden from LRI, IHD, CEV, COPD, and LC was

141   estimated between 0 and 95 years upwards in 5 year groupings.

142

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

143

144   Annual years of life lost (YLL) for each age and disease were estimated as a function of

145   premature mortality and age-specific life expectancy (LE) from the standard reference life table

146   from the GBD2016 (Equation 2) (GBD, 2017b).

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

148

149   We estimate the short-term health impacts during Diwali Fest in Delhi from exposure to

150   ambient $PM_{2.5}$ concentrations as all-cause premature mortality. The short-term health impacts

151   are accounted for within the long-term health impacts, and are used to indicate the variation in

152   the daily burden from acute responses (Héroux et al., 2015). We use the summary risk estimates

153   (γ) from Atkinson et al. (2014) of 1.04% (0.52–1.56) per 10 µg/m^3 change in daily mean $PM_{2.5}$

154   concentrations (C_d), with respect to a reference $PM_{2.5}$ concentration (C_r) of 0 µg/m^3. We assume

155   no upper concentration cutoff. India-specific risk functions for ambient $PM_{2.5}$ exposure do not

156   currently exist, however, the use of the summary risk estimate of 1.04% is conservative when

157   compared with the summary risk estimate of 1.2% from Levy et al. (2012) and 1.23% from

158   WHO (2013). Baseline mortality data are taken from the GBD2016 for India for all ages for

159   both genders combined (GBD, 2018). We convert these annual rates to daily rates (I_d) by

160   dividing by 365.25, consistent with previous work due to the lack of daily data (West et al.,

161   2007). We use first-three-day of Diwali Fest (320 µg/m^3) and October-November two-month

162   (183 µg/m^3) averaged daily-mean $PM_{2.5}$ concentrations during 2015-2018 from the U.S.

163   Embassy measurements for Delhi.

164                    $$RR_d = 1 + [\gamma \times (C_d - C_r) \times 0.1] \tag{3}$$

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

166   We use a linear exposure-response function with no cap on daily relative risk (RR_d),

167   similar to a previous work (van Donkelaar et al., 2011), estimating daily relative risks following

168   Equation 3. Daily premature mortality (M_d) is then estimated using Equation 4.

169   Using a logarithmic exposure-response function as in previous work (Crippa et al., 2016),

170   our estimates of short-term premature mortality are about 10% larger than with a linear

171   exposure-response function. To be conservative, we use the linear exposure-response function

172   in this study.

173

3. Results

3.1 Overview of PM$_{2.5}$ in Four Megacities

176   The locations of Delhi, Chennai, Hyderabad and Mumbai are shown in Fig. 1, together

177   with annual mean surface concentrations of PM$_{2.5}$ of anthropogenic origin over India in 2015

178   (van Donkelaar et al., 2015; van Donkelaar et al., 2011). Fig. 2 shows a calendar-view of daily

179   average PM$_{2.5}$ concentrations in the four cities during 2015-2018, and monthly statistics are

180   shown in Fig. S1. There is no clear inter-annual trend in PM$_{2.5}$ observed in these cities during

181   2015-2018. The Indian NAAQS classifies six different levels of air quality based on daily 24-

182   hour averaged PM$_{2.5}$ concentrations (Fig. 2). The two cleanest air quality levels, 'good' and

183   'satisfactory', are defined as healthy, and the others (PM$_{2.5}$ > 60 $\mu g/m^3$) are defined as

184   unhealthy (CPCB, 2014). Delhi suffers the worst air quality among these cities, and the air

185   quality levels are categorized as 'poor', 'very poor' or 'severe' for ~50% of the time. These

186   hazy days mostly occur during October-February. The air quality in Chennai and Hyderabad

187   is much better than Delhi, with few 'poor' air-quality days; and 'healthy' days counted up to

188 50% of the time in Hyderabad and most of the time in Chennai. Mumbai has better air quality

189 than Delhi. This may be due to its coastal climate, where surface $PM_{2.5}$ is often diluted by clean

190 air from the ocean. However, Mumbai still experiences about four months per year with air

191 quality of 'poor' standard or worse. The Diwali Fest and New Year festivals make the air

192 quality substantially worse in Delhi, as shown by the 'severe' days at the beginning of

193 November and January (Fig. 2a). This suggests that the fireworks during the festivals contribute

194 to an increase of $PM_{2.5}$ loading in Delhi significantly. However, there is no clear festival effect

195 observed in the other three cities. It is unclear why no festival effect is observed in these other

196 cities, although it may reflect lower firework use and more favourable meteorological

197 conditions for dispersion in coastal cities.

198   All cities suffered severe episodes of poor air quality, with maximum hourly $PM_{2.5}$

199 concentrations of 1676 µg/m^3, 1334 µg/m^3, 1107 µg/m^3 and 758 µg/m^3 in Delhi, Chennai,

200 Hyderabad and Mumbai, respectively. In Delhi, the maximum hourly $PM_{2.5}$, observed during

201 the Diwali Fest nights in 2016 and 2018, is ~70% higher than the highest level recorded in

202 Beijing (980 µg/m^3), China (San Martini et al., 2015; Wang et al., 2014; Zheng et al., 2015).

203 This strongly suggests that control of fireworks during the Diwali Fest would efficiently

204 mitigate short-term $PM_{2.5}$ exposure in Delhi. This is also implied by a previous study (Singh et

205 al., 2010), where a significant increase in particle loading by a factor of 2-6 compared with the

206 period before and after Diwali Fest was found in Delhi during 2002-2007. Extreme episodes in

207 other cities were observed at night-time (10 p.m.-2 a.m.) from the end of October to the

208 beginning of December. The shallow planetary boundary layer (PBL) at night and intensive

209 crop burning in this season are the likely reasons for these extremely high concentrations

210 (Tiwari et al., 2013). Fig. S2 shows that there is a clear decrease in the frequency of high $PM_{2.5}$

211 concentrations in all cities as the PBL height increases. We also observe an anti-correlation

212 between wind speed and $PM_{2.5}$ loading. With the same PBL height, $PM_{2.5}$ loading generally

213 decreases as wind speed increases, and $PM_{2.5}$ is generally less than $100 \, \mu g/m^3$ when the wind

214 speed is greater than 4 m/s in all cities (Fig. S2). This is because the higher PBL and larger

215 wind speed dilute the surface $PM_{2.5}$ (Chen et al., 2009; Mohan and Gupta, 2018).

216   In order to investigate the possible source regions of $PM_{2.5}$ for each city, we analyse the

217 relationship between $PM_{2.5}$ concentration and wind direction (Fig. 3). Delhi is influenced by

218 easterly and westerly/northwesterly winds, with high $PM_{2.5}$ concentrations ($>150 \, \mu g/m^3$) from

219 both directions. The westerly and northwesterly winds have the highest frequency (~33%) and

220 are associated with the most polluted episodes in Delhi. About 30% of the time $PM_{2.5}$

221 concentration in Delhi are higher than $150 \, \mu g/m^3$, ~50% of which is associated with a westerly

222 or northwestly wind. This indicates that crop biomass burning and desert dust could be major

223 sources of $PM_{2.5}$ in Delhi. Punjab and Haryana are located to the northwest of Delhi, and are

224 major sources of particles and gaseous precursors from crop burning during October-November

225 (Cusworth et al., 2018; Jethva et al., 2018; Rastogi et al., 2014), when the worst air quality is

226 observed in Delhi. Furthermore, previous modelling studies show significant increases ($> 50\%$)

227 in aerosol loading when the westerly and northwesterly wind transports dust from the Thar

228 Desert to Delhi during April-June (Kumar et al., 2014a; Kumar et al., 2014b). In Hyderabad,

229 another inland city, the easterly/westerly wind pattern is also dominant. The easterly wind

230 brings a substantial amount of $PM_{2.5}$ to Hyderabad, but the conditions are better than in Delhi,

231 with limited episodes of $PM_{2.5}$ concentration higher than $150 \, \mu g/m^3$. Chennai and Mumbai are

232 coastal cities with a prevailing onshore wind for 70-80% of the time which brings relatively

233 clean marine air masses. The $PM_{2.5}$ concentrations are generally lower than $75 \, \mu g/m^3$ when an

234 onshore wind is present. The offshore wind brings pollutants from inland regions to the cities,

235 but this occurs much less frequently (20-30%). These results indicate that there is a strong

236 interaction between meteorology and $PM_{2.5}$ pollution, and strong local characteristics are found

237   in each city. Detailed investigation of these local characteristics would be helpful in tailoring

238   an effective mitigation policy for each city.

239   **3.2 Seasonal and Diurnal Patterns of PM$_{2.5}$**

240   A distinct seasonal variation in the diurnal patterns is found, and this has different

241   characteristics in each city (Fig. 4). Generally, the climate in India is characterised by four

242   seasons: pre-monsoon/summer (March-May), monsoon (June-August), post-monsoon

243   (September-November) and winter (December-February). Notable inter-seasonal changes in

244   meteorology lead to significant differences in PM$_{2.5}$ loading. Benefitting from the cleansing

245   effect of precipitation in the monsoon season (Ghosh et al., 2015), the hourly PM$_{2.5}$ is generally

246   less than 50 µg/m^3 in the inland cities (Delhi and Hyderabad) and less than 30 µg/m^3 in the

247   coastal cities (Chennai and Mumbai). Apart from cleansing by precipitation, frequent deep

248   convection during summer monsoon in India can lift air pollutants near the surface to free

249   troposphere or even upper troposphere, as reported by previous modelling and observational

250   studies (Fadnavis et al., 2011; Kumar et al., 2015; Lelieveld et al., 2018). This transport process

251   dilutes air pollutants near the surface and could be one of the reasons that surface PM$_{2.5}$

252   concentration is the lowest during the monsoon season. Future works, with aircraft

253   observations and modelling, are needed to quantify the relative importance of wash out and

254   vertical transport in reducing concentrations of surface pollutants. Chennai benefits from

255   prevailing onshore winds, with low PM$_{2.5}$ loadings in both the pre-monsoon and monsoon

256   seasons (< 30 µg/m^3). As a result of unfavourable meteorological conditions for dispersion and

257   an increase in emissions from heating (Guttikunda and Calori, 2013; Guttikunda and Gurjar,

258   2012), winter is the most polluted season in all cities. The slow wind speeds and shallow PBL

259   (Fig. S2) can trap PM$_{2.5}$ in the surface layer and increase its concentration (Hu et al., 2019;

260   Zheng et al., 2015). The post-monsoon is the second most polluted season, with PM$_{2.5}$ higher

261 than the annual averages. This inter-seasonal variation is consistent with the observations

262 during 2013-2016 (Sreekanth et al., 2018) despite the rapid increase of anthropogenic

263 emissions in India over the past decade (Li et al., 2017), indicating the importance of

264 meteorology on the seasonal variation.

265 A clear diurnal pattern is found in all cities during winter, post-monsoon and pre-monsoon

266 seasons (Fig. 4). However, no clear diurnal pattern is found during the monsoon season due to

267 the influence of precipitation. The minimum $PM_{2.5}$ concentration during a day is generally

268 found at 3-4 p.m. local time, possibly resulting from the dilution effect of the fully developed

269 PBL in the afternoon (Fig. S3). $PM_{2.5}$ concentrations peak during the morning rush hour in all

270 cities, the peaks approach 280 $\mu g/m^3$ (Delhi), 90 $\mu g/m^3$ (Chennai), 115 $\mu g/m^3$ (Hyderabad) and

271 140 $\mu g/m^3$ (Mumbai) in winter, respectively. It is interesting that the morning rush hour

272 consistently shifts 1-2 hours later from around 8 a.m. (pre-monsoon) to 10 a.m. (winter) in

273 Delhi and Mumbai, and to 9 a.m. (winter) in Chennai and Hyderabad. A remarkably strong

274 $PM_{2.5}$ peak is found during morning rush hour in Chennai and Hyderabad, with hourly $PM_{2.5}$

275 increased by ~50% and ~30% in two hours, respectively. However, only a slight increase in

276 $PM_{2.5}$ concentration is observed in Delhi and Mumbai, with an increase of ~10% in winter.

277 These characteristics of $PM_{2.5}$ variation during morning rush hour may be related to the size of

278 the population of each city. According to the latest census of India, there are around 4.6 and

279 7.0 million citizens in Chennai and Hyderabad, respectively; but more than 10 million citizens

280 in Delhi and Mumbai (India Office of the Registrar General and Census Commissioner, 2011).

281 Our results suggest that there is much greater human activity and emissions during the night in

282 these two larger megacities leading to higher night-time $PM_{2.5}$ concentration but less variation

283 during the morning. The morning rush hour lasts longer until 10 a.m. in winter in these

284 megacities, in contrast to 9 a.m. in Chennai and Hyderabad. This is possibly because the busy

285 traffic, also alarger city size would prevent a smooth commute and lead to longer commuting

286   times (Alam and Ahmed, 2013; Srinivas, 2018). In addition, traffic is a major local source of

287    $PM_{2.5}$ (~45%) in Delhi (Sahu et al., 2011). These results suggest that developing a more

288   convenient and efficient public transport system and encouraging the usage could be a key to

289   mitigate $PM_{2.5}$ pollution, especially in the biggest cities. More work on source apportionment

290   is needed for each city to inform better targeted mitigation strategies.

291

3.3 Weekend Effect in Four Cities

293   We report the influence of a weekend effect on the diurnal patterns of $PM_{2.5}$ in these cities,

294   as shown in Fig. 5. No noticeable weekend effect is found in Delhi and Hyderabad. This is

295   similar to Beijing and Chengdu in China (San Martini et al., 2015), with the diurnal patterns of

296    $PM_{2.5}$ similar during weekdays and at the weekend. However, a notable weekend effect can be

297   found in Chennai and Mumbai. The difference in the diurnal pattern of $PM_{2.5}$ between weekday

298   and weekend is greatest before 10 a.m. A stronger morning rush hour is found in Chennai and

299   Mumbai on weekdays, with ~10% higher $PM_{2.5}$ than at the weekend. This indicates that the

300   decrease of traffic emissions in Mumbai and Chennai during weekend is probably the reason

301   of weekend effect, and control of traffic emissions could be an efficient measure for improving

302   air quality. In Chennai, $PM_{2.5}$ concentrations are about 5 $\mu g/m^3$ higher during night (12-5 a.m.)

303   at the weekend than on weekdays; in contrast, $PM_{2.5}$ concentration is about 5 $\mu g/m^3$ lower at

304   the weekend in Mumbai. These different weekend effects possibly indicate different life styles

305   and $PM_{2.5}$ sources in each city. Further modelling and emission flux studies are needed to better

306   understand the sources of $PM_{2.5}$ in each city.

3.4 Exposure to $PM_{2.5}$ and Health Impacts

13

308   We use these long-term in-situ observations to estimate the exposure of the population to

309    $PM_{2.5}$ in Delhi, Chennai, Hyderabad and Mumbai. The annual averaged $PM_{2.5}$ loading in these

310   cities is 110 µg/m^3, 33 µg/m^3, 56 µg/m^3 and 60 µg/m^3, respectively. The population-weighted

311   annual mean $PM_{2.5}$ loading is 72 µg/m^3 across the four cities, which is about 3.5 times higher

312   than the global population-weighted value (20 µg/m^3, van Donkelaar et al., 2010) and ~22%

313   higher than average Chinese city-level value (Zhang and Cao, 2015). The annual averaged

314    $PM_{2.5}$ loading in Delhi is much higher than all Chinese major cities in the last five years (Wang

315   et al., 2019). Fig. 6 shows the time integrated exposure, which indicates the proportion of time

316   that a citizen is exposed to $PM_{2.5}$ concentrations over a given level over the four years

317   measurement period. Citizens are exposed to unhealthy air quality ($PM_{2.5} > 60$ µg/m^3) for about

318   70% (Delhi), 15% (Chennai), 50% (Hyderabad) and 45% (Mumbai) of the time. The air quality

319   is especially unhealthy in Delhi where citizens are exposed to 'severe' $PM_{2.5}$ pollution (>250

320   µg/m^3) for about 10% of the time. It is noteworthy that citizens of all four cities are exposed to

321   air quality exceeding the 10 µg/m^3 WHO guideline nearly 100% of the time. $PM_{2.5}$ in all the

322   cities except Chennai severely exceeds the revised Indian NAAQS standards of an annual

323   average of 40 µg/m^3.

324   These continuous in-situ measurements give us an opportunity to make a robust

325   assessment of long-term health impacts on a city scale in India (Fig. 7). We estimate that long-

326   term ambient $PM_{2.5}$ exposure causes 10,200 (95% confidence interval: 6,800–14,300), 2,800

327   (1,500–4,100), 5,200 (3,100–7,400), and 9,500 (5,800–13,600) premature mortality each year

328   in Delhi, Chennai, Hyderabad, and Mumbai, respectively. Our premature mortality estimate

329   for Delhi is reasonably agreed (~10% negative bias) with a previous estimate from the

330   GBD2016 (GBD, 2016). We estimate that about 248,000 (168,000–340,700), 66,000 (37,400–

331   96,800), 125,000 (78,300–176,100), and 230,000 (145,200–321,700) years of life are lost each

332   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

357   In order to analyse the spatial representativeness of observations in U.S. diplomatic

358   missions in each city and the corresponding uncertainty, we extract surface $PM_{2.5}$

359   concentrations from a global high spatial resolution satellite-retrieved dataset (van Donkelaar

360   et al., 2015, http://fizz.phys.dal.ca/~atmos/martin/?page_id=140). The extracted dataset

361   includes the annual averaged (2015-2016) $PM_{2.5}$ concentration at locations of U.S. diplomatic

362   missions and their surrounding regions within a distance of 20-100 km. This satellite-retrieved

363   dataset is of high horizontal-resolution of 0.01 deg. $\times$ 0.01 deg. (lat-lon, about 1km $\times$ 1km).

364   The retrieved data has been validated and widely adopted for global health effect analysis in

365   previous studies (van Donkelaar et al., 2010; van Donkelaar et al., 2015). The standard

366   deviation and ratios of $PM_{2.5}$ concentrations between U.S. diplomatic missions' locations and

367   averages of surrounding regions are given in Fig. 8.

368   As shown in Fig. 8, the uncertainty in Chennai and Hyderabad is negligible, with

369   difference between U.S. diplomatic missions and surrounding regions less than 5%, and the

370   standard deviations increase slowly with the increase of distance from U.S. diplomatic missions

371   but always less than 5%. This indicates a relatively homogeneous spatial distribution of $PM_{2.5}$

372   concentrations in Chennai and Hyderabad. In Mumbai, the standard deviation varies between

373   5-7%, with the minimum at a distance of ~60 km. This may be due to the influence of nearby

374   large cities, such as Pune which is about 100 km away from Mumbai. The difference between

375   U.S. diplomatic mission in Mumbai and the surrounding regional average is less than 6% in

376   general, with the maximum underestimation of ~6% when the distance is about 40 km. This

377   indicates that the observations of U.S. diplomatic mission in Mumbai well represent the nearby

378   region, at least the region within 100 km. The representativeness of observations in the U.S.

379   Embassy of Delhi decreases as the distance increases. The U.S. Embassy's observations may

380   overestimate the $PM_{2.5}$ concentrations in Delhi compared with the regional average, but this

381   overestimation is less than 5% and with standard deviations less than 6% when the distance (or

382 region radius) is less than 60 km. However, the overestimation increases to ~10% with a

383 standard deviation of ~10% when the distance is 100 km. This indicates a good

384 representativeness of U.S. Embassy's observation for Delhi and its surrounding region within

385 60 km, but may overestimate the $PM_{2.5}$ concentration and the corresponding human exposure

386 by ~10% if using U.S. Embassy's observations to estimate the $PM_{2.5}$ human exposure in a

387 larger region of Delhi, such as with a radius of 100 km. This could be due to the higher

388 urbanization level of Delhi, leading to a higher pollution level in/near the city center.

389

390 **4. Conclusions and Discussion**

391 This study has estimated the health risks of long-term exposure to $PM_{2.5}$ based on in-situ

392 observations in four Indian megacities (Delhi, Hyderabad, Chennai and Mumbai) during 2015-

393 2018, and quantified the health risks of short-term exposure during Diwali Fest in Delhi for the

394 first time. We also summarized the local characteristics of seasonal and diurnal variations of

395 $PM_{2.5}$, and report the influence of a weekend effect on diurnal patterns. The results from this

396 study are valuable for modelling studies and helpful in tailoring city-specific mitigation

397 strategies.

398 Generally, substantial inter-seasonal variations in $PM_{2.5}$ are observed in the four cities,

399 with the highest concentration during winter and the lowest during the monsoon season, when

400 intensive wet scavenging lowers pollutant concentrations (Naja et al., 2014; Ojha et al., 2012).

401 Winter is the most polluted season as a consequence of the shallow PBL and increased

402 emissions from heating (Guttikunda and Calori, 2013; Guttikunda and Gurjar, 2012). Solid fuel

403 burning is a common form of household heating in winter over India (Dumka et al., 2019;

404 Jagadish and Dwivedi, 2018). To increase the efficiency of energy use and reduce $PM_{2.5}$

405 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 two hours 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

18

431 ageing and growth, which enhance the susceptibility to disease, unless stringent emission

432 control policies are implemented (Conibear et al., 2018b).

433   We have estimated the $PM_{2.5}$ exposure in the four cities with continuous observations, but

434 it is noteworthy that some other Indian cities experience more severe air pollution (WHO,

435 2016). Continuous, widespread pollutant measurements across India would provide more

436 complete information on regional pollutant characteristics and overall pollutant levels. More

437 detailed measurements of the physicochemical properties of $PM_{2.5}$ in major cities, e.g., their

438 composition and size distribution, would permit better characterisation of urban sources, and

439 provide the information needed to design appropriate mitigation strategies.

440

Author contributions

442   Y. C. and O. W. conceived the study. Y. C. performed the analysis and interpreted the results with

443 input from all co-authors. L. C. helped with the health effect assessment. The manuscript was written with

444 input from all co-authors.

Additional Information

446   The authors declare no competing financial interest.

References:

Alam, M.A., Ahmed, F., 2013. URBAN TRANSPORT SYSTEMS AND CONGESTION: A CASE STUDY OF INDIAN CITIES. Transport and Communications Bulletin for Asia and the Pacific 82.

Atkinson, R.W., Kang, S., Anderson, H.R., Mills, I.C., Walton, H.A., 2014. Epidemiological time series studies of $PM_{2.5}$ and daily mortality and hospital admissions: a systematic review and meta-analysis. Thorax 69, 660-665.

Burnett Richard, T., Pope, C.A., Ezzati, M., Olives, C., Lim Stephen, S., Mehta, S., Shin Hwashin, H., Singh, G., Hubbell, B., Brauer, M., Anderson, H.R., Smith Kirk, R., Balmes John, R., Bruce Nigel, G., Kan, H., Laden, F., Prüss-Ustün, A., Turner Michelle, C., Gapstur Susan, M., Diver, W.R., Cohen, A., 2014. An Integrated Risk Function for Estimating the Global Burden of Disease Attributable to Ambient Fine Particulate Matter Exposure. Environmental Health Perspectives 122, 397-403.

Chen, Y., Zhao, C., Zhang, Q., Deng, Z.Z., Huang, M.Y., Ma, X.C., 2009. Aircraft study of mountain chimney effect of Beijing, china. Journal of Geophysical Research: Atmospheres 114.

Chowdhury, S., Dey, S., 2016. Cause-specific premature death from ambient PM2.5 exposure in India: Estimate adjusted for baseline mortality. Environment International 91, 283-290.

Chowdhury, S., Dey, S., Smith, K.R., 2018. Ambient PM2.5 exposure and expected premature mortality to 2100 in India under climate change scenarios. Nature Communications 9, 318.

Chowdhury, S., Dey, S., Tripathi, S.N., Beig, G., Mishra, A.K., Sharma, S., 2017. "Traffic intervention" policy fails to mitigate air pollution in megacity Delhi. Environmental Science & Policy 74, 8-13.

Conibear, L., Butt, E.W., Knote, C., Arnold, S.R., Spracklen, D.V., 2018a. Residential energy use emissions dominate health impacts from exposure to ambient particulate matter in India. Nature Communications 9, 617.

Conibear, L., Butt, E.W., Knote, C., Arnold, S.R., Spracklen, D.V., 2018b. Stringent Emission Control Policies Can Provide Large Improvements in Air Quality and Public Health in India. GeoHealth 2, 196-211.

CPCB, 2009. National Ambient Air Quality Standards. Central Pollution Control Board, New Delhi, India.

CPCB, 2014. National Air Quality Index Report, Central Pollution Control Board, New Delhi, India., https://app.cpcbccr.com/AQI_India/ (last access: 25.01.2019).

Crippa, P., Castruccio, S., Archer-Nicholls, S., Lebron, G.B., Kuwata, M., Thota, A., Sumin, S., Butt, E., Wiedinmyer, C., Spracklen, D.V., 2016. Population exposure to hazardous air quality due to the 2015 fires in Equatorial Asia. Scientific Reports 6, 37074.

Cusworth, D.H., Mickley, L.J., Sulprizio, M.P., Liu, T., Marlier, M.E., DeFries, R.S., Guttikunda, S.K., Gupta, P., 2018. Quantifying the influence of agricultural fires in northwest India on urban air pollution in Delhi, India. Environmental Research Letters 13, 044018.

Dumka, U.C., Tiwari, S., Kaskaoutis, D.G., Soni, V.K., Safai, P.D., Attri, S.D., 2019. Aerosol and pollutant characteristics in Delhi during a winter research campaign. Environmental Science and Pollution Research 26, 3771-3794.

EPA, 2009. Standard operating procedure for the continuous measurement of particulate matter. (last access: 8 Nov. 2018).

EPA, 2015. List of designated reference and equivalent methods. (last access: 20 Nov. 2018).

Fadnavis, S., Buchunde, P., Ghude, S.D., Kulkarni, S.H., Beig, G., 2011. Evidence of seasonal enhancement of CO in the upper troposphere over India. International Journal of Remote Sensing 32, 7441-7452.

Gao, M., Beig, G., Song, S., Zhang, H., Hu, J., Ying, Q., Liang, F., Liu, Y., Wang, H., Lu, X., Zhu, T., Carmichael, G.R., Nielsen, C.P., McElroy, M.B., 2018a. The impact of power generation emissions on ambient PM2.5 pollution and human health in China and India. Environment International 121, 250-259.

Gao, M., Han, Z., Liu, Z., Li, M., Xin, J., Tao, Z., Li, J., Kang, J.E., Huang, K., Dong, X., Zhuang, B., Li, S., Ge, B., Wu, Q., Cheng, Y., Wang, Y., Lee, H.J., Kim, C.H., Fu, J.S., Wang, T., Chin, M., Woo, J.H., Zhang, Q., Wang, Z., Carmichael, G.R., 2018b. Air quality and climate change, Topic 3 of the Model Inter-Comparison Study for Asia Phase III (MICS-Asia III) – Part 1: Overview and model evaluation. Atmos. Chem. Phys. 18, 4859-4884.

Gao, M., Saide, P.E., Xin, J., Wang, Y., Liu, Z., Wang, Y., Wang, Z., Pagowski, M., Guttikunda, S.K., Carmichael, G.R., 2017. Estimates of Health Impacts and Radiative Forcing in Winter Haze in Eastern China through Constraints of Surface PM2.5 Predictions. Environmental Science & Technology 51, 2178-2185.

GBD, 2016. Global, regional, and national comparative risk assessment of 84 behavioural, environmental and occupational, and metabolic risks or clusters of risks, 1990–2016: a systematic analysis for the Global Burden of Disease Study 2016. The Lancet 390 1345-1422.

GBD, 2017a. Global Burden of Disease Study 2016 (GBD 2016) Population Estimates 1950-2016. http://ghdx.healthdata.org/record/global-burden-disease-study-2016-gbd-2016-population-estimates-1950-2016 (last access: 2025.2001.2019).

GBD, 2017b. Global Burden of Disease Study 2016 (GBD 2016) Reference Life Table. http://ghdx.healthdata.org/record/global-burden-disease-study-2016-gbd-2016-reference-life-table (lase access: 2025.2001.2019).

GBD, 2018. Institute for Health Metrics and Evaluation: GBD Compare Data Visualization. vizhub.healthdata.org/gbd-compare (lase access: 25.01.2019).

Ghosh, S., Biswas, J., Guttikunda, S., Roychowdhury, S., Nayak, M., 2015. An investigation of potential regional and local source regions affecting fine particulate matter concentrations in Delhi, India. Journal of the Air & Waste Management Association 65, 218-231.

Guttikunda, S.K., Calori, G., 2013. A GIS based emissions inventory at 1 km × 1 km spatial resolution for air pollution analysis in Delhi, India. Atmospheric Environment 67, 101-111.

Guttikunda, S.K., Gurjar, B.R., 2012. Role of meteorology in seasonality of air pollution in megacity Delhi, India. Environmental Monitoring and Assessment 184, 3199-3211.

Héroux, M.-E., Anderson, H.R., Atkinson, R., Brunekreef, B., Cohen, A., Forastiere, F., Hurley, F., Katsouyanni, K., Krewski, D., Krzyzanowski, M., Künzli, N., Mills, I., Querol, X., Ostro, B., Walton, H., 2015. Quantifying the health impacts of ambient air pollutants: recommendations of a WHO/Europe project. International Journal of Public Health 60, 619-627.

Hu, D., Chen, Y., Wang, Y., Daële, V., Idir, M., Yu, C., Wang, J., Mellouki, A., 2019. Photochemical reaction playing a key role in particulate matter pollution over Central France: Insight from the aerosol optical properties. Science of The Total Environment 657, 1074-1084.

Huang, J., Pan, X., Guo, X., Li, G., 2018. Health impact of China's Air Pollution Prevention and Control Action Plan: an analysis of national air quality monitoring and mortality data. The Lancet Planetary Health 2, e313-e323.

India Office of the Registrar General and Census Commissioner, 2011. Census of India, Minist. of Home Affairs, Gov. of India, New Delhi.

Jagadish, A., Dwivedi, P., 2018. In the hearth, on the mind: Cultural consensus on fuelwood and cookstoves in the middle Himalayas of India. Energy Research & Social Science 37, 44-51.

Jethva, H., Chand, D., Torres, O., Gupta, P., Lyapustin, A., Patadia, F., 2018. Agricultural Burning and Air Quality over Northern India: A Synergistic Analysis using NASA’s A-train Satellite Data and Ground Measurements. Aerosol and Air Quality Research 18, 1756-1773.

Jiang, J., Zhou, W., Cheng, Z., Wang, S., He, K., Hao, J., 2015. Particulate Matter Distributions in China during a Winter Period with Frequent Pollution Episodes (January 2013). Aerosol and Air Quality Research 15, 494-503.

Kumar, R., Barth, M.C., Madronich, S., Naja, M., Carmichael, G.R., Pfister, G.G., Knote, C., Brasseur, G.P., Ojha, N., Sarangi, T., 2014a. Effects of dust aerosols on tropospheric chemistry during a typical pre-monsoon season dust storm in northern India. Atmos. Chem. Phys. 14, 6813-6834.

Kumar, R., Barth, M.C., Pfister, G.G., Nair, V.S., Ghude, S.D., Ojha, N., 2015. What controls the seasonal cycle of black carbon aerosols in India? Journal of Geophysical Research: Atmospheres 120, 7788-7812.

Kumar, R., Barth, M.C., Pfister, G.G., Naja, M., Brasseur, G.P., 2014b. WRF-Chem simulations of a typical pre-monsoon dust storm in northern India: influences on aerosol optical properties and radiation budget. Atmos. Chem. Phys. 14, 2431-2446.

Lelieveld, J., Bourtsoukidis, E., Brühl, C., Fischer, H., Fuchs, H., Harder, H., Hofzumahaus, A., Holland, F., Marno, D., Neumaier, M., Pozzer, A., Schlager, H., Williams, J., Zahn, A., Ziereis, H., 2018. The South Asian monsoon—Pollution pump and purifier. Science.

Lelieveld, J., Evans, J.S., Fnais, M., Giannadaki, D., Pozzer, A., 2015. The contribution of outdoor air pollution sources to premature mortality on a global scale. Nature 525, 367.

Levy, J.I., Diez, D., Dou, Y., Barr, C.D., Dominici, F., 2012. A Meta-Analysis and Multisite Time-Series Analysis of the Differential Toxicity of Major Fine Particulate Matter Constituents. American Journal of Epidemiology 175, 1091-1099.

Li, C., McLinden, C., Fioletov, V., Krotkov, N., Carn, S., Joiner, J., Streets, D., He, H., Ren, X., Li, Z., Dickerson, R.R., 2017. India Is Overtaking China as the World's Largest Emitter of Anthropogenic Sulfur Dioxide. Scientific Reports 7, 14304.

Liu, D., Quennehen, B., Darbyshire, E., Allan, J.D., Williams, P.I., Taylor, J.W., Bauguitte, S.J.B., Flynn, M.J., Lowe, D., Gallagher, M.W., Bower, K.N., Choularton, T.W., Coe, H., 2015. The importance of Asia as a source of black carbon to the European Arctic during springtime 2013. Atmos. Chem. Phys. 15, 11537-11555.

Lv, B., Cai, J., Xu, B., Bai, Y., 2017. Understanding the Rising Phase of the PM2.5 Concentration Evolution in Large China Cities. Scientific Reports 7, 46456.

Lv, B., Liu, Y., Yu, P., Zhang, B., Bai, Y., 2015. Characterizations of PM2. 5 pollution pathways and sources analysis in four large cities in China. Aerosol Air Qual. Res 15, 1836-1843.

Marrapu, P., Cheng, Y., Beig, G., Sahu, S., Srinivas, R., Carmichael, G.R., 2014. Air quality in Delhi during the Commonwealth Games. Atmos. Chem. Phys. 14, 10619-10630.

Mohan, M., Gupta, M., 2018. Sensitivity of PBL parameterizations on PM10 and ozone simulation using chemical transport model WRF-Chem over a sub-tropical urban airshed in India. Atmospheric Environment 185, 53-63.

Mukherjee, A., Toohey, D.W., 2016. A study of aerosol properties based on observations of particulate matter from the U.S. Embassy in Beijing, China. Earth's Future 4, 381-395.

Naja, M., Mallik, C., Sarangi, T., Sheel, V., Lal, S., 2014. SO2 measurements at a high altitude site in the central Himalayas: Role of regional transport. Atmospheric Environment 99, 392-402.

Ojha, N., Naja, M., Singh, K.P., Sarangi, T., Kumar, R., Lal, S., Lawrence, M.G., Butler, T.M., Chandola, H.C., 2012. Variabilities in ozone at a semi-urban site in the Indo-Gangetic Plain region: Association with the meteorology and regional processes. Journal of Geophysical Research: Atmospheres 117.

Pope, C.A., Ezzati, M., Dockery, D.W., 2009. Fine-Particulate Air Pollution and Life Expectancy in the United States. New England Journal of Medicine 360, 376-386.

Rastogi, N., Singh, A., Singh, D., Sarin, M.M., 2014. Chemical characteristics of PM2.5 at a source region of biomass burning emissions: Evidence for secondary aerosol formation. Environmental Pollution 184, 563-569.

Sahu, S.K., Beig, G., Parkhi, N.S., 2011. Emissions inventory of anthropogenic PM2.5 and PM10 in Delhi during Commonwealth Games 2010. Atmospheric Environment 45, 6180-6190.

Sahu, S.K., Kota, S.H., 2017. Significance of PM2.5 Air Quality at the Indian Capital. Aerosol and Air Quality Research 17, 588-597.

San Martini, F.M., Hasenkopf, C.A., Roberts, D.C., 2015. Statistical analysis of PM2.5 observations from diplomatic facilities in China. Atmospheric Environment 110, 174-185.

Schnell, J.L., Naik, V., Horowitz, L.W., Paulot, F., Mao, J., Ginoux, P., Zhao, M., Ram, K., 2018. Exploring the relationship between surface PM2.5 and meteorology in Northern India. Atmos. Chem. Phys. 18, 10157-10175.

Sharma, M., Dixit, O., 2016. Comprehensive Study on Air Pollution and Green House Gases (GHGs) in Delhi. . DPCC.

Sharma, S.K., Mandal, T.K., Sharma, A., Jain, S., Saraswati, 2018. Carbonaceous Species of PM2.5 in Megacity Delhi, India During 2012–2016. Bulletin of Environmental Contamination and Toxicology 100, 695-701.

Singh, D.P., Gadi, R., Mandal, T.K., Dixit, C.K., Singh, K., Saud, T., Singh, N., Gupta, P.K., 2010. Study of temporal variation in ambient air quality during Diwali festival in India. Environmental Monitoring and Assessment 169, 1-13.

Sreekanth, V., Mahesh, B., Niranjan, K., 2018. Gradients in PM2.5 over India: Five city study. Urban Climate 25, 99-108.

Srinivas, A., 2018. How traffic flow affects travel time in Delhi and Mumbai. livemint.

Tiwari, S., Srivastava, A.K., Bisht, D.S., Parmita, P., Srivastava, M.K., Attri, S.D., 2013. Diurnal and seasonal variations of black carbon and PM2.5 over New Delhi, India: Influence of meteorology. Atmospheric Research 125-126, 50-62.

van Donkelaar, A., Martin Randall, V., Brauer, M., Kahn, R., Levy, R., Verduzco, C., Villeneuve Paul, J., 2010. Global Estimates of Ambient Fine Particulate Matter Concentrations from Satellite-Based Aerosol Optical Depth: Development and Application. Environmental Health Perspectives 118, 847-855.

van Donkelaar, A., Martin, R.V., Brauer, M., Boys, B.L., 2015. Use of Satellite Observations for Long-Term Exposure Assessment of Global Concentrations of Fine Particulate Matter. Environmental Health Perspectives 123, 135-143.

van Donkelaar, A., Martin, R.V., Levy, R.C., da Silva, A.M., Krzyzanowski, M., Chubarova, N.E., Semutnikova, E., Cohen, A.J., 2011. Satellite-based estimates of ground-level fine particulate matter during extreme events: A case study of the Moscow fires in 2010. Atmospheric Environment 45, 6225-6232.

Wang, J., Xing, J., Mathur, R., Pleim Jonathan, E., Wang, S., Hogrefe, C., Gan, C.-M., Wong David, C., Hao, J., 2017. Historical Trends in PM2.5-Related Premature Mortality during 1990–2010 across the Northern Hemisphere. Environmental Health Perspectives 125, 400-408.

Wang, Y., Chen, Y., 2019. Significant Climate Impact of Highly Hygroscopic Atmospheric Aerosols in Delhi, India. Geophysical Research Letters 0.

Wang, Y., Li, W., Gao, W., Liu, Z., Tian, S., Shen, R., Ji, D., Wang, S., Wang, L., Tang, G., Song, T., Cheng, M., Wang, G., Gong, Z., Hao, J., Zhang, Y., 2019. Trends in particulate matter and its chemical compositions in China from 2013–2017. Science China Earth Sciences.

Wang, Y., Yao, L., Wang, L., Liu, Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., Xin, J., 2014. Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China. Science China Earth Sciences 57, 14-25.

West, J.J., Szopa, S., Hauglustaine, D.A., 2007. Human mortality effects of future concentrations of tropospheric ozone. Comptes Rendus Geoscience 339, 775-783.

WHO, 2013. Health risks of air pollution in Europe – HRAPIE project: Recommendations for concentration–response functions for cost–benefit analysis of particulate matter, ozone and nitrogen dioxide. http://www.euro.who.int/__data/assets/pdf_file/0006/238956/Health_risks_air_pollution_HRAPIE_project.pdf?ua=238951 (last access: 238925.238901.232019).

WHO, 2016. WHO Global Urban Ambient Air Pollution Database (update 2016). Available: http://www.who.int/airpollution/data/cities-2016/en/, (last access: 08 Nov. 2018).

Yu, P., Rosenlof, K.H., Liu, S., Telg, H., Thornberry, T.D., Rollins, A.W., Portmann, R.W., Bai, Z., Ray, E.A., Duan, Y., Pan, L.L., Toon, O.B., Bian, J., Gao, R.-S., 2017. Efficient transport of tropospheric aerosol into the stratosphere via the Asian summer monsoon anticyclone. Proceedings of the National Academy of Sciences 114, 6972-6977.

Zhang, Y.-L., Cao, F., 2015. Fine particulate matter (PM2.5) in China at a city level. Scientific Reports 5, 14884.

Zheng, G.J., Duan, F.K., Su, H., Ma, Y.L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimoto, T., Chang, D., Pöschl, U., Cheng, Y.F., He, K.B., 2015. Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions. Atmos. Chem. Phys. 15, 2969-2983.
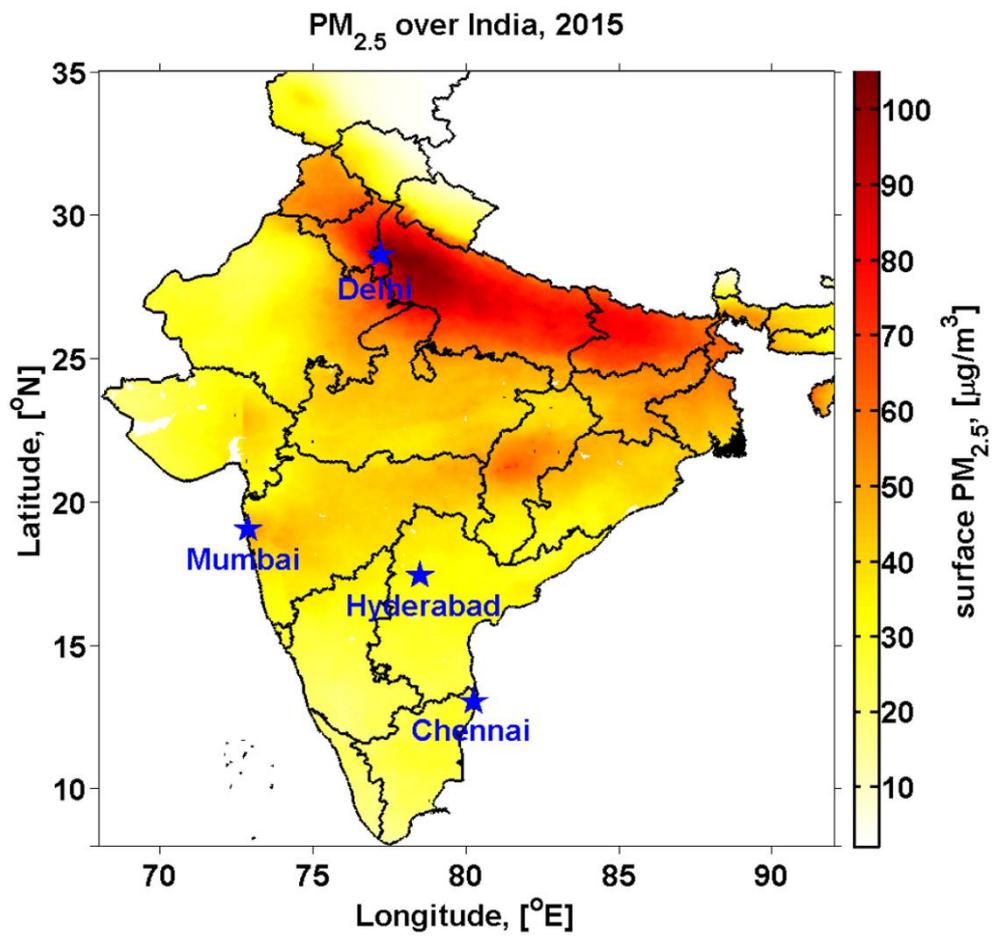
Figure 1. Map of Delhi, Chennai, Hyderabad and Mumbai. Surface annual (2015) average of PM$_{2.5}$ is retrieved from satellite observations with sea-salt and dust excluded and at a relative humidity of 35% (van Donkelaar et al., 2015).
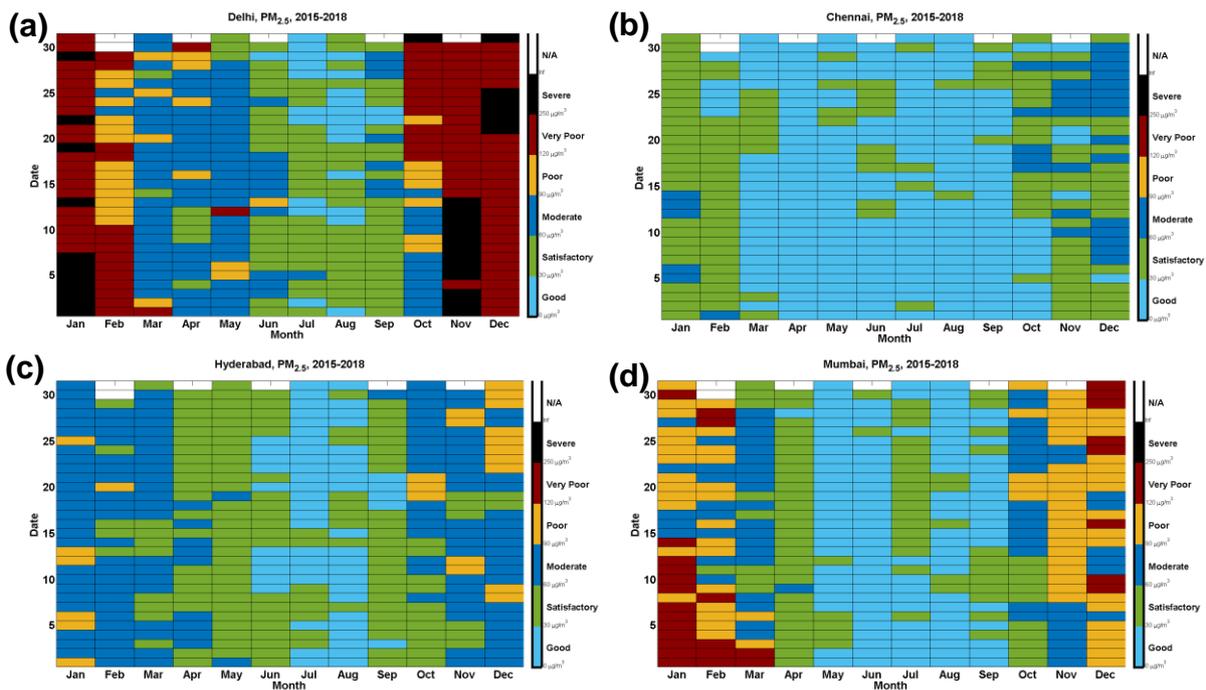
Figure 2. Calendar-view of daily PM_{2.5} air quality levels averaged over 2015-2018. (**a**) Delhi, (**b**) Chennai, (**c**) Hyderabad, and (**d**) Mumbai. The air quality levels are categorized following the Indian national air quality index definitions (https://app.cpcbccr.com/AQI_India).
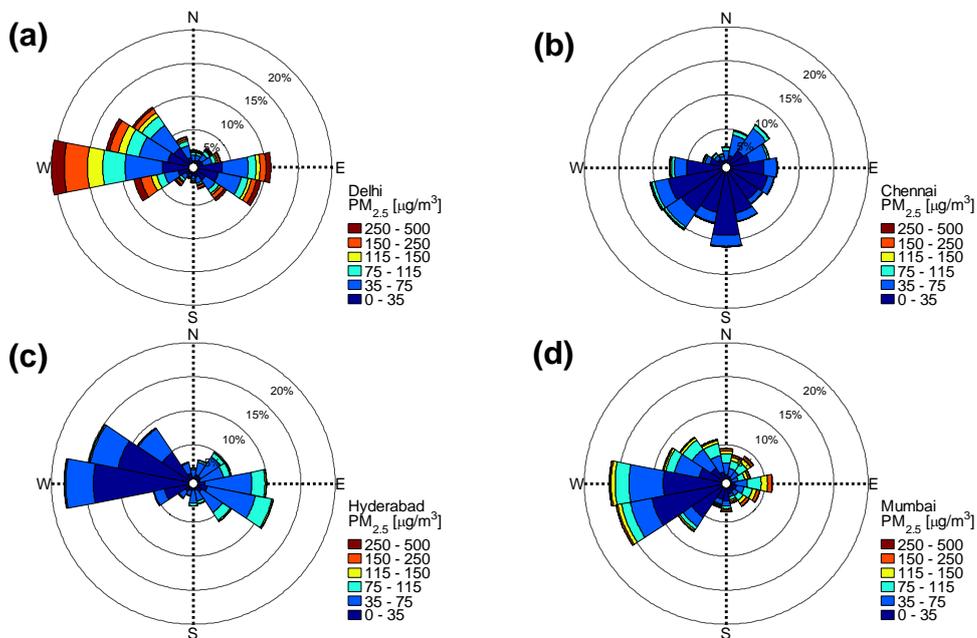
Figure 3. Frequency distributions of PM_{2.5} concentration as a function of wind direction. (**a**) Delhi, (**b**) Chennai, (**c**) Hyderabad, and (**d**) Mumbai.
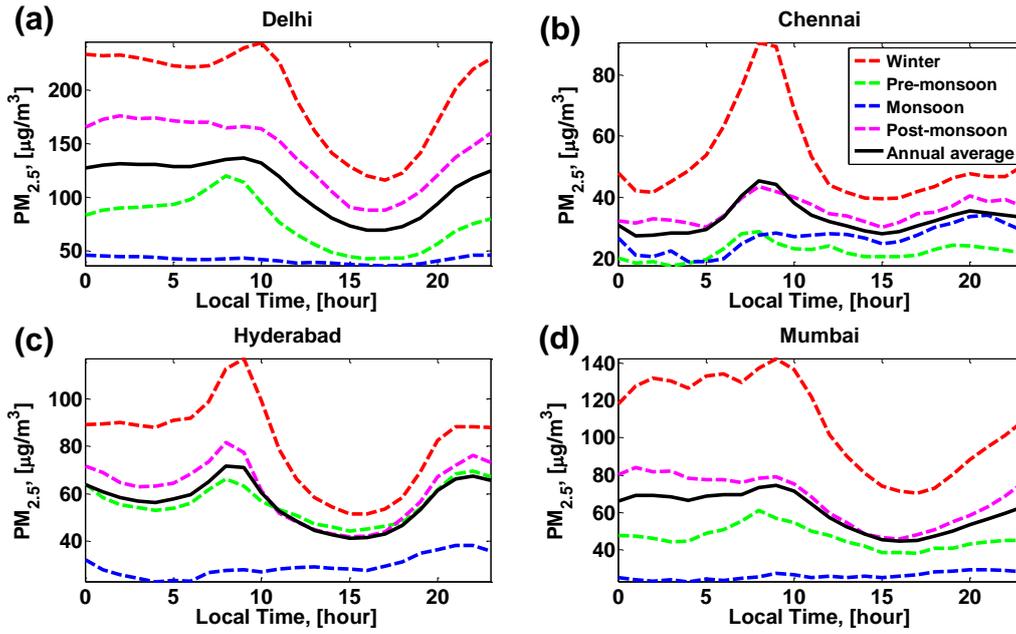
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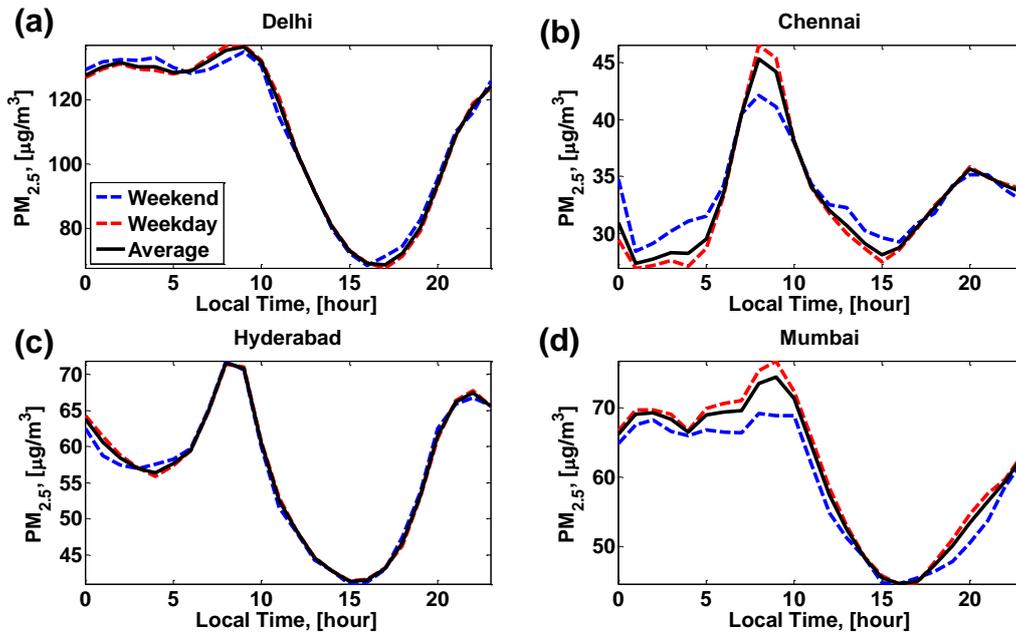
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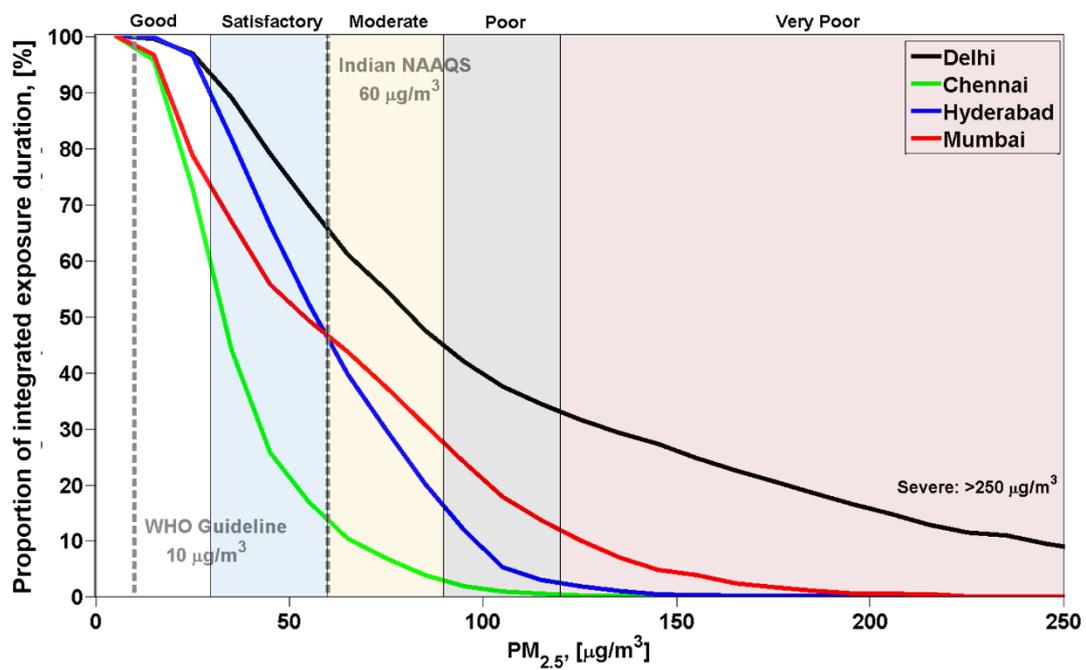
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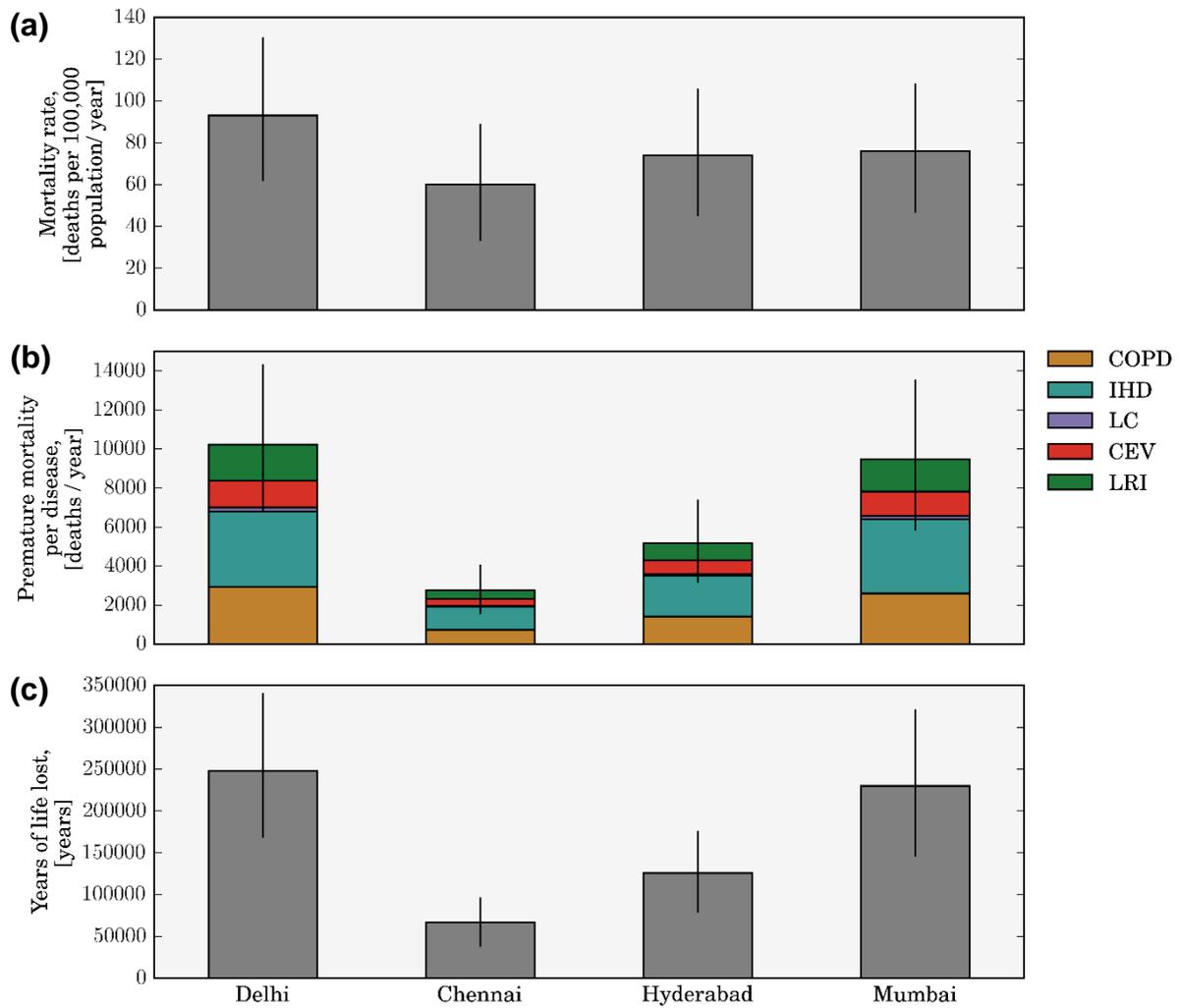
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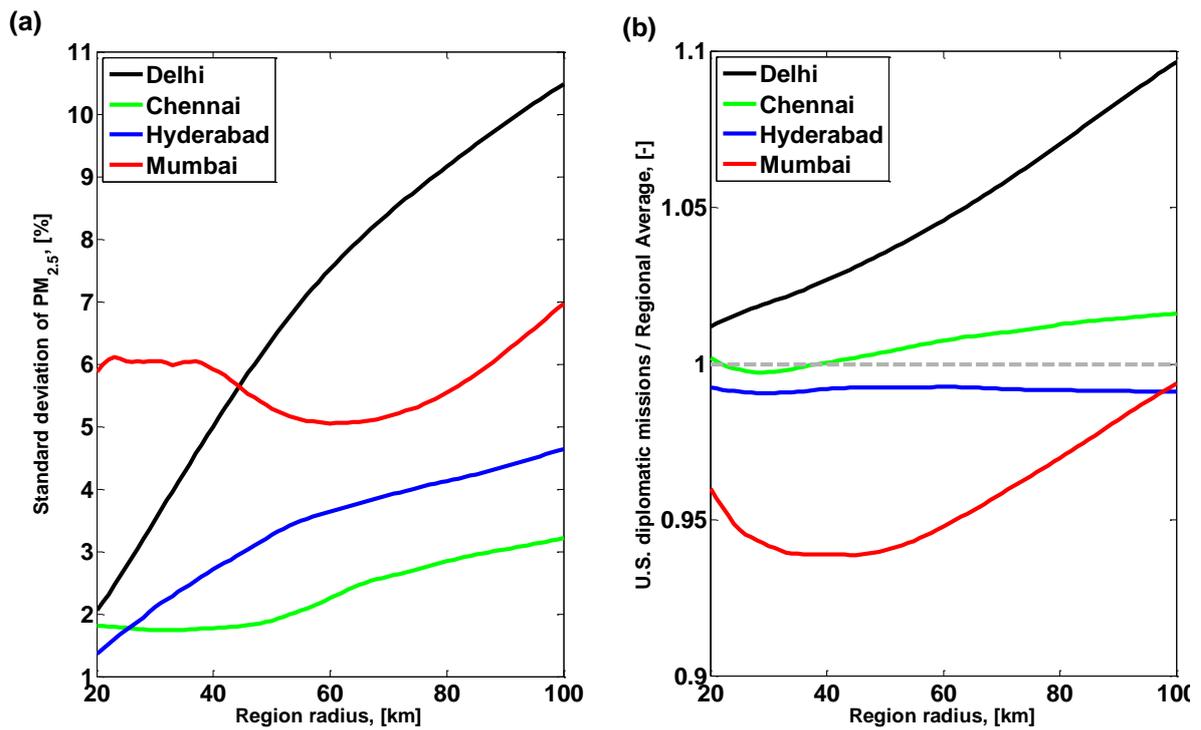
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Supporting Information

for

Local Characteristics of and Exposure to Fine Particulate Matter (PM$_{2.5}$) in Four Indian Megacities

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Table S1. Overview of PM$_{2.5}$ observations in four cities.

City	Lat. / Long. [ºN / ºE]	Population* [million]	Climate Description	Annual PM$_{2.5}$ [µg/m^3]	Unhealth/Valid[#] [days]
Delhi	28.60 / 77.19	11.0	Subtropical/continental	110	837/1373
Chennai	13.05 / 80.22	4.6	Tropical/coastal	33	133/1274
Hyderabad	17.44 / 78.47	7.0	Tropical/continental	56	528/1335
Mumbai	19.06 / 72.87	12.5	Tropical/coastal	60	565/1293

*source from census of India 2011 (India Office of the Registrar General and Census Commissioner, 2011)

[#]Valid days show how many days with valid data during March 2015 to December 2018. The unhealth-day is counted the days with daily-averaged PM$_{2.5}$ > 60 µg/m^3, following the definition of Indian NAAQS (CPCB, 2009).
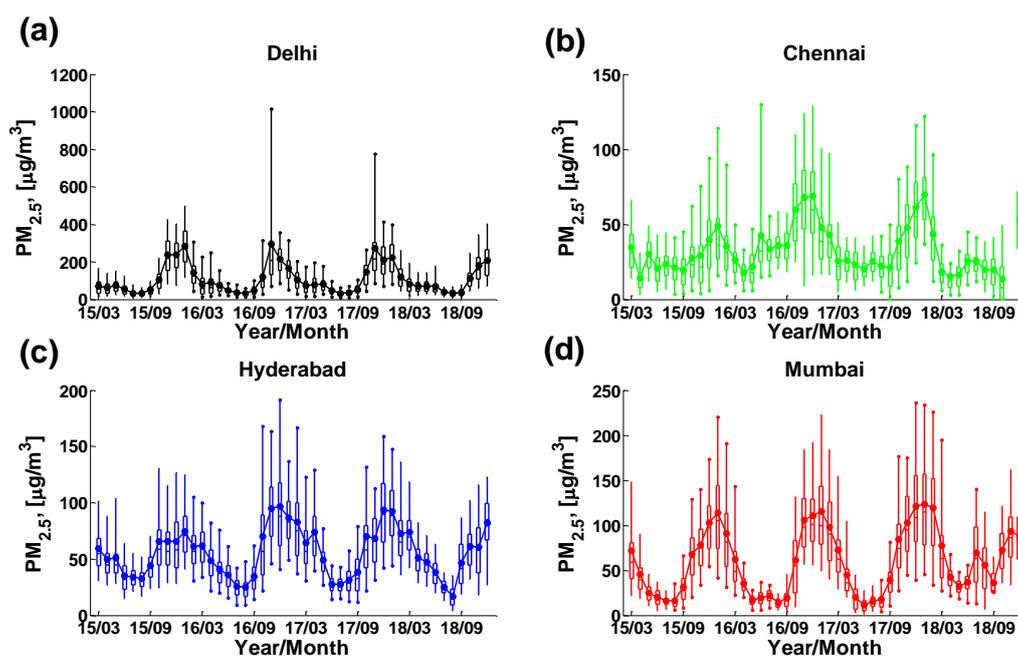
Figure S1. Monthly statistical overview of PM$_{2.5}$ hourly concentrations. (**a**) Delhi, (**b**) Chennai, (**c**) Hyderabad, and (**d**) Mumbai. The dots indicate the average value; short-scores in the middle indicate the median value; the boxes indicate the 25% and 75% percentage values; and the error bars indicate the 5% and 95% percentage values.
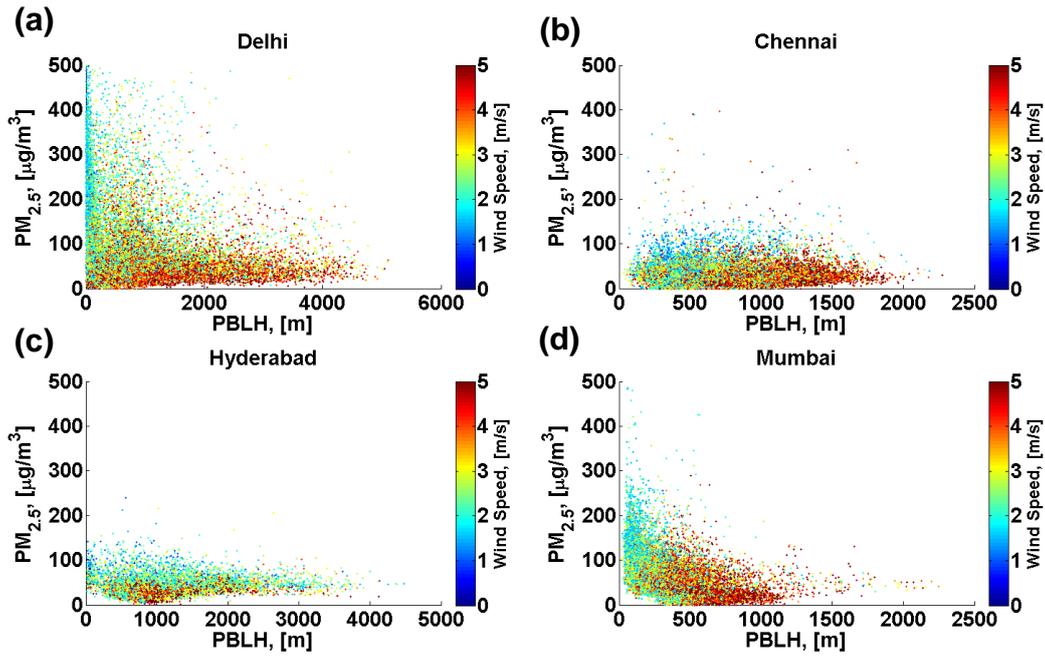
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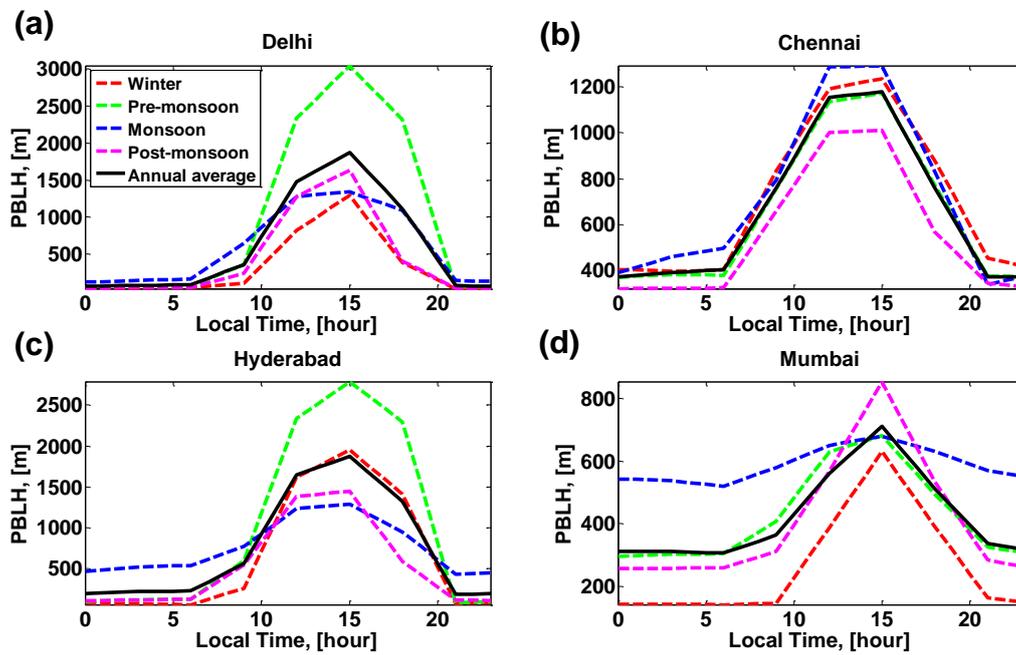

Figure S3. The averaged diurnal pattern of the height of PBL (PBLH) for each season. (**a**) Delhi, (**b**) Chennai, (**c**) Hyderabad, and (**d**) Mumbai.
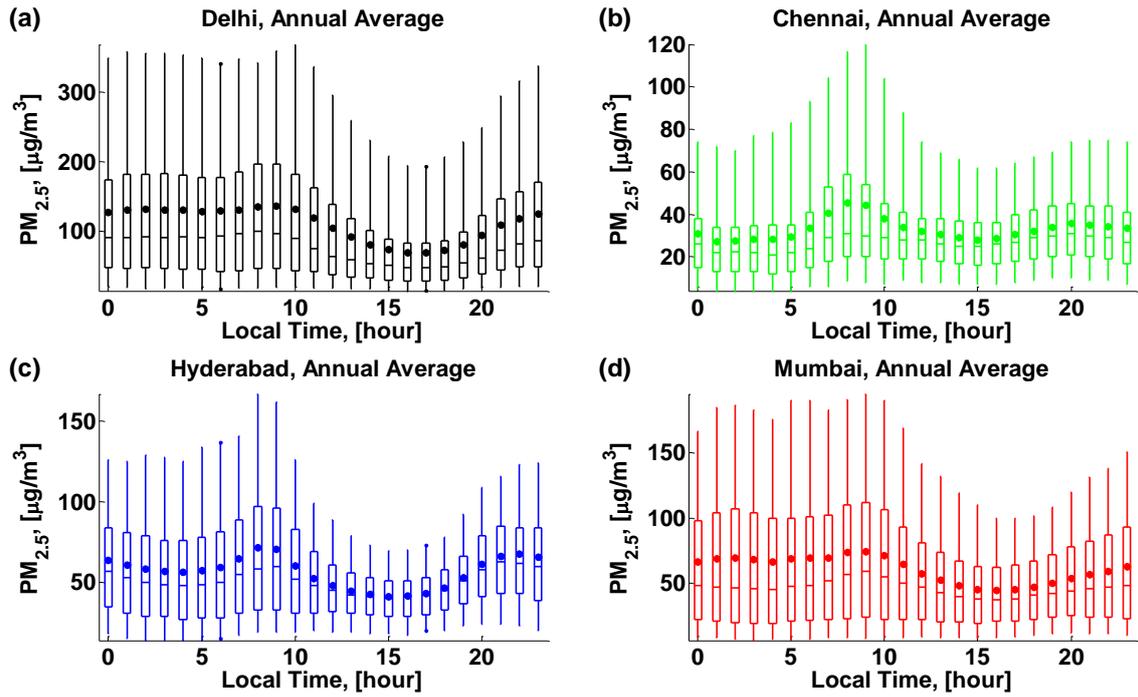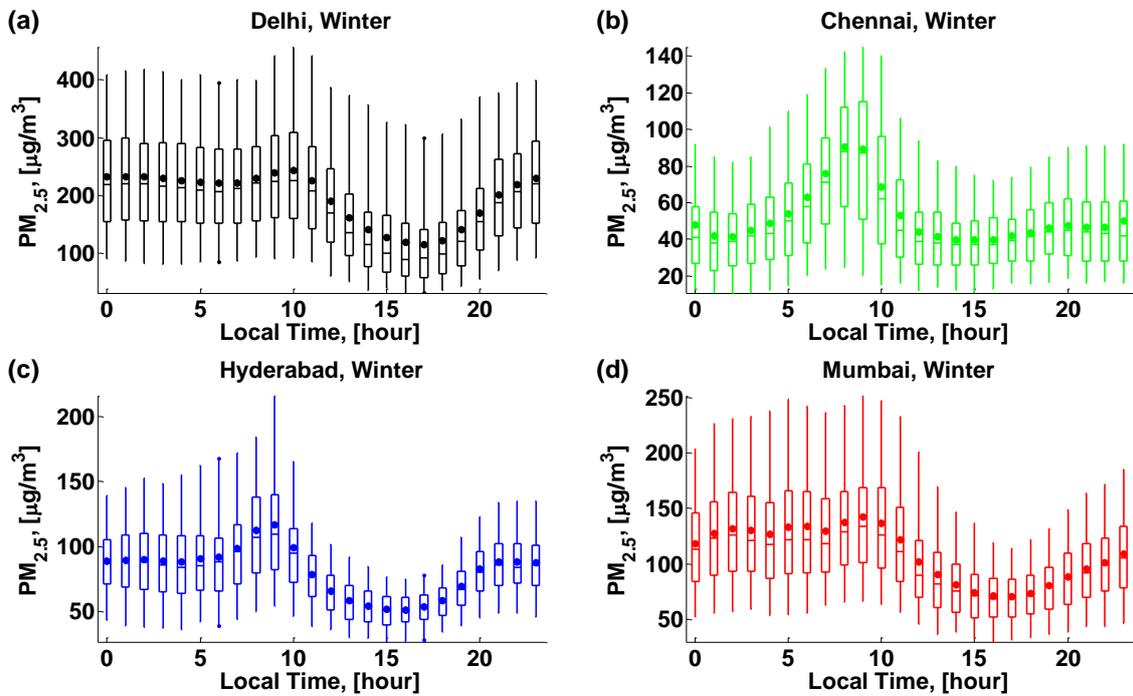
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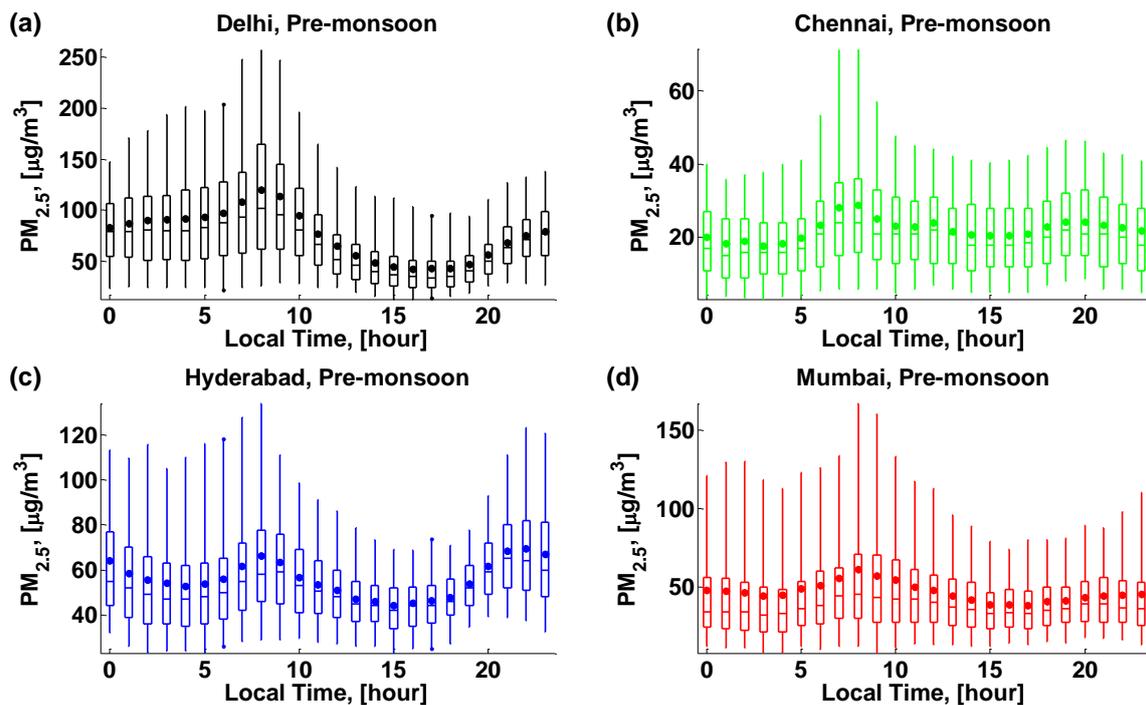**Figure S5. Same as Fig. S4, but during the winter season.**

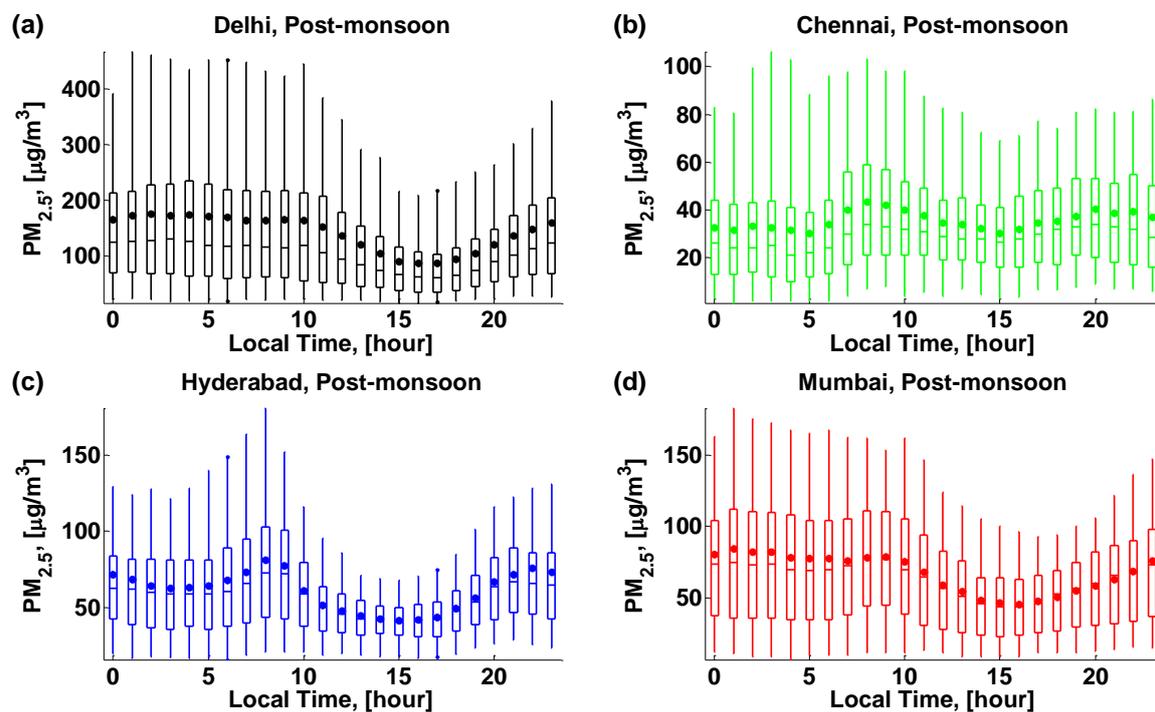**Figure S6. Same as Fig. S4, but during the pre-monsoon season.**

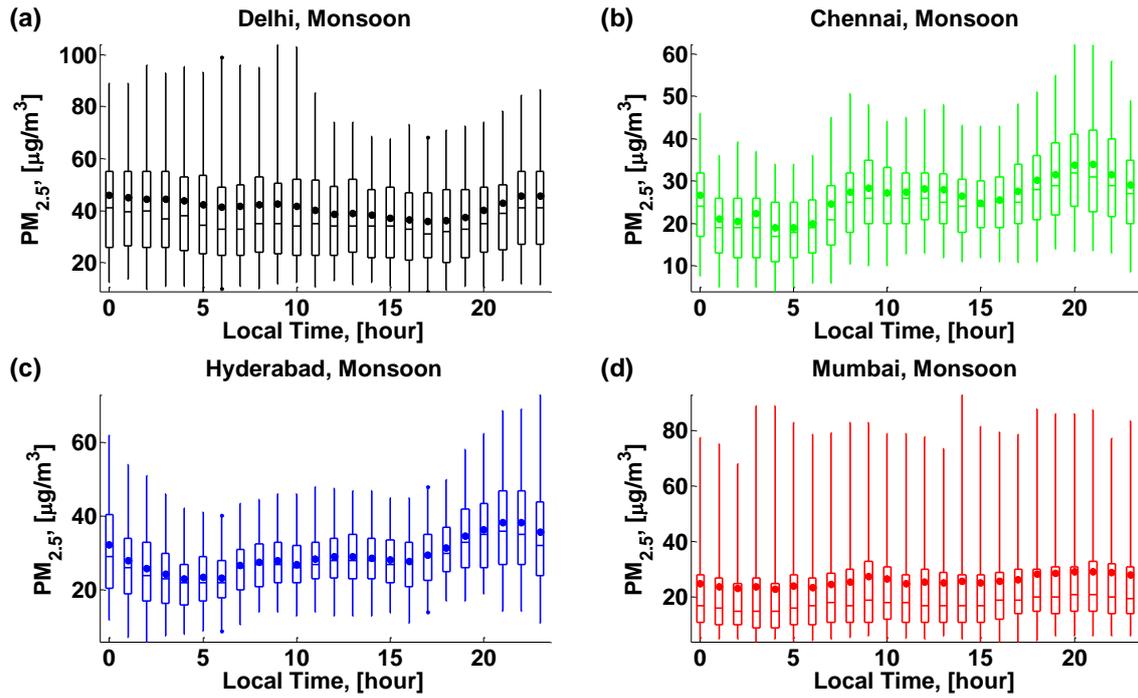**Figure S7. Same as Fig. S4, but during the post-monsoon season.**

Figure S8. Same as Fig. S4, but during the monsoon season.

Supplementary References:

CPCB, 2009. National Ambient Air Quality Standards. Central Pollution Control Board, New Delhi, India.

India Office of the Registrar General and Census Commissioner, 2011. Census of India, Minist. of Home Affairs, Gov. of India, New Delhi.