

1 Less atmospheric radiative heating by dust due to the synergy of 2 coarser size and aspherical shape

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8 **Abstract.** Mineral dust aerosols cool and warm the atmosphere by scattering and absorbing solar (short-wave: SW)
9 and thermal (long-wave: LW) radiation. However, significant uncertainties remain in dust radiative effects, largely
10 due to differences in the dust size distribution and spectral optical properties simulated in Earth system models. Dust
11 models typically underestimate the coarse dust load (more than 2.5 μm in a diameter) and assume a spherical shape,
12 which leads to an overestimate of the fine dust load (less than 2.5 μm) after the dust emissions in the models are scaled
13 to match observed dust aerosol optical depth at 550 nm (DAOD₅₅₀). Here, we improve the simulated dust properties
14 with datasets that leverage measurements of size-resolved dust concentration, asphericity factor, and refractive index
15 in a coupled global chemical transport model with a radiative transfer module. After the adjustment of size-resolved
16 dust concentration and spectral optical properties, the global and annual average of DAOD₅₅₀ from the simulation
17 increases from 0.023 to 0.029 and falls within the range of a semi-observationally-based estimate (0.030 ± 0.005).
18 The reduction of fine dust load after the adjustment leads to a reduction of the SW cooling at the Top Of the
19 Atmosphere (TOA). To improve agreement against a semi-observationally-based estimate of the radiative effect
20 efficiency at TOA, we find that a less absorptive SW dust refractive index is required for coarser aspherical dust. Thus,
21 only a minor difference is estimated for the net global dust radiative effect at TOA (-0.08 vs. $-0.00 \text{ W}\cdot\text{m}^{-2}$ on a global
22 scale). Conversely, our sensitivity simulations reveal that the surface warming is substantially enhanced near the
23 strong dust source regions (less cooling to -0.23 from $-0.60 \text{ W}\cdot\text{m}^{-2}$ on a global scale). Thus, less atmospheric radiative
24 heating is estimated near the major source regions (less heating to 0.15 from $0.59 \text{ W}\cdot\text{m}^{-2}$ on a global scale), because
25 of enhanced LW warming at the surface by the synergy of coarser size and aspherical shape.

26 1 Introduction

27 Mineral dust aerosols can both cool and warm the climate, but how much dust aerosols net influence global
28 climate is highly uncertain (Penner, 2019). Global dust modeling studies have suggested that mineral dust exerts
29 global and annual mean aerosol radiative effect (RE) between -0.6 and $+0.2 \text{ W m}^{-2}$ at the Top Of the Atmosphere
30 (TOA) and between -0.2 and -2.7 W m^{-2} at the surface (Miller and Tegen, 1998; Balkanski et al., 2007; Tanaka et
31 al., 2007; Takemura et al., 2009; Räisänen et al., 2013; Zhao et al., 2013; Albani et al., 2014; Colarco et al., 2014;
32 Heald et al., 2014; Di Biagio et al., 2020; Tuccella et al., 2020). Whereas a negative RE corresponds to the cooling of
33 the global system when the sunlight is reflected to space, a positive RE corresponds to an overall warming of the
34 Earth-atmosphere system by trapping incident short-wave (SW) and outgoing long-wave (LW) radiation. Radiative
35 effect by dust aerosols perturbs surface temperature, wind speed, rainfall, and vegetation cover, which may induce

36 feedback on dust emissions (Perlwitz et al. 2001; Miller et al., 2004a; Colarco et al., 2014). The climate feedback does
37 not only depend on RE at TOA or the surface alone but also on the difference to the value at TOA and surface, which
38 represents radiative heating within the atmosphere (Miller et al., 2004b; Yoshioka et al., 2007; Lau et al., 2009). The
39 large uncertainties in quantifying the dust RE in the models are mainly propagated from the large spatial heterogeneity
40 and temporal variability of mineral dust abundance and the physicochemical properties (e.g., size distribution, mineral
41 composition, and shape), as well as the ground surface characteristics and atmospheric properties (e.g., surface
42 reflectance, temperature, and atmospheric absorption) (Sicard et al., 2014; Lacagnina et al., 2015; Li and Sokolik,
43 2018). The model errors in dust size distribution and particle shape can lead to an overestimate of fine dust load after
44 the dust emissions in the models are scaled to match observed dust aerosol optical depth at 550 nm (DAOD₅₅₀). The
45 corresponding overestimate of SW cooling might be compensated for in models by using a refractive index that is too
46 absorbing (Di Biagio et al., 2019, 2020), which depends on the mineral composition of the dust. We regard “fine” and
47 “coarse” dust as dust particles with a diameter less than 2.5 μm (i.e., PM_{2.5}) and between 2.5 and 20 μm, respectively.
48 Below, we provide a brief discussion of the effects of the dust size distribution, particle shape, and mineral
49 composition on dust radiative effects.

50 First, there has been increased attention paid to the importance of accurately predicting the abundance of coarse
51 dust for the global energy balance (Kok et al., 2017; Song et al., 2018; Di Biagio et al., 2020; Adebisi and Kok, 2020).
52 The coarser particles are expected to be more prevalent closer to the source regions, as they fall much faster than finer
53 particles (Mahowald et al., 2014). For instance, the lifetime of dust aerosols larger than 30 μm in diameter is less than
54 12 h in most cases except in large haboobs (Ryder et al., 2013). Current models, however, cannot accurately simulate
55 observed transport of coarse dust particles across the Atlantic (Weinzierl et al., 2017; Ansmann et al., 2017), although
56 several hypotheses have been proposed to explain measurements of giant dust particles (larger than 63 μm in diameter)
57 relatively far from source regions (van der Does et al., 2018). The potential mechanism for long-range transport of
58 giant dust particles is that the uplift events of coarse dust can be induced by a nocturnal low-level jet or cold pool
59 outflow from mesoscale convective systems (i.e., haboobs) (Rosenberg et al., 2014; Ryder et al., 2019). At higher
60 elevation, electrostatic forces might retard the settling of coarse and giant dust particles and thus may facilitate the
61 transport of these particles over longer distances (Harrison et al., 2018; Toth et al., 2019). Other missing processes
62 that affect the transport and deposition of giant particles would also need to be incorporated into the models to
63 reproduce the measurements of the size distribution over the open ocean (van der Does et al., 2018). The coarse dust
64 particles scatter and absorb both the solar and thermal radiation, causing a net warming effect at TOA. In contrast, the
65 fine dust particles principally scatter SW radiation, causing a net cooling effect. Since coarse dust tends to warm the

66 climate, the underestimation of the abundance of coarse dust causes Earth system models to underestimate the
67 warming near the dust source regions.

68 Second, previous studies have shown that the SW radiative effect of dust asphericity on climate simulations is
69 minor on a global scale, partly because the larger DAOD is compensated for by the larger asymmetry parameter of
70 aspherical dust, which reduces the amount of radiation scattered backward to space (Räsänen et al., 2013; Colarco et
71 al., 2014). Moreover, non-spherical calcium-rich dust particles can be converted to spherical particles, due to
72 heterogeneous reactions with nitrate and sulfate on these particles, especially over polluted regions (Laskin et al.,
73 2005; Matsuki et al., 2005). As the plumes move downwind to the oceans, the dust aerosols can be aggregated with
74 sea salt in the marine boundary layer, which leads to more spherical shapes and larger sizes (Zhang and Iwasaka,
75 2004). However, the assumption of spherical shape in models leads to a substantial underestimation of the extinction
76 efficiency and thus DAOD near the strong source regions, mainly because the assumption of sphericity causes an
77 underestimation of the surface-to-volume ratio compared to aspherical dust (Kok et al., 2017, 2021; Hoshyaripour et
78 al., 2019; Tuccella et al., 2020). Radiative effect efficiency is often used for the evaluation of the models and is defined
79 as the gradient of a linear least squares fit applied to AOD and dust radiative effect at each two-dimensional (2-D)
80 grid box ($W \cdot m^{-2} AOD^{-1}$). Thus, the estimates of the dust radiative effect efficiency could be biased, in part, due to
81 large uncertainties associated with the spherical assumption on AOD retrieval (Zhou et al., 2020).

82 Third, the dust refractive index is often derived from measurements based on dust or individual mineral particles
83 (Bedidi and Cervelle, 1993; Long et al., 1993; Di Biagio et al., 2017, 2019; Stegmann & Yang, 2017). Indeed, most
84 dust particles are internal mixtures of various mineral compositions and irregular shapes (Reid, 2003; Wiegner et al.,
85 2009; Wagner et al., 2012). In desert soils, iron (Fe) oxides are generally hematite ($\alpha\text{-Fe}_2\text{O}_3$) and goethite (FeOOH),
86 which cause soil-derived dust absorption at ultraviolet (UV) and visible wavelengths (Sokolik and Toon, 1999;
87 Balkanski et al., 2007). These two minerals have distinct optical properties, which might cause various intensities of
88 SW absorption and thus RE of dust aerosols (Lafon et al., 2016). The dust complex refractive index in the LW also
89 depends on the particle mineralogical composition (Sokolik et al., 1998). Di Biagio et al. (2017) found a linear
90 relationship between the magnitude of the imaginary refractive index at 7.0, 9.2, and 11.4 μm and the mass
91 concentration of calcite and quartz absorbing at these wavelengths. However, the speciation of dust into its mineral
92 components inherently comprises uncertainties on soil mineralogy, mineral content in size-segregated dust particles,
93 and refractive index of mineral, partly due to the differences in prescribed parameters such as the particle size. The
94 atmospheric aging of Fe-containing aerosols can further modulate the optical properties of Fe oxides (Ito et al., 2018)
95 and organic carbon (Al-Abadleh, 2021), while the photochemical transformation of Fe oxides from lithogenic sources
96 due to atmospheric processing is relatively limited ($< 10\%$), compared to pyrogenic sources (Ito et al., 2019).

97 Here, we focus on the influence of the size-resolved abundance of aspherical dust on the aerosol radiative effects
98 in a coupled global chemical transport model (IMPACT) (Ito et al., 2020 and references therein) with a radiative
99 transfer module (RRTMG) (Iacono et al., 2008). We improve the accuracy of these simulations by correcting the bias
100 in size-resolved dust concentration with the Dust Constraints from joint Observational-Modelling-Experimental
101 analysis (DustCOMM) data set (Adebiyi et al., 2020), as well as by considering the aspherical shape (Huang et al.,
102 2020, 2021). We then explore the sensitivity to dust refractive index.

103 **2 Methods**

104 We examined the dust radiative effects using ten combinations of different numerical experiments that varied
105 (1) the simulated dust concentration and their size distribution, (2) particle shape, and (3) mineralogical composition
106 (Tables 1 and 2). Two RRTMG calculations used the hourly averaged aerosol concentrations calculated from one
107 IMPACT model simulation (E1 and E3) (denoted as “IMPACT”). The two sensitivity experiments were handled in
108 the RRTMG calculations performed with the distinction between spherical and non-spherical dust and different
109 refractive indices. We denoted “Sphere” when the RRTMG calculations used the spherical assumption on the particle
110 shape, while the IMPACT model considered asphericity in calculation of gravitational settling velocities. On the other
111 hand, we denoted “Asphere” when the dust asphericity was also considered in the RRTMG calculations. Subsequently,
112 the simulated dust concentration and the size distribution were adjusted to the semi-observationally-based
113 concentrations (Adebiyi and Kok, 2020) in another chemical transport model simulation, which was performed with
114 the five RRTMG calculations (E4, E5, E6, E8, and E9) (denoted as “DustCOMM”). The term “semi-observationally-
115 based” is used for DustCOMM, DAOD₅₅₀, and dust radiative effect efficiency when the estimates are based on the
116 combination of observations and models. We examined different refractive indices for the dust mineralogy to represent
117 the regional variations in refractive indices (denoted as “Mineral”, “DB17”, “DB19”, “V83”, “Less SW”, “More LW”,
118 “More SW”, and “Less LW”). Thus, the other three experiments (E2, E7, and E10) were calculated from the model
119 output with a post-processor. DustCOMM-Asphere-DB19-V83 (E2) was obtained from combination of DustCOMM-
120 Asphere-DB19-DB17 (E4) for SW and DustCOMM-Asphere-Mineral-V83 (E6) for LW. DustCOMM-Asphere-Less-
121 More (E7) was obtained from combination of DustCOMM-Asphere-Less-Less (E8) for SW and DustCOMM-
122 Asphere-More-More (E9) for LW. DustCOMM-Asphere-More-Less (E10) was obtained from combination of
123 DustCOMM-Asphere-More-More (E9) for SW and DustCOMM-Asphere-Less-Less (E8) for LW. These sensitivity
124 simulations and their radiative effects are summarized in Tables 1 and 2, respectively, with more details below. In
125 section 2.3, we describe the DustCOMM data set used to adjust (1) size-resolved abundance of dust concentration. In
126 section 2.4, we describe the adjustment factor of (2) particle shape for spectral optical properties. In section 2.5, we

127 describe differences in spectral refractive indices due to (3) different mineralogical compositions for the radiative flux
128 calculation.

129 **2.1 Aerosol chemistry transport model**

130 This study used the Integrated Massively Parallel Atmospheric Chemical Transport (IMPACT) model (Ito et
131 al., 2020 and references therein). Simulations were performed for the year 2016, using a horizontal resolution of
132 $2.0^{\circ} \times 2.5^{\circ}$ for latitude by longitude and 47 vertical layers. The chemical transport model was driven by the Modern
133 Era Retrospective analysis for Research and Applications 2 (MERRA-2) reanalysis meteorological data from the
134 National Aeronautics and Space Administration (NASA) Global Modeling and Assimilation Office (GMAO) (Gelaro
135 et al., 2017). Thus, the radiative feedback of the dust aerosol on the climate was not considered in this study.

136 The model simulated the emissions, chemistry, transport, radiation, and deposition of major aerosol species,
137 including mineral dust, black carbon (BC), particulate organic matter (POM), sulfate, nitrate, ammonium, and sea
138 spray aerosols, and their precursor gases. Dust emissions were dynamically simulated using a physically-based
139 emission scheme (Kok et al., 2014; Ito and Kok, 2017) with the soil mineralogical map (Journet et al., 2014; Ito and
140 Shi, 2016). Atmospheric processing of mineral dust aerosols, during transport, were projected for four distinct aerosol
141 size bins ($<1.26 \mu\text{m}$, $1.26\text{--}2.5 \mu\text{m}$, $2.5\text{--}5 \mu\text{m}$, and $5\text{--}20 \mu\text{m}$ of diameter). In this version of the IMPACT model, two
142 modes were used for sulfate aerosol (nuclei and accumulation mode), and two moments were predicted within each
143 mode (sulfate aerosol number and mass concentration) (Liu et al., 2005). The surface coating of sulfate on dust
144 aerosols occurred because of the condensation of sulfuric acid gas on their surfaces, coagulation with sulfate aerosol,
145 and formation in aqueous reactions within cloudy regions of the atmosphere (Liu et al., 2005). The heterogeneous
146 uptake of nitrate, ammonium, and water vapor by each aerosol for each size bin was interactively simulated in the
147 model following a hybrid dynamical approach (Feng and Penner, 2007). Five types of aerosols (i.e., dust, nucleated
148 sulfate, carbonaceous aerosols from fossil fuel combustion, carbonaceous aerosols from biomass burning, and sea salt)
149 were assumed to be externally mixed in each size bin for the computation of spectral optical properties (Xu and Penner,
150 2012). To derive atmospheric concentration of mineral composition for dust aerosol, “tagged” tracer was used for
151 each size-resolved mineral source. The direct emissions of dust were evenly distributed in mixing ratio throughout the
152 planetary boundary layer. The global scaling factor of dust emission was determined from the comparison of the model
153 results with ground-based AOD measurements near the dust source regions prior to the adjustment to the DustCOMM
154 (Kok et al., 2014; Ito and Kok, 2017). In recent review papers, multi-model evaluations of aerosol iron concentrations
155 and their solubilities have been comprehensively summarized on global and regional scales (Myriokefalitakis et al.,
156 2018; Ito et al., 2021).

157 To improve the accuracy of our simulations of mineral dust, we made several upgrades to the on-line emission
158 and gravitational settling schemes used in Ito et al. (2020). The dust emissions were extremely sensitive to soil
159 moisture, and thus the bias was adjusted with satellite observations (Ito and Kok, 2017). However, the satellite
160 measurements were only available every other day, depending on location. The Soil Moisture Active Passive (SMAP)
161 Level-4 Soil Moisture data product addressed these limitations by merging the satellite observations into a numerical
162 model of the land surface water and energy balance while considering the uncertainty of the observations and model
163 estimates (Reichle et al., 2019). In this work, we utilized the 3-hourly data of soil moisture derived from the SMAP
164 for barren and open shrublands separately (Reichle et al., 2018). To achieve this, we used the MODerate resolution
165 Imaging Spectroradiometer (MODIS) land cover map at 500 m resolution to calculate the fraction of barren and open
166 shrublands in each ground surface layer (Friedl et al., 2019)

167 Compared to the assumption on spherical shapes of aerosols, the dust asphericity increased aerodynamic drag
168 at a given volume and mass, and thus increased gravitational settling lifetime by about 20% (Huang et al., 2020). Here,
169 we implemented a globally averaged asphericity factor of 0.87 (Huang et al., 2020) to the gravitational settling scheme
170 for mineral dust. Nevertheless, the lifetime of the dust aerosol for the largest-size bin in the IMPACT model, even
171 after accounting for asphericity (1.4 days for 5–20 μm of diameter), was significantly shorter than an ensemble of
172 model results (2.1 ± 0.3 days for the mass mean diameter of 8.3 μm) (Kok et al., 2017). The impact of this
173 underestimate of atmospheric lifetime is explored using the DustCOMM data set, as was summarized in Table 2 (E3
174 – E4).

175 **2.2 Integration of IMPACT and RRTMG**

176 To improve the accuracy of our simulations of dust RE, we made upgrades to the radiative transfer calculations
177 (Ito et al., 2018 and references therein). In this study, we integrated the Rapid Radiative Transfer Model for GCMs
178 (RRTMG) online within the IMPACT model to calculate the radiative fluxes associated with atmospheric aerosols.
179 RRTMG is a radiative transfer code that calculates the SW and LW atmospheric fluxes (Iacono et al., 2008). Given
180 the size range of dust particles, scattering and absorption in the on-line model were described in terms of Mie theory.
181 Assuming homogeneous spherical particles, the spectral optical properties such as the mass extinction coefficient,
182 single scattering albedo, and asymmetry parameter were calculated using a look-up table as a function of refractive
183 index and size parameter (Xu and Penner, 2012). The impact of this spherical assumption is explored using aspherical
184 factor, as was summarized in Table 2 (E5 – E4).

185 The mineral dust particles were assumed to follow prescribed size distributions within each size bin (Liu et al.,
186 2005). In applying the look-up table, the size spectrum for mineral dust was divided into 30 sub bins (Wang and

187 Penner, 2009). As for the SW, the particle size increased with the uptake of sulfate, nitrate, ammonium, and water by
188 the aerosols (Xu and Penner, 2012). These coating materials on aerosol cores were treated as internally mixed with
189 each aerosol core in each size bin. Thus, the coating materials on dust only can reduce solar absorption of mineral
190 dust. Subsequently, these optical properties were used by the RRTMG to calculate RE based on dust mixing ratio
191 distributions in the IMPACT model. The dust RE was estimated as the difference in the calculated radiative fluxes
192 with all aerosols and with all aerosols except the dust aerosols coated with sulfate, nitrate, ammonium, and water for
193 each bin. As the LW scattering was not accounted for in the RRTMG, we multiplied the LW radiative fluxes by the
194 adjustment factors of 1.18 ± 0.01 and 2.04 ± 0.18 for the dry particles at the surface and TOA (Dufresne et al., 2002),
195 following Di Biagio et al. (2020). The larger adjustment factor at TOA reflects the fact that the upward LW radiation
196 emitted from the ground surface can be trapped through scattering and absorption compared to the surface.

197 The broadband direct and diffuse albedos for both the UV visible and visible IR were specified from the hourly
198 MERRA-2. The surface emissivity was based on the hourly MERRA-2. Long-lived greenhouse gas concentrations
199 were obtained from historical greenhouse gas concentrations for climate models (Meinshausen et al., 2017). Water
200 vapor concentrations were specified according to the MERRA-2. Cloud optical properties were calculated based on
201 the liquid and ice visible optical depths from the MERRA-2, prescribing effective radii of $10 \mu\text{m}$ for water droplets
202 and $25 \mu\text{m}$ for ice particles, respectively (Gettelman et al., 2010; Heald et al., 2014).

203 **2.3 DustCOMM dataset and sensitivity experiments to size-resolved dust concentration**

204 Dust Constraints from joint Observational-Modelling-experimental analysis (DustCOMM) is a dataset of
205 three-dimensional (3-D) dust properties obtained by combining observational, experimental, and modeling constraints
206 on dust properties. While details can be found in Adebisi et al. (2020) and Adebisi and Kok (2020), we provide a
207 brief overview here. First, DustCOMM's constraint on the 3-D dust size distribution combines dozens of previously
208 published in-situ measurements of dust size distributions taken during several field campaigns, with an ensemble of
209 climate model simulations. The framework used those in-situ measurements first to constrain the globally averaged
210 size distribution (Adebisi and Kok, 2020), which is used subsequently to adjust the bias in an ensemble of six global
211 model simulations (Adebisi et al., 2020). The constraints on dust size distribution range from $0.2 \mu\text{m}$ to $20 \mu\text{m}$ in
212 diameter, where a generalized analytical function describes the sub-bin distribution based on brittle fragmentation
213 theory (Kok, 2011). The second DustCOMM product – atmospheric dust mass loading – combines the constraints on
214 dust size distribution with constraints on dust extinction efficiency and dust aerosol optical depth (Adebisi et al., 2020).
215 The constraints on dust extinction efficiency used the single-scattering database of Meng et al. (2010) and leveraged
216 measurements of the dust index of refraction as well as accounts for the non-spherical shape of dust particles (Kok et

217 al., 2017). For this, we approximate dust as tri-axial ellipsoidal particles described by the globally representative
 218 values of measured dust aspect ratio (the length-to-width ratio), and the height-to-width ratio (HWR) obtained from
 219 Huang et al. (2020). Furthermore, the dust aerosol optical depth used to obtain the dust mass loading combines the
 220 semi-observationally-based dataset from Ridley et al. (2016) with information from four reanalysis products. This
 221 includes the MERRA-2, Navy Aerosol Analysis and Prediction System (NAAPS), Japanese Reanalysis for Aerosol
 222 (JRAero), and Copernicus Atmosphere Monitoring Service interim ReAnalysis (CAMSiRA) (Adebisi et al., 2020).
 223 The aerosol RE of mineral dust strongly depends on both the magnitude of dust load and the dust size distribution
 224 (Tegen and Lacis, 1996; Liao and Seinfeld, 1998). The DustCOMM data set contains total column loading (X, Y) and
 225 concentration of mineral dust resolved by season (T) and particle size (S) (Adebisi et al., 2020). To correct the bias in
 226 the seasonally averaged size-resolved dust emission in the IMPACT model, $E_{\text{IMPACT}}(X, Y, T, S)$, the sum of bin 1, bin
 227 2, and bin 3 dust emission flux was scaled by the seasonal mean of the ratio of the sum of bin 1, bin 2, and bin 3 dust
 228 column loading between the model, $L_{\text{IMPACT}}(X, Y, T, S)$, and DustCOMM, $L_{\text{DustCOMM}}(X, Y, T, S)$, at each 2-D grid
 229 box. The bias correction factor, $L_{\text{bias}}(X, Y, T)$, between the IMPACT model and DustCOMM data set is given by:

$$230 \quad L_{\text{bias}}(X, Y, T) = \sum_{S=1}^3 L_{\text{DustCOMM}}(X, Y, T, S) \div \sum_{S=1}^3 L_{\text{IMPACT}}(X, Y, T, S) \quad (1).$$

231 When the source function was used for high-latitude dust in the Northern Hemisphere, this led to substantially high
 232 emissions and thus RE over there, likely due to the influences from long-range transported dust. Therefore, the direct
 233 emissions of dust from the nine major source regions only (Kok et al., 2021) were adjusted using the DustCOMM
 234 data (Fig. 1). To adjust the size bias in dust emissions, the mass fraction of emitted dust for each bin was prescribed
 235 according to the size-resolved total column loading of DustCOMM at each 2-D grid box. The mass fraction for each
 236 size bin, $S_{\text{DustCOMM}}(X, Y, T, S)$ is given by:

$$237 \quad S_{\text{DustCOMM}}(X, Y, T, S) = L_{\text{DustCOMM}}(X, Y, T, S) \div \sum_{S=1}^4 L_{\text{DustCOMM}}(X, Y, T, S) \quad (2).$$

238 Thus, the dust emission flux after the adjustment, $E_{\text{DustCOMM}}(X, Y, T, S)$ is given by:

$$239 \quad E_{\text{DustCOMM}}(X, Y, T, S) = L_{\text{bias}}(X, Y, T) \times S_{\text{DustCOMM}}(X, Y, T, S) \times E_{\text{IMPACT}}(X, Y, T) \quad (3).$$

240 Overall, the IMPACT-simulated lifetime of the dust aerosol for the second-size bin (7.8 days 1.26–2.5 μm of diameter)
 241 was in good agreement with the ensemble of model results (8.5 ± 1.1 days for the mass mean diameter of 1.8 μm)
 242 (Kok et al., 2017). To correct the bias in the seasonally averaged 3-D dust size distribution after the transport, the mass
 243 fraction of dust concentration for each bin between 0.2 and 20.0 μm of diameter was scaled at each 3-D grid box prior
 244 to calculating the radiative fluxes using the RRTMG by the ratio of mass concentration of $\text{PM}_{2.5}$ (i.e., the sum of bin
 245 1 and bin 2) to each bin (Table 3).

246 **2.4 Asphericity factor for optical properties and sensitivity experiments to particle shape**

247 To account for the dust asphericity, an adjustment factor was applied to the spherical optical properties at each
248 dust size parameter and refractive index. The adjustment factors for the spectral optical properties of non-spherical
249 particles were calculated after Huang et al. (2021). The atmospheric aging of mineral dust can form a uniform coating
250 around the mineral core and therefore decrease particle asphericity during transport. This is implicitly considered in
251 the globally averaged shape distribution of dust (Huang et al., 2019). Specifically, Huang et al. (2021) combined
252 globally representative dust shape distributions (Huang et al., 2020) with a shape-resolved single-scattering database
253 (Meng et al., 2010). This database combines four computational methods (Mie theory, T-matrix method, discrete
254 dipole approximation, and an improved geometric optics method) to compute the single-scattering properties of non-
255 spherical dust for a wide range of shape descriptors. Huang et al. (2021) provided the look-up table containing optical
256 properties of non-spherical dust as functions of size parameter and refractive index.

257 The approximation of particles to spheres is evaluated by applying aspherical factors to the optical properties
258 of the mass extinction coefficient, single scattering albedo, and asymmetry parameter for SW, as well as absorption
259 fraction of extinction for the LW. At the same time, we maintained the consideration of asphericity on the gravitational
260 velocity and kept the dust concentrations unaltered between the spherical (denoted as “Sphere”) and aspherical
261 (denoted as “Asphere”) cases.

262 **2.5 Spectral refractive index and sensitivity experiments to mineralogical compositions**

263 The aerosol RE of mineral dust depends on mineralogical composition. For the sensitivity simulation to the
264 SW and LW refractive indices, we used the global mean of laboratory measurements of the refractive index from 19
265 natural soils from various source regions around the world in Di Biagio et al. (2019) (denoted as “DB19”) and in Di
266 Biagio et al. (2017) (denoted as “DB17”), respectively. To illustrate the regional heterogeneity of refractive index, the
267 refractive index obtained from 19 samples was aggregated into 9 main source regions, and the arithmetic mean was
268 calculated for each source region (Di Biagio et al., 2017, 2019). The regionally averaged imaginary parts of the
269 refractive indices at the wavelength of 0.52 μm and 9.7 μm showed large differences in SW and LW absorptivity,
270 respectively, between different samples collected at various geographical locations (Fig. 1).

271 The optical properties from the measurements for dust samples generated from 19 natural soils suggested a
272 considerable role of Fe oxides in determining the SW absorption (Di Biagio et al., 2019). The refractive indices for
273 mineral components were used for hematite, goethite (Bedidi and Cervelle, 1993), silicate particle group, quartz,
274 gypsum (CaSO_4) (Stegmann & Yang, 2017), and calcite (CaCO_3) (Long et al., 1993) in the simulations denoted as
275 “Mineral”. The hematite and goethite were treated separately according to the mineralogical map (Journet et al., 2014).

276 Consequently, hematite mass content averaged in the dust at emission (0.79% for fine and 0.50% for coarse from the
277 IMPACT simulation) was lower than goethite content (1.8% and 1.3%, respectively) on a global scale. In addition to
278 the primary emission of gypsum, CaSO_4 is secondarily formed due to the dissolution/precipitation of CaCO_3 in
279 thermodynamic equilibrium condition (Ito and Feng, 2010). To illustrate the difference in refractive index, the global
280 mean of the mineral composition was used for the comparison with DB19 (Fig. 1). The imaginary parts of the
281 refractive indices from mineralogical map were higher than DB19, resulting in a stronger absorption over the SW
282 spectrum.

283 The mineral dust LW refractive index also depends on its mineralogical composition (Sokolik et al., 1998; Di
284 Biagio et al., 2017). The LW refractive index of Volz (1983) has been widely used in climate models and satellite
285 remote sensing algorithms and thus was examined here (denoted as “V83”) (Song et al., 2018). The imaginary parts
286 of the refractive indices from V83 were higher than DB17, resulting in a stronger absorption over most of the LW
287 spectrum. To analyze the dependence of the results on less (more) absorptive SW and less (more) absorptive LW
288 refractive indices, we made further sensitivity simulations by varying the values of imaginary parts of the refractive
289 index within the range of values from Di Biagio et al. (2017, 2019) (10% or 90% percentiles for SW or LW,
290 respectively) (denoted as “Less” or “More”). The associated real parts with 10% or 90% percentile imaginary parts
291 for LW were calculated to account for the Kramers-Kronig relation (Lucarini et al., 2005).

292 **2.6 Semi-observationally-based dust SW and LW radiative effect efficiency**

293 To estimate dust radiative effect efficiency, aerosol and radiation remote sensing products have been used with
294 various methods (Table 4) (Zhang and Christopher 2003; Li et al. 2004; Christopher and Jones 2007; Brindley and
295 Russell 2009; Yang et al. 2009; Di Biagio et al. 2010; Hansell et al. 2010; Hansell et al. 2012; Song et al. 2018).

296 The instantaneous SW radiative effect efficiency at TOA is obtained from the linear regression of TOA
297 radiation flux versus AOD observations, although the values in low-dust periods can be substantially influenced by
298 other types of aerosols such as biomass burning (Li et al. 2004). This radiative effect efficiency corresponds to the
299 instantaneous value derived under the limited condition at the measurements (e.g., solar position, atmospheric
300 condition). From the extrapolation of the instantaneous value, the diurnal mean dust SW radiative effect efficiency at
301 the surface and TOA can be derived based on model calculations.

302 The LW radiative effect efficiency at TOA can be obtained from the linear regression of TOA radiation flux
303 versus AOD observations over the source regions (Brindley and Russell 2009). However, the observed outgoing LW
304 radiation is not only dependent on DAOD but also on other factors such as dust layer height, water vapor content, and
305 other types of aerosols. Thus, the LW radiative effect efficiency is estimated from the difference between observed

306 outgoing LW radiation and the dust-free outgoing LW radiation, which can be estimated using radiative transfer model
307 (Song et al., 2018).

308 Consequently, the semi-observationally-based estimates of the dust radiative effect efficiency could be biased,
309 in part, due to large uncertainties associated with the estimation method, the selection of cloud-free and dust-dominant
310 data, and dust physicochemical properties. To understand the sensitivity of the dust radiative effect efficiency to the
311 particle size distribution, asphericity, and refractive index of dust, radiative transfer computations have been carried
312 out in previous studies (Li et al., 2004; Song et al., 2018). Song et al. (2018) found that the combination of the coarser
313 dust particle size distribution and the more absorptive LW refractive index (V83) yielded the best simulation of the
314 dust LW radiative effect in comparison with the satellite flux observations (i.e., Clouds and the Earth's Radiant Energy
315 System (CERES)), compared to the less absorptive LW refractive index (DB17).

316 **3. Results and Discussions**

317 We evaluate our results from the sensitivity simulations against semi-observationally-based estimates of
318 $DAOD_{550}$ in section 3.1 and radiative effect efficiency for SW and LW in section 3.2 and section 3.3, respectively.
319 We focus this evaluation on the North Africa and the North Atlantic in boreal summer (June, July, and August) partly
320 because that is the region and season for which most observational constraints on dust radiative effects are available.
321 The better agreement is obtained for the less absorptive SW (Di Biagio et al., 2019) and the more absorptive LW
322 (Volz, 1983) dust refractive indices with adjustments of size-resolved dust concentration and particle shape. Our
323 improved simulation from IMPACT-Sphere-Mineral-V83 (E1) to DustCOMM-Asphere-DB19-V83 (E2)
324 substantially reduces the model estimates of atmospheric radiative heating by mineral dust near the major source
325 regions even though it induces only a minor difference in RE at TOA on a global scale (section 3.4). To elucidate the
326 differences in dust radiative effects between different simulations, the results from the sensitivity simulations in
327 conjunction with previous modeling studies are analyzed in section 3.5.

328 **3.1 Dust load and aerosol optical depth**

329 We compared our model estimates of $DAOD_{550}$ against semi-observationally-based data in box plots and
330 Taylor diagrams (Taylor, 2001) for the evaluation of the various model experiments against semi-observationally-
331 based estimates (Ridley et al., 2016; Adebisi et al., 2020) to provide a concise statistical summary of the bias,
332 correlation coefficient, root mean square errors, and the ratio of standard deviation (Fig. 2, Tables S1 and S2).
333 IMPACT-Sphere-Mineral-V83 (E1) simulations resulted in a significant underestimation of the global and annual
334 mean of $DAOD_{550}$ (0.023) (Fig. 2 and Table 3). After considering the dust asphericity for spectral optical properties,
335 we adjusted IMPACT-simulated dust loads against the constraints on dust load from the DustCOMM data set. This

336 adjustment led the simulated total dust load to increase from 25 Tg (E1) to 32 Tg (E2), which addressed the issue of
337 coarse dust underestimation and fine dust overestimation by the model (Fig. 3, Table 3). Consequently, the global and
338 annual mean of DAOD₅₅₀ from DustCOMM-Asphere-DB19-V83 (E2) simulation (0.029) fell within the range in the
339 semi-observationally-based estimate (0.030 ± 0.005) (Ridley et al., 2016) (Table 3). We found that the agreement in
340 the median with the semi-observationally-based estimate (0.127) was improved from IMPACT-Sphere-Mineral-DB17
341 (0.049) to DustCOMM-Asphere-DB19-V83 (0.117) (solid line within the box mark in Fig. 2d). We also found higher
342 DAOD₅₅₀ from E2 than E1 over East Asia and Bodele/Sudan in winter (Fig. 2, Table S2). The better agreement
343 suggested that DustCOMM-Asphere-DB19-V83 (E2) simulation was reasonably constrained by the DAOD₅₅₀ (Ridley
344 et al., 2016; Adebisi et al., 2020).

345 **3.2 Dust SW radiative effect efficiency**

346 Modeled estimates of clear-sky dust SW radiative effect efficiencies ($\text{W} \cdot \text{m}^{-2} \text{DAOD}_{550}^{-1}$) at the surface (Table
347 S3) and TOA (Table S4) were compared with estimates reported by regional studies based on satellite observations
348 over the North Africa and the North Atlantic (Fig. 4). Sensitivity simulations demonstrated that the radiative effect
349 efficiency strongly depended on the particle size, refractive index, and particle shape (Fig. 4). The adjustment of size-
350 resolved dust concentration and shape with the same refractive index led to overestimates of the SW radiative effect
351 efficiencies against semi-observationally-based data at TOA (from E1 to E6 in Fig. 4h), because coarser dust absorbs
352 more SW radiation efficiently than finer particles. Subsequently, the use of less absorptive SW refractive index with
353 DustCOMM-Asphere-DB19-V83 (E2) simulations led to a better agreement (from E6 to E2 in Fig. 4). On the other
354 hand, the use of much less (10% percentile) absorptive SW refractive index from DustCOMM-Asphere-Less-More
355 (E7) simulation deteriorated the agreement due to the underestimate of cooling at the surface (Fig. 4g). In contrast,
356 the use of a more absorptive SW refractive index from DustCOMM-Asphere-Mineral-V83 (E6) improved the
357 agreement at the surface. However, the semi-observationally-based estimates of diurnally averaged radiative effect
358 efficiency at the surface were derived from extrapolation of the instantaneous values, which would affect the
359 comparison due to differences in the methodologies between dust models (section 2.6). The differences in the model-
360 based estimates of radiative effect efficiency might arise from different data sets of the refractive index, size
361 distribution, and particle shape (Song et al., 2018).

362 **3.3 Dust LW radiative effect efficiency**

363 Modeled estimates of clear-sky dust LW (Fig. 5) radiative effect efficiencies ($\text{W} \cdot \text{m}^{-2} \text{DAOD}_{550}^{-1}$) at the surface
364 (Table S5) and TOA (Table S6) were compared with estimates reported by regional studies based on satellite

365 observations over North Africa and the North Atlantic. Sensitivity simulations demonstrated that the radiative effect
366 efficiency strongly depended on the particle size, refractive index, and particle shape (Fig. 5). Both the IMPACT-
367 Sphere-Mineral-V83 (E1) and DustCOMM-Asphere-DB19-V83 (E2) simulations yielded better agreement with semi-
368 observationally-based data at the surface and TOA, compared to the less absorptive LW dust refractive indices (E3,
369 E4, E5, and E7) (Fig. 5). The relatively high LW radiative effect efficiencies over western Africa were also consistent
370 with the semi-observationally-based data. On the other hand, the relatively low LW radiative effect efficiencies were
371 found over eastern Africa. Moving toward the northeastern side of the region, however, the associated uncertainties
372 in the semi-observationally-based values increased (Brindley and Russell 2009). The dust LW radiative effect
373 efficiency depends strongly on the vertical profile of dust concentration, temperature, and water vapor, which would
374 affect the comparison due to a high variability in these factors (section 2.6).

375 **3.4 Less atmospheric radiative heating by dust due to the synergy of coarser size and aspherical shape**

376 The Saharan dust cools the ground surface by reducing the solar radiation reaching the surface and warms the
377 atmosphere by absorbing solar radiation (Fig. 6). On the other hand, thermal emission by dust warms the surface and
378 cools the atmosphere (Fig. 7). Our sensitivity simulations showed that the annually averaged net instantaneous
379 radiative effect due to mineral aerosol (NET) ranged from -0.48 (DustCOMM-Asphere-Less-Less) to $+0.25$
380 (DustCOMM-Asphere-Mineral-V83) $\text{W}\cdot\text{m}^{-2}$ at TOA (Table 5). The net RE from both the IMPACT-Sphere-Mineral-
381 V83 ($-0.00 \text{ W}\cdot\text{m}^{-2}$) and DustCOMM-Asphere-DB19-V83 ($-0.08 \text{ W}\cdot\text{m}^{-2}$) simulations resulted within 98%
382 confidential interval of DustCOMM data set (-0.27 to $0.14 \text{ W}\cdot\text{m}^{-2}$).

383 The SW RE by dust outweighs the LW warming effect at the surface in the IMPACT-Sphere-Mineral-V83
384 (E1) simulation (Fig. 8). Consequently, the highly absorbing dust could play an important role in the aerosol radiative
385 forcing for the climate models to alter the West African monsoon, with the radiative heating concentrated in the dust
386 layer (Miller et al., 2004b; Lau et al., 2009). Our model results of dust RE from DustCOMM-Asphere-DB19-V83
387 (E2) simulation, however, suggested that the surface warming was substantially enhanced near the strong dust source
388 regions ($-0.23 \text{ W}\cdot\text{m}^{-2}$ on a global scale) (Fig. 8), compared to the IMPACT-Sphere-Mineral-V83 simulation (-0.60
389 $\text{W}\cdot\text{m}^{-2}$ on a global scale). Thus, our results demonstrated that the atmospheric radiative heating by mineral dust was
390 substantially reduced for DustCOMM-Asphere-DB19-V83 (E2) simulation ($0.15 \text{ W}\cdot\text{m}^{-2}$), compared to the IMPACT-
391 Sphere-Mineral-V83 (E1) simulation ($0.59 \text{ W}\cdot\text{m}^{-2}$).

392 3.5 Variability of dust radiative effect in different simulations

393 To elucidate the differences in dust radiative effects between the IMPACT-Sphere-Mineral-V83 (E1) and
394 DustCOMM-Asphere-DB19-V83 (E2) simulations and to explore the variability in different previous model estimates
395 (Fig. 9), the differences in annually averaged radiative effects of mineral dust from DustCOMM-Asphere-DB19-
396 DB17 (E4) simulation were shown in Fig. 10. A slope of one in Fig. 10 represented an identical change in both the
397 surface and TOA and thus corresponded to no change in radiative heating within the atmosphere. The distances from
398 the DustCOMM-Asphere-DB19-DB17 (E4) simulation demonstrated that large uncertainties existed for the size
399 distribution and spectral optical properties. Our sensitivity simulations revealed that the DustCOMM-Asphere-DB19-
400 V83 (E2) simulation led to a similar net RE at TOA to the IMPACT-Sphere-Mineral-V83 (E1) simulation but resulted
401 in less cooling at the surface (Fig. 9). This revision can be divided into (1) the size-resolved abundance (black
402 hexagons, E3 – E4, in Fig. 10), (2) SW refractive index (red diamonds, E6 – E4, in Fig. 10), and (3) particle shape
403 (red circles, E5– E4, in Fig. 10). Additionally, we show the sensitivity of dust RE to LW refractive index (DB17),
404 which was used by both Di Biagio et al. (2020) and Balkanski et al. (2021).

405 First, at TOA, the SW RE was more sensitive to the size-resolved abundance ($-0.17 \text{ W}\cdot\text{m}^{-2}$ at the vertical axis
406 of black hexagon in Fig. 10a), compared to LW ($0.00 \text{ W}\cdot\text{m}^{-2}$ at the vertical axis of black hexagon in Fig. 10b). Second,
407 this less SW cooling effect with coarser dust (E3 – E4) was partially compensated for by more SW cooling with the
408 use of the less absorptive SW refractive index (E4: $-0.32 \text{ W}\cdot\text{m}^{-2}$) than E6 ($0.02 \text{ W}\cdot\text{m}^{-2}$). Thirdly, the sensitivity of
409 SW RE to dust asphericity was rather minor ($0.04 \text{ W}\cdot\text{m}^{-2}$ at the vertical axis of red circle in Fig. 10a), partly because
410 the lower DAOD was compensated for by the lower asymmetry parameter of spherical dust, which enhanced the
411 amount of radiation scattered backward to space (Räisänen et al., 2013; Colarco et al., 2014). The partial compensation
412 led to a small enhancement of SW RE for the IMPACT-Sphere-Mineral-V83 (E1) simulation and thus the resulting
413 similar net RE to DustCOMM-Asphere-DB19-V83 (E2) at TOA (Fig. 9).

414 In contrast, at the surface, our sensitivity simulations demonstrated substantially different responses in the RE,
415 mostly because of LW warming effects (Fig. 9). The enhanced LW warming by coarser dust ($-0.08 \text{ W}\cdot\text{m}^{-2}$ at the
416 horizontal axis of black hexagon in Fig. 10b) was accompanied by the asphericity ($-0.15 \text{ W}\cdot\text{m}^{-2}$ at the horizontal axis
417 of red circle in Fig. 10b), because the enhancement of the absorption fraction of extinction due to asphericity was
418 larger at coarser size. The enhanced LW warming effects of each as well as the synergy was further amplified using
419 the more absorptive LW dust refractive index (Volz, 1983) (at the horizontal axis of red diamond in Fig. 10b). As a
420 result, our sensitivity simulations revealed that substantially less dust absorption at LW due to the underestimation of
421 the coarse dust load and the assumption of the spherical shape (IMPACT-Sphere-Mineral-V83) contributed to the less
422 surface warming, compared to DustCOMM-Asphere-DB19-V83 (Fig. 9).

423 A relatively good agreement of net RE by dust at TOA with both Di Biagio et al. (2020) ($-0.06 \text{ W}\cdot\text{m}^{-2}$) and
424 Balkanski et al. (2021) ($-0.02 \text{ W}\cdot\text{m}^{-2}$) could be obtained from both the IMPACT-Sphere-Mineral-V83 (E1: -0.00
425 $\text{W}\cdot\text{m}^{-2}$) and DustCOMM-Asphere-DB19-V83 (E2: $-0.08 \text{ W}\cdot\text{m}^{-2}$) simulations (Fig. 9 and Table 5). On the other hand,
426 our modeled dust net RE at the surface from DustCOMM-Asphere-DB19-V83 (E2: $-0.23 \text{ W}\cdot\text{m}^{-2}$) indicated much
427 less cooling than Di Biagio et al. (2020) ($-0.63 \text{ W}\cdot\text{m}^{-2}$), Balkanski et al. (2021) ($-1.01 \text{ W}\cdot\text{m}^{-2}$), and IMPACT-Sphere-
428 Mineral-V83 (E1: $-0.60 \text{ W}\cdot\text{m}^{-2}$). The synergy of coarser size and aspherical dust could contribute to the less surface
429 cooling of the DustCOMM-Asphere-DB19-V83 (E2), because of enhanced LW warming. At the same time, both Di
430 Biagio et al. (2020) and Balkanski et al. (2021) used DB17 and considered dust with diameters more than $20 \mu\text{m}$.
431 Thus, the more absorptive LW dust refractive index (V83, E6 for LW: $1.00 \text{ W}\cdot\text{m}^{-2}$) than DB17 (E4 for LW: 0.58
432 $\text{W}\cdot\text{m}^{-2}$) (E6 – E4 for LW: $0.42 \text{ W}\cdot\text{m}^{-2}$ at the horizontal axis of red diamond in Fig. 10b) could also contribute to the
433 less surface cooling, which might be partially compensated for in our model by the omission of dust with diameters
434 more than $20 \mu\text{m}$. Consequently, our estimate of atmospheric radiative heating by dust from DustCOMM-Asphere-
435 DB19-V83 (E2: $0.15 \text{ W}\cdot\text{m}^{-2}$) was lower than Di Biagio et al. (2020) ($0.63 \text{ W}\cdot\text{m}^{-2}$), Balkanski et al. (2021) (0.98
436 $\text{W}\cdot\text{m}^{-2}$), and IMPACT-Sphere-Mineral-V83 (E1: $0.59 \text{ W}\cdot\text{m}^{-2}$). Additionally, the hot and dry climate over brighter
437 desert surface exaggerates differences in RE at the surface between the models (Miller et al., 2014). The low humidity
438 allows dust particles to absorb LW radiation with reduced competition from water vapor, while high temperatures
439 within the boundary layer increase downward thermal emission by dust (Liao and Seinfeld, 1998). The reduction of
440 fine dust load after the adjustment leads to underestimates of the SW cooling at TOA. To improve agreement against
441 semi-observationally-based estimate of the radiative effect efficiency at TOA, the less absorptive SW dust refractive
442 index is required for coarser aspherical dust. Thus, uncertainties in the size-resolved dust concentration, particle shape,
443 and refractive index contribute to the diversity in the simulated dust RE at the surface.

444 4. Conclusions

445 Accurate estimates of the size-resolved dust abundance, their spectral optical properties, and their seasonality
446 in regional and vertical scales provide a step towards a more reliable projection of the climatic feedback of mineral
447 aerosols. The radiative effect efficiency depends on numerous variables in model simulations, including the spatial
448 distribution and temporal variation of size-resolved dust concentrations, the mass extinction coefficient, single
449 scattering albedo, and asymmetry parameter of dust. Since the models typically underestimate the coarse dust load
450 and overestimate the fine dust load, the sensitivity to the aerosol absorptivity might be considerably different from
451 previous studies. Thus, the model results should be re-evaluated against semi-observationally-based estimate of the
452 DAOD_{550} and dust radiative effect efficiency.

453 We improved the accuracy of the simulations by adjusting the bias in size-resolved aspherical dust
454 concentration with the DustCOMM data set. Alternatively, dust mineralogy might contribute to the underestimation
455 of modeled aerosol absorption compared to satellite observations (Lacagnina et al., 2015). This enhanced aerosol
456 absorption was examined by specifying the mineralogy with varying amounts of light-absorbing Fe oxides for SW.
457 The better agreement with the semi-observationally-based data of dust radiative effect efficiency was obtained using
458 the less absorptive SW dust refractive indices after the adjustments of dust sizes and shapes.

459 The diversity of modeled dust net RE at the surface ($-1.64 \text{ W}\cdot\text{m}^{-2}$ to $-0.20 \text{ W}\cdot\text{m}^{-2}$) is much larger than at TOA
460 ($-0.01 \text{ W}\cdot\text{m}^{-2}$ to $-0.60 \text{ W}\cdot\text{m}^{-2}$), partly because the refractive index is optimized to obtain reasonable agreement against
461 satellite observations of TOA radiation flux (e.g., CERES). The uncertainties in the size-resolved dust concentration,
462 particle shape, and refractive index contribute to the model diversity at the surface. DustCOMM-Asphere-DB19-V83
463 (E2) simulation resulted in less cooling at the surface by the synergy of coarser size and aspherical shape, compared
464 to IMPACT-Sphere-Mineral-V83 (E1) simulation (-0.23 vs. $-0.60 \text{ W}\cdot\text{m}^{-2}$ on a global scale). Consequently, the
465 atmospheric heating due to mineral dust was substantially reduced for the DustCOMM-Asphere-DB19-V83 (E2)
466 simulation ($0.15 \text{ W}\cdot\text{m}^{-2}$), compared to the intensified atmospheric heating from the IMPACT-Sphere-Mineral-V83
467 (E1) simulation ($0.59 \text{ W}\cdot\text{m}^{-2}$). The less intensified atmospheric heating due to mineral dust could substantially modify
468 the vertical temperature profile in Earth system models and thus has important implications for the projection of dust
469 feedback near the major source regions in the past and future climate changes (Kok et al., 2018). More accurate
470 estimates of semi-observationally-based dust SW and LW radiative effect efficiencies over strong dust source regions
471 are needed to narrow the uncertainty in the RE.

472 Currently, the model did not include dust particles above $20 \mu\text{m}$, but a substantial fraction of airborne dust near
473 source regions may be above this threshold (Ryder et al., 2019). Moreover, such large particles can be transported to
474 higher altitudes and longer distances than the model prediction. The higher the dust layer resides, the larger the dust
475 LW RE at TOA is estimated under the clear-sky conditions (Liao and Seinfeld, 1998). Marine sediment traps, which
476 are located underneath the main Saharan dust plume in the Atlantic Ocean, suggest that giant particles are dominated
477 by platy mica and rounded quartz particles (van der Does et al., 2016). Thus, mineral composition of the giant particles
478 could be different from the aerosol samples generated from soils in the laboratory by Di Biagio et al. (2017), which
479 may reflect less absorbing LW refractive index of DB17 than V83. Indeed, the dust sample was collected for V83
480 from rainwater after strong wind. On the other hand, the contribution of the LW scattering might be underestimated
481 in the models, as Di Biagio et al. (2020) noted that the adjustment factor was estimated for dust of diameter less than
482 $10 \mu\text{m}$ and thus might be a lower approximation of the LW scattering by coarse dust. Therefore, a better understanding
483 of the effect of such large particles beyond $20 \mu\text{m}$ and mineralogical composition on radiation balance remains a topic

484 of active research, given their potential to amplify the warming of the climate system. In such an extreme case as the
485 “Godzilla” dust storm over the North Africa and the tropical Atlantic in June 2020 (Francis et al., 2020), the dust
486 loading could be larger than that examined for this study, and our estimates of the warming effects might be
487 conservative during such events. However, to keep the giant particles in the atmosphere, the modeled deposition fluxes
488 should be reduced from the current model. Therefore, models should improve their ability to capture the evolution of
489 the dust size distribution as the plumes move downwind of the source regions.

490 **Code availability.**

491 The source code of the RRTMG has been obtained from the website at https://github.com/AER-RC/RRTMG_LW and
492 https://github.com/AER-RC/RRTMG_SW. The source code of the Kramers-Kronig relations has been obtained from
493 the website at [https://www.mathworks.com/matlabcentral/fileexchange/8135-tools-for-data-analysis-in-optics-
494 acoustics-signal-processing](https://www.mathworks.com/matlabcentral/fileexchange/8135-tools-for-data-analysis-in-optics-acoustics-signal-processing). The source code of the Taylor diagram has been obtained from the web site at
495 <https://www.mathworks.com/matlabcentral/fileexchange/20559-taylor-diagram>.

496 **Data availability.**

497 SMAP data have been obtained from the website at <https://nsidc.org/data/smap/smap-data.html>. MODIS land data
498 have been retrieved from the website at <https://ladsweb.modaps.eosdis.nasa.gov/>. MERRA-2 data have been provided
499 by the Global Modeling and Assimilation Office (GMAO) at NASA Goddard Space Flight Center
500 (<https://disc.gsfc.nasa.gov/datasets/>). The DustCOMM data are available at <https://dustcomm.atmos.ucla.edu/>. The
501 datasets supporting the conclusions of this article are included within the article and its supplement file.

502 **Supplement.**

503 The supplement related to this article is available online at:

504 **Author contributions.**

505 AI and JFK initiated the modeling collaboration with semi-observationally-based data sets. AI carried out the
506 modeling study. AAA, YH, JFK contributed semi-observationally-based data sets of DustCOMM and asphericity
507 factor. All authors read and approved the final manuscript.

508 **Competing interests.**

509 The authors declare that they have no competing interests.

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810 **Figure captions**

811 Figure 1. Imaginary part of the refractive index at (a) 0.52 μm , (b) SW, (c) 9.7 μm , and (d) LW. The refractive
812 index obtained from 19 samples was aggregated into 9 main source regions and the arithmetic mean was
813 calculated for each source region (Di Biagio et al., 2017, 2019). The global mean is used for others. The
814 coordinates of the nine source regions were: (S1) western North Africa (20°W – 7.5°E; 18°N – 37.5°N), (S2)
815 eastern North Africa (7.5°E – 35°E; 18°N – 37.5°N), (S3) the Sahel (20°W – 35°E; 0°N – 18°N), (S4) Middle
816 East / Central Asia (30°E – 70°E for 0°N – 35°N, and 30°E – 75°E for 35°N – 50°N), (S5) East Asia (70°E
817 – 120°E; 35°N – 50°N), (S6) North America (130°W – 80°W; 20°N – 45°N), (S7) Australia (110°E – 160°E;
818 10°S – 40°S), (S8) South America (80°W – 20°W; 0°S – 60°S), and (S9) Southern Africa (0°E – 40°E; 0°S
819 – 40°S).

820 Figure 2. The model better reproduced semi-observationally-based data of DAOD₅₅₀ after adjusting the size-
821 resolved dust load with DustCOMM and considering the dust asphericity. (a) semi-observationally-based
822 estimates of the DAOD₅₅₀ were averaged over 2004–2008 (Ridley et al., 2016; Adebisi et al., 2020). The
823 annually averaged model results were shown for (b) DustCOMM-Asphere-DB19-V83 (E2) and (c) the
824 differences between IMPACT-Sphere-Mineral-V83 (E1) and E2 simulations. (d) Comparison of seasonally
825 averaged DAOD₅₅₀ for semi-observationally-based (SOB) data, E1, E2, IMPACT-Asphere-DB19-DB17
826 (E3), and DustCOMM-Sphere-DB19-DB17 (E5). The square symbol represents the mean. The solid line
827 within the box mark shows the median. The boundaries of the box mark the 25th and 75th percentiles. The
828 whiskers above and below the box indicate the 1.5 × interquartile range, and the points indicate the outside
829 of the range. (e) Taylor diagram summarizing the statistics of the comparison against the seasonally averaged
830 regional DAOD₅₅₀ for the different experiments. The horizontal axis shows the standard deviation of the data
831 set or model prediction, the curved axis shows the correlation, and the green dashed lines denote the root-
832 mean-squared errors between the semi-observationally-based data and the model predictions. As such, the
833 distance between the semi-observationally-based data and the model predictions is a measure of the model's
834 ability to reproduce the spatiotemporal variability in the semi-observationally-based data. The coordinates
835 and the values of DAOD₅₅₀ at the 15 regions (marked in Fig. 3a) in summer were listed in Table S1. The
836 comparison for other seasons was presented in Table S2.

837 Figure 3. Model-simulated dust loads at fine (smaller than 2.5 μm of diameter) and coarse size ranges (larger
838 than 2.5 μm of diameter) before and after adjusting the size-resolved dust load with DustCOMM. Results
839 were shown for (a) fine dust from DustCOMM, (b) fine dust from IMPACT-Sphere-Mineral-V83 (E1), (c)
840 fine dust from DustCOMM-Asphere-DB19-V83 (E2), (d) coarse dust from DustCOMM, (e) coarse dust from

841 E1, and (f) coarse dust from E2 simulations. The parentheses represented the global dust burden (T_g). The
842 values of dust load at each bin were listed in Table 3.

843 Figure 4. Dust clear-sky SW radiative effect efficiency ($W \cdot m^{-2} DAOD^{-1}$). Semi-observationally-based data
844 at (a) the surface and (b) TOA were based on satellite observations (Yang et al. 2009; Li et al., 2004; Song
845 et al., 2018; Christopher and Jones, 2007). The model results were shown for (c) and (d) IMPACT-Sphere-
846 Mineral-V83 (E1), and (e) and (f) DustCOMM-Asphere-DB19-V83 (E2) simulations at the surface and TOA,
847 respectively. Comparison of seasonally averaged SW radiative effect efficiency for semi-observationally-
848 based (SOB) data and the different experiments at (g) the surface and (h) TOA. The square symbol represents
849 the mean. The solid line within the box mark shows the median. The boundaries of the box mark the 25th
850 and 75th percentiles. The whiskers above and below the box indicate the $1.5 \times$ interquartile range, and the
851 points indicate the outside of the range. Taylor diagram summarizing the statistics of the comparison against
852 the seasonally averaged regional SW radiative effect efficiency for the different experiments at (i) the surface
853 and (j) TOA. The horizontal axis shows the standard deviation of the data set or model prediction, the curved
854 axis shows the correlation, and the green dashed lines denote the root-mean-squared errors between the semi-
855 observationally-based data and the model predictions. As such, the distance between the semi-
856 observationally-based data and the model predictions is a measure of the model's ability to reproduce the
857 spatiotemporal variability in the semi-observationally-based data. The regionally averaged values were listed
858 in Tables S3 and S4 at the surface and TOA, respectively.

859 Figure 5. Dust clear-sky LW radiative effect efficiency ($W \cdot m^{-2} DAOD^{-1}$). Semi-observationally-based
860 estimates at (a) surface and (b) TOA were based on satellite observations (Song et al., 2018; Christopher and
861 Jones, 2007; Zhang and Christopher, 2003; Brindley and Russell, 2009; Yang et al., 2009). The model results
862 were shown for (c) and (d) IMPACT-Sphere-Mineral-V83 (E1), and (e) and (f) DustCOMM-Asphere-DB19-
863 V83 (E2) simulations at the surface and TOA, respectively. Comparison of seasonally averaged LW radiative
864 effect efficiency for semi-observationally-based (SOB) data and the different experiments at (g) the surface
865 and (h) TOA. The square symbol represents the mean. The solid line within the box mark shows the median.
866 The boundaries of the box mark the 25th and 75th percentiles. The whiskers above and below the box indicate
867 the $1.5 \times$ interquartile range, and the points indicate the outside of the range. Taylor diagram summarizing
868 the statistics of the comparison against the seasonally averaged regional SW radiative effect efficiency for
869 the different experiments at (i) the surface and (j) TOA. The horizontal axis shows the standard deviation of
870 the data set or model prediction, the curved axis shows the correlation, and the green dashed lines denote the
871 root-mean-squared errors between the semi-observationally-based data and the model predictions. As such,

872 the distance between the semi-observationally-based data and the model predictions is a measure of the
873 model's ability to reproduce the spatiotemporal variability in the semi-observationally-based data. The
874 regionally averaged values were listed in Tables S5 and S6 at the surface and TOA, respectively.

875 Figure 6. Dust SW radiative effect ($W \cdot m^{-2}$) and radiative heating of the atmosphere (i.e., the subtraction of
876 radiative effects from TOA to the surface in unit of $W \cdot m^{-2}$). The model results were shown for the simulations
877 for (a) IMPACT-Sphere-Mineral-V83 (E1) at the surface, (b) DustCOMM-Asphere-DB19-V83 (E2) at the
878 surface, (c) E1 in atmospheric column, (d) E2 in atmospheric column, (e) E1 at TOA, and (f) E2 simulations
879 at TOA. The numbers in parentheses represented the global mean.

880 Figure 7. Dust LW radiative effect ($W \cdot m^{-2}$) and radiative heating of the atmosphere (i.e., the subtraction of
881 radiative effects from TOA to the surface in unit of $W \cdot m^{-2}$). The model results were shown for the simulations
882 for (a) IMPACT-Sphere-Mineral-V83 (E1) at the surface, (b) DustCOMM-Asphere-DB19-V83 (E2) at the
883 surface, (c) E1 in atmospheric column, (d) E2 in atmospheric column, (e) E1 at TOA, and (f) E2 simulations
884 at TOA. The numbers in parentheses represented the global mean.

885 Figure 8. Dust net radiative effect ($W \cdot m^{-2}$) and radiative heating of the atmosphere (i.e., the subtraction of
886 radiative effects from TOA to the surface in unit of $W \cdot m^{-2}$). The model results were shown for the simulations
887 for (a) IMPACT-Sphere-Mineral-V83 (E1) at the surface, (b) DustCOMM-Asphere-DB19-V83 (E2) at the
888 surface, (c) E1 in atmospheric column, (d) E2 in atmospheric column, (e) E1 at TOA, and (f) E2 simulations
889 at TOA. The numbers in parentheses represented the global mean.

890 Figure 9. Variability of dust radiative effect ($W \cdot m^{-2}$) in different model simulations at the surface and TOA
891 for (a) total dust SW, (b) total dust LW, and (c) total dust NET. The annually averaged values were listed in
892 Table 5.

893 Figure 10. Radiative effect ($W \cdot m^{-2}$) of mineral dust due to various aerosol absorptivity at the surface and TOA
894 for (a) total dust SW, (b) total dust LW, and (c) total dust NET. The annually averaged values were listed in
895 Table 5. The dashed line represented a 1 : 1 correspondence and corresponded to no change in radiative
896 heating within the atmosphere.

Table 1. Summary of ten combinations of different numerical experiments compared in this study.

Number	Experiment	Size-resolved dust	Sphericity	SW refractive index	LW refractive index
E1	IMPACT-Sphere-Mineral-V83	IMPACT	Sphere	Mineralogical map ^d	Volz (1983)
E2 ^a	DustCOMM-Asphere-DB19-V83	DustCOMM ^b	Asphere ^c	Di Biagio et al. (2019)	Volz (1983)
E3	IMPACT-Asphere-DB19-DB17	IMPACT	Asphere ^c	Di Biagio et al. (2019)	Di Biagio et al. (2017)
E4	DustCOMM-Asphere-DB19-DB17	DustCOMM ^b	Asphere ^c	Di Biagio et al. (2019)	Di Biagio et al. (2017)
E5	DustCOMM-Sphere-DB19-DB17	DustCOMM ^b	Sphere	Di Biagio et al. (2019)	Di Biagio et al. (2017)
E6	DustCOMM-Asphere-Mineral-V83	DustCOMM ^b	Asphere ^c	Mineralogical map ^d	Volz (1983)
E7	DustCOMM-Asphere-Less-More	DustCOMM ^b	Asphere ^c	Less SW ^e	More LW ^g
E8	DustCOMM-Asphere-Less-Less	DustCOMM ^b	Asphere ^c	Less SW ^e	Less LW ^h
E9	DustCOMM-Asphere-More-More	DustCOMM ^b	Asphere ^c	More SW ^f	More LW ^g
E10	DustCOMM-Asphere-More-Less	DustCOMM ^b	Asphere ^c	More SW ^f	Less LW ^h

^aCombination of DustCOMM-Asphere-DB19-DB17 (E4) for SW and DustCOMM-Asphere-Mineral-V83 (E6) for LW.

^bSize-resolved dust concentration was adjusted with semi-observationally-based estimate (Adebiyi & Kok, 2020).

^cDust asphericity was considered in calculating the optical properties, which further assumed internal mixing of minerals (Huang et al., 2021) using a volume-weighted mixture for each size bin.

^dMineralogical composition of dust aerosol for each size was prescribed at emission by mineralogical map (Journet et al., 2014; Ito and Shi 2016). The more absorptive SW refractive indices (Bedidi and Cervelle, 1993; Stegmann & Yang, 2017; Long et al., 1993) were used for mineral dust, compared to the less absorptive global mean data set (Di Biagio et al., 2019).

^eLess absorptive SW refractive indices were calculated by varying the values of the imaginary parts of the refractive index within the range of values from Di Biagio et al. (2019) (10% percentile).

^fMore absorptive SW refractive indices were calculated by varying the values of the imaginary parts of the refractive index within the range of values from Di Biagio et al. (2019) (90% percentile).

^gMore absorptive LW refractive indices were calculated by varying the values of the imaginary parts of the refractive index within the range of values from Di Biagio et al. (2017) (90% percentile).

^hLess absorptive LW refractive indices were calculated by varying the values of the imaginary parts of the refractive index within the range of values from Di Biagio et al. (2017) (10% percentile).

Table 2. Summary of radiative effects estimated in this study.

SW radiative effect	LW radiative effect	Difference
Less absorptive SW, coarser particle size, & aspherical shape	Coarser particle size & aspherical shape	E2 – E1
Less absorptive SW & aspherical shape	Less absorptive LW & aspherical shape	E3 – E1
Size-resolved dust abundance	Size-resolved dust abundance	E3 – E4
Aspherical shape	Aspherical shape	E5 – E4
Mineralogical variability in refractive index (more absorptive SW)	Mineralogical variability in refractive index (more absorptive LW)	E6 – E4
Less absorptive SW (10% percentile)	More absorptive LW (90% percentile)	E7 – E4
Less absorptive SW (10% percentile)	Less absorptive LW (10% percentile)	E8 – E4
More absorptive SW (90% percentile)	More absorptive LW (90% percentile)	E9 – E4
More absorptive SW (90% percentile)	Less absorptive LW (10% percentile)	E10 – E4

Table 3. Annually averages of dust load (Tg), mass extinction efficiency ($\text{m}^2\cdot\text{g}^{-1}$), and DAOD_{550} at each bin on a global scale. The size-resolved dust concentration and shape in IMPACT-Sphere-Mineral-V83 (E1) simulation was adjusted to DustCOMM in DustCOMM-Asphere-DB19-V83 (E2) simulation. At the same time, we maintained the consideration of asphericity on the gravitational velocity and kept the dust concentrations unaltered between IMPACT-Sphere-Mineral-V83 (E1) and IMPACT-Asphere-DB19-DB17 simulations (E3).

Dust size bin	Dust load			Mass extinction efficiency				DAOD_{550}			
	E1	E2	DustCOMM	E1	E2	E3	DustCOMM	E1	E2	E3	DustCOMM
Bin 1 ^a	1.2	0.8	1.2 ± 0.7	2.11	3.41	3.33	3.06	0.0050	0.0055	0.0078	0.0070
Bin 2 (1.26–2.5 μm)	4.7	2.6	3.5 ± 2.1	0.73	1.25	1.21	1.22	0.0067	0.0064	0.0111	0.0084
Bin 3 (2.5–5 μm)	8.2	6.2	6.8 ± 3.8	0.37	0.59	0.57	0.57	0.0060	0.0071	0.0092	0.0077
Bin 4 (5–20 μm)	10.9	22.2	16.8 ± 9.0	0.23	0.24	0.29	0.19	0.0050	0.0104	0.0063	0.0063
Sum of 4 bins	25.0	31.8	28.4 ± 15.5	0.46	0.47	0.70	0.53	0.0227	0.0295	0.0345	0.0294

^aBin 1 in IMPACT-Sphere-Mineral-V83 (E1) is 0.1–1.26 μm , whereas bin1 in DustCOMM-Asphere-DB19-V83 (E2) and DustCOMM is 0.2–1.26 μm .

Table 4. Semi-observationally-based data set of clear-sky dust radiative effect efficiency at the surface and TOA.

Number	Region name	Season	Region coordinates	Aerosol type selection	AOD data
R1 ^a	Sahara Desert	Summer	15°–30°N, 10°W–30°E	No selection	OMI-MISR
R2 ^b	Tropical Atlantic	Summer	15°–25°N, 45°–15°W	MODIS effective radius peaks 0.8–0.9 μm	MODIS
R3 ^c	Tropical Atlantic	Summer	10°–30°N, 45°–20°W	CALIOP dust and polluted dust	CERES-CALIPSO-CloudSat-MODIS
R4 ^d	Atlantic Ocean	Summer	0°–30°N, 60°–10°W	Dust detection based on DAOD ₅₅₀ and fraction	MODIS
R5 ^{e,f}	North Africa	Summer	15°–35°N, 18°W–40°E	No selection	MISR ^e or SEVIRI ^f
R6 ^{e,f}	West Africa	Summer	16°–28°N, 16°–4°W	No selection	MISR ^e or SEVIRI ^f
R7 ^{e,f}	Niger-Chad	Summer	15°–20°N, 15°–22°E	No selection	MISR ^e or SEVIRI ^f
R8 ^{e,f}	Sudan	Summer	15°–22°N, 22°–36°E	No selection	MISR ^e or SEVIRI ^f
R9 ^{e,f}	Egypt-Israel	Summer	23°–32°N, 23°–35°E	No selection	MISR ^e or SEVIRI ^f
R10 ^{e,f}	North Libya	Summer	27°–33°N, 15°–25°E	No selection	MISR ^e or SEVIRI ^f
R11 ^{e,f}	South Libya	Summer	23°–27°N, 15°–25°E	No selection	MISR ^e or SEVIRI ^f
R12 ^g	Mediterranean	Summer	35.5°N, 12.6°E	Dust detection based on optical property	Ground-based measurements
R13 ^h	Cape Verde	Summer	16.7°N, 22.9°E	Dust detection based on brightness temperature	Ground-based measurements
R14 ⁱ	China	Spring	39°N, 101°E	Dust detection based on brightness temperature	Ground-based measurements

^aYang et al. (2009). ^bLi et al. (2004). ^cSong et al. (2018). ^dChristopher and Jones (2007). ^eZhang and Christopher (2003). ^fBrindley and Russell (2009). ^gDi Biagio et al. (2010). ^hHansell et al. (2010). ⁱHansell et al. (2012).

Table 5. Annually averages of short-wave (SW) ($W \cdot m^{-2}$), long-wave (LW) ($W \cdot m^{-2}$), and net radiative effect (NET) ($W \cdot m^{-2}$) at the surface, TOA, and atmospheric radiative heating on a global scale.

Number	Data	Total dust SW		Total dust LW		Total dust NET	
		TOA (surface) ^a	Atmosphere	TOA (surface) ^a	Atmosphere	TOA (surface) ^a	Atmosphere
E1	IMPACT-Sphere-Mineral-V83	-0.18 (-1.26)	1.07	+0.18 (0.66)	-0.48	-0.00 (-0.60)	0.59
E2	DustCOMM-Asphere-DB19-V83	-0.32 (-1.23)^b	0.91^b	+0.23 (1.00)^b	-0.77^b	-0.08 (-0.23)^b	0.15^b
E3	IMPACT-Asphere-DB19-DB17	-0.49 (-1.35)	0.86	+0.12 (0.50)	-0.38	-0.37 (-0.84)	0.48
E4	DustCOMM-Asphere-DB19-DB17	-0.32 (-1.23)^b	0.91^b	+0.12 (0.58)	-0.46	-0.20 (-0.65)	0.45
E5	DustCOMM-Sphere-DB19-DV17	-0.28 (-0.90)	0.62	+0.08 (0.43)	-0.34	-0.20 (-0.47)	0.28
E6	DustCOMM-Asphere-Mineral-V83	+0.02 (-1.61)	1.63	+0.23 (1.00)^b	-0.77^b	+0.25 (-0.62)	0.87
E7	DustCOMM-Asphere-Less-More	-0.54 (-0.98)	0.43	+0.16 (0.76)	-0.60	-0.38 (-0.22)	-0.16
E8	DustCOMM-Asphere-Less-Less	-0.54 (-0.98)	0.43	+0.06 (0.35)	-0.29	-0.48 (-0.36)	0.15
E9	DustCOMM-Asphere-More-More	-0.08 (-1.51)	1.43	+0.16 (0.76)	-0.60	+0.09 (-0.75)	0.84
E10	DustCOMM-Asphere-More-Less	-0.08 (-1.51)	1.43	+0.06 (0.35)	-0.29	-0.01 (-1.16)	1.15
	DustCOMM (Adebisi & Kok, 2020)	-0.59 to 0.17 ^c		+0.25 to 0.41 ^c		-0.27 to 0.14 ^c	
M1	Miller et al. (2004b)	-0.33 (-1.82)	1.49	+0.15 (0.18)	-0.03	-0.18 (-1.64)	1.46
M2	Tanaka et al. (2007)	-0.38 (-1.22)	0.84	+0.16 (0.57)	-0.41	-0.22 (-0.65)	0.43
M3	Yoshioka et al. (2007)	-0.92 (-1.59)	0.67	+0.31 (1.13)	-0.81	-0.60 (-0.46)	-0.14
M4	Takemura et al. (2009)	-0.10 (-0.38)	0.28	+0.09 (0.18)	-0.09	-0.01 (-0.20)	0.19
M5	Albani et al. (2014)	-0.38 (-1.20)	0.81	+0.15 (0.64)	-0.49	-0.23 (-0.56)	0.33
M6	Colarco et al. (2014)	-0.32 (-1.25)	0.93	+0.05 (0.30)	-0.25	-0.27 (-0.95)	0.68
M7	Di Biagio et al. (2020)	-0.29 (-1.17) ^d	0.88 ^d	+0.23 (0.48) ^d	-0.26 ^d	-0.06 (-0.69) ^d	0.63 ^d
M8	Balkanski et al. (2021)	-0.14 (-1.42)	1.28	+0.12 (0.41)	-0.29	-0.02 (-1.01)	0.98

^aThe parentheses represent the RE at the surface. ^bThe bold represents the combination of DB19 for SW and V83 for LW (i.e., DustCOMM-Asphere-DB19-V83). ^c98% confidential interval of DustCOMM data set is listed. ^dFor a comparison with our estimates, sum of single mode simulations from Di Biagio et al. (2019) is listed.

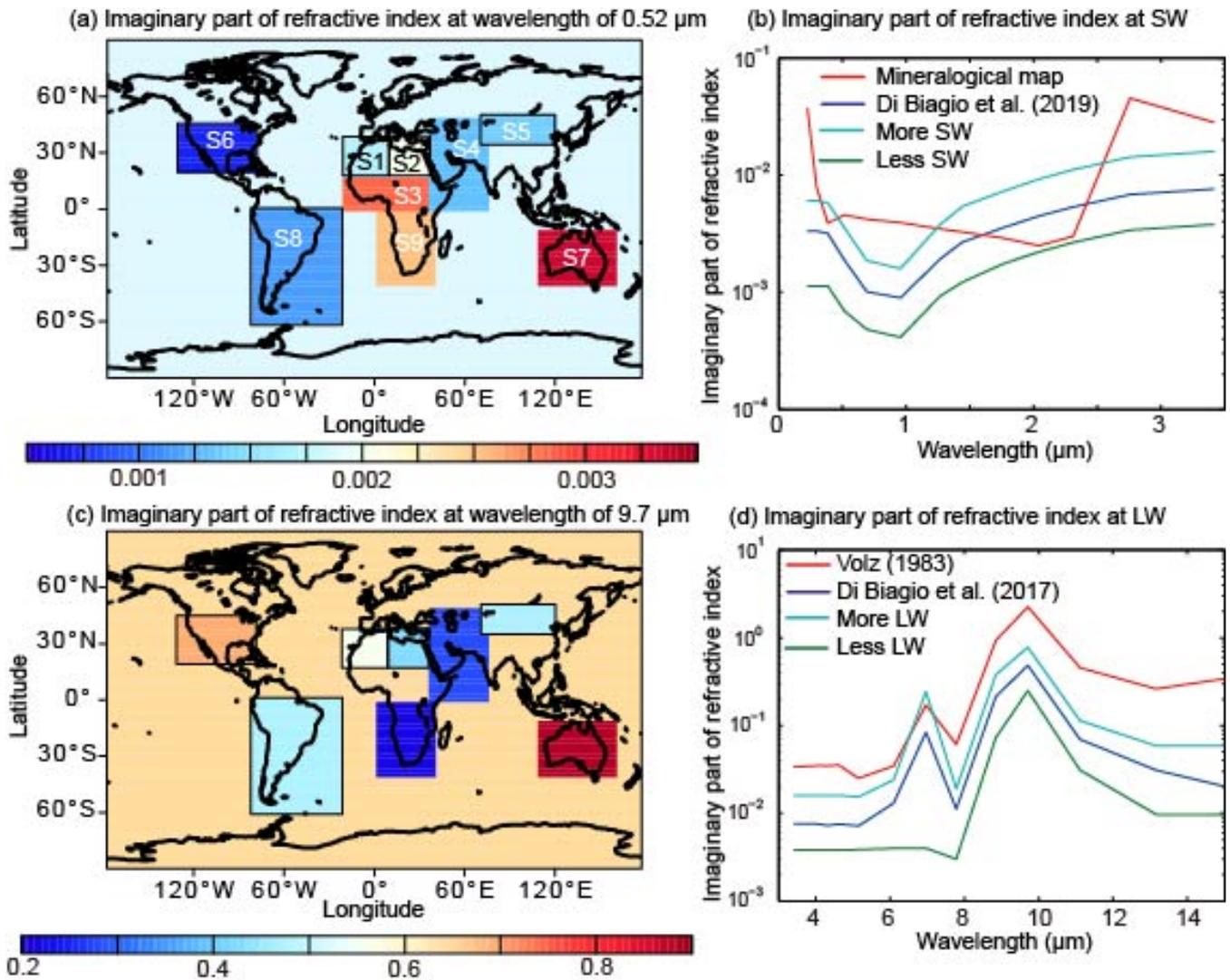


Figure 1. Imaginary part of the refractive index at (a) 0.52 μm, (b) SW, (c) 9.7 μm, and (d) LW. The refractive index obtained from 19 samples was aggregated into 9 main source regions and the arithmetic mean was calculated for each source region (Di Biagio et al., 2017, 2019). The global mean is used for others. The coordinates of the nine source regions were: (S1) western North Africa (20°W – 7.5°E; 18°N – 37.5°N), (S2) eastern North Africa (7.5°E – 35°E; 18°N – 37.5°N), (S3) the Sahel (20°W – 35°E; 0°N – 18°N), (S4) Middle East / Central Asia (30°E – 70°E for 0°N – 35°N, and 30°E – 75°E for 35°N – 50°N), (S5) East Asia (70°E – 120°E; 35°N – 50°N), (S6) North America (130°W – 80°W; 20°N – 45°N), (S7) Australia (110°E – 160°E; 10°S – 40°S), (S8) South America (80°W – 20°W; 0°S – 60°S), and (S9) Southern Africa (0°E – 40°E; 0°S – 40°S).

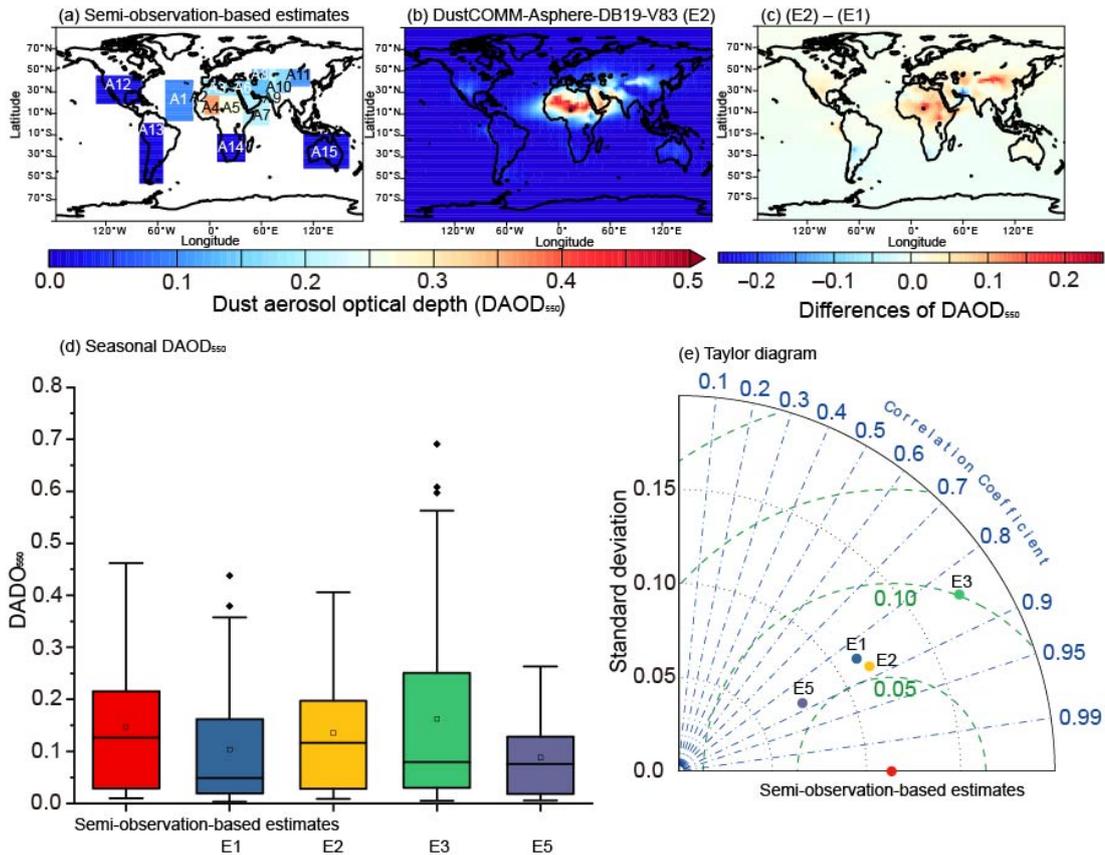


Figure 2. The model better reproduced semi-observationally-based data of DAOD₅₅₀ after adjusting the size-resolved dust load with DustCOMM and considering the dust asphericity. (a) semi-observationally-based estimates of the DAOD₅₅₀ were averaged over 2004–2008 (Ridley et al., 2016; Adebisi et al., 2020). The annually averaged model results were shown for (b) 15 DustCOMM-Asphere-DB19-V83 (E2) and (c) the differences between IMPACT-Sphere-Mineral-V83 (E1) and E2 simulations. (d) Comparison of seasonally averaged DAOD₅₅₀ for semi-observationally-based (SOB) data, E1, E2, IMPACT-Asphere-DB19-DB17 (E3), and DustCOMM-Sphere-DB19-DB17 (E5). The square symbol represents the mean. The solid line within the box mark shows the median. The boundaries of the box mark the 25th and 75th percentiles. The whiskers above and below the box indicate the $1.5 \times$ interquartile range, and the points indicate the outside of the range. (e) Taylor diagram summarizing the statistics of the comparison against the seasonally averaged regional DAOD₅₅₀ for the different experiments. The horizontal axis shows the standard deviation of the data set or model prediction, the curved axis shows the correlation, and the green dashed lines denote the root-mean-squared errors between the semi-observationally-based data and the model predictions. As such, the distance between the semi-observationally-based data and the model predictions is a measure of the model’s ability to reproduce the spatiotemporal variability in the semi-observationally-based data. The coordinates and the values of DAOD₅₅₀ at the 15 regions (marked in Fig. 2a) in summer were listed in Table S1. The comparison for other seasons was presented in Table S2.

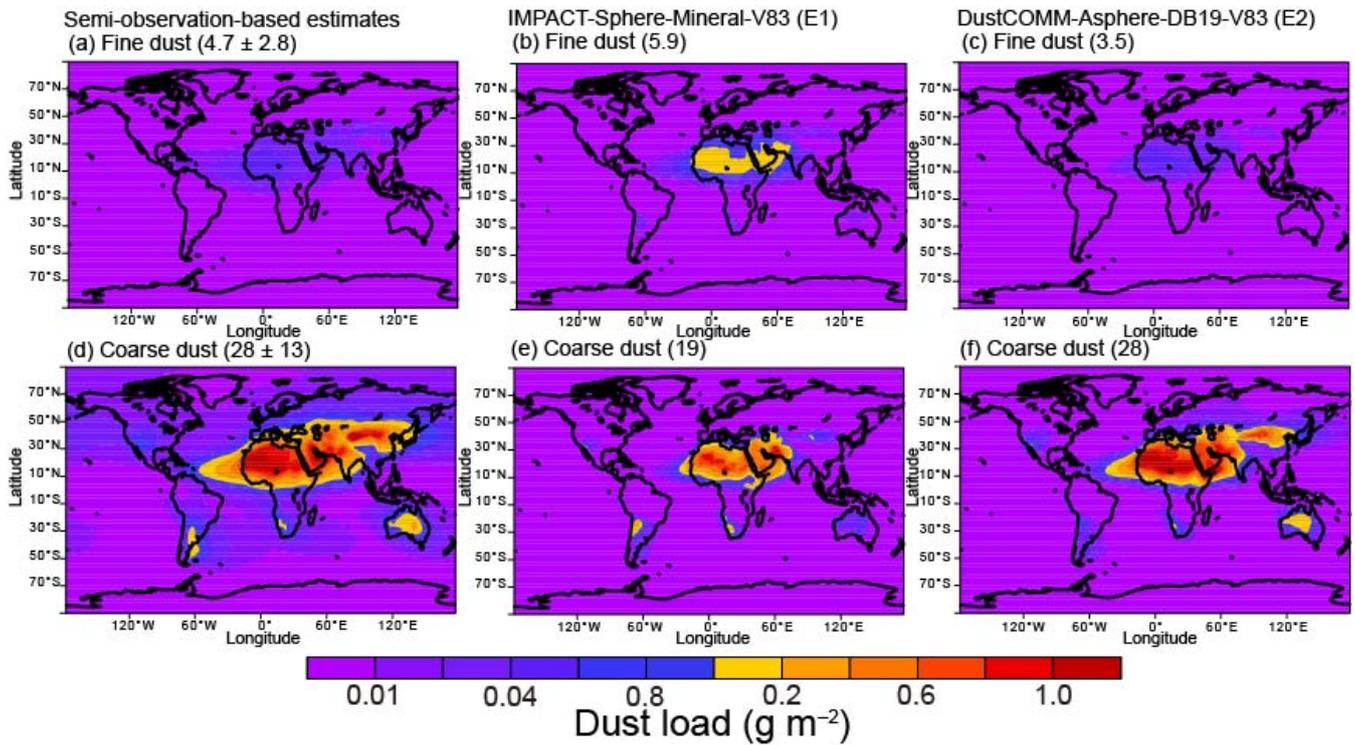


Figure 3. Model-simulated dust loads at fine (smaller than 2.5 μm of diameter) and coarse size ranges (larger than 2.5 μm of diameter) before and after adjusting the size-resolved dust load with DustCOMM. Results were shown for (a) fine dust from DustCOMM, (b) fine dust from IMPACT-Sphere-Mineral-V83 (E1), (c) fine dust from DustCOMM-Asphere-DB19-V83 (E2), (d) coarse dust from DustCOMM, (e) coarse dust from E1, and (f) coarse dust from E2 simulations. The parentheses represented the global dust burden (T_g). The values of dust load at each bin were listed in Table 3.

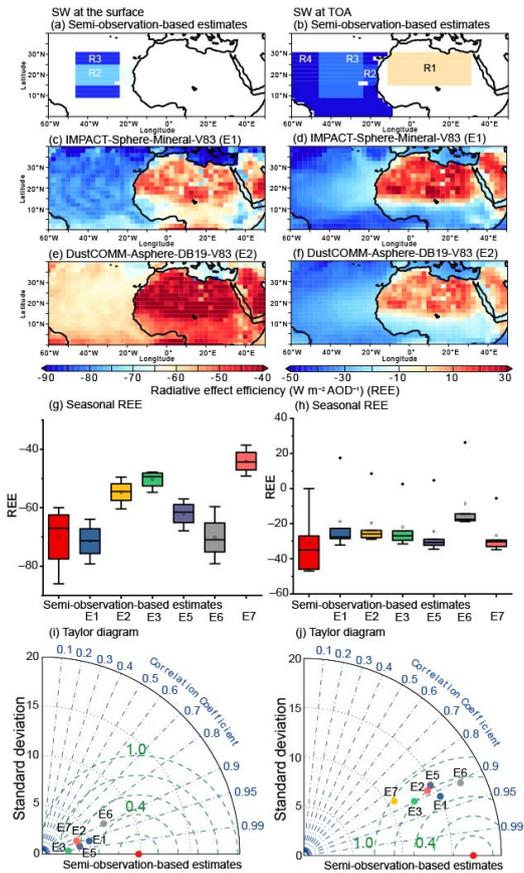


Figure 4. Dust clear-sky SW radiative effect efficiency ($W \cdot m^{-2} DAOD^{-1}$). Semi-observationally-based data at (a) the surface and (b) TOA were based on satellite observations (Yang et al. 2009; Li et al., 2004; Song et al., 2018; Christopher and Jones, 2007). The model results were shown for (c) and (d) IMPACT-Sphere-Mineral-V83 (E1), and (e) and (f) DustCOMM-Asphere-DB19-V83 (E2) simulations at the surface and TOA, respectively. Comparison of seasonally averaged SW radiative effect efficiency for semi-observationally-based (SOB) data and the different experiments at (g) the surface and (h) TOA. The square symbol represents the mean. The solid line within the box mark shows the median. The boundaries of the box mark the 25th and 75th percentiles. The whiskers above and below the box indicate the $1.5 \times$ interquartile range, and the points indicate the outside of the range. Taylor diagram summarizing the statistics of the comparison against the seasonally averaged regional SW radiative effect efficiency for the different experiments at (i) the surface and (j) TOA. The horizontal axis shows the standard deviation of the data set or model prediction, the curved axis shows the correlation, and the green dashed lines denote the root-mean-squared errors between the semi-observationally-based data and the model predictions. As such, the distance between the semi-observationally-based data and the model predictions is a measure of the model's ability to reproduce the spatiotemporal variability in the semi-observationally-based data. The regionally averaged values were listed in Tables S3 and S4 at the surface and TOA, respectively.

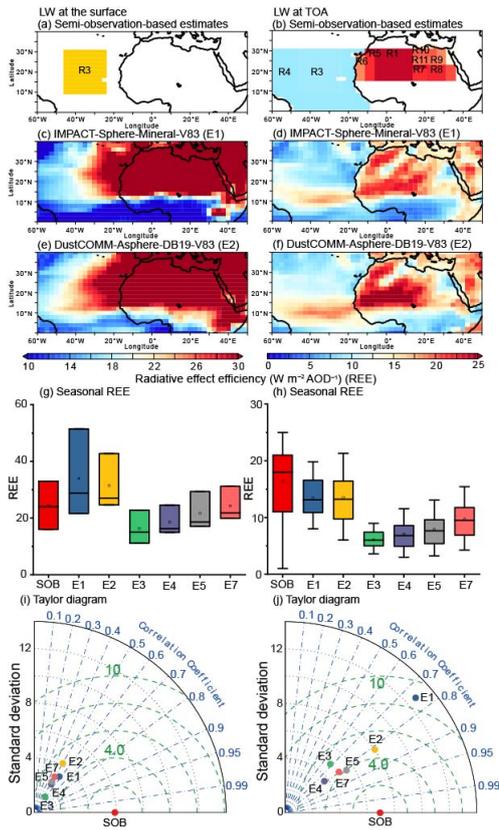
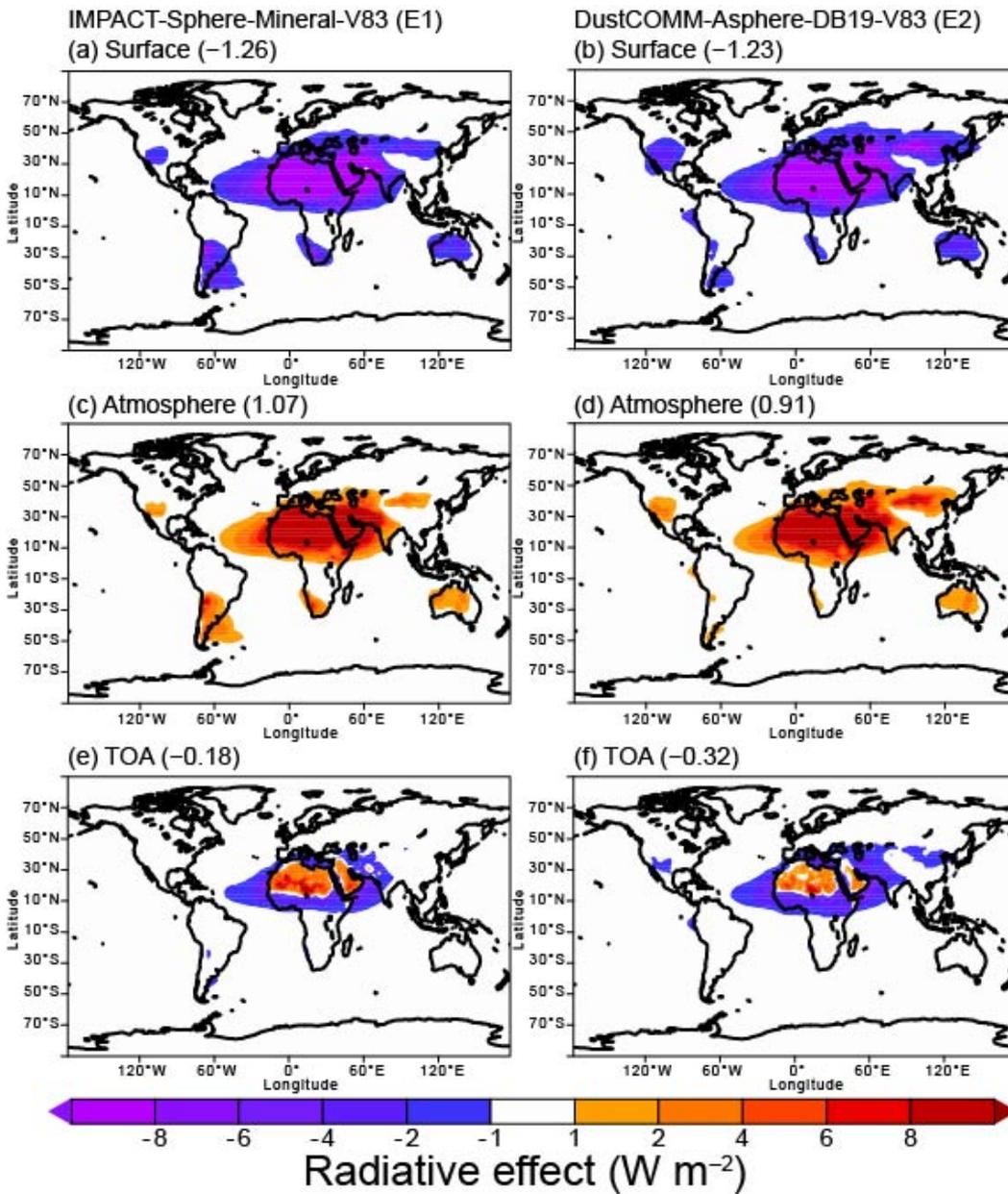


Figure 5. Dust clear-sky LW radiative effect efficiency ($\text{W} \cdot \text{m}^{-2} \text{ DAOD}^{-1}$). Semi-observationally-based estimates at (a) surface and (b) TOA were based on satellite observations (Song et al., 2018; Christopher and Jones, 2007; Zhang and Christopher, 2003; Brindley and Russell, 2009; Yang et al., 2009). The model results were shown for (c) and (d) IMPACT-Sphere-Mineral-V83 (E1), and (e) and (f) DustCOMM-Asphere-DB19-V83 (E2) simulations at the surface and TOA, respectively. Comparison of seasonally averaged LW radiative effect efficiency for semi-observationally-based (SOB) data and the different experiments at (g) the surface and (h) TOA. The square symbol represents the mean. The solid line within the box mark shows the median. The boundaries of the box mark the 25th and 75th percentiles. The whiskers above and below the box indicate the $1.5 \times$ interquartile range, and the points indicate the outside of the range. Taylor diagram summarizing the statistics of the comparison against the seasonally averaged regional SW radiative effect efficiency for the different experiments at (i) the surface and (j) TOA. The horizontal axis shows the standard deviation of the data set or model prediction, the curved axis shows the correlation, and the green dashed lines denote the root-mean-squared errors between the semi-observationally-based data and the model predictions. As such, the distance between the semi-observationally-based data and the model predictions is a measure of the model's ability to reproduce the spatiotemporal variability in the semi-observationally-based data. The regionally averaged values were listed in Tables S5 and S6 at the surface and TOA, respectively.



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Figure 6. Dust SW radiative effect ($\text{W}\cdot\text{m}^{-2}$) and radiative heating of the atmosphere (i.e., the subtraction of radiative effects from TOA to the surface in unit of $\text{W}\cdot\text{m}^{-2}$). The model results were shown for the simulations for (a) IMPACT-Sphere-Mineral-V83 (E1) at the surface, (b) DustCOMM-Asphere-DB19-V83 (E2) at the surface, (c) E1 in atmospheric column, (d) E2 in atmospheric column, (e) E1 at TOA, and (f) E2 simulations at TOA. The numbers in parentheses represented the global

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mean.

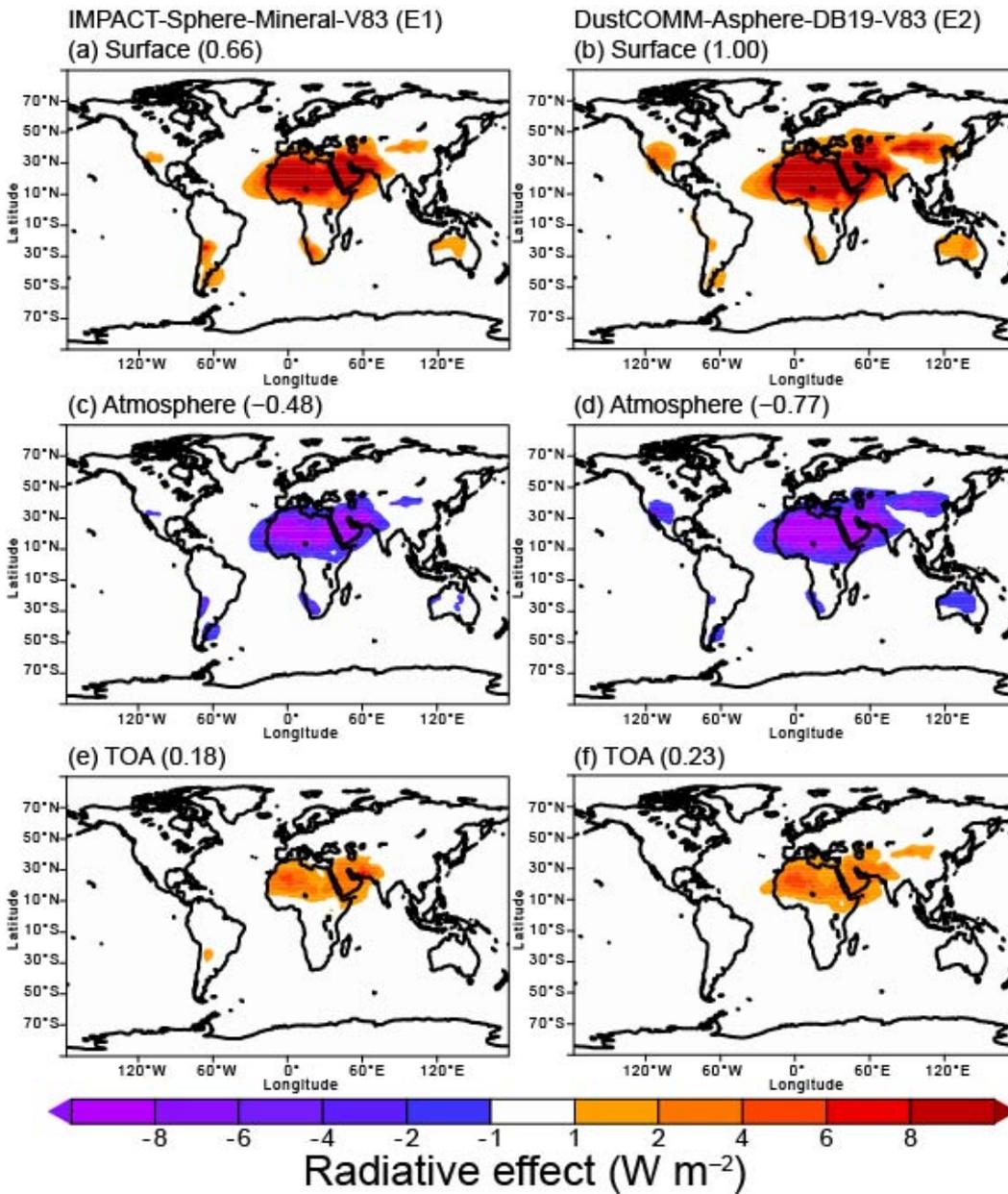


Figure 7. Dust LW radiative effect ($\text{W}\cdot\text{m}^{-2}$) and radiative heating of the atmosphere (i.e., the subtraction of radiative effects from TOA to the surface in unit of $\text{W}\cdot\text{m}^{-2}$). The model results were shown for the simulations for (a) IMPACT-Sphere-Mineral-V83 (E1) at the surface, (b) DustCOMM-Asphere-DB19-V83 (E2) at the surface, (c) E1 in atmospheric column, (d) E2 in atmospheric column, (e) E1 at TOA, and (f) E2 simulations at TOA. The numbers in parentheses represented the global mean.

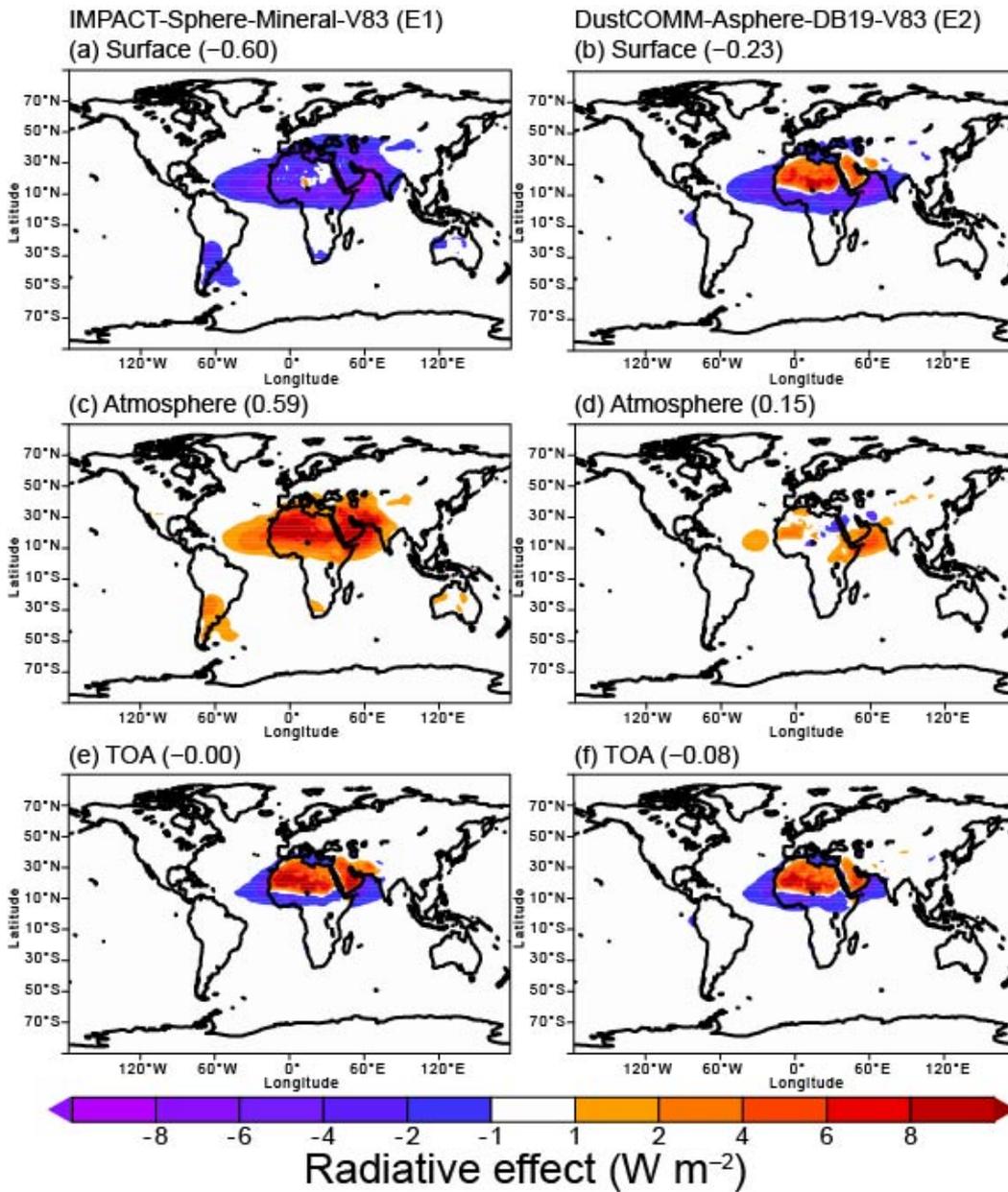
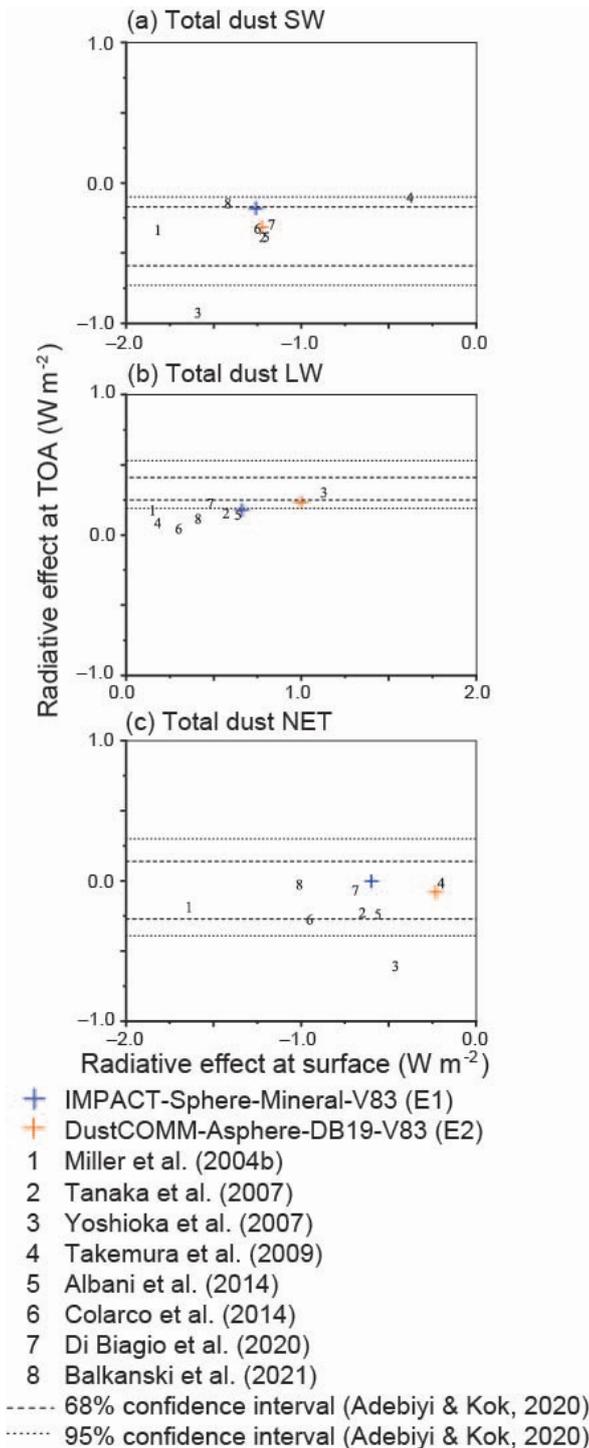


Figure 8. Dust net radiative effect ($\text{W}\cdot\text{m}^{-2}$) and radiative heating of the atmosphere (i.e., the subtraction of radiative effects from TOA to the surface in unit of $\text{W}\cdot\text{m}^{-2}$). The model results were shown for the simulations for (a) IMPACT-Sphere-Mineral-V83 (E1) at the surface, (b) DustCOMM-Asphere-DB19-V83 (E2) at the surface, (c) E1 in atmospheric column, (d) E2 in atmospheric column, (e) E1 at TOA, and (f) E2 simulations at TOA. The numbers in parentheses represented the global mean.



85 **Figure 9.** Variability of dust radiative effect ($W \cdot m^{-2}$) in different model simulations at the surface and TOA for (a) total dust SW, (b) total dust LW, and (c) total dust NET. The annually averaged values were listed in Table 5.

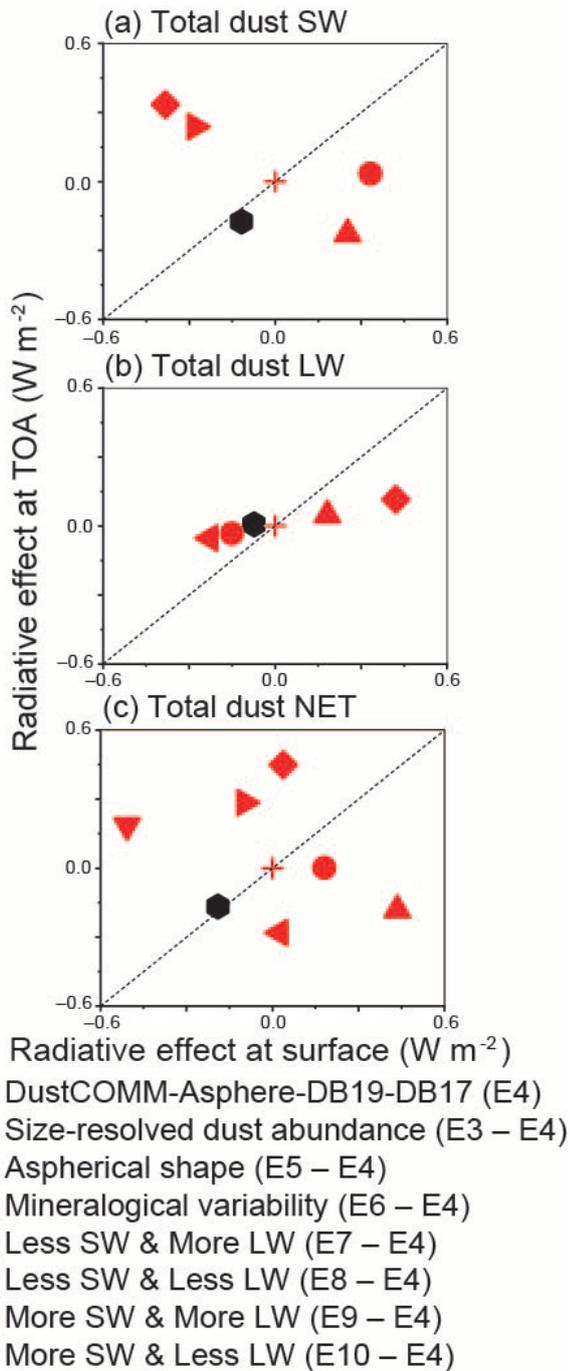


Figure 10. Radiative effect ($W \cdot m^{-2}$) of mineral dust due to various aerosol absorptivity at the surface and TOA for (a) total dust SW, (b) total dust LW, and (c) total dust NET. The annually averaged values were listed in Table 5. The dashed line represented a 1 : 1 correspondence and corresponded to no change in radiative heating within the atmosphere.