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### Municipal solid waste and dung cake burning: discoloring the Taj Mahal and human health impacts in Agra

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### Abstract

The Taj Mahal—an iconic World Heritage monument built of white marble—has become discolored with time, due, in part, to high levels of particulate matter (PM) soiling its surface (Bergin et al 2015 Environ. Sci. Technol. 49 808–812). Such discoloration has required extensive and costly treatment (2015 Two Hundred Sixty Second Report on Effects of Pollution on Taj Parliament of India Rajya Sabha, New Delhi) and despite previous interventions to reduce pollution in its vicinity, the haze and darkening persists (Bergin et al 2015 Environ. Sci. Technol. 49 808-812; 2015 Two Hundred Sixty Second Report on Effects of Pollution on Taj Parliament of India Rajya Sabha, New Delhi). PM responsible for the soiling has been attributed to a variety of sources including industrial emissions, vehicular exhaust and biomass burning, but the contribution of the emissions from the burning of open municipal solid waste (MSW) may also play an important role. A recent source apportionment study of fine particulate matter (PM2.5) at the Taj Mahal showed biomass burning emissions, which would include MSW emissions, accounted for nearly 40% of organic matter (OM)-a component of PM—deposition to its surface (Bergin et al 2015 Environ. Sci. Technol. 49 808–812); dung cake burning, used extensively for cooking in the region, was the suggested culprit and banned within the city limits (2015 Two Hundred Sixty Second Report on Effects of Pollution on Taj Parliament of India Rajya Sabha, New Delhi), although the burning of MSW, a ubiquitous practice in the area (Nagpure et al 2015 Environ. Sci. Technol. 49 12904–12), may play a more important role in local air quality. Using spatially detailed emission estimates and air quality modeling, we find that open MSW burning leads to about 150 ( $\pm$ 130) mg m<sup>-2</sup> yr<sup>-1</sup> of PM<sub>2.5</sub> being deposited to the surface of the Taj Mahal compared to about 12 ( $\pm$ 3.2) mg m<sup>-2</sup> yr<sup>-1</sup> from dung cake burning. Those two sources, combined, also lead to an estimated 713 (377–1050) premature mortalities in Agra each year, dominated by waste burning in socioeconomically lower status neighborhoods. An effective MSW management strategy would reduce soiling of the Taj Mahal, improve human health, and have additional aesthetic benefits.

### Introduction

The Taj Mahal in Agra, India is a UNESCO World Heritage Site that attracts millions of tourists each year. However, its surface has been soiled over time, discoloring its white marble façade. Studies have recognized that poor air quality is responsible for the soiling and discoloration [1, 4–7] and measures have been taken to curb the impact of local air pollution around the Taj Mahal including restricting vehicles near the complex, closing over 200 enterprises in Agra, requiring iron foundries to install scrubbers and filters on their smokestacks, prohibiting new polluting enterprises from being built within a defined buffer zone around the mausoleum, and most recently, banning cow dung cake burning as cooking fuel [2]. A recent source apportionment study of fine particulate matter (PM<sub>2.5</sub>, whose particles are less than 2.5  $\mu$ m in aerodynamic diameter) at the Taj Mahal found that biomass burning accounts for nearly 40% of all organic matter (OM) deposition to its surface [1]. Two sources of biomass burning PM2.5 in Agra, which would be included in the measurement of deposited OM, are the open combustion of municipal solid waste (MSW) and dung cake burning [3]. The high particulate matter (PM) loadings in Agra also reduce visibility, further impairing the aesthetic beauty of the Taj Mahal.

While the discoloration of the Taj Mahal and the deterioration of visibility may be the most immediately noticeable outcome of MSW and dung cake burning in the area, human health is of concern as well. The Global Burden of Disease (GBD) found that of 67 environmental factors associated with premature mortality, exposure to ambient PM pollution is the 5th leading cause of premature mortality in India after high blood pressure, indoor air pollution (which is also affected by dung cake burning), smoking and dietary risks [8]. Additionally, residential and commercial energy use, including biomass burning used for heating and cooking, is responsible for the largest impact on mortality linked to outdoor air pollution throughout India [9].

Rapid growth in Agra, coupled with a limited MSW management infrastructure, has resulted in less effective waste management that leaves large volumes of trash accumulating in the streets [3, 10]. Further, generated waste is openly and frequently burned on roadsides and in residential and commercial areas in Agra [3] and throughout India [10], leading to byproducts of poor combustion and increased pollutant emissions [11-13]. The Central Pollution Control Board of India estimated MSW-burning to contribute between 5% and 11% of primary PM emissions from sources within cities [14]. MSW emissions include combustion byproducts of plastics and other waste in addition to biomass, which can contain chlorinated dioxins, polyaromatic hydrocarbons organics, (PAHs), numerous volatile organic compounds (VOCs) and heavy metals including lead, cadmium and mercury [15, 16]. Health impacts specific to these toxic compounds are not specifically addressed in the GBD approach.

Dung cake burning used as cooking fuel has been more studied in Indian cities [17–19]; 11% of rural Indian households depend on cow dung as their primary cooking fuel [19]. Open MSW burning and dung cake burning tends to be more concentrated in areas of poorer populations [3, 20–24], exacerbating exposures to more vulnerable populations. MSW and dung cake emissions can also influence radiative balance and lead to regional and global change [11, 25, 26].

In this paper, the contributions of MSW and dung cake burning to ambient OM and BC (pollutants known to discolor surfaces [27]) concentrations in Agra, the deposition to and soiling of the Taj Mahal, and health impacts are assessed by quantifying location specific MSW and dung cake burning emissions, performing air quality and deposition modeling, and conducting a health impact assessment. Such information can be used to evaluate the potential benefits of policy interventions, including improved MSW collection management practices and the associated infrastructure in and around Agra.

### Methods

#### Open MSW and dung cake burning inventories

Waste burn rate inventories were generated in Agra using a recently developed field transect approach to quantify the spatial and temporal trends of open MSW burning [3]. In this method, researchers move along the transect (route/line) and record burning incidents, approximate weight, and composition of MSW in a predetermined distance from the line of the transect (route/line) (typically visible range is used as the distance). MSW burning incident density is then estimated by the total MSW burning incidents count and surveyed area. Two separate transect routes in Agra that covered 35 and 45 km<sup>2</sup>, respectively (SI figures 1 and 2), were used in this study over three days for each route between 30 May and 2 Jun, 2015 to quantify the waste burn density, composition, and the mass of waste burn. These surveys assessed MSW burning by socioeconomic status (SES) based on census data [18] at the neighborhood level and represented 14 neighborhoods of different SES (SI figure 1). Satellite-driven studies at the global scale cannot capture the very high levels of waste burning found in neighborhoods or near roads [9], thus the on-ground field approach is an important part of developing an improved PM emission inventory from MSW burning.

The open waste burn rate, TWB<sub>*i*</sub> (g-MSW day<sup>-1</sup>), within an electoral ward, *i*, from the SES-based waste burning rates is quantified by:

$$TWB_{i} = WBR_{lowSES} * POP_{i,lowSES} + WBR_{highSES} * (1 - POP_{i,lowSES})$$
(1)

where WBR<sub>lowSES</sub> = daily per capita waste burn rate of the low SES, POP<sub>*i*,lowSES</sub> = illiterate population within the ward as reported in the 2011 census [18], and WBR<sub>highSES</sub> = daily per capita waste burn of the high SES. Literacy was the primary indicator of SES used in this study; the total reported literacy rate in Agra is 64% [18]. Waste burn inventories were generated on an electoral ward basis and each ward



was modeled as its own emission grid, as were five additional zones (SI figure 3).

Data on the use of cow-dung cakes as fuel for food preparation data was assessed from the census [18]. The census gave the percentage of households at the ward/precinct level using different types of fuel for cooking. Annual per household consumption of cow dung was then multiplied with the number of households using cow dung as a fuel for cooking (SI figure 4) within each ward/precinct to determine electoralward based burning inventories, computed on an annual basis and then converted to daily average emission rates. Applying the same method, air quality impacts from two additional sources, firewood and crop residue, were also modeled for comparison.

### MSW and dung cake burn inventories to AERMOD dispersion modeling

Open MSW and dung cake burn rates were applied in AERMOD, a Gaussian plume dispersion model [28], to spatially characterize the ambient, annually averaged PM<sub>2.5</sub> concentrations from MSW and dung cake burning. AERMOD is a recommended regulatory air pollution dispersion model, but has limitations as it does not include atmospheric chemical processes or secondary pollution formation [28]. The findings presented here are specific source impacts from emissions within the study domain, i.e., background transport is not considered. Integrated hourly surface data from the National Climatic Data Center (NCDC) at the Agra Station from the National Oceanic and Atmospheric Administration (NOAA) and upper air data from the US National Weather Service (NWS) at the Delhi Station were used in AERMET, a meteorological input to AERMOD. Digital Elevation Models from the Global 30 Arc-Second Elevation (GTOPO30) were used in AERMAP, a terrain processing input to AERMOD.

OM and BC source emission rates from both MSW and dung cake burning were determined using emission factors from the literature [29, 30] (SI table 2). PM<sub>2.5</sub> component-specific emission factors for MSW burning used here are from measurements of trash burning in peri-urban communities near Mexico City at varying combustion stages [29]. Christian et al [29] found emission factors of OC = 5.3 ( $\pm$ 4.9) and  $BC = 0.65 \ (\pm 0.27) \, g \, kg^{-1}$  burned. These emission factors are within the reported range of  $0.04-9.97 \text{ g BC kg}^{-1}$  burned from recent measurements of trash burning in Nepal where some samples were enriched for specific compositions of plastic and foil [31], but lower than the reported range of 8.4–73.9 g OC kg<sup>-1</sup> burned. MSW emissions can vary significantly and have high uncertainties due to the composition of the waste and stage of combustion [13, 32]. Emission factors applied for dung cake burning were measured in households throughout the Indo-Gangetic Plain [30]. An OM/OC factor of 2.1 [33] was applied to the OC emission factors; OM is

related to OC as the former accounts for specific elements other than carbon associated with the organic compounds.

## Human health risk assessment from open MSW and dung cake burning emissions

Premature mortality attributable  $PM_{2.5}$ to (BC + OM) emissions from MSW and cow dung cake burning were determined using concentration response function (CRFs) based equations. Five major diseases-acute respiratory lung infection (ALRI), chronic obstructive pulmonary disease (COPD), ischemic heart disease (IHD), cerebrovascular disease (stroke) and lung cancer (LC)-associated with PM2.5 mortality risks were assessed in this study. COPD, IHD, stroke, and LC related mortality were determined for adults (age  $\geq 25$  years), while mortalities related to ALRI were estimated for children under five years of age. Disease-specific relative risk equations use a CRF, incidence rate for premature mortality, change (increment) in ambient pollution concentration, and exposed population to estimate the mortality. The CRFs data and equation (2) used integratedexposure response functions (IERs) to estimate specific health impacts [34].

$$\begin{cases} \operatorname{RR} = 1 + a \{ 1 - \exp[-b(\Delta C)^p] \}, & \text{for } C > C_o \\ \operatorname{RR} = 1, & \text{for } C \leqslant C_o \end{cases}$$

$$(2)$$

$$PAF = \frac{\sum_{i=1}^{n} P_i(RR_i - 1)}{\sum_{i=1}^{n} P_i(RR_i - 1) + 1}$$
(3)

$$P_h = B_i^* \text{PAF}^* P_i, \tag{4}$$

where RR is the relative risk or CRFs,  $\Delta C$  is the increase of ambient PM2.5 concentrations due to dung cake and MSW burning emissions,  $C_0$  is the baseline PM<sub>2.5</sub> concentration (considered 0 for this source impact application), and a, b, and p are parameters that determine the relationship of concentration to response and are discussed further in Burnett et al 2014 [34]. PAF is the population attributable fraction, i.e., the proportion of the disease incidence on the exposed population that can be attributed to the exposure,  $P_i$  is the fraction of the population in exposure category, *i*, and *n* is the number of exposure categories, where exposure categories were defined by five-year age increments with available CRFs.  $P_h$  is the premature mortality associated with PM2.5 exposure and  $B_i$  is the baseline population incidence of given health effects (i.e. death per 100 000). The exposed population within each modeling grid was retrieved from the 2015 Worldpop Database. A growth factor for the total population within the study domain for the Worldpop Database reported population compared to the 2014 projected population from the census [18] was used, as the modeling results presented are for 2014.

Also determined were disability adjusted life years (DALY), which estimate the current discounted value

of future years of health life lost due to morbidity and future year of human years of life lost (YLL) due to premature mortality. Since air pollutants are not a primary cause of mortality, but rather contributory, DALY can be a better indicator of health risks than premature mortality [35]. The DALYs are calculated as the total of the YLL due to premature mortality and years lost due to disability (YLD) because of morbidity. In this study we only estimated the premature deaths due to PM<sub>2.5</sub> emissions associated with biomass and MSW burning and thus considered YLL as the measure of DALYs. YLL were calculated using the following equation:

$$YLL = B_i * PAF * POP_i * LE, (5)$$

where POP<sub>i</sub> is the exposed population (i.e., the population within each modeled grid) and LE is the standard life expectancy at age of death (in years).

### Dry deposition to and pollutant covering of the Taj Mahal

Pollutant deposition to the surface of the Taj Mahal contributes to its browning [1], so the impacts of wet and dry deposition from MSW and dung cake emissions were quantified. Dry deposition rates were calculated using modeled concentrations, measured size distributions and size-dependent deposition velocities. Deposition velocity is a variable that incorporates the aerodynamic transport through the atmospheric surface layer, the transport across the quasi-laminar sublayer, and the uptake at the surface into a single parameter [36, 37]. Imaging from a scanning electron microscropy (SEM) (LEO 1530, Carl Zeiss Microscopy) and energy dispersive x-ray spectroscopy (Oxford Instruments  $X_{\text{max}}$  detectors) were used to measure the average particle size of carbonaceous PM species at the surface of the Taj Mahal [1]. The average particle size was found to be ~1  $\mu$ m.

The PM<sub>2.5</sub> component specific mass fluxes (g m<sup>-2</sup> s<sup>-1</sup>),  $F_i$ , of OM and BC to the surface of the Taj Mahal by dry deposition were found as:

$$F_{i}(t) = -V_{D,i}(d_{p,\text{ave}})^{*}[C_{i}(t)],$$
(6)

where  $V_D$  is the size-specific surface deposition velocity (m s<sup>-1</sup>) and  $d_{p,ave}$  is the average particle diameter. The pollutant concentration,  $[C_i(t)]$ , used here is the annual average, ambient pollutant concentration from open waste and dung cake burning at the Taj Mahal as determined in AERMOD. Wet deposition was considered in this analysis to account for rain, and the wet deposition loadings were small compared to dry deposition (see SI section 3 for a detailed assessment).

The fraction of the Taj Mahal's surface covered by pollutant deposition from MSW and dung cake burning emissions was also quantified from the modeled number of particles deposited per area of the surface and the total surface area of the aerosol deposited per



area of the surface. The number of particles per unit area (particles m<sup>-2</sup>), *N*, from each source and pollutant, *i*, was determined by:

$$N_i = \frac{\sigma_i}{\rho_i d_{p,\text{ave}}^3/6},\tag{7}$$

where  $\sigma_i$  (mg m<sup>-2</sup> yr<sup>-1</sup>) is the specific pollutant loading for each source,  $\rho_i$  is the pollutant (OM or BC) density [38, 39], and  $d_{p,ave}$  is the average particle diameter from on-site measurements (~1  $\mu$ m).

Combined with the average surface area per particle, the fractional cover of  $PM_{2.5}$  emissions from MSW and dung cake burning in one year,  $\Omega_i$ , was then calculated as:

$$\Omega_i = \frac{6\sigma_i}{\rho_i d_{p,\text{ave}}}.$$
(8)

### **Results and discussion**

### Open MSW and dung cake burning emissions to modeled concentrations throughout Agra and model evaluation

Employing the field transect method developed by Nagpure *et al* [3], the total average waste burn rate in Agra was estimated at 130 g MSW capita<sup>-1</sup> day<sup>-1</sup> with higher per capita burn rates observed in low SES areas (table 1). Burn rates were higher in the morning than the evening within the city, but showed less diurnal difference in the rural areas (areas outside of the city boundaries). If Agra's per capita average waste burn rate is applied to the entire population of India, the annual nationwide burn rate would be  $68000 \text{ Gg yr}^{-1}$ , consistent with model findings of Wiedinmyer et al of  $35000-75000 \text{ Gg yr}^{-1}$  for India [40]. The total cow dung cake burning emissions on a ward-by-ward basis within Agra were calculated from household fuel use data [17, 18] (SI figure 4) and ranged between  $0-9100 \text{ kg day}^{-1} \text{ ward}^{-1}$  within the study domain, compared to 490–25000 kg day<sup>-1</sup> ward<sup>-1</sup> from open waste burning (SI table 1). A report on sustainable solid waste management in India reported the average waste generation rate in Agra as 580 g MSW capita<sup>-1</sup> day<sup>-1</sup> [41]. Applying this MSW generation rate, the average burn rate of MSW in Agra is 23%, higher than the 5%-10% estimates from previous waste burning studies in Indian cities [10, 42, 43].

Applying emission factors from the literature [29, 30] in conjunction with observed burn rates resulted in annual combined emissions in Agra from open waste and dung burning to be 2500 ( $\pm$ 2200) kg yr<sup>-1</sup> and 150 ( $\pm$ 58) kg yr<sup>-1</sup> for the OM and BC components of PM<sub>2.5</sub>, respectively. Annual average PM<sub>2.5</sub> component concentrations due to open waste and dung cake burning throughout Agra, simulated by AERMOD, found concentrations at the Taj Mahal to be 4.1 ( $\pm$ 3.8) and 0.24 ( $\pm$ 0.10)  $\mu$ g m<sup>-3</sup> for OM and BC from MSW burning and 0.32 ( $\pm$ 9.1 × 10<sup>-2</sup>) and 0.019 ( $\pm$ 9.7 × 10<sup>-4</sup>)  $\mu$ g m<sup>-3</sup> for OM and BC from

**Table 1.** Diurnal per capita open MSW burn rates (g capita<sup>-1</sup> day<sup>-1</sup>) in Agra categorized by socioeconomic status (SES) using a recently developed field transect approach [3]. Higher per capita open waste burn rates were observed in regions of lower SES.

	Morning transect	Evening transect	Full day
High SES	73.0	20.9	93.9
Low SES	157	39.3	196
Rural areas	73.5	106	180

dung cake burning (figure 1 and SI figure 5). Uncertainty was assessed just for the emission factors as that is where much of the uncertainty lies due to variations in waste composition and stage of combustion. The calculation does not consider secondary formation of PM<sub>2.5</sub> due to gaseous emissions from those sources. These results were evaluated using measurements from a recent PM2.5 source apportionment study at the Taj Mahal that found that the contribution of biomass burning emissions to OM (which can be from a variety of combustion activities including wood, crop, dung and MSW burning) at the Taj Mahal to be  $12 \,\mu g \,\mathrm{m}^{-3}$  [1]. While the sum of the four sources assessed here (MSW, dung cake, firewood, and crop residue) is 5.9 ( $\pm$ 4.7)  $\mu$ g m<sup>-3</sup>, suggesting regional transport of additional OM, MSW is the highest contributor of modeled biomass burning sources (SI figure 6).

Maximum combined annual-averaged impacts on  $PM_{2.5}$  in Agra were 33 (±30)  $\mu g m^{-3}$  from MSW burning and 3.3 ( $\pm 0.90$ )  $\mu g m^{-3}$  from dung cake burning (figure 1 and SI figure 7). High levels were found in neighborhoods with lower SES where MSW and dung-cake burning are most prevalent. The contribution from open MSW burning is greater than for dung cake burning throughout Agra, except in the rural areas where dung cake burning is a primary fuel source for cooking [17, 18]. The combined annuallyaveraged ambient PM2.5 concentration averaged throughout Agra from open waste and dung cake burning was 4.3 ( $\pm$ 3.8)  $\mu$ g m<sup>-3</sup> for OM and 0.25  $(\pm 0.10) \,\mu \text{g m}^{-3}$  for BC. Recent ambient OC and elemental carbon concentration measurements throughout Agra have been reported between 10.2 ( $\pm$ 7.2)–30  $(\pm 13) \,\mu g \,\mathrm{m}^{-3}$  and 1.3  $(\pm 0.8)$ -4.0  $(\pm 1.5) \,\mu g \,\mathrm{m}^{-3}$ [32, 44], which suggest the source impact modeling results averaged over the study domain are in line with ambient measurements.

### Adverse health and premature mortality assessments

Estimation of premature mortality associated with  $PM_{2.5}$  (BC + OM) emissions from dung cake and MSW burning suggest that these two sources are responsible for 713 (377–1050) cases of premature mortalities from outdoor exposure in Agra annually, 380 (247–540) attributed to IHD, 231 (98–362) attributed to stroke, 94 (31–170) attributed to COPD, and 7 (1–12) attributed to LC for adults (age  $\geq 25$ 



years). Premature mortality due to ALRI from MSW and cow dung cake burning contributes an additional 1 (0–2) case (age  $\leq$  5 years) annually in Agra. For allcause mortality (i.e., ALRI, COPD, IHD, stroke and LC) attributable to PM<sub>2.5</sub> emissions from MSW and cow dung cake burning, the total human YLL is estimated at 10087 years (5480–14 520) from one year's exposure, where IHD (56%) is the highest contributor followed by stroke (32%), COPD (11%), and LC (1%).

#### Deposition and soiling of the Taj Mahal

The deposition of MSW and dung cake burning emissions to the Taj Mahal via dry and wet deposition was quantified using the simulated concentrations, along with observed size distributions and rainfall data. Detailed size distributions measured on-site showed the average surface area median diameter of the carbonaceous particles deposited to outdoor surfaces at the Taj Mahal to be  $\sim 1 \mu m$  [1], which was used in conjunction with deposition velocity relationships to derive a deposition velocity of 0.11 cm s<sup>-1</sup> [45]. Similar deposition velocities have been measured for particles of similar size and composition in previous studies in urban areas [46–49].

Estimated total annual combined  $PM_{2.5}$  dry deposition to the Taj Mahal is 150 (±130) mg m<sup>-2</sup> from open waste burning and 12 (±3.2) mg m<sup>-2</sup> from dung cake burning (table 2). The wet deposition loadings were small compared to dry deposition and detailed findings are available in SI section 3. While the mass loading of organic species, which contains light-absorbing brown carbon (BrC), is nearly eight times more than BC loading, BC is a strong light absorber [1, 50]. Emission factor measurements do not consider secondary formation, so this analysis is likely underestimating the total OM deposition from the two sources as both also have gaseous emissions [11, 32].

Additionally, the pollutant coverage of the Taj Mahal's surface was quantified to better gauge discoloration—if the fractional surface area coverage exceeds 1, its perceived color will likely be impacted. MSW burning emissions showed a fractional cover of 0.73 ( $\pm$ 0.67) while dung cake burning emissions contributed an additional 5.7 × 10<sup>-2</sup> ( $\pm$ 1.6 × 10<sup>-2</sup>) annually. Treatment cleanings have occurred four times since 1994. Given the time between cleanings, the influence of MSW and dung cake burning emissions is likely to exceed a fractional coverage of 1, suggesting their combined deposition will lead to surface discoloration.

### **Conclusions and implications**

Our model finds that open MSW-burning and dung cake burning led to estimated  $PM_{2.5}$  impacts of 4.3 and 0.34  $\mu$ g m<sup>-3</sup> (annually averaged) at the Taj Mahal, respectively, and up to 33 and 3.3  $\mu$ g m<sup>-3</sup> in Agra, with





**Figure 1.** Annual average fine particulate matter (PM<sub>2.5</sub>) concentrations in Agra from: (a), open MSW burning (b), dung cake burning. Modeled [PM<sub>2.5</sub>] at the Taj Mahal (depicted by the white star) was 4.3 ( $\pm$ 3.9)  $\mu$ g m<sup>-3</sup> from MSW emissions and 0.34 ( $\pm$ 9.1 × 10<sup>-2</sup>)  $\mu$ g m<sup>-3</sup> from dung cake burning emissions. These concentration profiles generated in AERMOD showed higher pollution from both forms of biomass burning concentrated in areas of lower socioeconomic status. Organic matter (OM) and black carbon (BC), the PM<sub>2.5</sub> components modeled, concentration profiles show the same spatial variation, but OM concentrations contribute more than BC to ambient PM<sub>2.5</sub> (SI figure 5).

**Table 2.** Comparison of the dry total organic matter (OM) and black carbon (BC) deposition (mg  $m^{-2}$ ) to the surface of the Taj Mahal from open MSW and dung cake burning in 2014.

	ОМ	BC	Total combined deposition
MSW	140 (±130)	$\begin{array}{c} 8.3(\pm 3.4)\\ 0.66(\pm 3.4\times10^{-2})\end{array}$	150 (±130)
DC	11.0 (±3.1)		12 (±3.2)

the highest levels in low SES neighborhoods. The increased OM and BC PM2.5 from those sources at the Taj Mahal lead to an increase of  $160 \text{ mg m}^{-2} \text{ yr}^{-1}$  of  $PM_{25}$  deposition to its surface, 150 mg m<sup>-2</sup> yr<sup>-1</sup> from open waste burning and 12 mg m<sup>-2</sup> yr<sup>-1</sup> from dung cake burning. The amount of PM2.5 deposited, along with the optical characteristics of the particles [1, 11, 13] lead to substantial soiling and discoloration of the Taj Mahal, and also reduced visibility, further degrading the aesthetic beauty of the site. A population, concentration-weighted exposure and health assessment finds that chronic exposure to MSW and dung burning related ambient PM2.5 was found to increase premature deaths by approximately 713 per year. While more difficult to quantify, acute exposures to the high PM<sub>2.5</sub> levels can have additional health impacts, e.g., to visitors.

Potential interventions can address the soiling of the Taj Mahal, degraded visibility, and human health in the area. In addition to improving ambient air quality, the recently promulgated ban on dung cake burning can improve indoor air quality, magnifying the estimated health benefits beyond those found based on improving ambient air quality alone. However, the benefits from its proposed implementation will be dependent upon more than 50 000 homes using cleaner sources for cooking [51, 52]. Better MSW management and prevention of garbage-burning in Agra were explored previously [53] but were not considered as high impact options to protect the Taj Mahal and public health. This paper indicates that preventing MSW burning can have a higher impact compared to the recently enacted dung cake burning ban on reducing  $PM_{2.5}$  concentrations affecting health and  $PM_{2.5}$ deposition that soils the Taj Mahal. Policies and action to reduce MSW burning should therefore be considered in the portfolio of actions to preserve the Taj and improve urban public health in Agra, particularly in low SES areas where people are disproportionately exposed to MSW and dung cake burning emissions.

Interventions leading to better waste management have not been a high priority in previous efforts to address air pollution in Indian cities. Agra Municipality has shown the initiative to implement policies designed to reduce soiling of the Taj Mahal, including limiting mobile source emissions near the landmark, banning polluting enterprises nearby, and prohibiting dung cake burning. Our results suggest that implementing a better waste management infrastructure [53] can be a high impact action that can improve ambient air quality in Agra, decrease soiling of the Taj Mahal and reduce adverse health outcomes.

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### Author contributions

AGR, MHB, AR, SNT planned the research. RML, LL developed the models applied in the study and quantified the surface deposition. ASN, AR, AGR developed the waste burning inventory methodology. ASN, RML conducted the on-site waste burn sampling, developed the emissions inventories, and performed the health impact assessments. MHB and SNT performed on-site detailed particle size measurements. RML, ASN, AGR wrote the manuscript.

#### References

- Bergin M H et al 2015 The discoloration of the Taj Mahal due to particulate carbon and dust deposition *Environ. Sci. Technol.* 49 808–12
- [2] Kumar A 2015 Two Hundred Sixty Second Report on Effects of Pollution on Taj, Report No. 262 Parliament of India Rajya Sabha, New Delhi
- [3] Nagpure A S, Ramaswami A and Russell A 2015 Characterizing the spatial and temporal patterns of open burning of municipal solid waste (MSW) in Indian cities *Environ. Sci. Technol.* 49 12904–12
- [4] Goyal P and Singh M P 1990 The long-term concentration of sulphur dioxide at Taj Mahal due to the Mathura Refinery *Atmos. Environ.* 24B 407–11
- [5] Raghavan N, Goyal P and Basu S 1983 A gaussian model for predicting SO<sub>2</sub> concentration in the city of Agra Atmos. Environ. 17 2199–203
- [6] Sharma R K and Gupta H O 1993 Dust pollution at the Taj Mahal—a case study Proc. Int. RILEM/UNESCO Congress (Paris: UNESCO Headquarters)
- [7] Hicks B B and Manju K 1987 Marble discoloration at the Taj Mahal: a proposed explanation ICOMOS 8th General Assembly and Int. Symp. (Washington, DC)
- [8] Cohen 2013 Global burden of disease 2010 Institute for Health Metrics and Evaluation (www.cseindia.org/userfiles/global\_ burden\_aaron.pdf)
- [9] Lelieveld J et al 2015 The contribution of outdoor air pollution sources to premature mortality on a global scale Nature 525 367–71
- [10] Gupta S, Mohan K, Prasad R, Gupta S and Kansal A 1998 Solid waste management in India: options and opportunities *Resour. Conservation Recycling* 24 137–54
- Shamjad P M et al 2015 Contribution of brown carbon to direct radiative forcing over the indo-gangetic plain Environm. Sci. Technol. 49 10474–81
- [12] Stockwell C E et al 2016 Nepal ambient monitoring and source testing experiment (NAMaSTE): emissions of trace gases and light-absorbing carbon from wood and dung cooking fires, garbage and crop residue burning, brick kilns, and other sources Atmos. Chem. Phys. Discuss. 16 11043–81
- [13] Vreeland H et al 2016 Chemical characterization and toxicity of particulate matter from roadside trash combustino in urban India Environ. Sci. Technol. 147 22–30



- [14] CPCB 2010 Air Quality Monitoring, Emission Inventory and Source Apportionment Study for Indian Cities (New Delhi, India: C.P.C. Board)
- [15] Allsopp M, Costner P and Johnston P 2001 Incineration and human health *Environ. Sci. Pollut. Res.* 8 141–5
- [16] McKay G 2002 Dioxin characterisation, formationand minimisation during municipal solid waste (MSW) incineration: review *Chem. Eng. J.* 86 343–68
- [17] Balakrishnan K et al 2010 IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Household Use of Solid Fuels and High-Temperature Frying vol 95, Agency for Research on Cancer
- [18] 2011 Census of India 2011: provisional population totals India data sheet Office of the Registrar General Census Commissioner, India
- [19] Das S K 2012 Energy Sources of Indian Households for Cooking and Lighting Report No. 542, National Sample Survey Office, Ministry of Statistics and Programme Implementation, Government of India
- [20] Sharholy M et al 2008 Municipal solid waste management in Indian cities—a review Waste Manag. 28 459–67
- [21] Guerrero L A, Maas G and Hogland W 2013 Solid waste management challenges for cities in developing countries *Waste Manag.* 33 220–32
- [22] Sujauddin M, Huda S M and Hoque A T 2008 Household solid waste characteristics and management in Chittagong, Bangladesh Waste Manag. 28 1688–95
- [23] Ramaswami A, Baidwan N K and Nagpure A S 2016 Exploring social and infrastructural factors affecting open burning of municipal solid waste (MSW) in Indian cities: a comparative case study of three neighborhoods of Delhi Waste Manag Res. accepted (doi:10.1177/0734242X16659924)
- Hoornweg D and Bhada-Tata P 2012 What a Waste: a Global Review of Solid Waste Management (Washington, DC: W. Bank)
- [25] Ramanathan V and Carmichael G 2008 Global and regional climate changes due to black carbon Nat. Geosci. 1 221–7
- [26] Tripathi S N 2005 Aerosol black carbon radiative forcing at an industrial city in northern India *Geophys. Res. Lett.* 32 L08802
- [27] Hamilton R S and Mansfield T A 1991 Airborne particulate elemental carbon: its sources, transport and contribution to dark smoke and soiling *Atmos. Environ.* 25A 715–23
- [28] AERMOD E 2004 *Description of Model Formulation* North Carolina, US Environmetal Protection Agency
- [29] Christian T J et al 2010 Trace gas and particle emissions from domestic and industrial biofuel use and garbage burning in central Mexico Atmos. Chem. Phys. 10 565–84
- [30] Saud T et al 2012 Emission estimates of organic and elemental carbon from household biomass fuel used over the Indo-Gangetic Plain (IGP), India Atmos. Environ. 61 212–20
- [31] Jayarathne T et al 2016 Nepal ambient monitoring and source testing experiment (NAMaSTE): emissions of particulate matter from wood and dung cooking fires, brick kilns, generators, trash and crop residue burning Atmos. Chem. Phys. in preparation
- [32] Pachauri T et al 2013 Characteristics and sources of carbonaceous aerosols in PM<sub>2.5</sub> during wintertime in Agra, India Aerosol. Air Qual. Res. 13 977–91
- [33] Russell L M 2003 Aerosol organic-mass-to-organic-carbon ratio measurements *Environ. Sci. Technol.* 37 2982–7
- [34] Burnett R T 2015 An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure *Environ. Health Perspect.* An Integrated Risk Function for Estimating the Global Burden of Disease Attributable to Ambient Fine Particulate Matter Exposure 122 397–403
- [35] Murray C J 1994 Quantifying the burden of disease: the technical basis for disability-adjusted life years *Bull. World Health Org.* 72 429–45
- [36] Seinfeld J H and Pandis S N 2006 Atmospheric Chemistry and Physics (New York: Wiley)



- [37] Ramaswami A, Milford J B and Small M J 2005 Integrated Environmental Modeling: Pollutant Transport, Fate, and Risk in the Environment (New York: Wiley)
- [38] Bond T C *et al* 2013 Bounding the role of black carbon in the climate system: a scientific assessment *J. Geophys. Res.* 118 5380–552
- [39] Turpin B J and Lim H-J 2001 Species contributions to PM<sub>2.5</sub> mass concentrations: revisiting common assumptions for estimating organic mass Aerosol Sci. Technol. 35 602–10
- [40] Wiedinmyer C, Yokelson R J and Gullett B K 2014 Global emissions of trace gases, particulate matter, and hazardous air pollutants from open burning of domestic waste *Environ. Sci. Technol.* 48 9523–30
- [41] Annepu R K 2012 Sustainable Solid Waste Management in India (New York: Columbia University)
- [42] Yedla S and Parikh J 2001 Economic evaluation of a landfill system with gas recovery for municipal solid waste management: a case study *Int. J. Environ. Pollut.* 15 433–47
- [43] Wate S R 2010 Air Quality Assessment, Emissions Inventory and Source Apportionment Studies: Mumbai National Environmental Engineering Research Institute, Central Pollution Control Board, New Delhi
- [44] Villalobos A M et al 2015 Source apportionment of carbonaceous fine particulate matter (PM<sub>2.5</sub>) in two contrasting cities across the Indo-Gangetic Plain Atmos. Pollut. Res. 6 398–405

- [45] Sehmel G A and Hodgson W H 1978 Model for Predicting Dry Deposition of Particles and Gases to Environmental Surfaces Battelle Pacific Northwest Labs., Richland, WA
- [46] Davidson C I et al 1985 Dry deposition of sulfate onto surrogate surfaces J. Geophys. Res. 90 2123–30
- [47] Mitchell R, Maher B A and Kinnersley R 2010 Rates of particulate pollution deposition onto leaf surfaces: temporal and inter-species magnetic analyses *Environ. Pollut.* 158 1472–8
- [48] Zufall M J et al 1998 Airborne concentrations and dry deposition fluxes of particulate species to surrogate surfaces deployed in southern lake michigan Environ. Sci. Technol. 32 1623–8
- [49] Thatcher T L and Layton D W 1995 Deposition, resuspension, and penetration of particles within a residence *Atmos. Environ.* 29 1487–97
- [50] Kirchstetter T W 2004 Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon *J. Geophys. Res.* 109 D21208
- [51] Viswanathan B and Kumar K S K 2005 Cooking fuel use patterns in India: 1983–2000 Energy Policy 33 1021–36
- [52] Goldemberg J et al 2004 A global clean cooking fuel initiative Energy Sustainable Dev. 8 5–12
- [53] 2006 Detailed Project Report for Solid Waste Management in Agra, Uttar Pradesh Regional Centre for Urban & Environmental Studies, Government of India