Radiative absorption enhancement of dust mixed with anthropogenic pollution over East Asia

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ABSTRACT: The particle mixing state plays a significant yet poorly quantified role in aerosol radiative forcing, especially for the mixing of dust (mineral absorbing) and anthropogenic pollution (black carbon absorbing) over East Asia. We have investigated the absorption enhancement of mixed-type aerosols over East Asia by using the Aerosol Robotic Network observations and radiative transfer model calculations. The mixed-type aerosols exhibit significantly enhanced absorbing ability than the corresponding unmixed dust and anthropogenic aerosols, as revealed in the spectral behavior of absorbing aerosol optical depth, single scattering albedo, and imaginary refractive index. The aerosol radiative efficiencies for the dust, mixed-type, and anthropogenic aerosols are -101.0, -112.9 and -98.3 $Wm^{-2}\tau^{-1}$ at the bottom of the atmosphere (BOA), -42.3, -22.5 and -39.8 $Wm^{-2}\tau^{-1}$ at the top of the atmosphere (TOA), and 58.7, 90.3 and 58.5 $Wm^{-2}\tau^{-1}$ in the atmosphere (ATM), respectively. The BOA cooling and ATM heating efficiencies of the mixed-type aerosols are significantly higher than those of the unmixed aerosol types over the East Asia region, resulting in atmospheric stabilization. In addition, the mixed-type aerosols correspond to a lower TOA cooling efficiency, indicating that the cooling effect by the corresponding individual aerosol components is partially counteracted. We conclude that the interaction between dust and anthropogenic pollution not only represents a viable aerosol formation pathway but also results in unfavorable dispersion conditions, both exacerbating the regional air pollution in East Asia. Our results highlight the necessity to accurately account for the mixing state of aerosols in atmospheric models over East Asia, in order to better understand the formation mechanism for regional air pollution and to assess its impacts on human health, weather, and climate.

1 Introduction

Atmospheric aerosols or particulate matter (PM) profoundly affect the energy budget of the earth-atmosphere system directly by interfering with the radiative transfer and indirectly by modifying cloud formation (Twomey, 1977; Charlson and Schwartz, 1992; Fan et al., 2007; Wang et al., 2011). However, the assessment of the aerosol radiative effects is limited because of the inherent difficulties associated with observations and model simulations (Stevens and Bony, 2013; Kok et al., 2017). In particular, an accurate quantification of the mixing state of absorbing aerosols poses a great challenge in the estimation of the aerosol direct radiative forcing (Haywood and Boucher, 2000; He et al., 2015). Currently, the assessment of the aerosol direct and indirect radiative forcing represents large uncertainty in the prediction of future climate by anthropogenic activities (IPCC, 2007; IPCC, 2013). Moreover, aerosol mixing state significantly affects atmospheric dynamics (Ramanathan and Carmichael, 2008).

Atmospheric aerosols are typically internally and/or externally mixed during their lifetimes (Jacobson, 2001; Zhang and Zhang, 2005; Zhang et al., 2008; Khalizov et al., 2009a; Pagels et al., 2009; Taylor et al., 2015). The East Asian region is experiencing persistent heavy air pollution conditions in the present day (Guo et al., 2014; Zhang et al., 2015; Wang et al., 2016). Black carbon (BC) is one of the major anthropogenic pollutants in this region that exerts significant environmental and climatic effects because of the strong absorption of solar radiation (Wang et al., 2012; Peng et al., 2016), which is further enhanced by the "Lensing effect" (e.g., Jacobson, 2001; Cui et al., 2016). East Asia is also the largest dust source region second to the Saharan Desert (Huang et al., 2014; Huang et al., 2015; Tian et al., 2015). As a result, coarse mode dust particles are frequently mixed with anthropogenic pollution along

69   their transport pathway in East Asia (Noh et al., 2012; Logan et al., 2013; Guo et al.,

70   2017; Hara et al., 2017). The potential anthropogenic influence on dust has been

71   investigated close to the dust source regions (Huang et al., 2010; Bi et al., 2017).

72   Lower single scattering albedo (SSA) of mixed dust plumes has been assessed in

73   previous studies (Kim et al., 2005; Khatri et al., 2014). Li et al. (2015) have studied

74   the SSA spectral curvature of the East Asian aerosol mixtures using Aerosol Robotic

75   Network (AERONET) products and model simulations. The observations from a

76   sun–sky radiometer and a lidar have been applied to identify the presence of Asian

77   dust in mixed aerosol plumes at several East Asian monitoring sites (Noh et al., 2017).

78   Those earlier studies on optical properties and radiative effects of the East Asian dust

79   and anthropogenic aerosol mixtures have promised reduction of the uncertainties in

80   estimating the aerosol radiative effects.

81   Internal mixing of coarse mode dust with fine mode anthropogenic aerosols has

82   been suggested by observations in the Asian Aerosol Characterization Experiment

83   (ACE-Asia) (Seinfeld et al., 2004) and recent studies (Sugimoto et al., 2015; Wang et

84   al., 2017), although the mechanism leading to the mixing has yet to be elucidated.

85   Internal mixing of dust with anthropogenic pollution likely occurs via condensation of

86   low-volatility organic and inorganic compounds, particle-phase reactions, and

87   coagulation with other aerosol types (Zhang et al., 1996; Zhao et al., 2006; Qiu et al.,

88   2011). In addition, dust particles provide reactive surfaces for catalytic conversion of

89   sulfur dioxide to sulfate, which has been suggested as a key mechanism for severe

90   haze formation in China (Zhang et al., 2015; Li et al., 2017a). Atmospheric

91   measurements using electron microscopy have identified BC and certain soluble

92   aerosols on the surface of dust particles (Tobo et al., 2010; Ma et al., 2012; Li et al.,

93   2014). Pan et al. (2017) have studied the morphology change of East Asian dust

94   mixed with anthropogenic aerosols and showed the possibility evidence for the

95   occurrence of aqueous-phase reactions.

96   The aerosol mixing state significantly affects the radiative effects (Jacobson,

97   2001; Khalizov et al., 2009b; Xue et al., 2009). Several previous studies have shown

98   that the amount of solar radiation reaching the Earth surface through mixtures of

99   mineral dust and other absorbing aerosols is considerably reduced compared to that

100   through dust-only aerosols (Derimian et al. 2008; Obregón et al. 2015). Researchers

101   have reported that the radiative efficiency of non-dust aerosols is higher than that of

102   dust aerosols at two urban Asian cities of Gwangju and Beijing (Noh et al., 2012; Yu

103   et al., 2016). Maximum radiative efficiency under unpolluted conditions has been

104   found by comparing aerosol radiative effects under pristine and polluted conditions

105   (Chen et al., 2016). Aerosol radiative efficiency has been found to be strongly

106   influenced by aerosol absorbing ability (e.g., SSA) and size of fine mode particles in

107   Central China (Zhang et al., 2017).

108   Although the mixing state of dust and anthropogenic aerosols considerably

109   affects aerosol radiative effects, further studies are urgently demanded to better

110   understand the key role that the East Asian aerosol mixtures play in the formation

111   mechanism of regional air pollution. In this present work, we have extensively

112   investigated the radiative absorption enhancement by the East Asian aerosol mixtures

113   on the basis of long-term AERONET observations and SBDART model simulations.

114   We classified the dust, mixed-type, and anthropogenic aerosols according to the

115   aerosol SSA spectral behavior and Ångström exponent parameter. The optical and

116   microphysical properties of the various aerosol types were analyzed, with emphasis

117   on the absorption enhancement by mixed-type aerosols. The mechanism leading to the

118   radiative absorption enhancement by the mixed-type aerosols and their impacts on

119 regional air pollution and climate have been discussed. Our results suggest that the

120 East Asian aerosol mixtures result in a more stable atmosphere that is unfavorable for

121 diffusion and dispersion of the atmospheric pollutants.

122 **2 Data and methodology**

123 **2.1 AERONET data**

124 The aerosol optical and microphysical data used in this research were originally

125 from the Aerosol Robotic Network (AERONET) (Holben et al., 1998). To ensure the

126 data quality, we only analyzed the cloud-screened, quality-assured Level 2.0 inversion

127 data. We further constrained the data with solar zenith angle between $50\,°$ and $80\,°$ to

128 avoid possible inversion errors and surface albedo smaller than 0.5 to exclude

129 seasonal snow-covered surfaces. The AERONET spectral products are available at the

130 wavelengths of 440, 675, 870, and 1020 nm, respectively. Previously, Dubovik et al.

131 (2000) have evaluated the uncertainty of the AERONET products.

132 All available worldwide measurement sites from the Aerosol Robotic Network

133 (AERONET) program with a sample number of greater than 100 were included in the

134 present work. Considering the regional representativeness and data availability, 11

135 sites (Table S1 and Fig. S1) were selected to represent the various types of the East

136 Asian aerosol mixtures. For example, the sample numbers of Beijing and Xianghe

137 were randomly reduced to a one-fourth of the total numbers because these two sites

138 are close to each other and the total sample numbers of the two sites are significantly

139 larger than the others.

140 **2.2 Radiative forcing and efficiency calculations**

141 The aerosol direct radiative forcing (ΔF) is defined as follows:

142 $$\Delta F = (F^{\downarrow} - F^{\uparrow}) - (F_0^{\downarrow} - F_0^{\uparrow}) \tag{1}$$

143 where F and F_0 are radiative fluxes under the aerosol-free and aerosol-laden

conditions respectively, the upward and downward arrows denote the directions of the radiative fluxes. The AERONET products include the aerosol radiative forcing and efficiency, in addition to the optical and microphysical parameters. However, the radiative forcing in the AERONET products was not exactly calculated using Equation (1). Instead, we calculated the aerosol direct radiative forcing and efficiency by using the widely adopted Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998). The aerosol radiative efficiency (ΔF^{eff}) is defined as the aerosol direct radiative forcing per unit aerosol optical depth (AOD):

$$\Delta F^{eff} = \Delta F / AOD_{0.55} \tag{2}$$

where $AOD_{0.55}$ is the 550 nm AOD. However, the aerosol direct radiative forcing is not linearly dependent on AOD (e.g., Wu et al., 2015). To exclude possible errors from aerosol loading in calculating the aerosol radiative efficiency in Equation (2), the 550 nm AOD was set to unity ($AOD_{0.55}$=1.0) in SBDART model calculations. The data processing procedure is presented in Fig. 1 and a detailed discussion of aerosol radiative forcing and efficiency calculation has been provided elsewhere (Tian et al., 2018).

2.3   Aerosol classification

The SSA spectral behavior and the Ångström exponent parameter were applied to identify aerosol mixtures (Fig. 1). The main absorbing component of the anthropogenic pollutants is BC, which exhibits strong absorption throughout a broad wavelength range (Bond and Bergstrom, 2006). Dust aerosols are also an absorbing medium because of the iron-bearing minerals such as hematite, goethite and magnetite, which enable the dust aerosols to absorb strongly in the UV and short visible wavelengths and weakly in the near infrared (Schuster et al., 2016). Thus, the dust aerosols exhibit a monotonically increasing SSA trend with increasing wavelength,

169   while the anthropogenic pollutants (the absorption of which is mainly contributed by

170   BC) show a monotonically decreasing SSA spectra (Bergstrom et al., 2007; Giles et

171   al., 2012; Tian et al., 2017). The absorption of aerosol mixtures is contributed by both

172   dust and BC, leading to a non-monotonic SSA spectra (Khatri et al., 2014). The

173   characteristic non-monotonic SSA spectral behavior of mixed-type aerosols provides

174   a useful approach to identify the mixed-type aerosols. SSA curvature of greater than

175   0.1, which has been suggested for "moderate mixing" (Li et al., 2015), was applied

176   for the mixed-type aerosol. We employed an additional criterion, Ångström exponent,

177   which is related to the aerosol size and the widely used in aerosol classification (Eck

178   et al., 2010; Derimian et al., 2016), to further constrain the aerosol classification.

179   Results show that Ångström exponent values are smaller for coarse mode dust

180   aerosols (lower than 0.7 in the present study), larger for fine mode anthropogenic

181   aerosols (greater than 1.4 in this research), and range from 0.8 and 1.3 for the

182   mixed-type aerosols, respectively.

183   The imaginary refractive index at 440 nm (k_{440}, in the visible bands) versus the

184   average of the imaginary refractive indices at 675, 870, and 1020 nm (k_{rnir}, in the red

185   and near-infrared bands) is shown in Fig. 2. Anthropogenic aerosols with the

186   absorption mainly contributed by BC particles exhibit a flat imaginary refractive

187   index (Bond and Bergstrom, 2006), while dust with the components such as hematite

188   absorbs strongly in UV but weakly in longer wavelengths (Hsu and Matijević, 1985;

189   Schuster et al., 2016). Hence, the data of anthropogenic aerosols are scattered along

190   the 1:1 line (Fig. 2). Dust aerosols show stronger absorption at the visible wavelength

191   than at the red and near-infrared wavelengths, so the data are scattered above the 1:1

192   line. The data for most of the mixed-type aerosols lie above the 1:1 line and on the

193   right side of the $k_{rnir} = 0.0042$ threshold, which is suggested by Schuster et al. (2016)

194 to separate dust ($k_{mir} < 0.0042$) and biomass burning ($k_{mir} > 0.0042$) aerosols. The

195 mixed-type aerosols show the strongest absorption and most of the sites with a

196 mixed-type aerosol sample number of 50 and higher are located in East Asia. Hence,

197 our method quantitatively separated various aerosol types.

3   Spectral behavior of the East Asian aerosol mixtures

199   Extensive investigations of the spectral optical properties were carried out to

200 discuss the enhanced absorbing ability of the East Asian aerosol mixtures. Dust

201 aerosols show the highest spectral AOD and the anthropogenic aerosols exhibit the

202 lowest spectral AOD (Fig. 3a). The spectral dependence of dust aerosols is nearly

203 invariant across the wavelength spectrum while the dependence of anthropogenic

204 aerosols is relatively high, which is relevant to aerosol size and the Ångström

205 parameter (Ångström, 1929). The mixed-type aerosols have smaller AOD than dust

206 aerosols, but higher absorption aerosol optical depth (AAOD) throughout the

207 wavelength band of 440 to 1020 nm (Fig. 3b). Dust aerosols exhibit higher AAOD at

208 the 440 nm wavelength than that of anthropogenic aerosols.

209   As expected by using our SSA spectral classification method, dust aerosols show

210 a monotonic increasing SSA trend with increasing wavelength, while anthropogenic

211 aerosols exhibit an opposite trend. In contrast, the SSA of the mixed-type aerosols

212 peaks at the wavelength of 675 nm (Fig. 3c). Interestingly, the mixed-type aerosols

213 exhibit the lowest SSA value that cannot be predicted by our classification method

214 because the classification method considers the spectral trend rather than the value of

215 SSA. This indicates enhanced absorption for the mixed-type aerosols. The spectral

216 average SSA of the dust, mixed-type, and anthropogenic aerosols are 0.94, 0.89, and

217 0.93, respectively. Hence, internal mixing of the East Asian aerosol mixtures yields

218 the lowest SSA that is distinct from the corresponding individual aerosol types.

219   The dust aerosols exhibit the highest value for the real part of the complex

220   refractive index, while the anthropogenic aerosols show the lowest value (Fig. 3e).

221   Interestingly, the real refractive index of the mixed-type aerosols is close to that of

222   dust aerosols, indicating high scattering of the mixed-type aerosols. The spectral

223   imaginary refractive index for anthropogenic aerosols is nearly constant (Fig. 3f),

224   which is characteristic of this aerosol type. However, the imaginary refractive index

225   of anthropogenic aerosols is much lower than that of BC aerosols (approximately 0.6

226   in Bond and Bergstrom (2006)), because the majority of anthropogenic aerosol

227   components are non-absorbing aerosols such as sulfate and nitrate salts. The spectral

228   imaginary refractive index of dust aerosols is similar to the AERONET dust

229   climatology over Africa and the Middle East (Schuster et al., 2016), with stronger

230   absorption at the visible wavelength (440 nm). The mixed-type aerosols exhibit the

231   highest imaginary refractive index, especially at the visible wavelength, where the

232   imaginary refractive index of the mixed-type aerosols (0.0159) is more than twice that

233   of anthropogenic aerosols (0.0078).

234   Hence, the absorbing ability of the East Asian aerosols is significantly enhanced

235   due to the mixing process, in light of the lowest SSA and the highest AAOD and

236   imaginary refractive index for the mixed-type aerosols.

237   **4   Enhanced radiative absorption by East Asian aerosol mixtures**

238   To investigate the radiative effects caused by the enhanced absorbing ability of

239   the East Asian aerosol mixtures, we calculated the average aerosol direct radiative

240   efficiency at the bottom of the atmosphere (BOA), at the top of the atmosphere (TOA),

241   and in the atmosphere (ATM) (Fig. 4), respectively. The mixed-type aerosols exhibit

242   lower spectral average AOD (0.48) than dust aerosols (0.64) (Fig. 3a), but show

243   comparable radiative forcing relative to dust aerosols at BOA (-72.7 and -73.6 Wm^{-2}

244 for the mixed-type and dust aerosols, respectively) (Fig. 4a). This feature is explained

245 by higher BOA cooling efficiency of the mixed-type aerosols (-112.9 $Wm^{-2}\tau^{-1}$) than

246 dust (-101.0 $Wm^{-2}\tau^{-1}$) (Fig. 4b). The radiative absorption enhancement is evident for

247 the TOA and ATM forcing: the mixed-type aerosols exhibit the highest ATM radiative

248 forcing (55.0 Wm^{-2}) and the lowest absolute TOA forcing (-17.8 Wm^{-2}). For

249 comparison, we calculated the aerosol radiative efficiency of various aerosol types to

250 rule out the effect of aerosol loading. The mixed-type aerosols exhibit the highest

251 BOA cooling efficiency, the highest ATM heating efficiency (90.3 $Wm^{-2}\tau^{-1}$), and the

252 lowest TOA cooling efficiency (-22.5 $Wm^{-2}\tau^{-1}$).

253 The average BOA radiative efficiency of dust aerosols (-101.0 $Wm^{-2}\tau^{-1}$) in the

254 present study (Fig. 4b) falls in the range of -96.1 to -127.0 $Wm^{-2}\tau^{-1}$ by Yu et al. (2016),

255 but lower than the result of -124.6 ± 12.2 $Wm^{-2}\tau^{-1}$ by Noh et al. (2012). Note that

256 results by Noh et al. (2012) and Yu et al. (2016) are likely biased due to a non-linear

257 dependence between aerosol direct radiative forcing and AOD, which is avoided in

258 the present study. Our previous work (Tian et al., 2018), which used the same

259 radiative efficiency calculation method but a different aerosol classification approach

260 from the present study, obtained a similar BOA radiative efficiency of -102.3 $Wm^{-2}\tau^{-1}$

261 at the Semi-Arid Climate and Environment Observatory of Lanzhou University

262 (SACOL), Northwest China.

263 The spatial distributions of the aerosol radiative efficiency for BOA, ATM, and

264 TOA are presented in Figs. 5-7, respectively. Only those sites with a sample number

265 of greater than 50 were averaged and included in the figures. Despite the fact that

266 AERONET sites are unavailable in some remote areas, the worldwide aerosol

267 distributions are well captured by the AERONET observations. The mixed-type

268 aerosols are distributed in East Asia, India and around the Saharan Desert regions.

269   The mixed-type aerosols exhibit a higher BOA radiative cooling efficiency than

270   that of dust and anthropogenic aerosols over the East Asia region (Fig. 5). The BOA

271   radiative cooling efficiency over India is also high, but the difference of various

272   aerosol types is small. Biomass burning aerosols over Africa exhibit the highest BOA

273   cooling efficiency in the globe, which may be explained by the different combustion

274   compositions and processes as described in Eck et al. (2010) and Garcá et al. (2012).

275   Anthropogenic aerosols over North America and dust aerosols around the Sahara

276   Desert show a relatively lower BOA cooling efficiency.

277   The mixed-type aerosols also exhibit a higher ATM radiative heating efficiency

278   than dust and anthropogenic aerosols over East Asia (Fig. 6). The ATM radiative

279   heating efficiency over India is high for all aerosol types. The ATM radiative heating

280   efficiency is high over South Africa, where biomass burning aerosols dominate. On

281   the other hand, the ATM radiative heating efficiency over North America and around

282   the Sahara Desert regions is relatively smaller.

283   The enhanced BOA cooling and ATM heating efficiencies reveal that the

284   mixed-type aerosols exhibit higher BOA cooling and ATM heating effects than those

285   of the unmixed dust and anthropogenic aerosols with the same aerosol loading. The

286   enhanced BOA cooling and ATM heating effects lead to a cooler surface and warmer

287   atmosphere and restrain the development of the planetary boundary layer, resulting in

288   a more stable atmosphere that is unfavorable for dispersion of atmospheric gaseous

289   and PM pollutants. Noting that mixed-type aerosols occur frequently in East Asia and

290   their occurrence can reach as high as fifty percent over some locations (Li et al.,

291   2015). Hence, the mixed-type aerosols likely play a significant role in enhancing air

292   pollution over East Asia. In addition, the mixed-type aerosols show lower TOA

293   radiative cooling efficiency than dust and anthropogenic aerosols over the East Asia

294 region (Fig. S1). The reduced TOA cooling efficiency indicates that the East Asian

295 aerosol mixtures partially counteract the cooling effect of the Earth-atmosphere

296 system by the corresponding individual components.

297 **5   Discussions**

298   The aerosol direct radiative efficiency strongly depends on solar zenith angle

299 (e.g., Derimian et al., 2016). To investigate the influence of solar zenith angle on the

300 result of the present study, the aerosol radiative efficiency as a function of solar zenith

301 angle were calculated for various aerosol types (Fig. 7). Note that the AERONET data

302 used in the present study is only available between $50°$ to $80°$ solar zenith angles. The

303 BOA radiative cooling and ATM heating efficiencies (absolute value of radiative

304 efficiency) decrease with increasing solar zenith angle (decreasing cosine of solar

305 zenith angle), while the TOA cooling efficiency increase with increasing solar zenith

306 angle. The mixed-type aerosols exhibit higher BOA cooling efficiency, higher ATM

307 heating efficiency, and lower TOA cooing efficiency than those of unmixed dust and

308 anthropogenic aerosols. The dust aerosols exhibit both higher BOA and TOA cooling

309 efficiency than the anthropogenic aerosols, leading to small difference in the ATM

310 heating efficiency between the two aerosol types.

311   Aerosol absorption has been suggested as a key factor that determines the aerosol

312 radiative effects (Li et al., 2010). The aerosol radiative efficiencies as a function of

313 SSA and imaginary refractive index for various aerosol types were calculated to

314 investigate the effect of aerosol absorbing on the aerosols radiative efficiency (Figs. 8

315 and 9). The BOA cooling efficiency and ATM heating efficiency increase with

316 increasing absorption (i.e., decreasing SSA and increasing imaginary refractive index)

317 and the TOA cooling efficiency decrease with increasing absorption. However, the

318 dependences between radiative efficiency and SSA are stronger than those between

319 radiative efficiency and the imaginary refractive index for BOA, TOA, and ATM. The

320 dependences between radiative efficiency and SSA are approximately linear, but the

321 dependences between radiative efficiency and imaginary refractive index become less

322 apparent with increasing imaginary refractive index. The strong dependence between

323 aerosol radiative efficiency and SSA has also been shown over Central China and

324 desert and semi-desert regions of northwestern China (Xin et al., 2016; Zhang et al.,

325 2017).

326   We also examined the effects of the fraction of fine and coarse mode on aerosol

327 radiative efficiency (Fig. 10). The BOA cooling and ATM heating efficiencies (TOA

328 cooling efficiency) initially increase (decreases) with increasing fine mode fraction

329 (FMF), when FMF is lower than 0.3 and coarse mode dust aerosols dominate. Then

330 the BOA cooling and ATM heating efficiencies (TOA cooling efficiency) reach a peak

331 (bottom) in the FMF range of 0.3 to 0.5. Finally the BOA cooling and ATM heating

332 efficiency (TOA cooling efficiency) begin to decrease (increase) when FMF is greater

333 than 0.5, where fine mode anthropogenic aerosols become dominate. Overall, the

334 moderate mixing of dust with fine mode anthropogenic pollutants, which is classified

335 as mixed-type aerosols in the present study, is responsible for the enhanced radiative

336 absorption. A previous study (Li et al., 2015) has revealed that moderate mixing of

337 East Asian dust with fine mode pollutants is responsible for high SSA spectral

338 curvature, which suggests well-mixed aerosol mixtures.

339 **6 Conclusions**

340   The mixing state of atmospheric aerosols plays a significant yet poorly

341 quantified role in determining the aerosol optical properties and radiative effects. In

342 the East Asia region, coarse mode dust and fine mode anthropogenic pollution are

343 typically mixed externally and/or internally in the atmosphere. The mixing of dust

with anthropogenic aerosols exerts a significant influence on aerosol absorption and radiative efficiency. We present an extensive investigation of the radiative effects of the East Asian aerosol mixtures.

The mixed-type aerosols exhibit significantly higher AAOD, lower SSA, and higher imaginary refractive index than those of unmixed dust and anthropogenic aerosols, showing significantly enhanced absorption. The absorption enhancement is most evident at wavelength 440 nm, where the imaginary refractive index of the mixed-type aerosols (0.0159) is more than twice that of anthropogenic aerosols (0.0078). The mixed-type aerosols also exhibit a unique non-monotonic SSA trend, which provides a characteristic signature for identifying these aerosols.

The values of the aerosol radiative efficiencies for dust, mixed-type and anthropogenic aerosols are -101.0, -112.9 and -98.3 $Wm^{-2}\tau^{-1}$ for BOA, -42.3, -22.5 and -39.8 $Wm^{-2}\tau^{-1}$ for TOA, and 58.7, 90.3 and 58.5 $Wm^{-2}\tau^{-1}$ for ATM, respectively. The mixed-type aerosols exhibit significantly higher BOA radiative cooling efficiency and ATM heating efficiency than those of dust and anthropogenic aerosols over East Asia. These enhanced BOA cooling and ATM heating efficiencies reveal that the mixed-type aerosols exhibit stronger BOA cooling and ATM heating effects than those of unmixed dust and anthropogenic aerosols for a given aerosol loading, resulting in a more stable atmosphere that is unfavorable for the diffusion and dispersion of the gaseous and PM pollution. Hence, our results suggest that the mixed-type aerosols likely play a significant role in enhancing the air pollution in East Asia, because the mixing of dust and anthropogenic aerosols occurs frequently in this region. In addition, the mixed-type aerosols show lower TOA cooling efficiency, indicating that the mixed-type aerosols partially counteract the cooling effect of the earth-atmosphere system by the corresponding individual components. Since dust

369   particles play a catalytic role in the conversion of sulfur dioxide to sulfate (Zhang et

370   al., 2015; Li et al., 2017a), the interaction between dust and anthropogenic aerosols

371   also provides a possible mechanism for the observed efficient internal mixing and

372   enhanced radiative absorption over East Asia.

373   Multiple factors have been suggested to be responsible for severe air pollution in

374   East Asia, including the interaction between BC aerosols and the atmospheric

375   boundary layer (Ding et al., 2016; Peng et al., 2016; Li et al., 2017b), rapid secondary

376   aerosol formation during severe haze events (Wang et al., 2016), weakening of the

377   East Asian monsoon circulation (Wu et al., 2016), and climate change (Cai et al,

378   2017). Our results indicate that interaction between dust and anthropogenic pollution

379   does not only represent a plausible pathway for PM formation and internal mixing but

380   also results in unfavorable dispersion conditions (i.e., increased atmospheric stability),

381   both exacerbating regional air pollution in East Asia. Clearly, future studies are

382   necessary to more accurately assess the mixing state of aerosols in atmospheric

383   models, in order to better understand the formation mechanism for air pollution over

384   East Asian and to assess its impacts on human heath, weather, and climate (Zhang et

385   al., 2015).

386   **7   Data availability**

387   The original sun photometer data are available from the AERONET website

388   (http://aeronet.gsfc.nasa.gov/). The radiative flux data for the worldwide AERONET

389   sites calculated using the SBDART model and all data for the figures and table in the

390   present research are available from the authors upon request.

391   **Competing interests**. The authors declare that they have no conflict of interest.

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669 **Figure Captions**

670 **Figure 1.** Diagram of the radiative efficiency calculation and aerosol classification

671 **Figure 2.** The imaginary refractive index at 440 nm versus the average of the imaginary refractive

672 indices at 675, 870, and 1020 nm for dust, mixed-type, and anthropogenic aerosols over

673 worldwide AERONET sites. Only those sites with a sample number of 50 and higher were

674 averaged and shown in the figure.

675 **Figure 3.** Spectral behavior of (a) AOD, (b) AAOD, (c) SSA, (d) asymmetry factor, (e) real part of

676 the complex refractive index, and (f) imaginary part of the complex refractive index for dust,

677 mixed-type and anthropogenic aerosols averaged from East Asian sites.

678 **Figure 4.** (a) Radiative forcing and (b) radiative efficiency of the dust, mixed-type, and

679 anthropogenic aerosols averaged from East Asian sites.

680 **Figure 5.** Aerosol radiative efficiency at BOA: (a) dust aerosols, (b) anthropogenic aerosols, and

681 (c) mixed-type aerosols.

682 **Figure 6.** Aerosol radiative efficiency in ATM: (a) dust aerosols, (b) anthropogenic aerosols, and

683 (c) mixed-type aerosols.

684 **Figure 7.** Aerosol direct radiative efficiency as a function of the cosine of solar zenith angle for

685 the dust, mixed-type and anthropogenic aerosols: (a) BOA, (b) ATM, and (c) TOA.

686 **Figure 8.** Aerosol direct radiative efficiency as a function of SSA for the dust, mixed-type and

687 anthropogenic aerosols: (a) BOA, (b) ATM, and (c) TOA.

688 **Figure 9.** Aerosol direct radiative efficiency as a function of imaginary refractive index for the

689 dust, mixed-type and anthropogenic aerosols: (a) BOA, (b) ATM, and (c) TOA.

690 **Figure 10.** Aerosol direct radiative efficiency as a function of FMF in the East Asian region: (a)

691 BOA, (b) ATM, and (c) TOA. The average radiative efficiencies of the dust, mixed-type and

692 anthropogenic aerosols are also plotted.

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697   **Figure 1.** Diagram of the radiative efficiency calculation and aerosol classification

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Figure 2. The imaginary refractive index at 440 nm versus the average of the imaginary refractive indices at 675, 870, and 1020 nm for dust, mixed-type, and anthropogenic aerosols over worldwide AERONET sites. Only those sites with a sample number of 50 and higher were averaged and shown in the figure.

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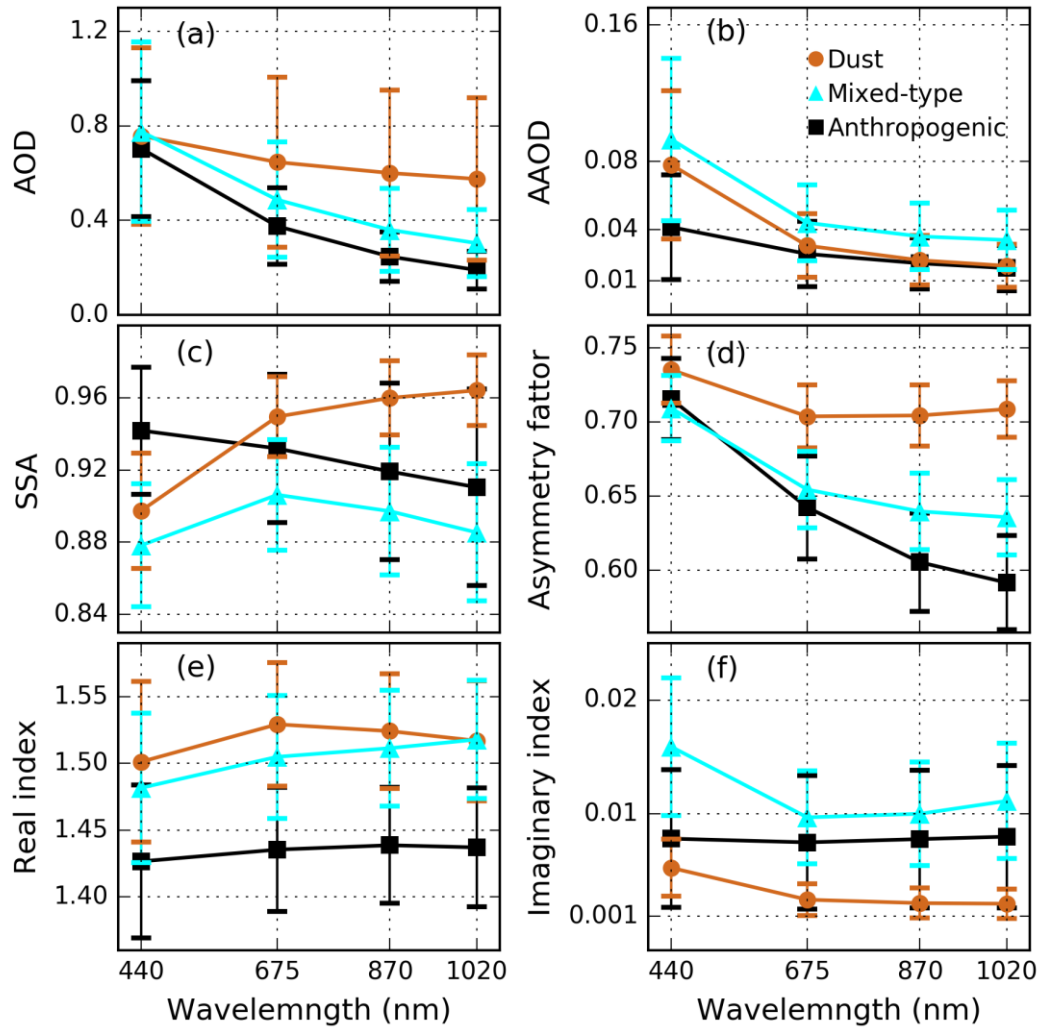
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Figure 3. Spectral behavior of (a) AOD, (b) AAOD, (c) SSA, (d) asymmetry factor, (e) real part of

the complex refractive index, and (f) imaginary part of the complex refractive index for dust,

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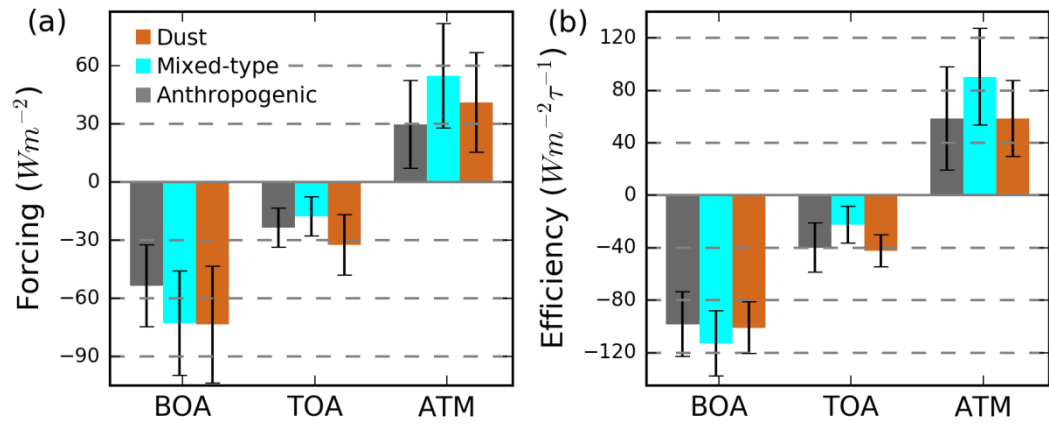
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714 **Figure 4.** (a) Radiative forcing and (b) radiative efficiency of the dust, mixed-type, and

715 anthropogenic aerosols averaged from East Asian sites.

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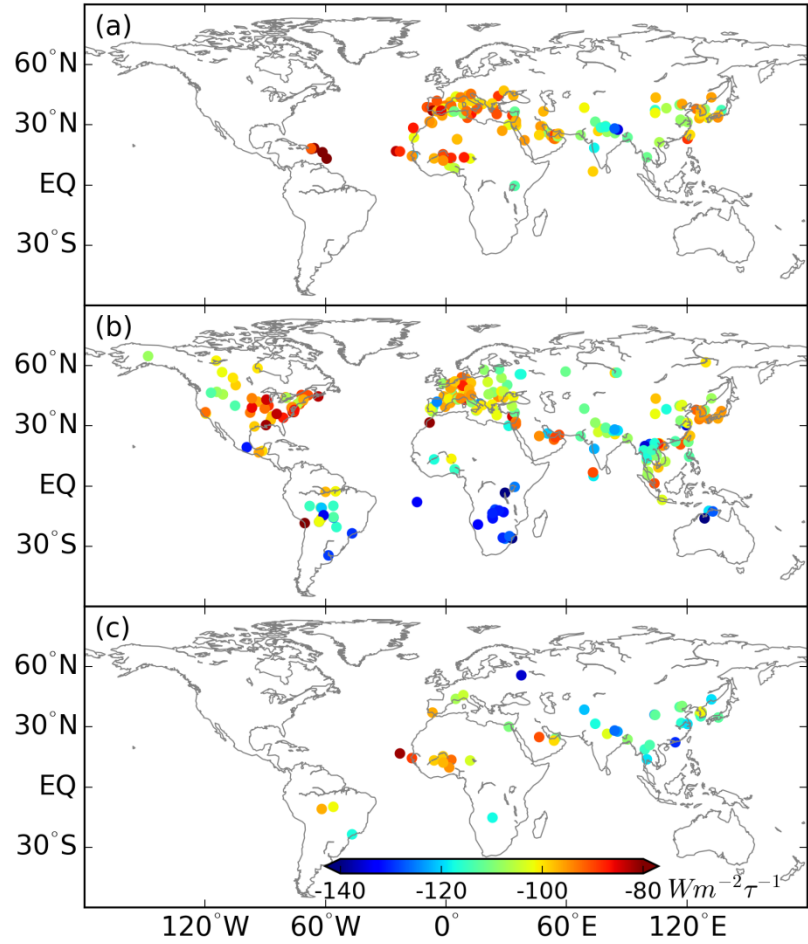
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Figure 5. Aerosol radiative efficiency at BOA: (a) dust aerosols, (b) anthropogenic aerosols, and

(c) mixed-type aerosols.

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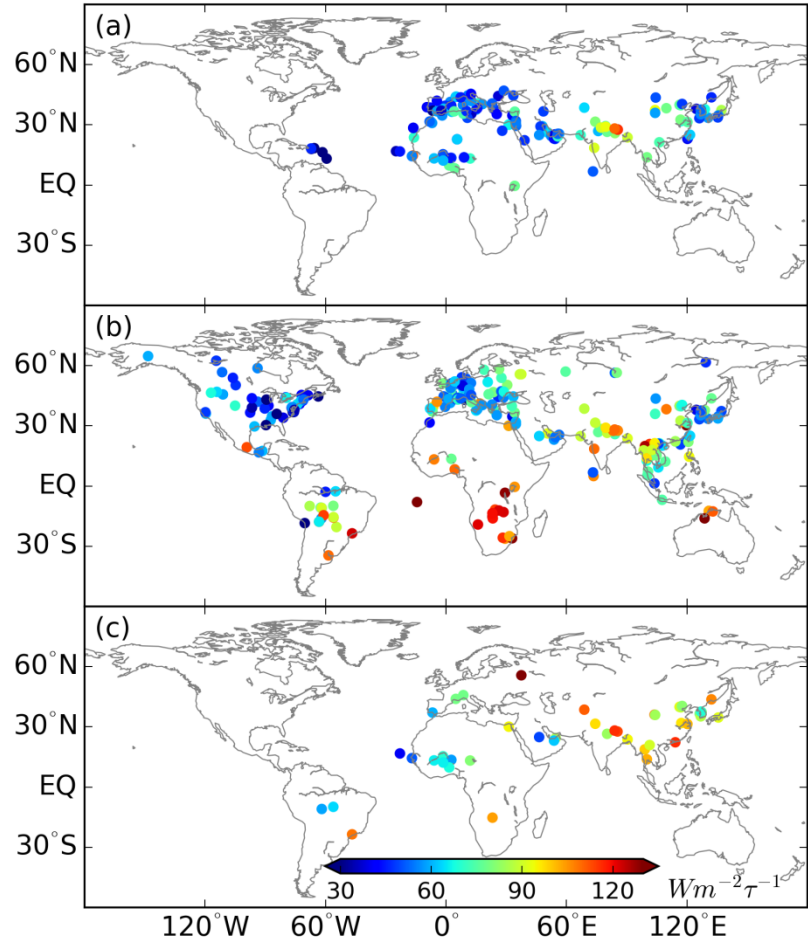
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Figure 6. Aerosol radiative efficiency in ATM: (a) dust aerosols, (b) anthropogenic aerosols, and

(c) mixed-type aerosols.

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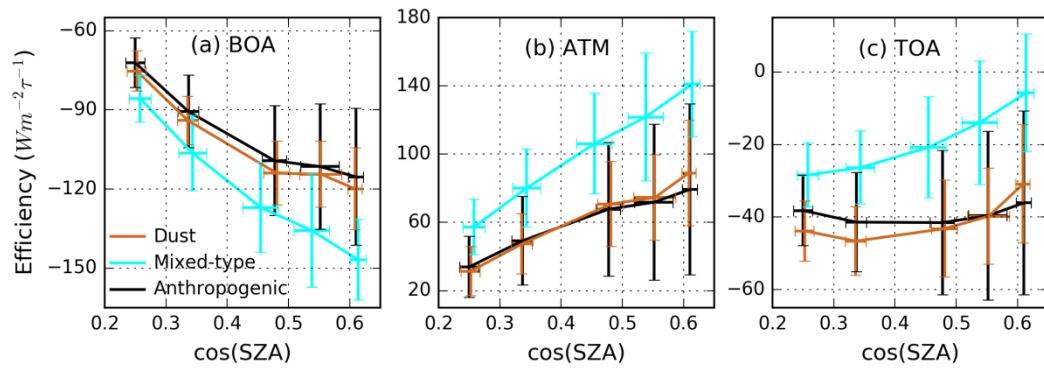
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730 **Figure 7.** Aerosol direct radiative efficiency as a function of the cosine of solar zenith angle for

731 the dust, mixed-type and anthropogenic aerosols: (a) BOA, (b) ATM, and (c) TOA.

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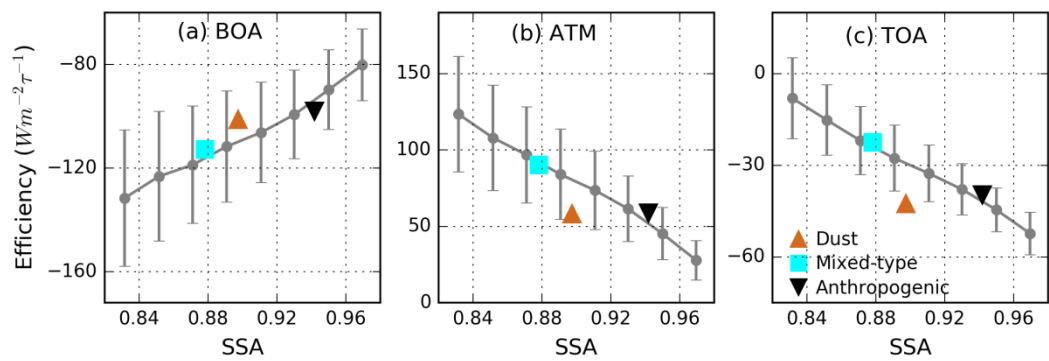
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735 **Figure 8.** Aerosol direct radiative efficiency as a function of SSA for the dust, mixed-type and

736 anthropogenic aerosols: (a) BOA, (b) ATM, and (c) TOA.

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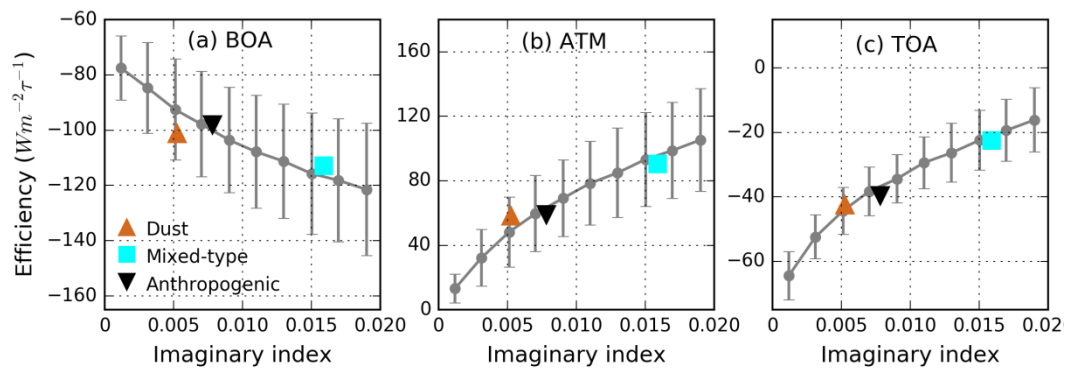
740

741 **Figure 9.** Aerosol direct radiative efficiency as a function of imaginary refractive index for the

742 dust, mixed-type and anthropogenic aerosols: (a) BOA, (b) ATM, and (c) TOA.

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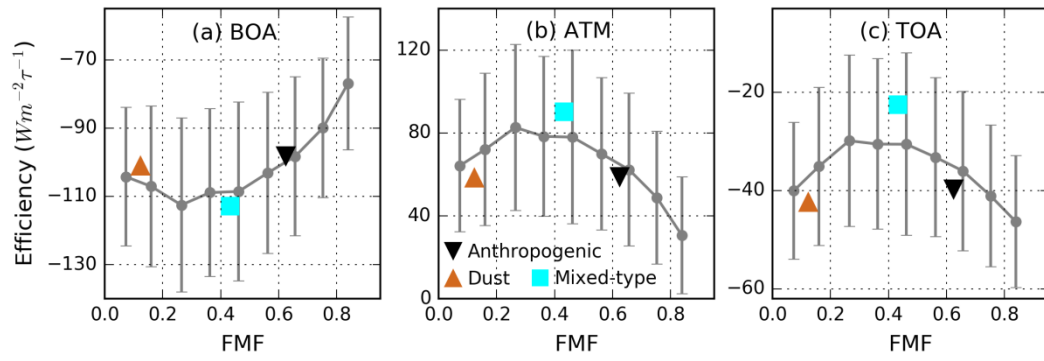
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746 **Figure 10.** Aerosol direct radiative efficiency as a function of FMF in the East Asian region: (a)

747 BOA, (b) ATM, and (c) TOA. The average radiative efficiencies of the dust, mixed-type and

748 anthropogenic aerosols are also plotted.

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