Less atmospheric radiative heating by dust due to the synergy of coarser size and aspherical shapedust with coarser size

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Abstract. Mineral dust aerosols cool and warm the atmosphere by scattering and absorbing both solar (short-wave: SW) and thermal (long-wave: LW) radiation. However, significantlarge uncertainties remain in dust radiative effects, largely due to differences in the dust size distribution and spectral optical properties simulated in Earth system models. Dust models typically underestimate the coarse dust load (more than 2.5 μm in a diameter) and assume a spherical shape, which leads to an overestimate of the fine dust load (less than 2.5 μm) after the dust emissions in the models are scaled to match observed dust aerosol optical depth at 550 nm ($DAOD_{550}$). Here, we improve the simulated dust properties with datasets that leverage measurements of size-resolved dust concentration, and asphericity factor, and refractive index (improved simulation) in a coupled global chemical transport model (IMPACT) with a radiative transfer module (RRTMG) (default simulation). After the adjustment of size-resolved dust concentration and spectral optical properties, Tthe global and annual average of dust aerosol optical depth at 550 nm ($DAOD_{550}$) from the improved simulation increases from 0.023 to(0.029), and falls within the range of a semi-observationally-based estimate (0.030 ± 0.005), in contrast to that (0.023) of the default simulation. The reduction of fine dust load after the adjustment leads to a reduction of the SW cooling at the Top Of the Atmosphere (TOA). To Iimproved agreement against a semi-observationally-based estimate of the radiative effect efficiency at TOA, we find that was obtained using a less absorptive SW and more absorptive LW dust refractive indexices is required for coarser aspherical dust. Thus, only a minor difference is estimated for the . Our sensitivity simulations reveal that the improved simulation leads to a similar net global dust radiative effect at the Top Of Atmosphere (TOA) on a global scale to the default simulation (–0.08 vs. –0.009 $W \cdot m^{-2}$ on a global scale) but . Conversely, our sensitivity simulations reveal that the surface warming is substantially enhanced near the strong dust source regionsthe net global dust radiative effect results in less cooling at the surface (less cooling to –0.23 vs.from –0.6088 $W \cdot m^{-2}$ on a global scale). Thus, less atmospheric radiative heating is estimated near the major source regions (less heating to 0.15 from 0.59 $W \cdot m^{-2}$ on a global scale), because of enhanced LW warming at the surface by the synergy of coarser size and aspherical dustshape. Our results thus suggest less atmospheric radiative heating due to aspherical dust with coarser size over the major source regions (0.15 vs. 0.79 $W \cdot m^{-2}$ on a global scale).

1 Introduction

Mineral dust aerosols can both cool and warm the climate, but how much dust aerosols net influence global climate is highly uncertain (Penner, 2019). Global dust modeling studies have suggested that mineral dust exerts an global and annual mean aerosol radiative effect (RE) between –0.6 and +0.2 W m^{-2} at the Top Of the Atmosphere (TOA) and between −0.2 and −2.7 W m^{-2} at the surface (Miller and Tegen, 1998; Balkanski et al., 2007; Tanaka et

al., 2007; Takemura et al., 2009; Räisänen et al., 2013; Zhao et al., 2013; Albani et al., 2014; Colarco et al., 2014; Heald et al., 2014~~; Tuccella et al., 2019~~; Di Biagio et al., 2020; Tuccella et al., 2020). Whereas a negative RE corresponds to the cooling of the global system when the sunlight is reflected to space, a positive RE corresponds to an overall warming of the Earth-atmosphere system by trapping incident short-wave (SW) and outgoing long-wave (LW) radiation. Radiative effect by dust aerosols perturbs surface temperature, wind speed, rainfall, and vegetation cover, which may induce feedback on dust emissions (Perlwitz et al. 2001; Miller et al., 2004a; Colarco et al., 2014). The climate feedback does not only depend ~~only~~ on RE at TOA or the surface alone~~,~~ but also on ~~its~~ the difference to the ~~surface~~ value at TOA and surface, which represents radiative heating within the atmosphere (Miller et al., 2004b; Yoshioka et al., 2007; Lau et al., 2009). The large uncertainties in quantifying the dust RE in the models are mainly propagated from the large spatial heterogeneity and temporal variability of mineral dust abundance and the physicochemical properties (e.g., size distribution, mineral composition, and shape), as well as the ground surface characteristics and atmospheric properties (e.g., surface reflectance, temperature, and atmospheric absorption) (Sicard et al., 2014; Lacagnina et al., 2015; Li and Sokolik, 2018). The model errors in dust size distribution and particle shape can lead to an overestimate of fine dust load after the dust emissions in the models are scaled to match observed dust aerosol optical depth at 550 nm ($DAOD_{550}$). The corresponding overestimate of SW cooling might be compensated for in models by using a refractive index that is too absorbing (Di Biagio et al., 2019, 2020), which depends on the mineral composition of the dust. We regard "fine" and "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. Below, we provide a brief discussion of the effects of the dust size distribution, particle shape~~mineral composition~~, and mineral composition ~~particle shape~~ on dust radiative effects.

~~Recently~~First, there has been increased attention paid to the importance of accurately predicting the abundance of coarse dust for the global energy balance (Kok et al., 2017; Song et al., 2018; Di Biagio et al., 2020; Adebiyi and Kok, 2020). The coarser particles are expected to be more prevalent closer to the source regions, as they fall ~~downward~~ much faster than finer particles (Mahowald et al., 2014). For instance, the lifetime of dust aerosols larger than 30 µm in diameter is less than 12 h in most cases except in large haboobs (Ryder et al., 2013). Current models, however, cannot accurately simulate observed transport of coarse dust particles across the Atlantic (Weinzierl et al., 2017; Ansmann et al., 2017), although several hypotheses have been proposed to explain measurements of giant dust particles (larger than 63 µm in diameter) relatively far from source regions (van der Does et al., 2018). ~~For instance,~~ The potential mechanism for long-range transport of giant dust particles is that the uplift events of coarse dust can be induced by a nocturnal low-level jet or cold pool outflow from mesoscale convective systems (i.e., haboobs) (Rosenberg~~,~~ et al., 2014; Ryder et al., 2019). At higher elevation, electrostatic forces ~~and dust asphericity~~ might retard

the settling of coarse and giant dust particles and thus may facilitate the transport of these particles over longer distances (Harrison et al., 2018; Toth et al., 2019~~; Huang et al., 2020a~~). Other missing processes that affect the transport and deposition of giant particles would also need to be incorporated into the models to reproduce the measurements of the size distribution over the open ocean (van der Does et al., 2018). The coarse dust particles scatter and absorb both the solar ~~(short wave: SW)~~ and thermal ~~(long wave: LW)~~ radiation, causing a net warming effect at TOA. In contrast, the fine dust particles principally scatter SW radiation, causing a net cooling effect. Since coarse dust tends to warm the climate, the underestimation of the abundance of coarse dust causes Earth system models to underestimate the warming near the dust source regions. ~~On the other hand, model errors due to the underestimated coarse dust load and corresponding warming might be compensated for in models by using a refractive index that is too absorbing (Di Biagio et al., 2019, 2020), and which depends on the mineral composition of the dust.~~

Second, ~~The dust refractive index is often derived from measurements based on dust or individual mineral particles (Bedidi and Cervelle, 1993; Long et al., 1993; Di Biagio et al., 2017, 2019; Stegmann & Yang, 2017). Indeed, most dust particles are internal mixtures of various mineral compositions and irregular shapes (Reid, 2003; Wiegner et al., 2009; Wagner et al., 2012). In desert soils, iron (Fe) oxides are generally hematite (α Fe_2O_3) and goethite (FeOOH), which cause soil-derived dust absorption at ultraviolet (UV) and visible wavelengths (Sokolik and Toon, 1999; Balkanski et al., 2007). These two minerals have distinct optical properties, which might cause various intensities of shortwave absorption and thus RE of dust aerosols (Lafon et al., 2016). However, the speciation of dust into its mineral components inherently comprises uncertainties on soil mineralogy, Fe content in mineral, and refractive index of Fe oxides, partly due to the differences in prescribed parameters. The atmospheric aging of Fe-containing aerosols can further modulate the optical properties of Fe oxides (Ito et al., 2018), while the photochemical transformation of Fe oxides from lithogenic sources due to atmospheric processing is relatively limited (< 10%), compared to pyrogenic sources (Ito et al., 2019).~~

~~P~~previous studies have shown that ~~the assumption of spherical shape in models leads to a substantial underestimation of the extinction efficiency and thus dust aerosol optical depth (DAOD), mainly because the assumption of sphericity causes an underestimation of the surface-to-volume ratio compared to aspherical dust (Kok et al., 2017; Hoshyaripour et al., 2019; Tuccella et al., 2019). Overall,~~ the ~~net~~ SW radiative effect of dust asphericity on climate simulations is minor on a global scale, partly because the larger DAOD is compensated for by the larger asymmetry parameter of aspherical dust, which reduces the amount of radiation scattered backward to space (Räisänen et al., 2013; Colarco et al., 2014). Moreover, non-spherical calcium-rich dust particles can be converted to spherical particles, due to heterogeneous reactions with nitrate and sulfate on these particles, especially over polluted regions (Laskin et al., 2005; Matsuki et al., 2005). As the plumes move downwind to the oceans, the dust aerosols can be

aggregated with sea salt in the marine boundary layer, which leads to more spherical shapes and larger sizes (Zhang and Iwasaka, 2004). However, the assumption of spherical shape in models leads to a substantial underestimation of the extinction efficiency and thus DAOD near the strong source regions, mainly because the assumption of sphericity causes an underestimation of the surface-to-volume ratio compared to aspherical dust (Kok et al., 2017, 2021; Hoshyaripour et al., 2019; Tuccella et al., 2020). Radiative effect efficiency is often used for the evaluation of the models and is defined as the gradient of a linear least squares fit applied to AOD and dust radiative effect at each two-dimensional (2-D) grid box ($W \cdot m^{-2}$ AOD^{-1}). Thus, the estimates of the dust radiative effect efficiency could be biased, in part, due to large uncertainties associated with the spherical assumption on AOD retrieval (Zhou et al., 2020).

Third, the dust refractive index is often derived from measurements based on dust or individual mineral particles (Bedidi and Cervelle, 1993; Long et al., 1993; Di Biagio et al., 2017, 2019; Stegmann & Yang, 2017). Indeed, most dust particles are internal mixtures of various mineral compositions and irregular shapes (Reid, 2003; Wiegner et al., 2009; Wagner et al., 2012). In desert soils, iron (Fe) oxides are generally hematite (α-Fe_2O_3) and goethite (FeOOH), which cause soil-derived dust absorption at ultraviolet (UV) and visible wavelengths (Sokolik and Toon, 1999; Balkanski et al., 2007). These two minerals have distinct optical properties, which might cause various intensities of SW absorption and thus RE of dust aerosols (Lafon et al., 2016). The dust complex refractive index in the LW also depends on the particle mineralogical composition (Sokolik et al., 1998). Di Biagio et al. (2017) found a linear relationship between the magnitude of the imaginary refractive index at 7.0, 9.2, and 11.4 μm and the mass concentration of calcite and quartz absorbing at these wavelengths. However, the speciation of dust into its mineral components inherently comprises uncertainties on soil mineralogy, mineral content in size-segregated dust particles, and refractive index of mineral, partly due to the differences in prescribed parameters such as the particle size. The atmospheric aging of Fe-containing aerosols can further modulate the optical properties of Fe oxides (Ito et al., 2018), while the photochemical transformation of Fe oxides from lithogenic sources due to atmospheric processing is relatively limited (< 10%), compared to pyrogenic sources (Ito et al., 2019).

Here, we focus on the influence of the size-resolved abundance of aspherical dust on the aerosol radiative effects in a coupled global chemical transport model (IMPACT) (Ito et al., 2020 and references therein) with a radiative transfer module (RRTMG) (Iacono et al., 2008) ~~(i.e., default simulation)~~. We improve the accuracy of these simulations by correcting the bias in size-resolved dust concentration with the Dust Constraints from joint Observational-Modelling-Experimental analysis (DustCOMM) data set (Adebiyi et al., 2020) ~~(i.e., improved simulation)~~, as well as by considering the aspherical shape (Huang et al., 2020a, 2020b). We then explore the sensitivity to dust refractive index. ~~After evaluating the model outputs against semi-observation-based DAOD at 550 nm ($DAOD_{550}$) and radiative effect efficiencies, the improved simulation suggests much less atmospheric radiative~~

~~heating over the major source regions, compared to the default simulation (0.15 vs. 0.79 W·m^{-2} on a global scale). The term "semi-observation-based" is used for DAOD$_{550}$ and dust radiative effect efficiency when the estimates are based on the combination of observations and models.~~

2 Methods

~~Since most models underestimate the coarse dust load differently (Ansmann et al., 2017; Adebiyi and Kok, 2020), the sensitivity of dust radiative effects to optical properties in the present study might be considerably is also different from previous other studies. In our study, we regard "fine" and "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. Here, w~~We examined the dust radiative effects using ~~seven~~ ten different numerical experiments that varied (1) the simulated dust concentration and their size distribution, (2) particle shape, and (3) mineralogical composition (Tables 1 and 2). Two experiments used the dust concentrations calculated from the ~~forward~~ IMPACT model with the finer dust size (denoted as "IMPACT~~fine~~"). Subsequently, the simulated dust concentration and the size distribution were adjusted to the semi-observationally-based concentrations (Adebiyi and Kok, 2020) in the other eight experiments with the coarser dust size (denoted as "DustCOMM~~coarse~~"). The term "semi-observationally-based" is used for DustCOMM, DAOD$_{550}$, and dust radiative effect efficiency when the estimates are based on the combination of observations and models. Five sensitivity experiments were handled in the RRTMG calculations performed with different refractive indices and hourly averaged aerosol concentrations with the two chemical transport model simulations of "IMPACT" and "DustCOMM". The other three experiments were calculated from the model output with a post-processor. T~~he default~~ wo simulations used the spherical assumption on the particle shape (denoted as "Sphere"), whereas the dust asphericity was considered in the other ~~six~~ eight experiments (denoted as "Asphere"). We then examined different refractive indices for the dust mineralogy to represent the regional variations in refractive indices (denoted as ~~"region",~~ "~~m~~Mineral", "DB17", "DB19", "V83", ~~or~~ "Less SW", "More LW~~absorption~~", "More SW", and "Less LW"). These sensitivity simulations and their radiative effects are summarized in Tables 1 and 2, respectively, with more details below. In section 2.3, we describe the DustCOMM data set used to adjust (1) size-resolved abundance of dust concentration. In section 2.4, we describe the adjustment factor of (2) particle shape for spectral optical properties. In section 2.5, we describe differences in spectral refractive indices due to (3) different mineralogical compositions for the radiative flux calculation.

2.1 Aerosol chemistry transport model

This study used the Integrated Massively Parallel Atmospheric Chemical Transport (IMPACT) model (Ito et al., 2020 and references therein). Simulations were performed for the year ~~of~~ 2016, using a horizontal resolution of 2.0°×2.5° for latitude by longitude and 47 vertical layers. The chemical transport model was driven by ~~used~~ the Modern Era Retrospective analysis for Research and Applications 2 (MERRA-2) reanalysis meteorological data from the National Aeronautics and Space Administration (NASA) Global Modeling and Assimilation Office (GMAO) (Gelaro et al., 2017). The radiative feedback of the dust on the climate model simulation can be predicted by a separate version of the model (Penner et al., 2018).

The model simulated the emissions, chemistry, transport, radiation, and deposition of major aerosol species, including mineral dust, black carbon (BC), particulate organic matter (POM), sulfate, nitrate, ammonium, and sea spray aerosols, and their precursor gases. Atmospheric processing of mineral dust aerosols, during transport, were projected for four distinct aerosol size bins (<1.26 μm, 1.26–2.5 μm, 2.5–5 μm, and 5–20 μm of diameter). In this version of the IMPACT model, two modes were used for sulfate aerosol (nuclei and accumulation mode), and two moments were predicted within each mode (sulfate aerosol number and mass concentration) (Liu et al., 2015). The surface coating of sulfate on dust aerosols occurred as a result of the condensation of sulfuric acid gas on their surfaces, coagulation with sulfate aerosol, and formation in aqueous reactions within cloudy regions of the atmosphere (Liu et al., 2015). The heterogeneous uptake of nitrate, ammonium, and water vapor by each aerosol for each size bin was interactively simulated in the model following a hybrid dynamical approach (Feng and Penner, 2007). Five types of aerosols (i.e., dust, nucleated sulfate, carbonaceous aerosols from fossil fuel combustion, carbonaceous aerosols from biomass burning, and sea salt) were assumed to be externally mixed in each size bin for the computation of spectral optical properties (Xu and Penner, 2012). Dust emissions were dynamically simulated using a physically-based emission scheme (Kok et al., 2014; Ito and Kok, 2017) with the soil mineralogical map (Journet et al., 2014; Ito and Shi, 2016). To derive atmospheric concentration of mineral composition for dust aerosol, "tagged" tracer was used for each size-resolved mineral source. The direct emissions of dust wea~~r~~e evenly distributed in mixing ratio throughout the planetary boundary layer. The global scaling factor of dust emission was determined from the comparison of the model results with ground-based AOD measurements near the dust source regions prior to the adjustment to the DustCOMM (Kok et al., 2014; Ito and Kok, 2017). In recent review papers, multi-model evaluations of aerosol iron concentrations and their solubilities have been comprehensively summarized on global and regional scales (Myriokefalitakis et al., 2018; Ito et al., 2021).

To improve the accuracy of our simulations of mineral dust, we made several upgrades to the on-line emission and gravitational settling schemes used in Ito et al. (2020). The dust emissions were extremely sensitive to soil

moisture, and thus the bias was adjusted with satellite observations (Ito and Kok, 2017). However, the satellite measurements were only available every other day, depending on location. The Soil Moisture Active Passive (SMAP) Level-4 Soil Moisture data product addressed these limitations by merging the satellite observations into a numerical model of the land surface water and energy balance while considering the uncertainty of the observations and model estimates (Reichle et al., 2019). In this work, we utilized the 3-hourly data of soil moisture derived from the SMAP for barren and open shrublands~~,~~ separately (Reichle et al., 2018). To achieve this, we used the MODerate resolution Imaging Spectroradiometer (MODIS) land cover map at 500 m resolution to calculate the fraction of barren and open shrublands in each grand surface layer (Friedl et al., 2019)

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

2.2 Integration of IMPACT and RRTMG

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

The mineral dust particles were assumed to follow prescribed size distributions within each size bin (Liu et al., 2015). In applying the look-up table, the size spectrum for mineral dust was divided into 30 sub bins (Wang and Penner, 2009). As for the SW, the particle size increased with the uptake of sulfate, nitrate, ammonium, and water by the aerosols (Xu and Penner, 2012). These coating materials were treated as internally mixed with dust aerosol in each

size bin and thus can reduce solar absorption of mineral dust. ~~As the LW scattering was not accounted for in the RRTMG, we used the 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), following Di Biagio et al. (2020) who noted that this adjustment was estimated for dust of diameter less than 10 μm and thus might be a lower approximation of the LW scattering by coarse dust. The larger adjustment factor at TOA reflects the fact that the upward LW radiation emitted from the ground surface can be reduced through scattering and absorption, compared to the surface.~~ Subsequently, these optical properties were used by the RRTMG to calculate RE based on dust mixing ratio distributions in the IMPACT model. The dust RE was estimated as the difference in the calculated radiative fluxes with all aerosols and with all aerosols except the dust aerosols for each bin. As the LW scattering was not accounted for in the RRTMG, we multiplied the LW radiative fluxes by the 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), following Di Biagio et al. (2020). The larger adjustment factor at TOA reflects the fact that the upward LW radiation emitted from the ground surface can be trapped through scattering and absorption compared to the surface.

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

2.3 ~~Description of~~ DustCOMM dataset and sensitivity experiments to size-resolved dust concentration

Dust Constraints from joint Observational-Modelling-experiMental analysis (DustCOMM) is a dataset of three-dimensional (3-D) dust properties obtained by combining observational, experimental, and modeling constraints on dust properties. While details can be found in Adebiyi et al. (2020) and Adebiyi and Kok (2020), we provide a brief overview here. First, DustCOMM's constraint on the 3-D dust size distribution combines dozens of previously published in-situ measurements of dust size distributions~~,~~ taken during several field campaigns, with an ensemble of climate model simulations. The framework used those in-situ measurements first to constrain the globally averaged size distribution (Adebiyi and Kok, 2020), which is used subsequently to ~~bias~~ adjust~~ment~~ the bias in an ensemble of six global model simulations (Adebiyi et al., 2020). The constraints on dust size distribution range~~s~~ from 0.2 μm to 20 μm in diameter, where a generalized analytical function describes the sub-bin distribution based on brittle fragmentation theory (Kok, 2011). The second DustCOMM product – ~~-~~atmospheric dust mass loading – ~~-~~combines

the constraints on dust size distribution with constraints on dust extinction efficiency and dust aerosol optical depth (Adebiyi et al., 2020). The constraints on dust extinction efficiency used the single-scattering database of Meng et al. (2010) and leveraged measurements of the dust index of refraction as well as accounts for the non-spherical shape of dust particles (Kok et al., 2017). For this, we approximate dust as tri-axial ellipsoidal particles described by the globally- representative values of measured dust aspect ratio (the length-to-width ratio), and the height-to-width ratio (HWR) obtained from Huang et al. (2020a). Furthermore, the dust aerosol optical depth used to obtain the dust mass loading combines the semi-observationally-based dataset from Ridley et al. (2016) with information from four reanalysis products. This includes the MERRA-2, Navy Aerosol Analysis and Prediction System (NAAPS), Japanese Reanalysis for Aerosol (JRAero), and Copernicus Atmosphere Monitoring Service interim ReAnalysis (CAMSiRA) (Adebiyi et al., 2020).

The aerosol RE of mineral dust strongly depends on both the magnitude of dust load and the dust size distribution (Tegen and Lacis, 1996; Liao and Seinfeld, 1998). The DustCOMM data set contains total column loading and concentration of mineral dust resolved by season and particle size (Adebiyi et al., 2020). To correct the bias in the seasonally averaged size-resolved dust emission in the IMPACT model, the sum of bin 1, bin 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 column loading between the model and DustCOMM at each 2-D grid box. When the source function was used for high-latitude dust in the Northern Hemisphere, this led to substantially high emissions and thus RE over there, likely due to the influences from long-range transported dust. Therefore, the direct emissions of dust from the nine major source regions only (Kok et al., 2021) were adjusted using the DustCOMM data (Fig. 1). To adjust the size bias in dust emissions, the mass fraction of emitted dust for each bin was prescribed according to the size-resolved total column loading of DustCOMM at each 2-D grid box. Overall, the IMPACT-simulated lifetime of the dust aerosol for the second-size bin (7.8 days 1.26–2.5 μm of diameter) was in good agreement with the ensemble of model results (8.5 ± 1.1 days for the mass mean diameter of 1.8 μm) (Kok et al., 2017). To correct the bias in the seasonally averaged 3-D dust size distribution after the transport, the mass 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 to calculating the radiative fluxes using the RRTMG by the ratio of mass concentration of $PM_{2.5}$ (i.e., the sum of bin 1 and bin 2) to each bin (Table 3).

2.4 ~~Description of a~~Asphericity factor for optical properties and sensitivity experiments to particle shape

To account for the dust asphericity, an adjustment factor was applied to the spherical optical properties at each dust size parameter and refractive index. The adjustment factors for the spectral optical properties of non-spherical particles were calculated after Huang et al. (2020b). The atmospheric aging of mineral dust can form a uniform coating

around the mineral core and therefore decrease particle asphericity during transport. This is implicitly considered in the globally averaged shape distribution of dust (Huang et al., 2019). Specifically, Huang et al. (2020b) combined globally representative dust shape distributions (Huang et al., 2020a) with a shape-resolved single-scattering database (Meng et al., 2010). This database combines four computational methods (Mie theory, T-matrix method, discrete dipole approximation, and an improved geometric optics method) to compute the single-scattering properties of non-spherical dust for a wide range of shape descriptors. Huang et al. (2020b) provided the look-up table containing optical properties of non-spherical dust as functions of size parameter and refractive index.

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

2.5 Spectral refractive index and sensitivity experiments to mineralogical compositions

The aerosol RE of mineral dust depends on mineralogical composition. For the sensitivity simulation to the SW and LW refractive indices, we used the global mean of laboratory measurements of the refractive index from 19 natural soils from various source regions around the world in Di Biagio et al. (2019) (denoted as "DB19") and in Di Biagio et al. (2017) (denoted as "DB17"), respectively. To illustrate the regional heterogeneity of refractive index, 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 regionally averaged imaginary parts of the refractive indices at the wavelength of 0.52 μm and 9.7 μm showed large differences in SW and LW absorptivity, respectively, between different samples collected at various geographical locations (Fig. 1).

The optical properties from the measurements for dust samples generated from 19 natural soils suggested a considerable role of Fe oxides in determining the SW absorption (Di Biagio et al., 2019). The refractive indices for mineral components were used for hematite, goethite (Bedidi and Cervelle, 1993), silicate particle group, quarts, gypsum ($CaSO_4$) (Stegmann & Yang, 2017), and calcite ($CaCO_3$) (Long et al., 1993) in the simulations denoted as "Mineral". The hematite and goethite were treated separately according to the mineralogical map (Journet et al., 2014). Consequently, hematite mass content averaged in the dust at emission (0.79% for fine and 0.50% for coarse from the IMPACT simulation) was lower than goethite content (1.8% and 1.3%, respectively) on a global scale. In addition to the primary emission of gypsum, $CaSO_4$ is secondarily formed due to the dissolution/precipitation of $CaCO_3$ in thermodynamic equilibrium condition (Ito and Feng, 2010). To illustrate the difference in refractive index, the global

mean of the mineral composition was used for the comparison with DB19 (Fig. 1). The imaginary parts of the refractive indices from mineralogical map were higher than DB17, resulting in a stronger absorption over the SW spectrum.

The mineral dust LW refractive index also depends on its mineralogical composition (Sokolik et al., 1998; Di Biagio et al., 2017). The LW refractive index of Volz (1983) has been widely used in climate models and satellite remote sensing algorithms and thus was examined here (denoted as "V83") (Song et al., 2018). The imaginary parts of the refractive indices from V83 were higher than DB19, resulting in a stronger absorption over most of the LW spectrum. To analyze the dependence of the results on less (more) absorptive SW and less (more) absorptive LW refractive indices, we made further sensitivity simulations by varying the values of imaginary parts of the refractive index within the range of values from Di Biagio et al. (2017, 2019) (10% or 90% percentiles for SW or LW, respectively) (denoted as "Less" or "More"). The associated real parts with 10% or 90% percentile imaginary parts for LW were calculated to account for the Kramers-Kronig relation (Lucarini et al., 2005).

~~**2.5 Sensitivity simulations**~~

~~First, the aerosol RE of mineral dust strongly depends on both the magnitude of dust load and the dust size distribution (Tegen and Lacis, 1996; Liao and Seinfeld, 1998). The DustCOMM data set contains total column loading and concentration of mineral dust resolved by season and particle size (Adebiyi et al., 2020). To adjust the seasonal dust load in the model, the sum of bin 1, bin 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 column loading between the model and DustCOMM at each 2-D grid box. When the source function was used for high latitude dust in the Northern Hemisphere, this led to substantially high emissions and thus RE over there, likely due to the influences from long range transported dust. Therefore, the direct emissions of dust from the nine major source regions only (Kok et al., 2020) were adjusted using the DustCOMM data (Fig. 1). To adjust the size bias in dust emissions, the mass fraction of emitted dust for each bin was prescribed according to the size resolved total column loading of DustCOMM at each 2-D grid box. Overall, the IMPACT simulated lifetime of the dust aerosol for the second size bin (7.8 days 1.26–2.5 μm of diameter) was in good agreement with the ensemble of model results (8.5 ± 1.1 days for the mass mean diameter of 1.8 μm) (Kok et al., 2017). To maintain the consistency in the $DAOD_{550}$, the mass fraction of dust concentration for each bin between 0.2 and 20.0 μm of diameter were adjusted at each 3-D grid box prior to calculating the radiative fluxes using the RRTMG based on the ratio of mass concentration of $PM_{2.5}$ (i.e., sum of bin 1 and bin 2) to each bin (Table 3).~~

~~Second, the approximation of particles to spheres is evaluated applying aspherical factors to the optical properties of the mass extinction coefficient, single scattering albedo, and asymmetry parameter for SW, as well as~~

~~absorption fraction of extinction for the LW. At the same time, we maintained the consideration of asphericity on the gravitational velocity and kept the dust concentrations unaltered between the spherical (default) and aspherical (fine-global) cases.~~

~~Third, the aerosol RE of mineral dust also depends on mineralogical composition. For the sensitivity simulation to refractive index, we used the global mean of laboratory measurements of the refractive index from 19 natural soils from various source regions around the world (Di Biagio et al., 2017, 2019) (denoted as "global"). To investigate the regional heterogeneity of refractive index, regional mean of refractive index was examined (denoted as "region"). The regionally averaged imaginary parts of the refractive indices at wavelength of 0.52 μm and 9.7 μm showed large differences in SW and LW absorptivity, respectively, between different samples collected at various geographical locations (Fig. 1). 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 was used for other areas. The mineral dust LW refractive index also depends on its mineralogical composition (Sokolik et at., 1998; Di Biagio et al., 2017). The LW refractive index of Volz (1983) has been widely used in climate models and satellite remote sensing algorithms and thus was examined here (denoted as "mineral"). To analyze the dependence of the results on less absorptive SW and more absorptive LW refractive indices, we made further sensitivity simulations by varying the values of imaginary parts of the refractive index within the range of values from Di Biagio et al. (2017, 2019) (10% and 90% percentiles for SW and LW, respectively) (denoted as "absorption"). The associated real parts with the 90% percentile imaginary parts for LW were calculated to account for the Kramers-Kronig relation (Lucarini et al., 2005).~~

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

~~Radiative effect efficiency is defined as the gradient of a linear least squares fit applied to the $DAOD_{550}$ and dust radiative effect at each 2-D grid box ($W \cdot m^{-2}$ $DAOD_{550}^{-1}$).~~ To estimate dust radiative effect efficiency, aerosol and radiation remote sensing products have been used with various methods (Table 4) (Zhang and Christopher 2003; Li et al. 2004; Christopher and Jones 2007; Brindley and Russell 2009; Yang et al. 2009; Di Biagio et al. 2010; Hansell et al. 2010; Hansell et al. 2012; Song et al. 2018).

The instantaneous SW radiative effect efficiency at TOA is obtained from the linear regression of TOA radiation flux versus AOD observations, although the values in low-dust periods can be substantially influenced by other types of aerosols such as biomass burning (Li et al. 2004). This radiative effect efficiency corresponds to the instantaneous value derived under the limited condition at the measurements (e.g., solar position, atmospheric

condition). From the extrapolation of the instantaneous value, the diurnal mean dust SW radiative effect efficiency at the surface and TOA can be derived based on model calculations.

The LW radiative effect efficiency at TOA can be obtained from the linear regression of TOA radiation flux versus AOD observations over the source regions (Brindley and Russell 2009). However, the observed outgoing ~~longwave~~ LW radiation is not only dependent on DAOD but also on other factors such as dust layer height, water vapor content, and other types of aerosols. Thus, the LW radiative effect efficiency is estimated from the difference between observed outgoing ~~longwave~~ LW radiation and the dust-free outgoing ~~longwave~~ LW radiation, which can be estimated using radiative transfer model (Song et al., 2018).

Consequently, the semi-observationally-based estimates of the dust radiative effect efficiency could be biased, in part, due to large uncertainties associated with the estimation method ~~such as spherical assumption on AOD retrieval (Zhou et al., 2020)~~, the selection of cloud-free and dust-dominant data, and dust physicochemical properties. To understand the sensitivity of the dust radiative effect efficiency to the particle size distribution, asphericity, and refractive index of dust, radiative transfer computations have been carried out in previous studies (Li et al., 2004; Song et al., 2018). Song et al. (2018) found that the combination of the coarser dust particle size distribution and the more absorptive LW refractive index (V83) yielded the best simulation of the dust LW radiative effect in comparison with the satellite flux observations (i.e., CERES), compared to the less absorptive LW refractive index (DB17).

3. Results and Discussions

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

3.1 Dust load and aerosol optical depth

We compared our model estimates of $DAOD_{550}$ against semi-observationally-based data in box plots and Taylor diagrams (Taylor, 2001) for the evaluation of the various model experiments against semi-observationally-based estimates (Ridley et al., 2016; Adebiyi et al., 2020) to provide a concise statistical summary of the bias, correlation coefficient, root mean square errors, and the ratio of standard deviation (Fig. 2, Tables S1 and S2). ~~Because assuming spherical dust underestimates the mass extinction efficiency by ~30% (Kok et al., 2017),~~ IMPACT-Sphere-Mineral-V83 (E1) ~~default~~ simulations resulted in a significant underestimation of the global and annual mean of $DAOD_{550}$ (0.023) (Fig. 2 and Table 3). The lower $DAOD_{550}$ from E2 than E1 was mostly found over East Asia and Bodele/Sudan in winter (Fig. ~~3~~2, Table S2). After considering the dust asphericity for spectral optical properties ~~(fine-global)~~, we adjusted IMPACT-simulated dust loads against the constraints on dust load ~~constrains of~~ from the DustCOMM data set. This adjustment led the simulated total dust load to increase from 25 Tg (E1~~default~~) to 32 Tg (E2~~improved simulation~~), which addressed the issue ~~that~~ of ~~model underestimated the~~ coarse dust underestimation and fine dust overestimation by the model ~~portion~~ (Fig. ~~3~~2, Table 3). ~~After all~~Consequently, the global and annual mean of $DAOD_{550}$ from DustCOMM-Asphere-DB19-V83 (E2)~~the improved~~ simulation (0.029) fell within the range in the semi-observationally-based estimate (0.030 ± 0.005) (Ridley et al., 2016) (Table 3). We found that the agreement in the median with the semi-observationally-based estimate (0.127)~~s~~ ~~in summer~~ was improved from IMPACT-Sphere-Mineral-DB17 (0.049) ~~default~~ to DustCOMM-Asphere-DB19-V83 (0.117)~~improved simulations, with the correlation coefficients (root mean square errors) increasing (decreasing) from 0.86 (0.08) to 0.91 (0.06), respectively~~ (solid line within the box mark in Fig. ~~3~~2d~~, Table S1, Fig. S1~~). We also found improvements over East Asia and Bodele/Sudan in winter ~~the other three seasons~~ (Table S2). The better agreement suggested that DustCOMM-Asphere-DB19-V83 (E2)~~the improved~~ simulation was reasonably constrained by the $DAOD_{550}$ (Ridley et al., 2016; Adebiyi et al., 2020).

3.2 Dust SW radiative effect efficiency

Modeled estimates of clear-sky dust SW radiative effect efficiencies ($W \cdot m^{-2}$ $DAOD_{550}^{-1}$) at the surface (Table S3) and TOA (Table S4) were compared with estimates reported by regional studies based on satellite observations over the N~~n~~orth Africa and the North Atlantic (Fig. 4). Comparisons for other regions and seasons were also ~~presented~~ included for SW in Figs. 4(g), 4(h), 4(i), and 4(j)~~S2~~. Sensitivity simulations demonstrated that the radiative effect efficiency strongly depended on the particle size, refractive index, and particle shape (Fig. 4~~ and Fig. S3~~). The adjustment of size-resolved dust concentration and shape with the same refractive index ~~worsened agreement~~led to overestimates of the SW radiative effect efficiencies against semi-observationally-based data ~~for the correlation~~

~~coefficients (root mean square errors) decreasing (increasing) from 0.96 (17)~~at TOA (from E1~~, default simulations)~~ to ~~0.94 (26) (from~~ E6~~coarse mineral) (~~ in Fig. ~~S2~~4h). Subsequently, the use of less absorptive SW refractive index with DustCOMM-Asphere-DB19-V83 (E2)~~the improved~~ simulations led to a better agreement ~~for the correlation coefficients (root mean square errors) increasing (decreasing) to 0.95 (22) (from the improved simulation) (~~from E6 to E2 in Fig. ~~S2~~4). ~~However~~On the other hand, the use of much less (10% percentile) absorptive SW refractive index from DustCOMM-Asphere-Less-More (E7)~~coarse absorption~~ simulation ~~(Fig. S2e)~~ deteriorated the agreement due to the underestimate of cooling at the surface (Fig. 4g). In contrast, the use of a more absorptive SW refractive index from DustCOMM-Asphere-Mineral-V83 (E6) improved the agreement at the surface. However, the semi-observationally-based estimates of diurnally averaged radiative effect efficiency at the surface were derived from extrapolation of the instantaneous values, which would affect the comparison due to differences in the methodologies between dust models (section 2.6). The differences in the model-based estimates of radiative effect efficiency might arise from different data sets of the refractive index, size distribution, and particle shape (Song et al., 2018).

3.3 Dust LW radiative effect efficiency

Modeled estimates of clear-sky dust LW (Fig. 5) radiative effect efficiencies ($W \cdot m^{-2}$ $DAOD_{550}^{-1}$) at the surface (Table S5) and TOA (Table S6) were compared with estimates reported by regional studies based on satellite observations over N~~n~~orth Africa and the North Atlantic. Comparisons for other regions and seasons were also ~~presented~~ included for LW Figs. 5(g), 5(h), 5(i), and 5(j)~~S4~~. Sensitivity simulations demonstrated that the radiative effect efficiency strongly depended on the particle size, refractive index, and particle shape (Fig. 5 ~~and Fig. S5~~). ~~The~~ Both the IMPACT-Sphere-Mineral-V83 (E1) and DustCOMM-Asphere-DB19-V83 (E2)~~improved~~ simulations yielded better agreement with semi-observationally-based data at the surface and TOA, compared to the less absorptive LW dust refractive indices~~, with the correlation coefficients (root mean square errors) increasing (decreasing) from 0.57 (12) (from default simulation) to 0.58 (6.3) (from the improved simulation)~~ (E3, E4, E5, and E7) (Fig. 5~~S4~~). ~~Substantial differences in dust LW radiative effect efficiencies between the default and the improved simulations were found at the surface over the major source regions. The improved simulation showed better agreements with semi-observation-based estimates of LW radiative effect efficiencies at the surface over tropical Atlantic (21 vs. 24 $W \cdot m^{-2}$ $DAOD^{-1}$) and China (33 vs. 31–35 $W \cdot m^{-2}$ $DAOD^{-1}$) compared to the default simulation (8.8 and 22 $W \cdot m^{-2}$ $DAOD^{-1}$, respectively).~~ The relatively high LW radiative effect efficiencies over western Africa were also consistent with the semi-observationally-based data. On the other hand, the relatively low LW radiative effect efficiencies were found over eastern Africa. Moving toward the northeastern side of the region, however, the associated uncertainties in the semi-observationally-based values increased (Brindley and Russell 2009). The dust LW

radiative effect efficiency depends strongly on the vertical profile of dust concentration, temperature, and water vapor, which would affect the comparison due to a high variability in these factors (section 2.6).

3.~~4~~5 Atmospheric ~~Dust~~ radiative heating ~~effect~~by dust due to coarser size and aspherical shape

The Saharan dust cools the ground surface by reducing the solar radiation reaching the surface and warms the atmosphere by absorbing solar radiation (Fig. 6). On the other hand, thermal emission by dust warms the surface and cools the atmosphere (Fig. 7). Our sensitivity simulations showed that the annually averaged net instantaneous radiative effect due to mineral aerosol (NET) rangeds from –0.~~3~~38 (DustCOMM-Asphere-Less-Less~~coarse-absorption~~) to +0.25 (DustCOMM-Asphere-Mineral-V83~~coarse-mineral~~) W·m^{-2} at TOA (Table 5). The net RE from ~~B~~both the IMPACT-Sphere-Mineral-V83 (~~–0.00 W·m^{-2}~~)~~default~~ and DustCOMM-Asphere-DB19-V83 (–0.08 W·m^{-2})~~improved~~ simulations resulted within ~~similar NET RE (–0.09 and –0.08 W·m^{-2}, respectively) to~~ 98% confidential interval of DustCOMM data set (–0.~~11~~27 ~~±~~ to 0.1~~0~~4 W·m^{-2}).

The SW RE by dust outweighs the ~~longwave~~ LW warming effect at the surface in the ~~default~~ IMPACT-Sphere-Mineral-V83 (E1) simulation (Fig. 8). Consequently, the highly absorbing dust could play an important role in the aerosol radiative forcing for the climate models to alter the West African monsoon, with the radiative heating concentrated in the dust layer (Miller et al., 2004b; Lau et al., 2009). Our model results of dust RE from DustCOMM-Asphere-DB19-V83 (E2)~~improved~~ simulation, however, suggested that the surface warming was substantially enhanced near the strong dust source regions~~much less cooling~~ (−0.23 W·m^{-2} on a global scale) ~~at the surface~~(Fig. 8), compared to the V83~~default~~ simulation (−0.~~60~~88 W·m^{-2} on a global scale)~~, despite similar RE at TOA on a global scale~~. Thus, our results demonstrated that the atmospheric radiative heating ~~due to~~by mineral dust was substantially reduced for DustCOMM-Asphere-DB19-V83 (E2)~~the improved~~ simulation (0.15 W·m^{-2}), compared to the IMPACT-Sphere-Mineral-V83 (E1) ~~default~~ simulation (0.~~7~~59 W·m^{-2}). ~~The improved simulation was obtained from the evaluation against semi-observation-based estimates of DAOD$_{550}$, SW, and LW radiative effect efficiencies.~~

3.5 Synergy of coarser size and aspherical shape

To ~~investigate~~ elucidate the differences in dust radiative effects between the IMPACT-Sphere-Mineral-V83 (E1) ~~default~~ and DustCOMM-Asphere-DB19-V83 (E2)~~improved~~ simulations (Fig. 9), the differences in annually averaged radiative effects of mineral dust from DustCOMM-Asphere-DB19-DB17 (E4)~~coarse-global~~ simulation were shown in Fig. 10~~9~~. A slope of one in Fig. 10~~9~~ represented an identical change in both the surface and TOA~~,~~ and thus corresponded to no change in radiative heating within the atmosphere. The distances from the DustCOMM-Asphere-DB19-DB17 (E4)~~coarse-global~~ simulation demonstrated that ~~the~~ large uncertainties existed for the size distribution

and spectral optical properties~~refractive index~~. Our sensitivity simulations revealed that the DustCOMM-Asphere-DB19-V83 (E2)~~the improved~~ simulation led to a similar ~~enhancement of NET~~net RE at TOA to the IMPACT-Sphere-Mineral-V83 (E1) ~~default~~ simulation ~~(+0.12 vs. +0.11 W·m^{-2} at the vertical axis of red star and black star, respectively, in Fig. 9c)~~ but result~~eds~~ in less cooling at the surface ~~(+0.42 vs. −0.20 W·m^{-2} at the horizontal axis in Fig. 9c)~~ (Fig. 9). This revision can be divided into (1) the size-resolved abundance ~~for both SW and LW~~ (black ~~triangles~~ hexagons in Fig. 10 ~~in Fig. 109a and Fig. 910b, respectively~~), (2) ~~mineralogical variability~~refractive index ~~for SW (red diamonds in Fig. 9a) and for LW~~ (red diamonds and triangles~~in Fig. 9b~~), and (3) particle shape (red circles).

First, at TOA, the SW RE was more sensitive to the size-resolved abundance (–0.17 W·m^{-2} at the vertical axis of black hexagon~~triangle~~ in Fig. 109a), compared to LW (0.0~~1~~0 W·m^{-2} at the vertical axis of black hexagon~~triangle~~ in Fig. 109b) ~~(Liao and Seinfeld, 1998)~~. Second, this less SW cooling effect with coarser dust was partially compensated for by more SW cooling by the less absorptive SW refractive index (0.3~~4~~3 W·m^{-2} at the vertical axis of red diamond in Fig. 109a). Thirdly, the sensitivity of SW RE to dust asphericity was rather minor (0.04 W·m^{-2} at the vertical axis of red circle in Fig. 10a), partly because the lower DAOD was compensated for by the lower asymmetry parameter of spherical dust, which enhanced the amount of radiation scattered backward to space (Räisänen et al., 2013; Colarco et al., 2014). The partial compensation led to a small enhancement of SW RE for the IMPACT-Sphere-Mineral-V83 (E1) ~~default~~ simulation ~~(0.13 W·m^{-2} at the vertical axis of black star in Fig. 9a)~~ and thus the resulting similar ~~NET~~ net RE to DustCOMM-Asphere-DB19-V83 (E2) at TOA (Fig. 9~~c~~).

In contrast, at the surface, our sensitivity simulations demonstrated substantially different responses in the RE, mostly because of LW warming effects (~~+0.42 vs. −0.20 W·m^{-2} at the horizontal axis in~~ Fig. ~~10~~9~~b~~). The ~~enhanced SW cooling by the more absorbing dust (−0.38 W·m^{-2} at the horizontal axis of red diamond in Fig. 9a) was mostly compensated for by the reduced SW cooling with coarser dust (0.12 W·m^{-2} at the horizontal axis of black triangle in Fig. 9a) and asphericity. This led to minor change in SW cooling by the default simulation (−0.03 W·m^{-2} at the horizontal axis of black star in Fig. 9a). In contrast,~~ enhanced LW warming by coarser dust (–0.08 W·m^{-2} at the horizontal axis of black hexagon in Fig. 10b)~~more absorptive LW refractive index (+0.42 W·m^{-2} at the horizontal axis of red diamond in Fig. 9b)~~ was accompanied ~~with enhanced LW warming~~ by the ~~coarser dust (0.07 W·m^{-2} at the horizontal axis of black triangle in Fig. 9b) and~~ asphericity (–0.1~~5~~3 W·m^{-2} at the horizontal axis of red circle~~from the difference between black triangle and black star~~ in Fig. 109b), because the enhancement of the absorption fraction of extinction due to asphericity was larger at coarser size. Th~~ie~~s enhanced LW warming effects of each as well as the synergy was further amplified using the more absorptive LW dust refractive index (Volz, 1983) (at the horizontal axis of red diamond in Fig. 10b). As a result, our sensitivity simulations revealed that substantially less dust absorption at LW due to the underestimation of the coarse dust load and the assumption of the spherical shape (IMPACT-Sphere-

Mineral-V83) contributed to the less surface warmingthe reduced radiative heating within the atmosphere, compared to DustCOMM-Asphere-DB19-V83 (Fig. 9) (less than the dashed line of red star in Fig. 9c) by the improved simulation (−0.30 W·m^{-2}), compared to the enhanced radiative heating (more than the dashed line of black star in Fig. 9c) by default simulation (0.34 W·m^{-2}).

A relatively good agreement of net RE by dust at TOA with Di Biagio et al. (2020) (–0.06 W·m^{-2}) could be obtained from both the IMPACT-Sphere-Mineral-V83 (E1) and DustCOMM-Asphere-DB19-V83 (E2) simulations (Fig. 9 and Table 5). On the other hand, our modeled dust net RE at the surface from DustCOMM-Asphere-DB19-V83 (E2) was much larger than Di Biagio et al. (2020) (–0.63 W·m^{-2}) and IMPACT-Sphere-Mineral-V83 (E1). The synergy of coarser size and aspherical dust could contribute to the less surface warming of the DustCOMM-Asphere-DB19-V83 (E2). At the same time, the more absorptive LW dust refractive index (V83) than DB17 (red diamond in Fig. 10b) could also contribute to the less surface warming, which might be partially compensated for in our model by the omission of dust with diameters in excess of 20 μm. Consequently, our estimate of atmospheric radiative heating by dust from DustCOMM-Asphere-DB19-V83 (E2) was lower than Di Biagio et al. (2020) (0.63 W·m^{-2}) and IMPACT-Sphere-Mineral-V83 (E1). Additionally, the hot and dry climate over brighter desert surface exaggerates differences in RE at the surface between the models (Miller et al., 2014). The low humidity allows dust particles to absorb LW radiation with reduced competition from water vapor, while high temperatures within the boundary layer increase downward thermal emission by dust (Liao and Seinfeld, 1998). The reduction of fine dust load after the adjustment leads to underestimates of the SW cooling at TOA. To improve agreement against semi-observationally-based estimate of the radiative effect efficiency at TOA, the less absorptive SW dust refractive index is required for coarser aspherical dust. Thus, uncertainties in the size-resolved dust concentration, particle shape, and refractive index contribute to the diversity in the simulated dust RE at the surface.

4. Conclusions

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

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

The diversity of modeled dust net RE at the surface (–1.74 W·m^{-2} to –0.20 W·m^{-2}) is much larger than at TOA (–0.01 W·m^{-2} to –0.61 W·m^{-2}), partly because the refractive index is optimized to obtain reasonable agreement against satellite observations of TOA radiation flux (e.g., CERES). The uncertainties in the size-resolved dust concentration, particle shape, and refractive index contribute to the model diversity at the surface. DustCOMM-Asphere-DB19-V83 (E2)The improved simulation led to similar net global dust radiative effect at TOA on a global scale to IMPACT-Sphere-Mineral (E1)the default simulation (−0.08 vs. −0.009 W·m^{-2}) but resulted in less cooling at the surface by the synergy of coarser size and aspherical shape, compared to IMPACT-Sphere-Mineral-V83 (E1) simulation (–0.23 vs. –0.88 W·m^{-2} on a global scale). Consequently, the atmospheric heating due to mineral dust was substantially reduced for the DustCOMM-Asphere-DB19-V83 (E2) improved simulation (0.15 W·m^{-2}), compared to the intensified atmospheric heating from the IMPACT-Sphere-Mineral-V83 (E1)default IMPACT simulation (0.759 W·m^{-2}). The less intensified atmospheric heating due to mineral dust could substantially modify the vertical temperature profile in Earth system models, and thus has important implications for the projection of dust feedback near the major source regions in the past and future climate changes (Kok et al., 2018). More accurate estimates of semi-observationally-based dust SW and LW radiative effect efficiencies over strong dust source regions are needed to narrow the uncertainty in the RE.

Currently, the model did not include dust particles above 20 μm, but a substantial fraction of airborne dust near source regions may be above this threshold (Ryder et al., 2019). Moreover, such large particles can be transported to higher altitudes and longer distances than the model prediction. The higher the dust layer resides, the larger the dust LW RE at TOA is estimated under the clear-sky conditions (Liao and Seinfeld, 1998). Marine sediment traps, which are located underneath the main Saharan dust plume in the Atlantic Ocean, suggest that giant particles are dominated by platy mica and rounded quartz particles (van der Does et al., 2016). Thus, mineral composition of the giant particles could be different from the aerosol samples generated from soils in the laboratory by Di Biagio et al. (2017), which may reflect less absorbing LW refractive index of DB17 than V83. Indeed, the dust sample was collected for V83 from rainwater after strong wind. On the other hand, the contribution of the LW scattering might be underestimated

in the models, as Di Biagio et al. (2020) noted that the adjustment factor was estimated for dust of diameter less than 10 µm and thus might be a lower approximation of the LW scattering by coarse dust. Therefore, a better understanding of the effect of such large particles beyond 20 µm and mineralogical composition on radiation balance remains a topic of active research, given their potential to amplify the warming of the climate system. In such an extreme case as the "Godzilla" dust storm over the North Africa and the tropical Atlantic in June 2020 (Francis et al., 2020), the dust loading could be larger than that examined for this study, and our estimates of the warming effects might be conservative during such events. However, to keep the giant particles in the atmosphere, the modeled deposition fluxes should be reduced from the current model. Therefore, models should improve their ability to capture the evolution of the dust size distribution as the plumes move downwind of the source regions.

Code availability.

The source code of the RRTMG hasve been obtained from the website~~web site~~ at https://github.com/AER-RC/RRTMG_LW and https://github.com/AER-RC/RRTMG_SW. The source code of the Kramers-Kronig relations hasve been obtained from the website~~web site~~ at 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 https://www.mathworks.com/matlabcentral/fileexchange/20559-taylor-diagram.

Data availability.

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

Supplement.

The supplement related to this article is available online at:

Author contributions.

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

Competing interests.

The authors declare that they have no competing interests.

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Figure captions

Figure 1. Imaginary part of the refractive index at ~~a wavelength of 0.52~~SW and ~~9.7 μm~~LW for the nine main source regions and the global mean for others. 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 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. 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)default, (c) fine dust from DustCOMM-Asphere-DB19-V83 (E2)improved, (d) coarse dust from DustCOMM, (e) coarse dust from E1, default and (f) coarse dust from E2improved simulations. The parentheses represented the global dust burden (Tg). The values of dust load at each bin were listed in Table 3.~~

Figure ~~3~~2. The model better reproduced semi-observationally-based data of $DAOD_{550}$ ~~in boreal summer (June, July, and August)~~ after adjusting the size-resolved dust load with DustCOMM and considering the dust asphericity. (a) semi-observationally-based estimates of the $DAOD_{550}$ were averaged over 2004–2008 (Ridley et al., 2016; Adebiyi et al., 2020). The annually averaged model results were shown for (b) ~~default~~ DustCOMM-Asphere-DB19-V83 (E2)~~,~~ and (c) the differences between~~improved~~ IMPACT-Sphere-Mineral-V83 (E1) and E2~~, and (d) fine-global~~ simulations. (d) Comparison of seasonally averaged $DAOD_{550}$ 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 × 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 $DAOD_{550}$ 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 of the fifteen regions were: (A1) Mid-Atlantic (4°–40°N, 50°–20°W), (A2) African West Coast (10°–34°N, 20°–5°W), (A3) Northern Africa (26°–40°N, 5°W–30°E), (A4) Mali/Niger (10°–26°N, 5°W–10°E), (A5) Bodele/Sudan (10°–26°N, 10°–40°E), (A6) Northern Middle East (26°–40°N, 30°–50°E), (A7) Southern Middle East (0°–26°N, 40°–67.5°E), (A8) Kyzyl Kum (26°–50°N, 50°–67.5°E), (A9) Thar (20°–50°N, 67.5°–75°E), (A10) Taklamakan (30°–50°N, 75°–92.5°E), (A11) Gobi (36°–50°N, 92.5°–115°E), (A12) North America (20°–45°N, 80°–130°W), (A13) South America (0°–55°S, 80°–55°W), (A14) Southern Africa (10°–35°S, 10°–40°E), and (A15) Australia (10°–40°S, 110°–160°E). The values of $DAOD_{550}$ at the 15 regions (marked in Fig. 3a) in summer were listed in Table S1. ~~The scatter plot was shown in Fig. S1. The summer season was shown because most semi-observation-based data on dust radiative effects were available.~~ 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 (Tg). The values of dust load at each bin were listed in Table 3.

Figure 4. ~~Summertime~~ ~~D~~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)~~default~~, and (e) and (f) DustCOMM-Asphere-DB19-V83 (E2)~~improved~~ simulations at the surface and TOA, respectively. (g) Comparison of seasonally averaged SW radiative effect efficiency for semi-observationally-based (SOB) data and the different experiments. 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 × interquartile range, and the points indicate the outside of the range. (h) Taylor diagram summarizing the statistics of the comparison against the seasonally averaged regional SW radiative effect efficiency 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 regionally averaged values were listed in Tables S3 and S4 at the surface and TOA, respectively. ~~The scatter plot was shown in Fig. S2. The results from other sensitivity simulations were also shown in Fig. S3.~~

Figure 5. ~~Summertime d~~Dust clear-sky LW radiative effect efficiency ($W \cdot m^{-2}$ $DAOD_{550}^{-1}$). 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)~~default~~, and (e) and (f) DustCOMM-Asphere-DB19-V83 (E2)~~improved~~ simulations at the surface and TOA, respectively. (g) Comparison of seasonally averaged LW radiative effect efficiency for semi-observationally-based (SOB) data and the different experiments. 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 × interquartile range, and the points indicate the outside of the range. (h) Taylor diagram summarizing the statistics of the comparison against the seasonally averaged regional SW radiative effect efficiency 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 regionally averaged values were listed in Tables S5 and S6 at the surface and TOA, respectively. ~~The scatter plot was shown in Fig. S4. The results from other sensitivity simulations were also shown in Fig. S5.~~

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

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

(e) E1~~default~~ at TOA, and (f) E2~~improved~~ simulations at TOA. The numbers in parentheses represented the global mean.

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

Figure 9. Radiative effect ($W \cdot m^{-2}$) of mineral dust due to different studies 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.

Table 1. Summary of ~~sevt~~en ~~simulations~~ experiments compared in this study.

Number~~Experiment~~	~~Simulation~~Experiment	Size-resolved dust	Sphericity	SW refractive index	LW refractive index
E~~xperiment~~ 1	IMPACT-Sphere-Mineral-V83~~DB17Default~~	IMPACT	Spherical	Mineralogical map[d]	Volz (1983)~~Di Biagio et al. (2017)~~
E~~xperiment~~ 2[a]	DustCOMM-Asphere-DB19-V83~~Improved~~	DustCOMM[b]	Aspherical[c]	Di Biagio et al. (2019)	Volz (1983)
E~~xperiment~~ 3	IMPACT~~Fine~~-Asphere-DB19-DB17~~global~~	IMPACT	Aspherical[c]	Di Biagio et al. (2019)	Di Biagio et al. (2017)
E~~xperiment~~ 4	DustCOMM~~Coarse~~-Asphere-DB19-DB17~~global~~	DustCOMM[b]	Aspherical[c]	Di Biagio et al. (2019)	Di Biagio et al. (2017)
E5	DustCOMM-Sphere-DB19-DB17	DustCOMM[b]	Spherical	Di Biagio et al. (2019)	Di Biagio et al. (2017)
~~Experiment 5~~	~~DustCOMMCoarse-Asphere-Rregion~~	~~DustCOMM[b]~~	~~Aspherical[e]~~	~~Regional mean[e]~~	~~Regional mean[e]~~
E~~xperiment~~ 6	DustCOMM~~Coarse~~-Asphere-M~~m~~ineral-V83	DustCOMM[b]	Aspherical[c]	Mineralogical map[d]	Volz (1983)
E~~xperiment~~ 7	DustCOMM~~Coarse~~-Asphere-Less-More~~absorption~~	DustCOMM[b]	Aspherical[c]	Less SW[f~~e~~]	More LW[g]
E8	DustCOMM-Asphere-Less-Less	DustCOMM[b]	Aspherical[c]	Less SW[e]	Less LW[h]
E9	DustCOMM-Asphere-More-More	DustCOMM[b]	Aspherical[c]	More SW[f]	More LW[g]
E10	DustCOMM-Asphere-More-Less	DustCOMM[b]	Aspherical[c]	More SW[f]	Less LW[h]

[a]Combination of DustCOMM-Asphere-DB19-DB17 (E~~xperiment~~ 4) for SW and DustCOMM-Asphere-Mineral-V83 (E6~~xperiment 7~~) for LW.

[b]Size-resolved dust concentration was adjusted with semi-observationally-based estimate (Adebiyi & Kok, 2020).

[c]Dust asphericity was considered in calculating the optical properties~~refractive index~~, which further assumed internal mixing of minerals (Huang et al., 2020b) using a volume-weighted mixture for each size bin.

[d]Mineralogical 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).

~~[e]Regional mean of refractive index was calculated for the nine main source regions (Fig. 1). The global mean was used for other areas.~~

[f]Less absorptive SW refractive indies 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).

[f]More absorptive SW refractive indies 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).

[g]More 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).

[h]Less 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	~~More absorptive LW, c~~Coarser particle size~~,~~ & aspherical shape	E~~xperiment~~ 2 – E~~xperiment~~ 1
Less absorptive SW & aspherical shape	Less absorptive LW & ~~A~~aspherical shape	E~~xperiment~~ 3 – E~~xperiment~~ 1
Size-resolved dust abundance	Size-resolved dust abundance	E~~xperiment~~ 3 – E~~xperiment~~ 4
Aspherical shape	Aspherical shape	E5 – E4
~~Regional variability in refractive index~~	~~Regional variability in refractive index~~	~~Experiment 5 – Experiment 4~~
Mineralogical variability in refractive index (more absorptive SW)	Mineralogical variability in refractive index (more absorptive LW)	E~~xperiment~~ 6 – E~~xperiment~~ 4
Less absorptive SW (10% percentile)	More absorptive LW (90% percentile)	E~~xperiment~~ 7 – E~~xperiment~~ 4
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 ($m^2 \cdot g^{-1}$), and $DAOD_{550}$ at each bin on a global scale. The size-resolved dust concentration and shape in IMPACT-Sphere-Mineral-V83 (E1)~~default~~ simulation was adjusted to DustCOMM in DustCOMM-Asphere-DB19-V83 (E2)~~improved~~ 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)~~the default~~ and IMPACT-Asphere-DB19-DB17~~fine global~~ simulations (E3).

Dust size bin	Dust load			Mass extinction efficiency				$DAOD_{550}$			
	E1~~Default~~	E2~~Improved~~	DustCOMM	E1~~Default~~	E2~~Improved~~	E3~~Fine global~~	DustCOMM	E1~~Default~~	E2~~Improved~~	E3~~Fine global~~	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

[a]Bin 1 in IMPACT-Sphere-Mineral-V83 (E1)~~the default model~~ is 0.1–1.26 μm, whereas bin1 in DustCOMM-Asphere-DB19-V83 (E2)~~the improved model~~ 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

[a]Yang et al. (2009). [b]Li et al. (2004). [c]Song et al. (2018). [d]Christopher and Jones (2007). [e]Zhang and Christopher (2003). [f]Brindley and Russell (2009). [g]Di Biagio et al. (2010). [h]Hansell et al. (2010). [i]Hansell 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
Experiment 1	IMPACT-Sphere-Mineral-V83DB17Default	–0.18 (–1.326)	1.07	+0.09 18 (0.3766)	–0.2948	–0.009 (–0.8860)	0.579
Experiment 2	DustCOMM-Asphere-DB19-V83Improved	**–0.32 (–1.23)**[b]	**0.91**[b]	**+0.23 (1.00)**[b]	**–0.77**[b]	**–0.08 (–0.23)**[b]	**0.15**[b]
Experiment 3	IMPACTFine-Asphere-DB19-DB17global	–0.49 (–1.35)	0.86	+0.12 (0.50)	–0.38	–0.37 (–0.84)	0.48
Experiment 4	DustCOMMCoarse-Asphere-DB19-DB17global	**–0.32 (–1.23)**[b]	**0.91**[b]	+0.121 (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
Experiment 5	**DustCOMM-AsphereCoarse-Rregion**	–0.33 (–1.20)	0.87	+0.11 (0.53)	–0.43	–0.23 (–0.67)	0.44
Experiment 6	DustCOMM-AsphereCoarse-mMineral-V83	+0.02 (–1.61)	1.63	**+0.23 (1.00)**[b]	**–0.77**[b]	+0.25 (–0.62)	0.87
Experiment 7	DustCOMM-AsphereCoarse-Less-Moreabsorption	–0.54 (–1.0.98)	0.43	+0.16 (0.76)	–0.60	–0.38 (–0.22)	–0.16
E8	DustCOMM-Asphere-Less-Less	–0.54 (–1.0.98)	0.43	+0.06 (0.35)	–0.29	–0.48 (–0.36)	0.15
E9	DustCOMM-Asphere-More-More	–0.08 (–1.1.51)	1.43	+0.16 (0.76)	–0.60	+0.09 (–0.75)	0.84
E10	DustCOMM-Asphere-More-Less	–0.08 (–1.1.51)	1.43	+0.06 (0.35)	–0.29	–0.01 (–1.16)	1.15
	DustCOMM (Adebiyi & Kok, 2020)	–0.59 to 0.17[c] –0.43 ± 0.21		+0.25 to 0.41[c] ± 0.32 ± 0.08		–0.27 to 0.14[c] –0.11 ± 0.10	

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]

[a]The parentheses represent the RE at the surface. [b]The bold represents the combination of DB19~~coarse global~~ for SW and V83~~coarse mineral~~ for LW (i.e., DustCOMM-Asphere-DB19-V83~~improved simulation~~). [c]98% confidential interval of DustCOMM data set is listed. [d]For a comparison with our estimates, sum of single mode simulations from Di Biagio et al. (2019) is listed.

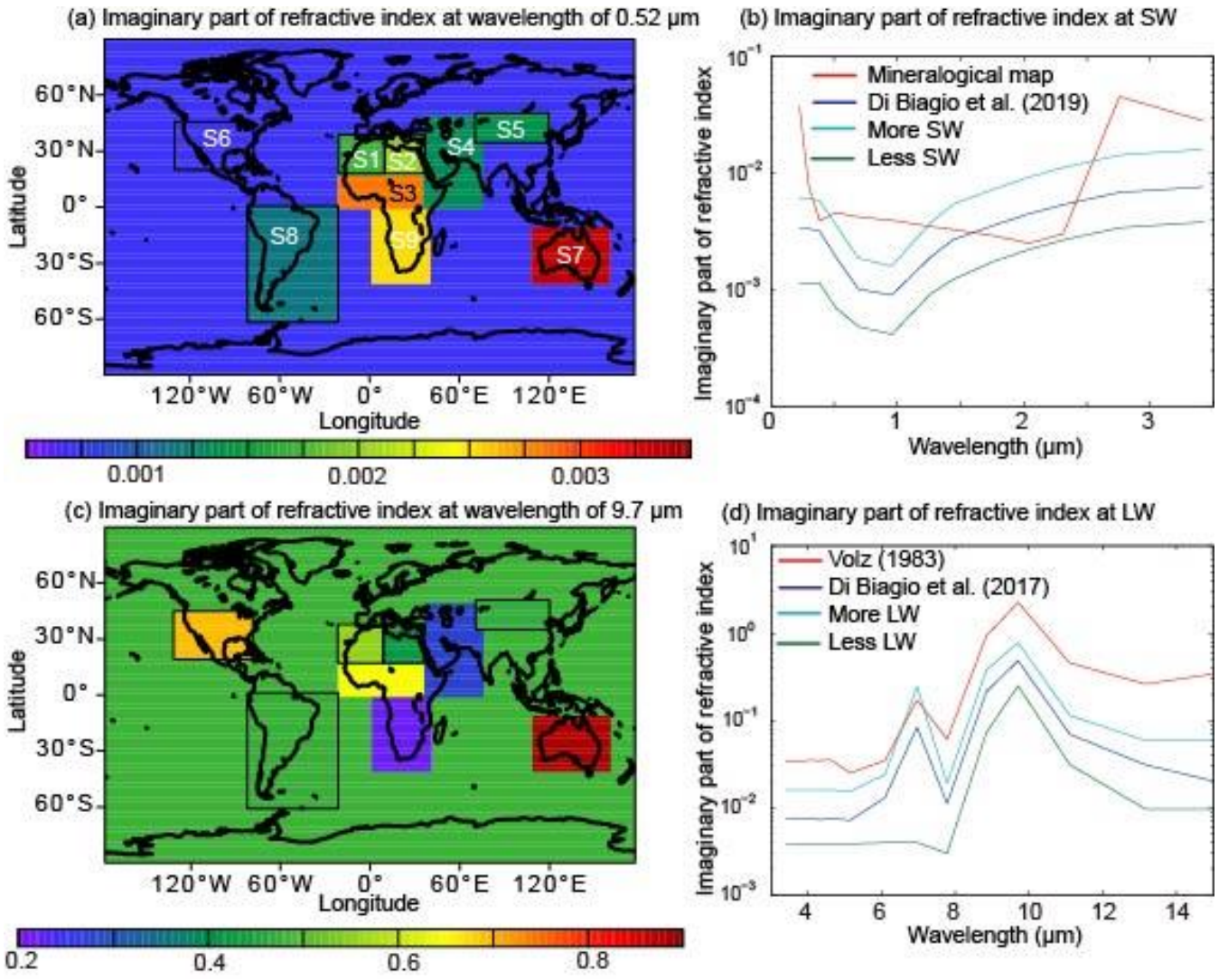


Figure 1. Imaginary part of the refractive index at ~~a wavelength of 0.52~~SW and ~~9.7 μm~~LW for the nine main source regions and the global mean for others. The refractive index obtained from 19 samples was aggregated into 9 main source regions and the arithmetic mean was calculated for each source region (Di Biagio et al., 2017, 2019). The 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).

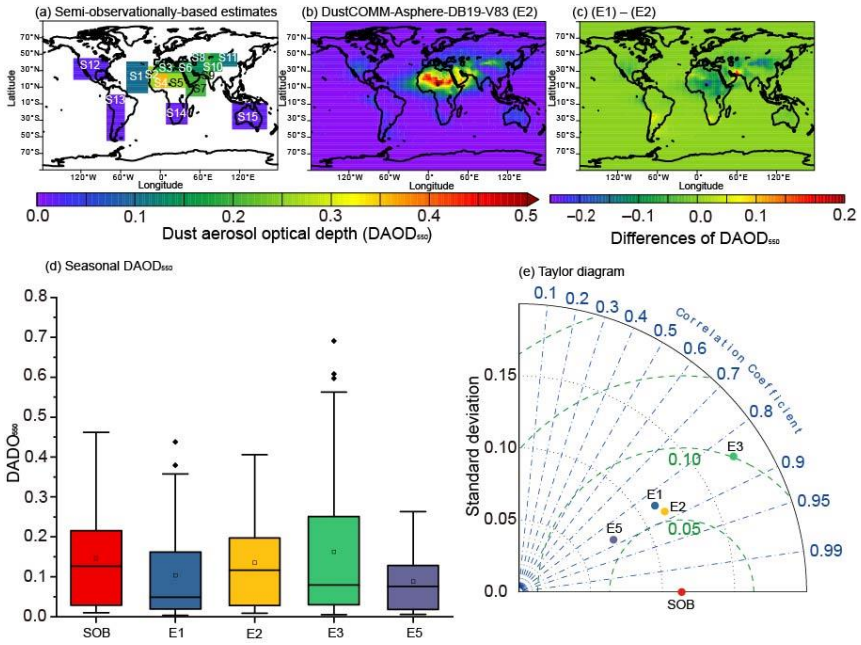
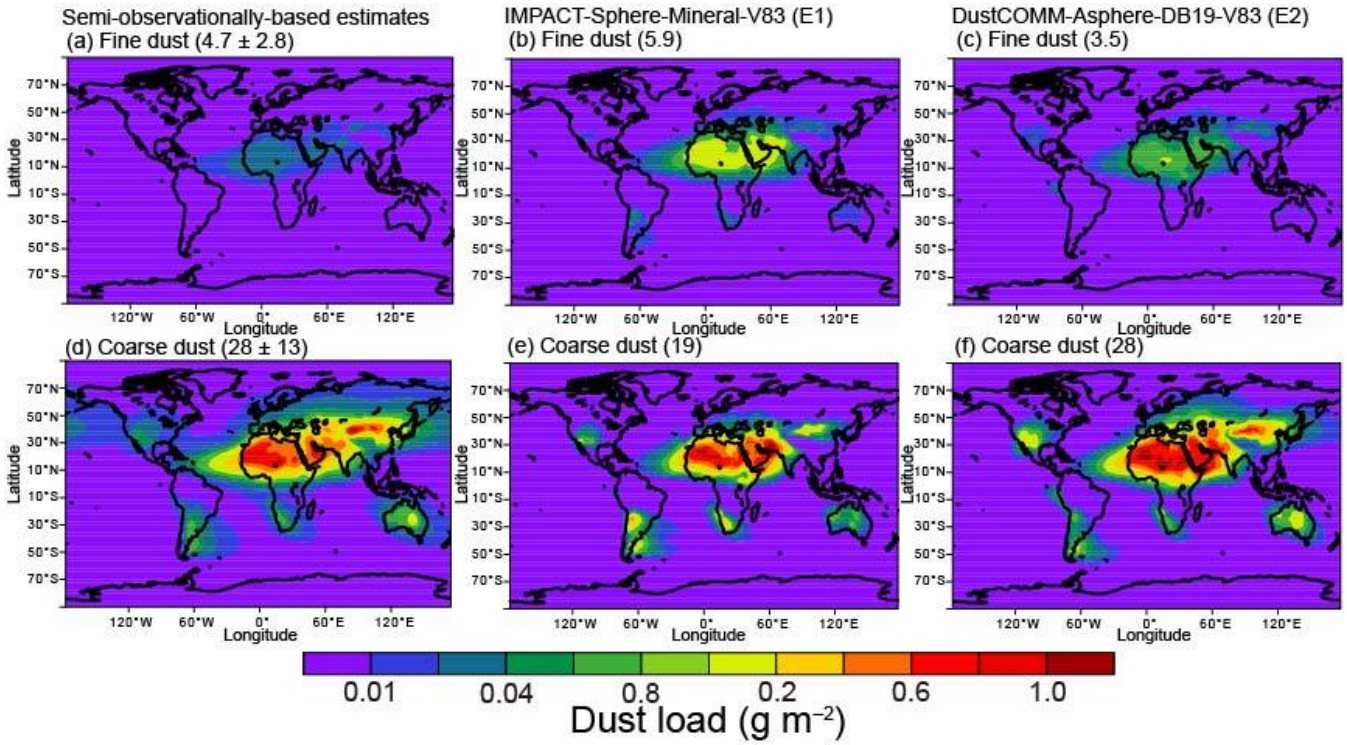


Figure ~~3~~2. The model better reproduced semi-observationally-based data of $DAOD_{550}$ ~~in boreal summer (June, July, and August)~~ after adjusting the size-resolved dust load with DustCOMM and considering the dust asphericity. (a) semi-observationally-based estimates of the $DAOD_{550}$ were averaged over 2004–2008 (Ridley et al., 2016; Adebiyi et al., 2020). The annually averaged model results were shown for (b) ~~default~~DustCOMM-Asphere-DB19-V83 (E2)~~,~~ and (c) the differences between~~improved~~ IMPACT-Sphere-Mineral-V83 (E1) and E2~~, and (d) fine global~~ simulations. (d) Comparison of seasonally averaged $DAOD_{550}$ 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 × 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 $DAOD_{550}$ 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 of the fifteen regions were: (A1) Mid-Atlantic (4°–40°N, 50°–20°W), (A2) African West Coast (10°–34°N, 20°–5°W), (A3) Northern Africa (26°–40°N, 5°W–30°E), (A4) Mali/Niger (10°–26°N, 5°W–10°E), (A5) Bodele/Sudan (10°–26°N, 10°–40°E), (A6) Northern Middle East (26°–40°N, 30°–50°E), (A7) Southern Middle East (0°–26°N, 40°–67.5°E), (A8) Kyzyl Kum (26°–50°N, 50°–67.5°E), (A9) Thar (20°–50°N, 67.5°–75°E), (A10) Taklamakan (30°–50°N, 75°–92.5°E), (A11) Gobi (36°–50°N, 92.5°–115°E), (A12) North America (20°–45°N, 80°–130°W), (A13) South America (0°–55°S, 80°–55°W), (A14) Southern Africa (10°–35°S, 10°–40°E), and (A15) Australia (10°–40°S, 110°–160°E). The values of $DAOD_{550}$ at the 15 regions (marked in Fig. ~~3~~2a) in summer were

listed in Table S1. ~~The scatter plot was shown in Fig. S1. The summer season was shown because most semi-observation-based data on dust radiative effects were available.~~ The comparison for other seasons was presented in Table S2.



35 **Figure ~~2~~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)~~default~~, (c) fine dust from DustCOMM-Asphere-DB19-V83 (E2)~~improved~~, (d) coarse dust from DustCOMM, (e) coarse dust from E1, ~~default~~ and (f) coarse dust from E2~~improved~~ simulations. The parentheses represented the global dust burden (Tg). The values of dust load at each bin were listed in Table
40 3.

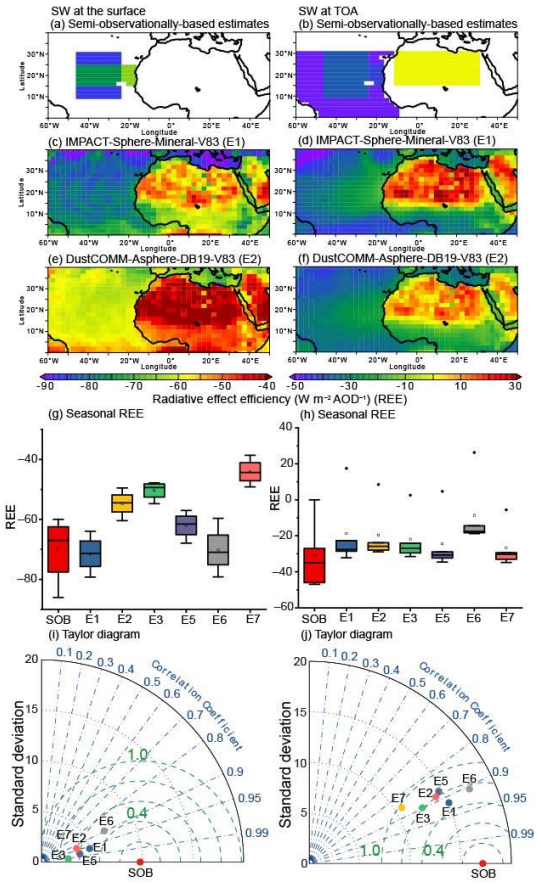


Figure 4. ~~Summertime~~ ~~D~~dust clear-sky SW radiative effect efficiency (W·m^{-2} DAOD^{-1}). Semi-observationally-based data at (a) the surface and (b) TOA were based on satellite observations (Yang et al. 2009; Li et al., 2004: Song et al., 2018; Christopher and Jones, 2007). The model results were shown for (c) and (d) IMPACT-Sphere-Mineral-V83 (E1)~~default~~, and (e) and (f) DustCOMM-Asphere-DB19-V83 (E2)~~improved~~ simulations at the surface and TOA, respectively. (g) Comparison of seasonally averaged SW radiative effect efficiency for semi-observationally-based (SOB) data and the different experiments. 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 × interquartile range, and the points indicate the outside of the range. (h) Taylor diagram summarizing the statistics of the comparison against the seasonally averaged regional SW radiative effect efficiency 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 regionally averaged values were listed in Tables S3 and S4 at the surface and TOA, respectively. ~~The scatter plot was shown in Fig. S2. The results from other sensitivity simulations were also shown~~

~~in Fig. S3.~~

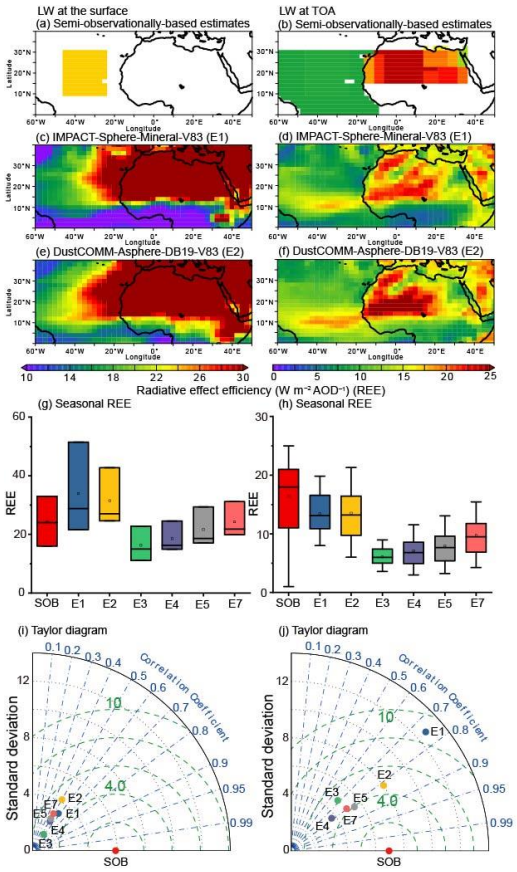


Figure 5. ~~Summertime d~~Dust clear-sky LW radiative effect efficiency (W·m^{-2} DAOD$_{550}^{-1}$). Semi-observationally-based estimates at (a) surface and (b) TOA were based on satellite observations (Song et al., 2018; Christopher and Jones, 2007; Zhang and Christopher, 2003; Brindley and Russell, 2009; Yang et al., 2009). The model results were shown for (c) and (d) IMPACT-Sphere-Mineral-V83 (E1)~~default~~, and (e) and (f) DustCOMM-Asphere-DB19-V83 (E2)~~improved~~ simulations at the surface and TOA, respectively. (g) Comparison of seasonally averaged LW radiative effect efficiency for semi-observationally-based (SOB) data and the different experiments. 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 × interquartile range, and the points indicate the outside of the range. (h) Taylor diagram summarizing the statistics of the comparison against the seasonally averaged regional SW radiative effect efficiency 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 regionally averaged values were listed in Tables S5 and S6 at the surface and TOA, respectively. ~~The scatter plot was shown in Fig. S4. The results from other sensitivity simulations were also shown in Fig. S5.~~

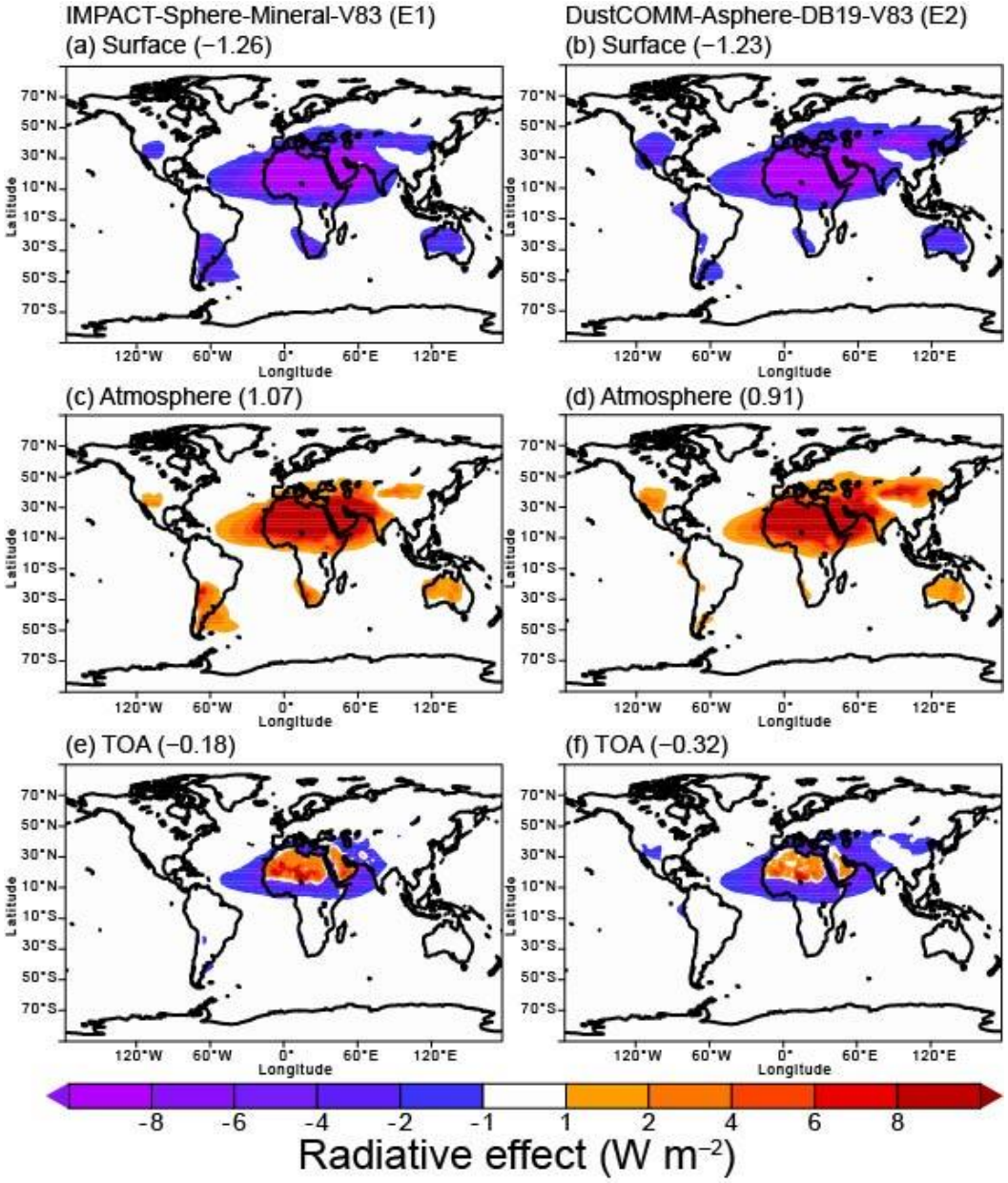


Figure 6. Dust SW radiative effect (W·m^{-2}) and radiative heating of the atmosphere (i.e., the subtraction of radiative effects from TOA to the surface in unit of W·m^{-2}). The model results were shown for the simulations for (a) IMPACT-Sphere-Mineral-V83 (E1)~~default~~ at the surface, (b) DustCOMM-Asphere-DB19-V83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

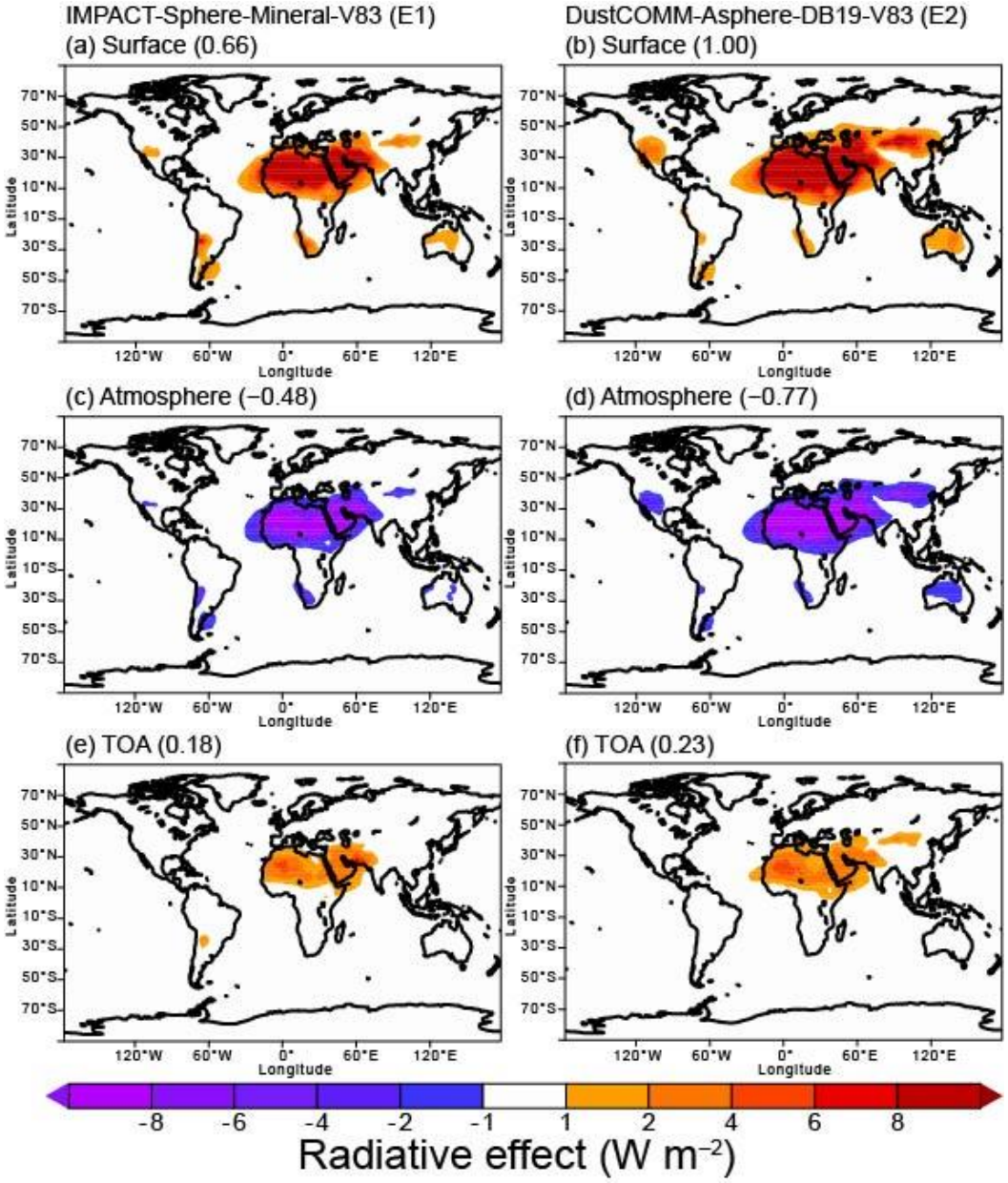


Figure 7. Dust LW radiative effect ($W \cdot m^{-2}$) and radiative heating of the atmosphere (i.e., the subtraction of radiative effects from TOA to the surface in unit of $W \cdot m^{-2}$). The model results were shown for the simulations for (a) IMPACT-Sphere-Mineral-V83 (E1)~~default~~ at the surface,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

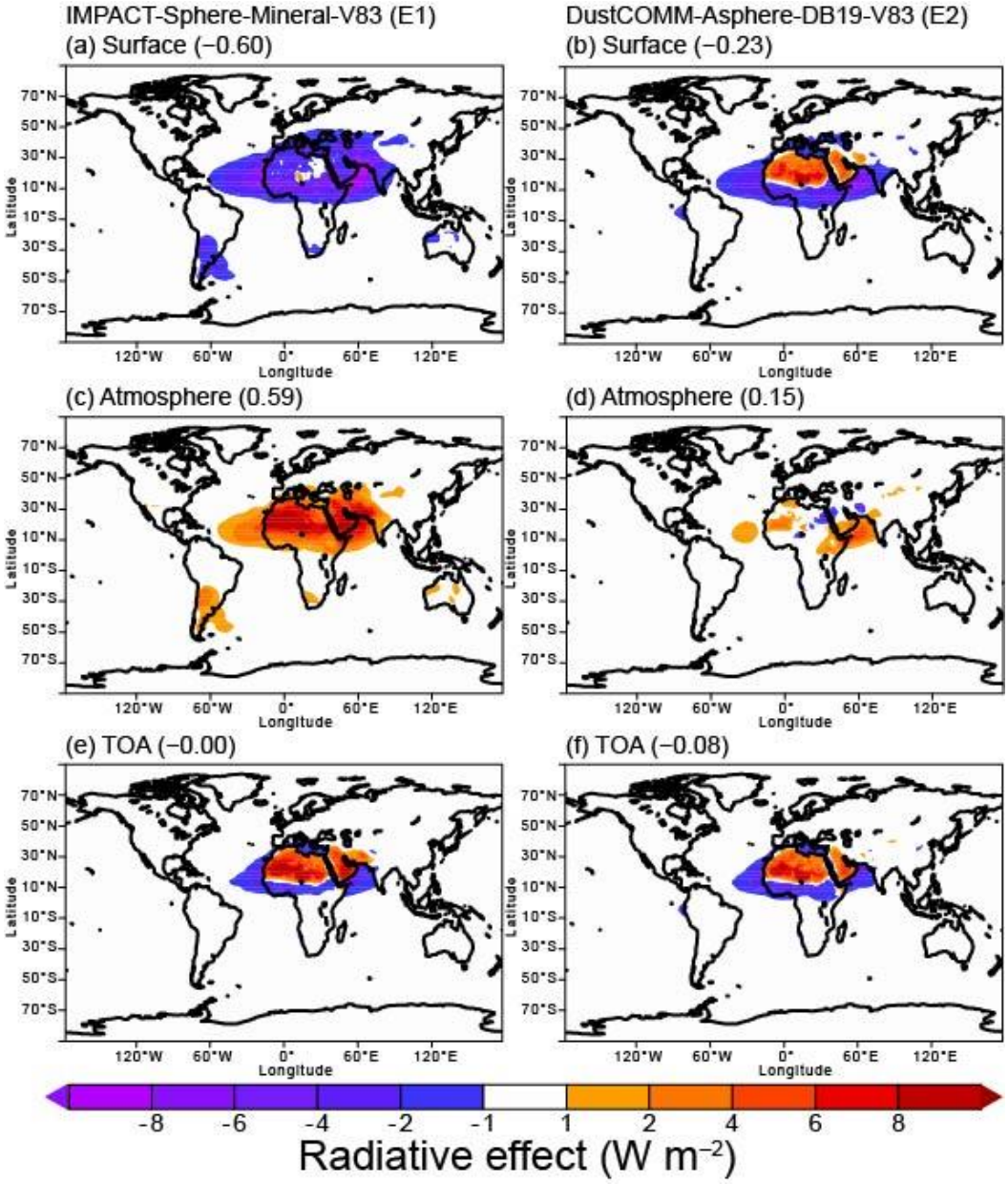


Figure 8. Dust radiative effect (W·m^{-2}) and radiative heating of the atmosphere (i.e., the subtraction of radiative effects from TOA to the surface in unit of W·m^{-2}). The model results were shown for the simulations for (a) IMPACT-Sphere-Mineral-V83 (E1)~~default~~ at the surface, (b) DustCOMM-Asphere-DB19-V83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

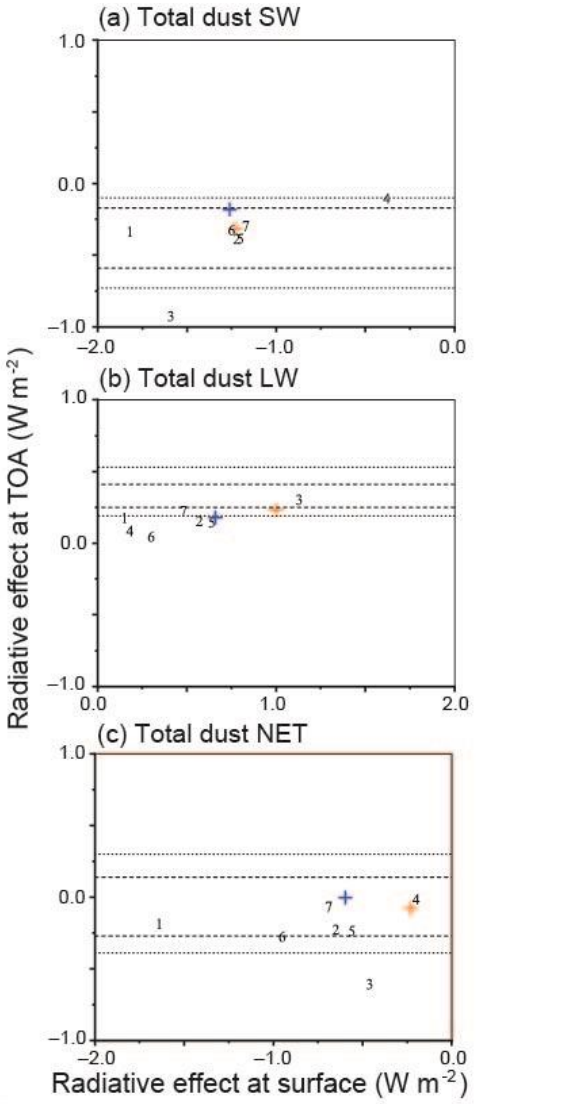


Figure 9. Radiative effect ($W \cdot m^{-2}$) of mineral dust due to different studies at the surface and TOA for (a) total dust SW, (b) total dust LW, and (c) total dust NET. The annually averaged values were listed in Table 5.

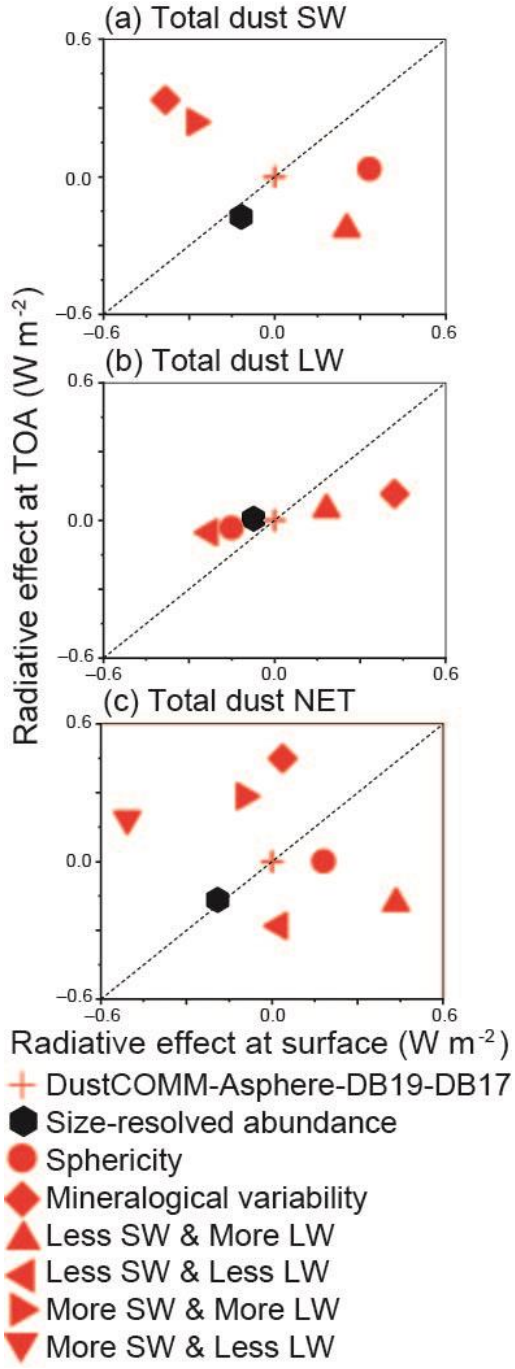


Figure 910. Radiative effect ($W \cdot m^{-2}$) of mineral dust due to various aerosol absorptivity at the surface and TOA for (a) total dust SW, (b) total dust LW, and (c) total dust NET. The annually averaged values were listed in Table 5. The dashed line represented a 1 : 1 correspondence and corresponded to no change in radiative heating within the atmosphere.