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# 1 The Discoloration of the Taj Mahal due to 2 Particulate Carbon and Dust Deposition

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14 Burning, Trash/Refuse Burning

15

**16 Abstract**

17 The white marble domes of the Taj Mahal are iconic images of India that attract millions of  
18 visitors every year. Over the past several decades the outer marble surfaces of the Taj Mahal  
19 have begun to discolor with time and must be painstakingly cleaned every several years.  
20 Although it has been generally believed that the discoloration is in some way linked with poor air  
21 quality in the Agra region, the specific components of air pollution responsible have yet to be  
22 identified. With this in mind, ambient particulate matter (PM) samples were collected over a one-  
23 year period and found to contain relatively high concentrations of light absorbing particles that  
24 could potentially discolor the Taj Mahal marble surfaces, that include black carbon (BC), light  
25 absorbing organic carbon (brown carbon, BrC), and dust. Analyses of particles deposited to  
26 marble surrogate surfaces at the Taj Mahal indicate that a large fraction of the outer Taj Mahal  
27 surfaces are covered with particles that contain both carbonaceous components and dust. We  
28 have developed a novel approach that estimates the impact of these deposited particles on the  
29 visible light surface reflectance, which is in turn used to estimate the perceived color by the  
30 human eye. Results indicate that deposited light absorbing dust and carbonaceous particles (both  
31 BC and BrC from the combustion of fossil fuels and biomass) are responsible for the surface  
32 discoloration of the Taj Mahal. Overall, the results suggest that the deposition of light absorbing  
33 particulate matter in regions of high aerosol loading are not only influencing cultural heritage but  
34 also the aesthetics of both natural and urban surfaces.

35

## 36 Introduction

37 On the timescale of several years the outer marble surfaces of the Taj Mahal become discolored  
38 and must be cleaned in a time consuming process (1). Figure 1 shows a cleaned section of a  
39 marble Mosque Dome at the Taj Mahal (where cleaning involves applying a layer of clay and  
40 removing the clay after it

41 dries, followed by rinsing  
42 the surface with clean  
43 water) directly next to an  
44 area being prepared for  
45 cleaning. An obvious  
46 contrast is seen between  
47 the clean, white marble  
48 surface and the darker  
49 discolored surface of the  
50 Marble Dome. Many  
51 measures have been



**FIGURE 1. A clean surface and an area being prepared for cleaning on a marble Mosque dome at the Taj Mahal**

52 undertaken to avoid the impact of local air pollution, including restricting traffic within 1 km of  
53 the grounds and limiting the emissions of industrial pollution in the city of Agra, where the Taj  
54 Mahal is located. While detailed scientific studies have not been reported in the literature, past  
55 efforts focusing on the discoloration have hypothesized that local air quality is responsible and  
56 suggestions have included surface reactions with gas-phase  $\text{SO}_2$ , as well as aqueous phase  
57 chemistry linked with the deposition of fog droplets, and water condensation (2), as well as dust

58 deposition (1). Despite efforts to keep the outer surfaces of the Taj Mahal white, it continues to  
59 become discolored with time, and the reason for the discoloration is not currently understood.  
60 Recent work has reported poor air quality throughout the Indo-Gangetic plain (3,4), including  
61 relatively high concentrations of particulate matter in Agra (5-7). Particulate matter in the region  
62 includes the light absorbing components black carbon (BC), light absorbing organic carbon (a  
63 fraction of which can absorb light preferentially in the UV region and is often termed brown  
64 carbon, BrC), and dust (8). Both organic carbon and dust have the potential to preferentially  
65 absorb solar light in the blue region of the spectrum, which can give the atmosphere a brown hue  
66 and has thus been dubbed the Atmospheric Brown Cloud (9). The presence of these light  
67 absorbing aerosols, and in particular those that can take on a dark hue against a light colored  
68 background, suggest that the deposition of ambient particulate matter may be playing a role in  
69 the discoloration of the outer white marble surfaces of the Taj Mahal.

70

## 71 **Experimental Methods**

72

73 **Ambient Particulate Sampling and Analyses.** In order to determine the influence of PM on the  
74 Taj Mahal, ambient aerosol sampling was conducted for a roughly one-year period beginning on  
75 Nov. 5, 2011 and continuing through June, 2012 just prior to the monsoon season. Filters were  
76 collected every 6<sup>th</sup> day each month for both PM<sub>2.5</sub> (fine particulate matter having diameters less  
77 than 2.5  $\mu\text{m}$ ) and total suspended particulate matter (TSP), and analyzed for major anions,  
78 organic (OC) and elemental carbon (EC), and trace elements. The PM<sub>2.5</sub> cut-point was  
79 established using an upstream cyclone, while the TSP directly sampled ambient air. Portions of  
80 each filter were combined to make monthly composites that were extracted and analyzed for  
81 source specific trace organic compounds using GCMS. The trace organic concentrations were

82 used to estimate source contributions to particulate organic carbon using chemical mass balance  
83 (CMB) modeling (10). Additional information on the sampling and chemical analyses is  
84 presented in the Supplementary Material section (sections S1 and S2).

85  
86 **Marble Deposition Target Sampling and Analyses.** In addition to ambient samples, several  
87 pre-cleaned marble deposition targets (with dimensions 2 cm x 2 cm x 0.5 cm) were placed  
88 outdoors within roughly 300 m of the main Taj Mahal dome. Both the air sampling equipment  
89 and targets were located in a section of the Taj Mahal that was accessible only to staff of the  
90 Archaeological Survey of India (ASI), and had very little foot traffic. Pre-cleaned marble  
91 cuboids were fastened to Taj Mahal structures with double-sided tape at a variety of locations,  
92 and exposed from April to June 2012. Some of the marble samples were placed horizontally and  
93 others vertically. Prior to, and after exposure, the marble samples were placed in sealed, pre-  
94 cleaned petri dishes and stored in a freezer to avoid degradation of deposited particles.

95 Scanning Electron Microscopy (SEM) (LEO 1530, Carl Zeiss Microscopy) and Energy  
96 Dispersive X-ray (EDX) Spectroscopy (Oxford Instruments X<sub>max</sub> detector) were carried out on  
97 two horizontally facing marble targets. Images were taken at many different magnifications to  
98 capture the particles having sizes ranging from 100 nm to 100 μm. The particle sizes and shapes  
99 were accessed through SEM images using image processing in a Matlab program. EDX analyses  
100 were carried out on the same marble targets on ~ 1000 particles. The information gained from the  
101 SEM/EDX analyses allowed for the estimation of the particle number and surface area  
102 concentration, and chemical composition as a function of area of the marble target. This  
103 information was then used, as described in the next sections, to estimate the change in color of

104 the marble surface. More information on the SEM/EDX analyses is given in the Supplementary  
 105 Material Sections S1 and S2.

106

107 **Linking Deposited Particles to Marble Surface Color.** In order to estimate the impact of  
 108 particles deposited to the marble substrate on the perceived color change of the surface, we  
 109 developed an approach that estimates the influence of deposited particles on wavelength-  
 110 dependent surface reflectance. The method builds on previous work that estimated the influence  
 111 of particles deposited to plant leaves on available photosynthetically active radiation (11). First,  
 112 we use SEM/EDX analyses of particles deposited to the marble targets to estimate the optical  
 113 depth of deposited particles as a function of wavelength ( $\tau_\lambda$ ) as follows:

$$114 \quad \tau_\lambda = \frac{\pi}{4} \sum_{i=1}^n A c_i D_{p,i}^2 [Q_{s\lambda} + Q_{a\lambda}]_i \quad (1)$$

115 Where  $A c$  is the areal particle number concentration (number of particles deposited per area of  
 116 the marble surface) for each size bin,  $i$ ,  $D_p$  is the particle diameter for deposited particles, and  
 117  $Q_{s\lambda}$  and  $Q_{a\lambda}$  are the wavelength dependent Mie scattering and absorption efficiencies that are  
 118 determined based on particle size and composition.

119

120 After estimating the optical depth, the wavelength dependent single scattering albedo,  $\omega_\lambda$  (ratio  
 121 of light scattering to extinction) is estimated as:

$$122 \quad \omega_\lambda = \frac{\sum_{i=1}^n A c_i D_{p,i}^2 Q_{s\lambda,i}}{\sum_{i=1}^n A c_i D_{p,i}^2 [Q_{s\lambda} + Q_{a\lambda}]_i} \quad (2)$$



123 The single scattering albedo is a key parameter that determines the relative amount of light  
124 absorption that occurs over the white marble surface. For white, scattering only particles the  
125 single scattering albedo is near 1.0 and the surface reflectance of a white surface will not change.

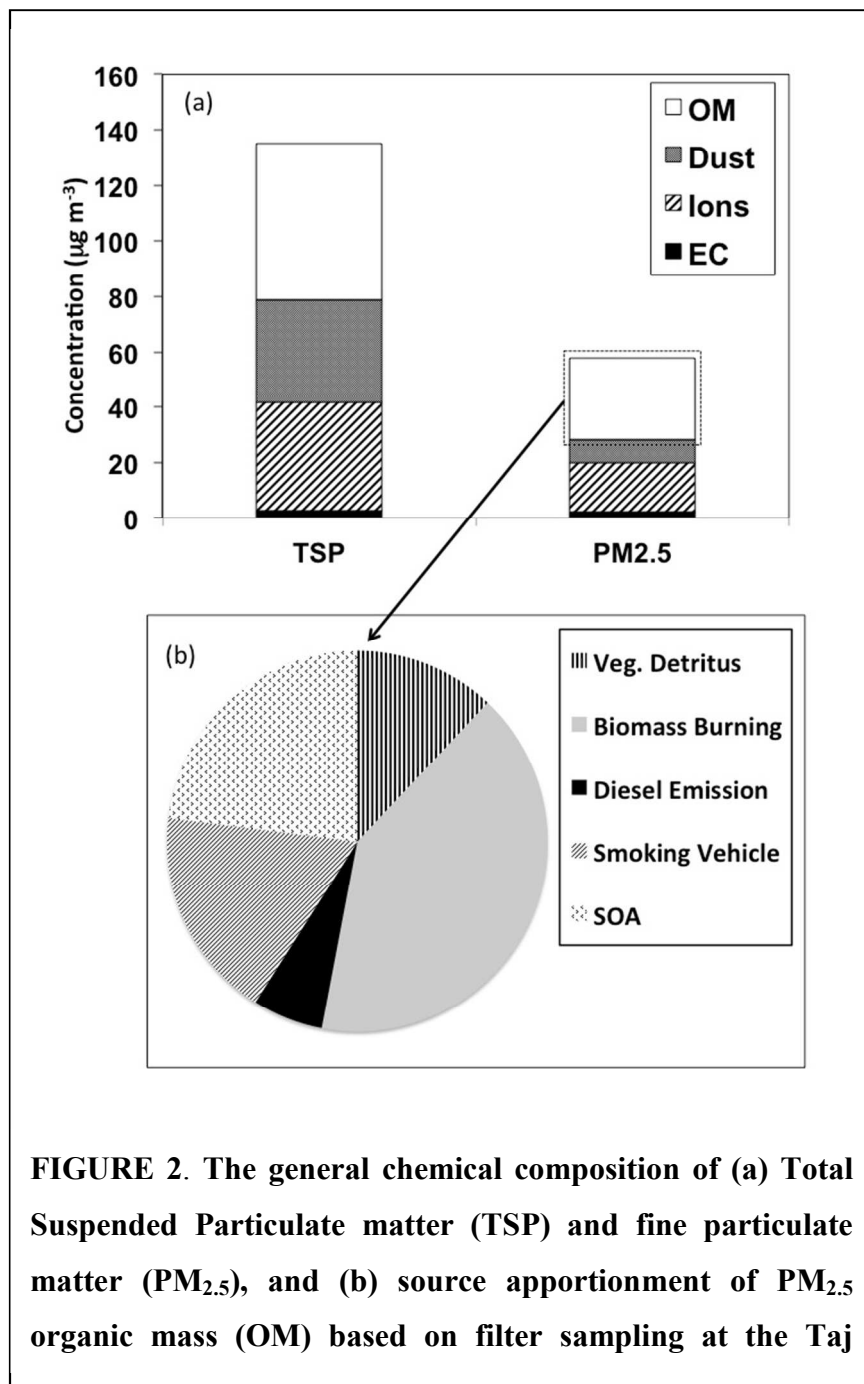
126  
127 The change in the surface reflectance of the white marble surface is calculated using SBDART, a  
128 radiative transfer model (12) with input values including  $\tau_\lambda$  and  $\omega_\lambda$  estimated from equations 1,  
129 and 2 as well as the asymmetry parameter (relative amount of light scattering in the forward  
130 direction) as a function of wavelength using Mie theory (13).

131  
132 In order to determine the perceived color change of the white marble surface based on particle  
133 deposition, and the related change in spectral surface reflectance we used the model described by  
134 D'Andrade and Romney (14) to convert spectral reflectance to perceived color in the Munsell  
135 color system. The Munsell color system is based on three components that include the value  
136 (lightness/darkness), hue (color), and chroma (purity/saturation). The model used to estimate the  
137 perceived color of the marble surface with deposited particles uses the spectral reflectance from  
138 the radiative transfer model of the marble surface loaded with particles to estimate the Munsell  
139 color. The Munsell color estimate also takes into consideration the human eye response as a  
140 function of wavelength of incident light (the approach is described in more detail in the  
141 supplementary information, section S3).

142

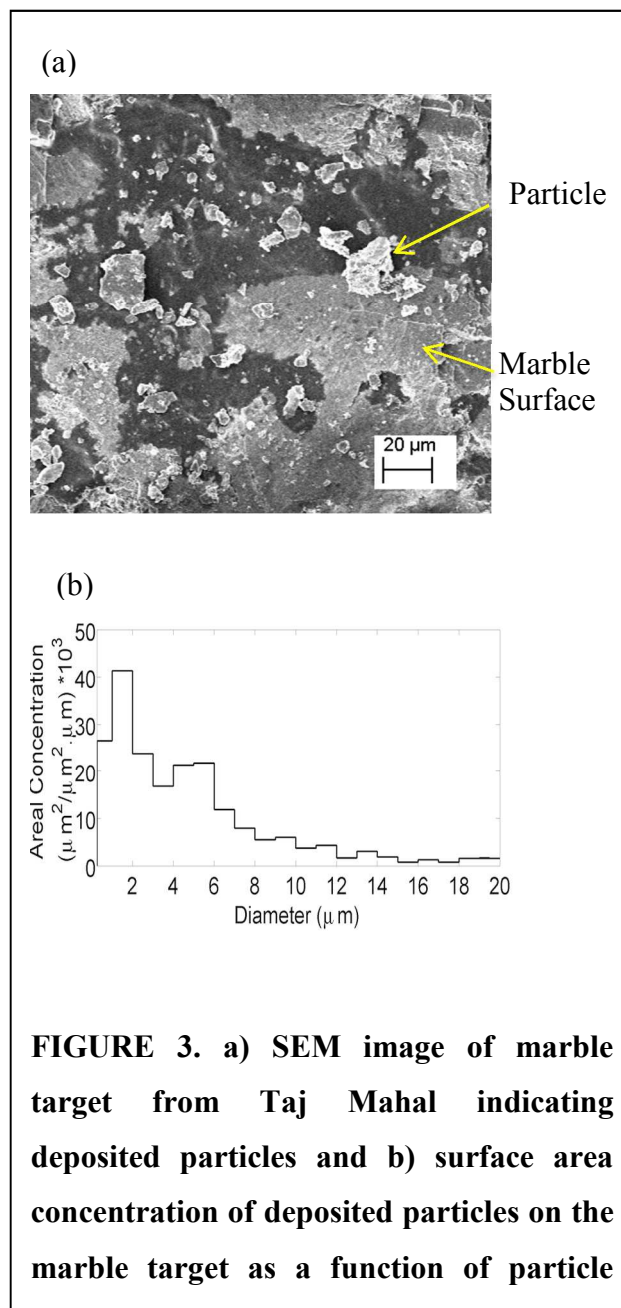
143 **Results and Discussion**

144 **Ambient Particulate Concentrations.** Figure 2a shows the average mass concentrations of  
 145 particulate organic carbon mass (OM), ions, dust and elemental carbon (EC) for both TSP and  
 146 PM<sub>2.5</sub> over the sampling  
 147 period. The mean daily  
 148 concentrations (and standard  
 149 deviations) of both TSP and  
 150 PM<sub>2.5</sub> are 135 (55) and 60  
 151 (39)  $\mu\text{g m}^{-3}$ . The values are  
 152 significantly higher than the  
 153 annual World Health  
 154 Organization (WHO) PM  
 155 guidelines for PM<sub>10</sub> and  
 156 PM<sub>2.5</sub> of 20  $\mu\text{g m}^{-3}$  and 10  
 157  $\mu\text{g m}^{-3}$  highlighting the poor  
 158 air quality in the region. The  
 159 fraction of particulate matter  
 160 greater than 2.5  $\mu\text{m}$  is ~60%,  
 161 and is due in large part to the  
 162 coarse mode dust that  
 163 increases from 15% of the  
 164 PM<sub>2.5</sub> mass fraction to 30%



**FIGURE 2.** The general chemical composition of (a) Total Suspended Particulate matter (TSP) and fine particulate matter (PM<sub>2.5</sub>), and (b) source apportionment of PM<sub>2.5</sub> organic mass (OM) based on filter sampling at the Taj

165 of the TSP mass. In addition to dust, other  
166 PM components that absorb light in the  
167 visible spectrum, and hence have the  
168 possibility to influence the color of the outer  
169 white marble surfaces, are elemental carbon  
170 (EC) that is responsible for 2% of the TSP  
171 mass, as well as OM that accounts for 39% of  
172 the TSP mass. Estimates of the sources of  
173 OM in the  $PM_{2.5}$  mass fraction shown in  
174 Figure 2b indicate that biomass burning, a  
175 known source of BrC, is responsible for  
176 roughly half of the OM with significant  
177 contributions from vehicular emissions. It  
178 should be pointed out that biomass burning  
179 OM can be from a variety of activities  
180 including the combustion of wood and dung,  
181 crop residue, and the burning of trash and  
182 refuse that is ubiquitous in the region. The  
183 prevalence of light absorbing aerosols in  
184 Agra (i.e. elemental and organic carbon, and dust) suggests that PM deposition to the white  
185 marble surfaces may be responsible for the observed discoloration of the outer Taj Mahal  
186 structures including the famous Taj Mahal dome.  
187



188 **Size and Composition of Particles Deposited to Marble Targets.** Figure 3a shows a Scanning  
189 Electron Microscopy (SEM) image of a marble deposition target that was placed horizontally at  
190 the Taj Mahal, and was exposed for a roughly two month period during the pre-monsoon season  
191 of 2012. The surface area concentration (surface area of particles per unit marble surface area per  
192 micron) distribution of particles as a function of particle size deposited to the marble target  
193 (Figure 3b) shows peaks in particle size at roughly 1-2  $\mu\text{m}$ , and an additional mode at 4-5  $\mu\text{m}$ .  
194 The fraction of the surface covered by particles is estimated to be  $\sim 30\%$ . Approximately 70% of  
195 the deposited particle surface area is for particles having diameters greater than 2  $\mu\text{m}$ , indicating  
196 that a large fraction of the particle surface area concentration is due to the deposition of coarse  
197 particles. The dominance of coarse particles is due to both the relatively high concentration of  
198 dust particles measured in Agra, as well as the fact that the dry deposition velocity of coarse ( $\sim 5$   
199  $\mu\text{m}$ ) particles is roughly 100 times greater than that of accumulation mode ( $\sim 1.0$   $\mu\text{m}$ ) aerosol  
200 particles (15). It is likely that dry deposition is the dominant mode of particle transport to the Taj  
201 surface given the relatively low amount of precipitation in Agra during the Fall through Spring,  
202 when particle loadings are high and the summer monsoon rainfall is not occurring. It is also  
203 important to note that water insoluble particles (such as dust, BC, and a fraction of OM) are  
204 likely not easily removed from the Taj Mahal surface by precipitation wash-off once deposited.  
205 This is based on similar observations of the build-up of water insoluble particles on leaf surfaces  
206 observed in the Yangtze delta region of China, an area that also experiences high PM loadings  
207 (11).  
208  
209 The EDX analyses indicate that more than 70% of particles are primarily crustal in origin, with  
210 spectra dominated by crustal elements. The crustal particles were typically in the coarse (having

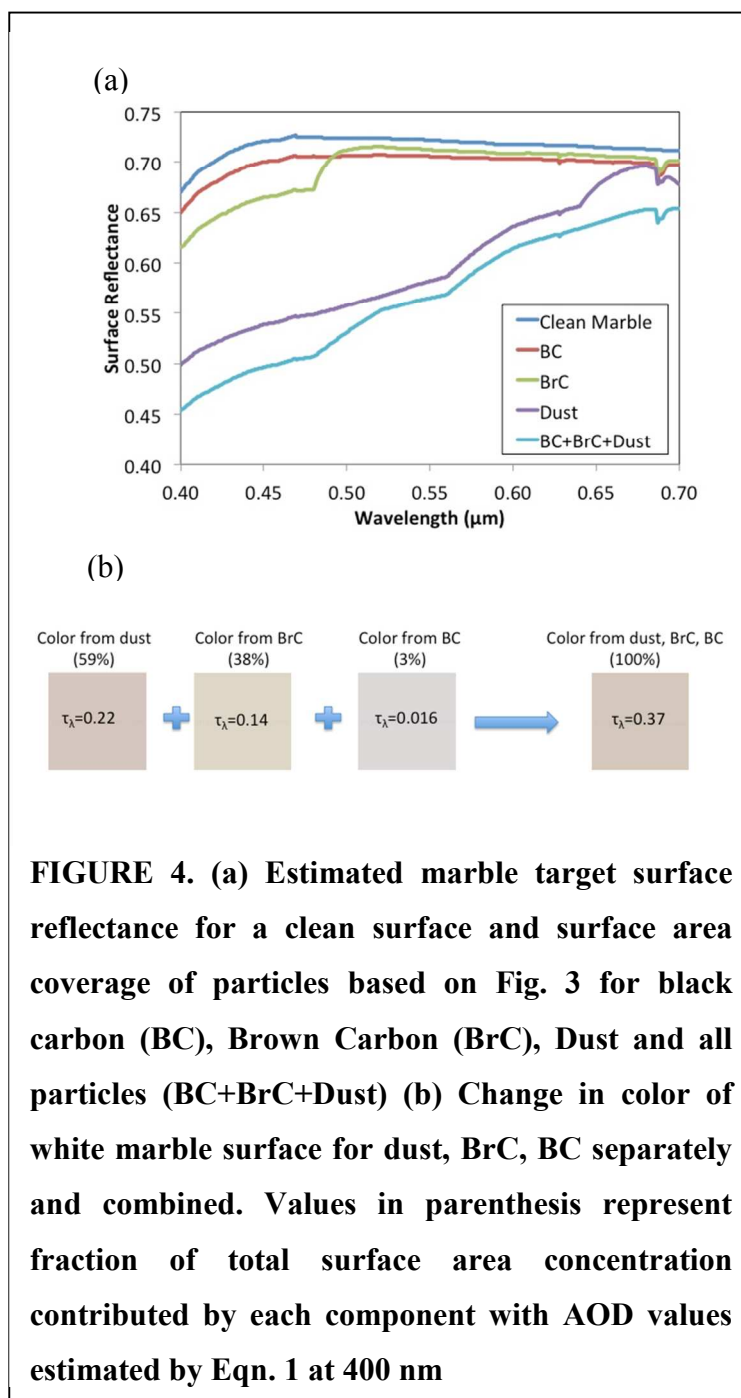
211 diameters greater than  $\sim 2\text{-}3\ \mu\text{m}$ ) size fraction. Particles having diameters less than  $2\ \mu\text{m}$ , which  
212 account for roughly 30% of the deposited particle surface area, also contained significant  
213 amounts of carbon likely from the sources of OM highlighted in CMB results in Figure 2b. More  
214 detailed information on the EDX analyses, including example spectra of particle EDX analyses  
215 are given in the supplementary section (S2).

216

217 **Deposited Particles and Perceived Color.** To estimate  $\tau_\lambda$  we assume that particles having  
218 diameters less than  $3\ \mu\text{m}$  are composed primarily of light absorbing organic carbon (BrC) with  
219 wavelength dependent refractive indices reported by Liu et al. (16). This assumption is based on  
220 the fact that ambient filters indicated that roughly half of the  $\text{PM}_{2.5}$  was carbonaceous in nature,  
221 combined with the fact that we did not observe the presence of major ion related elements (i.e. S)  
222 deposited to the marble targets, but did see a dominance of carbon particles in the less than  $3\ \mu\text{m}$   
223 particle sizes. We also assume that 10% of the particles less than  $3\ \mu\text{m}$  are black carbon (BC)  
224 particles with a refractive index commonly used for soot particles (13). It is likely that we  
225 overestimate the influence light absorption by BrC and BC since we assume that all of the  
226 particles less than  $3\ \mu\text{m}$  are carbonaceous and not elemental or ionic in nature. Aerosol optical  
227 depth contributions at 400 nm for dust, BrC and BC are estimated to be 0.222, 0.144 and 0.016  
228 respectively, highlighting the importance of light extinction by all three components. The single  
229 scattering albedo at 400 nm and 700 nm is estimated to be 0.64 and 0.95, indicating that a  
230 significant amount of light absorption occurs at near-ultraviolet wavelengths preferentially to  
231 that at the longer 700 nm wavelength.

232

233 Figure 4a shows the surface reflectance of a pure marble surface as well as the estimated surface  
 234 reflectance for several cases including the influence from each light absorbing particulate  
 235 component separately (BC, BrC and  
 236 dust), and the case when all  
 237 components are combined. As seen  
 238 in the plot, BC absorbs across all  
 239 wavelengths evenly, with brown  
 240 carbon showing preferential  
 241 absorption at shorter wavelengths  
 242 near 400 nm. Dust substantially  
 243 reduces the surface reflectance at all  
 244 wavelengths, and preferentially so at  
 245 shorter wavelengths due to the  
 246 presence of hematite, which absorbs  
 247 at blue wavelengths. When  
 248 combined, dust, BrC, and BC are  
 249 estimated to substantially alter the  
 250 surface reflectance with enhanced  
 251 absorption (i.e. lower values of  
 252 surface reflectance) at shorter  
 253 wavelengths.



**FIGURE 4. (a) Estimated marble target surface reflectance for a clean surface and surface area coverage of particles based on Fig. 3 for black carbon (BC), Brown Carbon (BrC), Dust and all particles (BC+BrC+Dust) (b) Change in color of white marble surface for dust, BrC, BC separately and combined. Values in parenthesis represent fraction of total surface area concentration contributed by each component with AOD values estimated by Eqn. 1 at 400 nm**

255 Figure 4b shows the estimated change in color of the white surface for deposition of the light  
256 absorbing particles measured in Figure 3b over the two-month period the targets were exposed.  
257 Results indicate that separately each component contributes to the color change of the Taj Mahal  
258 white marble surfaces. For BC alone (which we estimate to account for 3% of the total particle  
259 surface area) the color change results in a greyish color given that the change in surface  
260 reflectance is proportionally similar at each wavelength. Both BrC (~30% of the total particle  
261 surface area) and dust influence color with preferential absorption in the UV resulting in  
262 yellowish-brown hues. When combined, the perceived color of the surface shifts towards darker  
263 shades of yellow-brown. It should be pointed out that our sample targets were mounted for a  
264 relatively short time (~2 months) compared to the typical time between cleanings of the outer Taj  
265 Mahal surfaces (several years), and therefore it may be expected that the perceived color of the  
266 marble target would be attenuated compared to that of the white surfaces of the Taj Mahal.  
267 Indeed the marble target surface did appear somewhat lighter in color as compared to the color  
268 estimates in Figure 4b, although qualitatively were similar. There are several uncertainties in  
269 estimating perceived color including the loading, particle size, and optical properties. Analyses  
270 (included in section S3) suggest that results are moderately sensitive to both particle loading and  
271 size. For example, assuming an uncertainty of 50% for aerosol loading only moderately  
272 influences the perceived color and does not change the conclusions that both dust and  
273 carbonaceous particles contribute to the perceived color change of the Taj Mahal. Overall, the  
274 results indicate that light absorbing particles play an important role in the discoloration of the Taj  
275 Mahal surface and that dust, as well as BC and BrC that are primarily from biomass combustion,  
276 trash/refuse burning, and mobile sources, all make significant contributions to the discoloration.

277 This work further suggests that the deposition of light absorbing particulate matter to both  
278 natural and human-made surfaces results in a substantial discoloration in regions of high aerosol  
279 loading. The discoloration impacts not only cultural artifacts but also the aesthetics of the  
280 environment through the modification of surface albedo, and hence perceived color. The  
281 measurement/modeling approach developed in this paper allows surface color changes to be  
282 estimated based on the relative amounts of light absorbing particles deposited to surfaces, and  
283 can be used to develop future control strategies to prevent the discoloration of the environment  
284 by particle deposition, that will also improve air quality.

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289 Supporting information is associated with this paper and includes details related to filter and  
290 marble surrogate surface sampling (S1), chemical analyses on both the filters and marble  
291 surrogate surfaces (S2), as well as the approach to estimate perceived color based on surface  
292 reflectance (S3). The information is available free of charge via the internet at  
293 <http://pubs.acs.org>.

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