Modeling impacts of dust mineralogy on **fast climate response**

Deleted: Earth's Radiation and Climate

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Abstract

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16 Mineralogical composition drives dust impacts on Earth's climate systems. However, most climate 17 models still use homogeneous dust, without accounting for the temporal and spatial variation in 18 mineralogy. To quantify the radiative impact of resolving dust mineralogy on Earth's climate, we 19 implement and simulate the distribution of dust minerals (i.e., illite, kaolinite, smectite, hematite, 20 calcite, feldspar, quartz, and gypsum) from Claquin et al. (1999) (C1999) and activate their interaction with radiation in the GFDL AM4.0 model. Resolving mineralogy reduces dust 21 22 absorption compared to the homogeneous dust used in the standard GFDL AM4.0 model that 23 assumes a globally uniform hematite volume content of 2.7% (HD27). The reduction in dust 24 absorption results in improved agreement with observation-based single scattering albedo (SSA), 25 radiative fluxes from CERES (the Clouds and the Earth's Radiant Energy System), and land 26 surface temperature from CRU (Climatic Research Unit), compared to the baseline HD27 model 27 version. It also results in distinct radiative impacts on Earth's climate over North Africa. Over the 28 19-year (from 2001 to 2019) modeled period during JJA (June-July-August), the reduction in dust 29 absorption in AM4.0 leads to a reduction of over 50% in net downward radiation across the Sahara 30 and approximately 20% over the Sahel at top of atmosphere (TOA) compared to the baseline HD27 31 model version. The reduced dust absorption weakens the atmospheric warming effect of dust 32 aerosols and leads to an alteration in land surface temperature, resulting in a decrease of 0.66 K 33 over the Sahara and an increase of $0.7 \, \text{K}$ over the Sahel. The less warming in the atmosphere 34 suppresses ascent and weakens the monsoon inflow from the Gulf of Guinea. This brings less 35 moisture to the Sahel, which combined with decreased ascent induces a reduction of precipitation. To isolate the effect of reduced absorption, compared to resolving spatial and temporal mineralogy, 36 37 we carry out a simulation where the hematite volume content of homogeneous dust is reduced 38 from 2.7% to 0.9% (HD09). The dust absorption (e.g., single scattering albedo) of HD09 is 39 comparable to that of the mineralogically speciated model on a global mean scale, albeit with a 40 lower spatial variation that arises solely from particle size. Comparison of the two models indicates that the spatial inhomogeneity in dust absorption resulting from resolving mineralogy does not 41 42 have significant impacts on Earth's radiation and climate, provided there is a similar level of dust 43 absorption on a global mean scale before and after resolving dust mineralogy. However, 44 uncertainties related to emission and distribution of minerals may blur the advantages of resolving 45 minerals to study their impact on radiation, cloud properties, ocean biogeochemistry, air quality,

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- and photochemistry. On the other hand, lumping together clay minerals (i.e., illite, kaolinite, and
- 62 smectite), but excluding externally mixed hematite and gypsum, appears to provide both
- 63 computational efficiency and relative accuracy. Nevertheless, for specific research, it may be
- 64 necessary to fully resolve mineralogy to achieve accuracy.

1 Introduction

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Soil dust aerosols emitted from erodible land surfaces, hereafter referred to as dust, are the most 66 67 abundant aerosol component in the atmosphere in terms of dry mass. Dust has significant impacts on 68 the Earth's climate systems (atmosphere, ocean, cryosphere) due to its interaction with terrestrial and 69 solar radiation (Sokolik and Toon, 1999), cloud microphysics (Guo et al., 2021), tropospheric 70 chemistry (Bian and Zender, 2003; Paulot et al., 2016), and oceanic and terrestrial 71 biogeochemistry(Mahowald, 2011; Evans et al., 2019; Dunne et al., 2020). In addition, dust particles 72 deposited on snow and ice decrease surface reflectivity and accelerate snowmelt (Skiles et al., 2018; 73 Réveillet et al., 2022). Dust can influence Earth's radiative energy budget through different pathways; 74 among them: 1) directly by interacting with both solar and terrestrial radiation (i.e., direct radiative 75 effects, hereafter referred to as DRE) 2) by radiatively influencing the thermal dynamical structure of 76 atmosphere and thereby clouds (i.e., semi-direct radiative effect) and 3) indirectly by altering cloud 77 reflectivity (cloud albedo effect) and lifetime (cloud lifetime effect). Unfortunately, the quantitative 78 estimate of dust DRE at the top of atmosphere (TOA) is largely uncertain (Claquin et al., 1998; Miller 79 et al., 2014; Kok et al., 2017; Song et al., 2022). A significant part of this uncertainty has been attributed 80 to neglecting variations in dust mineralogical composition and its evolution during transport (Li et al., 81 2021).

The magnitude of dust impacts on the Earth's climate systems depends on its mineralogical composition, as has been shown in multiple studies. In the ShortWave (SW), dust absorption depends on the iron oxides content. Sokolik and Toon (1999) suggested that a small amount of iron oxides internally mixed with less absorptive minerals is able to reverse the sign of DRE_{SW} at TOA from negative (cooling effect) to positive (warming effect). Multiple studies have confirmed the importance of iron oxides to the dust DREsw (Balkanski et al., 2007; Li et al., 2021; Obiso et al., 2023). In LongWave (LW) spectrum, absorption and DRE_{LW} depend on the abundance of quartz, calcite, and clays in coarse and super-coarse modes (Di Biagio et al., 2017; Sokolik and Toon, 1999). As a result, resolving dust mineralogy allows to better understand the impact of dust DRE, such as the fast response

of the land surface temperature, as opposed to the slow response of sea surface temperature that will

Deleted: As a result, resolving dust mineralogy allows to better understand the impact of dust DRE, such as the fast response (i.e., the response with fixed sea surface temperature) of the Earth's surface radiation (Persad et al., 2014), land surface temperature, precipitation (Ming et al., 2010), and atmospheric circulation. Dust absorption can also affect the slow climate response (i.e., the response including changes in sea surface temperature) of the Earth surface temperature (Miller and Tegen, 1998), precipitation, ocean heat transport (Ocko et al., 2014), which may affect hurricane genesis (Strong et al., 2018), and induce feedback (Miller et al., 2004; Miller et al., 2014).

104 not be studied here. This fast temperature response will affect precipitation, and atmospheric 105 circulation (Ming et al., 2010; Persad et al., 2014). 106 In addition, resolving dust mineralogy is also crucial for studying heterogeneous reactions of acid gases 107 with dust aerosols. For example, the uptake of HNO₃, NO₃⁻, N₂O₅ on dust particles is suggested to be 108 limited by alkalinity that comprises calcium and magnesium carbonates (Song and Carmichael, 2001; 109 Paulot et al., 2016). These reactions will modify the composition of dust particles and subsequently 110 changing their hygroscopicity, cloud condensation nucleation (CCN), and ice nucleation activities 111 (Kelly et al., 2007), and thereby further affecting precipitation (Rosenfeld et al., 2001). Moreover, 112 heterogeneous reactions with mineral dust could significantly affect tropospheric photochemical 113 oxidation cycles, causing up to 10% reduction in O3 concentrations in dust source regions and nearby 114 (Dentener et al., 1996). Among the different minerals, K-Feldspar appears to dominate ice nucleation, 115 despite being a minor component of aeolian dust (Atkinson et al., 2013; Harrison et al., 2019), although 116 other minerals such as quartz may also contribute (Chatziparaschos et al., 2023). The key factor 117 controlling the production and removal of pollutants and the damages by acid rain is the pH of 118 raindrops, which has been observed to increase due to its dependency on Ca-rich dust (Grider et al., 119 2023). 120 Despite the potential importance of resolving dust mineralogy in various aspects, current climate 121 models tend to use a fixed mineralogy without considering the temporal and spatial variations in 122 dust mineralogical composition. To test the importance of resolving dust mineralogy on the fast 123 climate response (e.g., surface temperature response over land, atmospheric circulation and 124 precipitation response) through its interactions with SW and LW radiation (i.e., through dust DRE), 125 dust mineralogy has been implemented and simulated in the GFDL AM4.0 model (Zhao et al., 126 2018a, b), including its on-line interactions with radiation. Following the pioneering work of 127 Claquin et al. (1999) (C1999), we consider the emission, transport and interactions with radiation 128 and deposition of eight minerals: illite, kaolinite, smectite, hematite, calcite, feldspar, quartz and 129 gypsum. Following the recent launch of the Earth Surface Mineral Dust Source Investigation 130 (EMIT) instrument specifically designed to retrieve global distribution of dust mineralogy over 131 dust sources (Green et al., 2020), there have been coordinated efforts to represent dust mineralogy 132 and investigate DRE of mineral-speciated dust in climate models, in particular in Li et al. (2021), 133 Gonçalves Ageitos et al. (2023), and Obiso et al. (2023). However, to the best of our knowledge, 134 there have been no studies investigating the fast climate impact of dust while accounting for its

mineral speciation. Our work contributes to these efforts by incorporating dust mineralogy into the

136 GFDL models, and it is distinguished by extending its investigation to the fast climate response of

137 <u>mineral-speciated</u> dust. The impacts of dust mineralogy on other aspects, such as sea surface

138 temperature and slow climate response, heterogeneous reactions, and ice nucleation ability, will

139 be examined in future studies.

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140 Section 2 provides the description of the GFDL AM4.0 model and dust mineralogy

implementation. Section 3 describes our experimental design. In Section 4, we calculate mineral

142 optical properties, activate the interaction of minerals with radiation in GFDL AM4.0 and compare

modeled dust optical properties with observations. Section 5 presents the impacts of resolving dust

mineralogy on Earth's radiation and climate with a focus on the North Africa, as well as their

evaluations. In Section 6, we investigate the influences of reducing the number of mineral tracers.

Section 7 provides a summary of the study along with the main conclusions.

2 Model and Data

2.1 Model description

We conduct a series of experiments with GFDL AM4.0 (Zhao et al., 2018a, b) over the period

150 2001-2019. These experiments use the AMIP protocol, where sea surface temperature (SST) and

151 sea-ice are imposed based upon average monthly observations (see Gates, 1992 for details).

152 Observed gridded SST and sea-ice concentration boundary conditions are from the reconstructions

153 of Taylor et al., (2000). Historical reconstructions of monthly solar spectral irradiances are from

154 Matthes et al., (2017). For radiation calculations, global monthly mean concentrations of

 $155 \qquad \text{greenhouse gases (GHGs), including nitrous oxide (N$_2$O), and ozone-depleting substances (ODSs,$

156 including CFC-11, CFC-12, CFC-113, and HCFC-22) are specified from Meinshausen et al.,

157 (2017). The solar irradiances and GHG databases are standard for CMIP6. Longwave (LW)

scattering of aerosols is not accounted for in the model.

159 In AM4.0, dust emission is calculated interactively following the parameterization of Ginoux et al.

160 (2001) with a threshold of wind erosion and global scaling factor of 3.5 $m s^{-1}$ and 0.2 $\mu g s^2 m^{-5}$,

respectively. Dust size is represented by five bins with diameter ranging from $0.2 \,\mu m$ to $20 \mu m$

162 (bin1: 0.2 - 2 μm, bin2: 2 - 3.6 μm, bin3: 3.6 - 6 μm, bin4: 6 - 12 μm, bin5: 12 - 20 μm). The

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corresponding source fractions have been updated from 0.1, 0.225, 0.225, 0.225 and 0.225 to 170 171 values of 0.04, 0.14, 0.19, 0.49, and 0.14 for the <u>five bins. These updated source functions allocate</u> 172 more fraction to coarser size bins, following the suggested Brittle Fragmentation Theory (BFT) as 173 proposed by Kok et al. (2011). Dust mineral composition in the standard AM4.0 is considered as 174 uniform, with no temporal and spatial variations; in other words, dust Refractive Index (RI) is 175 temporally and spatially homogeneous (case referred to as homogeneous dust hereafter). The dust 176 RI in the standard AM4.0 is taken from Balkanski et al., (2007), assuming a fixed hematite content 177 of 2.7% by volume (HD27), which was calculated for the internal mixture of hematite and five 178 other minerals (calcite, quartz, illite, kaolinite and montmorillonite) using the Maxwell Garnett 179 mixing rule (see details in Balkanski et al., 2007). The decision to fix the hematite content for dust 180 particles at 2.7% was made during the development of the previous GFDL Climate Model CM3 181 (Donner et al., 2011). This decision was prompted by the discovery that dust absorption was 182 unrealistically high (by a factor 3) in CM2 (Delworth et al., 2006) compared to AERONET 83 observations (Balkanski et al., 2007). In CM3, the conjunction of a sharp decrease of black carbon 84 (strong aerosol absorber) with a new emission inventory and the switch to more scattering dust 185 had a negative effect on precipitation bias, and late 20th century warming (see Donner et al., 2011 186 for details). To mitigate this bias, the selection of 2.7% hematite was adopted in CM3, as well as 187 in the subsequent GFDL models. The control run conducted with the homogeneous dust in the 188 standard AM4.0 model is labeled as HD27 as described in Table 2.

2.2 Dust mineralogy implementation

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Claquin et al. (1999) (C1999) is the earliest study providing a soil mineralogy map oriented toward atmospheric and climate modelling. The soil map provides the mineral mass fractions present in the clay and silt size ranges for eight different minerals, namely: illite, smectite, kaolinite, calcite, quartz, feldspars, gypsum, and hematite. In this study, we implement the eight minerals from the soil mineralogy map provided by C1999 in GFDL AM4.0 to resolve dust mineralogy. The soil map is based on soil analyses that are usually done after wet sieving, which disperse mineral aggregates into small particles. This dispersal is particularly relevant for the phyllosilicates,

In addition, we conduct simulations assuming homogeneous dust with hematite content of 0.9%

by volume, with RI from Balkanski et al. (2007). Similar to HD27, this experiment, labeled as

HD09 in Table 2, does not account for the temporal and spatial variations in dust mineralogy.

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Deleted:) (C1999) is the earliest study providing a soil mineralogy map oriented toward atmospheric and climate modelling. The soil map provides the mineral mass fractions present in the clay and silt size ranges for eight different minerals, namely: illite, smectite, kaolinite, calcite, quartz, feldspars, gypsum, and hematite. In this study, we implement the eight minerals from the soil map provided by C1999, in GFDL AM4.0 to resolve dust (for more details see Ginoux et al. 2023, in preparation). The soil map is based on soil analyses that are usually done after wet sieving, which disperse mineral aggregates into small particles. We apply an algorithm based on Brittle Fragmentation Theory (BFT: Kok, 2011) to reconstruct the mineral aggregates emitted from the original undispersed soils. The mass density of the eight minerals, along with a brief description of their importance to Earth's climate, are listed in Table 1. The density of minerals impacts their settling velocity, which is relevant to the removal of particles in the atmosphere. Iron oxides, represented by hematite in C1999, have larger density than other minerals, so that hematite deposits more quickly and is not able to be transported to remote regions when not aggregated or internally mixed with lighter clay minerals. Moreover, hematite is the strongest absorber in the visible. As such, the correct representation of hematite content in dust aerosols is critical in improving the representation of dust interaction with SW radiation in climate models. Considering that iron oxides are usually found as accretions in other minerals, particularly phyllosilicates (clay minerals): illite, kaolinite and smectite (Zhang et al., 2015; Panta et al., 2023), we partition hematite into two portions: internally mixed hematite and externally mixed hematite, in terms of their mixing state with clay minerals. For accretions, we take the 5% mass fraction of hematite in the mixture at emission as a reference to estimate the amount of internally mixed hematite (following Gonçalves Ageitos et al., 2023). Specifically, hematite is internally mixed with clay minerals for the portion of its mass fraction at emission that is less than 5%, while it is considered externally mixed for the portion that exceeds 5%. typically found in the form of aggregates in soils. They are detected in the atmosphere with higher proportions at coarser (silt) sizes than those reported in the soil maps (Perlwitz et al., 2015b; Perez Garcia-Pando et al., 2016). These recent studies also show that the Brittle Fragmentation Theory (BFT; Kok, 2011) represents a practical framework to generate the emitted particle size distribution based on the dispersed soil PSD, which facilitates the utilization of soil mineralogy maps. In our simulations, we employ BFT to reconstruct the mineral aggregates emitted from the original undispersed soils, following the methods described in Gonçalves Ageitos et al., 2023. The mass density of the eight minerals, along with a brief description of their importance to Earth's climate, are listed in Table 1. The density of minerals impacts their settling velocity, which is relevant to the removal of particles in the atmosphere. Goethite and hematite are the two major types of iron oxides present in soils. Goethite is less absorptive than hematite and is not resolved in C1999. So, iron oxides are represented by hematite in this study. Hematite has larger density than other minerals, so that hematite deposits more quickly and is not able to be transported to remote regions when not aggregated or internally mixed with lighter clay minerals. Moreover, among the minerals considered here, hematite is the strongest absorber at ultraviolet (UV) and visible wavelengths, while it does not have noticeable absorption at infrared wavelengths (IR) compared to other minerals (Sokolik and Toon, 1999). As such, the correct representation of hematite content in dust aerosols is critical in improving the representation of dust interaction with SW radiation in climate models. All minerals are considered to be externally mixed, except for iron oxides. A large part of the emitted flux of iron oxides is considered to be internally mixed with other minerals, e.g., in the form of accretions in phyllosilicates, in line with observational evidence and previous modeling studies (Kandler et al., 2009; Perlwitz et al., 2015a; Zhang et al., 2015; Panta et al., 2023). As suggested by Gonçalves Ageitos et al. (2023), we define two different types of tracers for the iron oxides: one set of tracers carries the mass of the hematite that constitutes small accretions in clay minerals (i.e., internally mixed with clay minerals), are allowed to be up to 5 % of the masses of their host minerals at emission (Perlwitz et al., 2015a; Gonçalves Ageitos et al., 2023). Given the low fractional mass of hematite compared to their host minerals, we assume that these accretions do not change the density of their host particles. These internally mixed accretions form the largest fraction of the emitted hematite. Another smaller set of tracers carries the mass of the remaining fraction of hematite, which is considered to be externally mixed with the other minerals, including the internal mixtures of hematite with clay.

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In addition to the similar roles of clay minerals in carrying iron oxides, the optical properties of the three clay minerals are very similar, and the optical properties of their external mixture are found to be almost identical to their internal mixture (see Section S1 in the Supplement). This finding suggests the use of a single mineral species to represent all three clay minerals in their interaction with radiation to reduce computational cost. Therefore, the optical properties of one single mineral (clay433) are used to represent the optical properties of all three clay minerals. The clay433 represents a mixed mineral comprising three clay minerals: illite, kaolinite and smectite, with mass fraction of 40%, 30%, and 30%, respectively (see detailed descriptions in Supplementary Section S1). This simplification streamlines the calculations of optical properties for internal mixtures of hematite and the three clay minerals (illite, kaolinite, and smectite), reducing it from an internal mixture of four minerals (hematite, illite, kaolinite and smectite) to an internal mixture of two minerals (hematite and clay433).

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The optical properties of the internal mixture of hematite and clay433 are calculated using three mixing rules: volume weighted average (VOL-mixing), Maxwell-Garnett mixing rule (MGmixing) and Bruggeman mixing rule (BM-mixing). Generally, VOL-mixing is used for a quasihomogeneous mixture, that is when the components have similar refractive index. For cases involving dominant homogeneous host with small inclusions of contrasting composition, MGmixing is appropriate. BM-mixing is suitable for mixtures that the inclusions virtually occupy the entire volume of the particle, and the host disappears. The detailed discussion regarding the three mixing rules and their applications can be found in Liu and Daum (2008) and Markel (2016). The appropriate selection of mixing rules is important for the determination of the optical properties of the mixtures. Therefore, we incorporate all three mixing rules in this study. These calculations are performed for various volume mixing fractions of hematite with respect to clay433, to construct a lookup table (LUT) for each mixing rule. The optical properties of each mineral as well as the internal mixtures of hematite and clay433 are calculated offline using Mie code with a spherical shape assumption. As all other minerals have similar SW absorption, internal or external mixing does not change their absorption properties. So, we assume all other minerals to be externally mixed. More details about optical properties of minerals will be discussed in Section 4.

Overall, we implement nine types of mineral tracers: seven non-hematite minerals along with distinguished internal and external hematite, as listed in Table 1. Each type is distributed across

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five size bins. As a result, 45 mineral tracers have been incorporated in AM4.0 to account for dust

326 mineralogy.

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Table 1. The list of minerals considered in this study and their importance to Earth's climate. Mineral-dependent mass densities are defined following Table 1 in Gonçalves Ageitos et al. (2023), in which the references of mineral densities are listed.

Minerals	Density	Importance		
	(kg/m^3)			
1. Hematite (int.)	2570	It is the strongest visible absorber. It is internally mixed with clay minerals when its mass fraction at emission < 5%.		
2. Hematite (ext.)	4770 ⁽¹⁾	It is externally mixed for the part of emitted mass fraction > 5%.		
Three clay minerals:		They are the most abundant mineral components in clay-sized		
3. Illite	2570	(diameter $< 2\mu m$) minerals.		
Smectite	2570	They are internally mixed with internal hematite.		
Kaolinite	2630			
Clay in BM-RT	2590	The three clay minerals are lumped together as one mineral species 'clay' in the BM-RT experiment in Section 6.		
6. Calcite	2710	It is important for chemistry. (e.g., heterogeneous reaction with acidic gases and formation of sulfate and nitrates on the surface of dust particles, and cloud droplet pH)		
7. Feldspar	2680	A fraction of feldspar (K-feldspar) is important for ice nucleation		
8. Quartz	2670	It is the most abundant mineral component in silt-sized (diameter:2-63 \(\mu m\)) minerals. It is important for LW absorption and ice nucleation.		
9. Gypsum	2308	It possibly has impact on chemistry, but the impact is likely unimportant given the low abundance globally.		

(1) We use the mean of hematite and goethite densities for hematite, as in Gonçalves Ageitos et al. (2023).

2.3 AERONET Dust SSA

The AERONET Version 3 Level 2.0 Almucantar <u>inversion</u> retrievals (Giles et al., 2019; Sinyuk et al., 2020) from 2000 to 2020 are screened for dust events following the methodology in Gonçalves Ageitos et al. (2023) and Obiso et al. (2023). This screening process aims to select dust-dominated events and filter out the AERONET scenes contaminated by other absorbing aerosols. The criteria that are applied to AERONET retrievals to screen dust events are: 1) hourly retrievals from AERONET are considered to represent dust when the fine volume fraction is small (below 15%), 2) the SSA increases from 440 nm to 675 nm (a feature that distinguishes dust from other species, see Dubovik et al., 2002), and 3) the mean of the imaginary index at red and near-infrared

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- 339 wavelengths (675, 870 and 1020 nm) is lower than 0.0042 (as higher values would indicate the
- 340 presence of absorbing black and brown carbon, following Schuster et al., 2016). We calculate
- 341 AERONET SSA in the visible by averaging AERONET retrieved SSA at two visible wavelengths
- 342 (0.44 μm and 0.67 μm) weighted by solar spectrum, following Eq. (2).

2.4 Laboratory Dust SSA

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- The lab measured dust SSA at 550 nm is obtained from Di Biagio et al. (2019) (DB-2019 hereafter),
 - in which dust SSA was directly retrieved from scattering and absorption measurements. We
- 346 acknowledge the limits of laboratory measurements, where the dust samples are not aerosols
- 347 present in the atmosphere, but instead are reemitted in the lab from soil samples collected from
- 348 various source regions. Consequently, the laboratory measurements in DB-2019 do not account
- for dust aerosols transported from other regions to the regions of interest. In addition, in contrast
- 350 to the modeled dust diameter range of 0.2 μm to 20 μm , DB-2019 measures dust particles with a
- 351 diameter ranging from 0.2 μm up to $10\mu m$.

352 2.5 CERES Data

- 353 To compare modeled fluxes at TOA with observations, we use the Clouds and the Earth's Radiant
- Energy System (CERES) Energy Balanced and Filled (EBAF) Edition-4.2 data (Loeb et al., 2018).
- 355 The standard CERES level-3 products provide clear-sky fluxes by averaging all CERES footprints
- 356 within a region that are completely free of clouds. Therefore, there are many missing regions in
- 357 monthly mean clear-sky TOA flux maps because completely cloud-free conditions are not always
- 358 observed at the CERES footprint scale (~20 km at nadir). In contrast to the standard CERES level-
- 359 3 products, CERES_EBAF product infers clear-sky fluxes from clear portions of partly cloudy
- 360 CERES footprints thereby producing a clear-sky TOA flux climatology free of any missing regions
- 361 (details in Loeb et al., 2018). Starting from CERES_EBAF_Ed4.1, the product also provides clear-
- 362 sky flux estimates for the total region (i.e., the total CERES footprints) by combining CERES
- 363 observations and radiative transfer calculations, which represents clear-sky flux with clouds
- 364 removed from the entire atmospheric column of CERES footprints. These clear-sky fluxes for the
- 365 total region are defined in a way that is more consistent with how clear-sky fluxes are represented
- 366 in climate models (for details see CERES_EBAF_Ed4.1 Data Quality Summary). In this study,
- 367 the monthly mean TOA 'Clear-Sky Flux Estimate for Total Region' variables in the
- 368 CERES_EBAF_Ed4.2 product, the most recent version of the product, are used to compare with

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- modeled monthly mean clear-sky flux at TOA. The comparisons allow us to examine the agreement of modeled clear-sky fluxes from different experiments with observations. The
- 372 comparison results will be shown in section 5.1.

2.6 CRU TS Data

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- 374 The CRU TS (Climatic Research Unit gridded Time Series) dataset provides high-resolution (0.5°
- 375 latitude \times 0.5° longitude) climate dataset over land except Antarctica. The dataset is based on
- 376 extensive networks of weather stations going back to 1901(Harris et al., 2020). This dataset has
- 377 been widely used in various research areas since its first release in 2000. The mean 2-meter
- 378 temperature (TMP) and precipitation rate (PRE) variables from CRU TS v4.07 are used to evaluate
- our model simulations. The results will be shown in section 5.2 and section 5.3.

3 Experimental Design

Table 2. List of experiments and their description. Experiments are named based on the type of dust used or the mixing rules for minerals applied in each experiment.

Experiments	Dust or Minerals	Description	Optics	
HD27	HD27	Dust refractive index is spatially and temporally uniform. Dust is assumed to contain 2.7% of hematite by volume. Its optical properties are used to represent dust in the standard GFDL AM4.0 model.	% of used (2007)	
HD09	HD09	Dust refractive index is spatially and temporally uniform. Dust is assumed to contain 0.9% of hematite by volume.	Balkanski et al. (2007)	
VOL	VOL-mixing	Soil mineralogy from C1999 is implemented in AM4.0. Hematite (int.) is internally mixed with clay minerals following the volume-weighted mean mixing rule.	Scanza et al. (2015)	
MG	MG-mixing	Soil mineralogy from C1999 is implemented in AM4.0. Hematite (int.) is internally mixed with clay minerals following the Maxwell Garnett mixing rule.	Scanza et al. (2015)	
BM	BM-mixing	Soil mineralogy from C1999 is implemented in AM4.0. Hematite (int.) is internally mixed with clay minerals following the Bruggeman mixing rule.	Scanza et al. (2015)	
BM-RT	BM-mixing	Three experiments are performed step by step to reduce the number of mineral tracers. 1) BM-LC experiment: following BM experiment, illite, kaolinite and smectite are lumped together as one tracer 'clay'.	Scanza et al. (2015)	

BM-LCRH experiment: following BM-LC, the tracer of external mixed hematite is removed, its mass is combined with internal mixed hematite. BM-LCRHRG experiment: following BM-LCRH, gypsum tracer is removed, and its mass is proportionally added to all other remaining minerals.

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We conduct a total of six experiments using the GFDL AM4.0 model, with each experiment's description provided in Table 2. Two of these experiments serve as control runs in which dust aerosols are represented with temporally and spatially fixed composition in the model. The first control run, referred to as HD27, represents how dust aerosols are implemented in the standard GFDL AM4.0 model (Zhao et al., 2018a). The second control run is the HD09, in which dust is more scattering than that in the standard AM4.0 model (i.e., HD27) due to its reduced hematite volume fraction from 2.7% to 0.9%.

389 390 The other three experiments, namely VOL, MG, and BM, resolve dust mineralogy and activate 391 their interaction with radiation. These three experiments incorporate 45 mineral tracers for nine

types of mineral tracers distributed over five size bins as described in Section 2.2. Additionally, we conduct the BM-RT experiments, which consist of three sub-experiments: BM-LC, BM-LCRH,

BM-LCRHRG. These experiments aim to explore the potential of reducing mineral tracers, which

can improve the model computational efficiency. The results will be discussed in Section 6.

Each of the experiments ran for 19 years, from 2001 to 2019. We consider the 19-year runs of the experiment as a group of simulations, containing 19 members of one-year simulation. The two control runs (i.e., HD27 and HD09), combined with the three mineral-resolved experiments (i.e., VOL, MG, and BM), form a total of six contrasting pairs. In this study, for each contrasting pair, we define the anomaly as the group mean difference (based on 19-year mean) between mineral-

resolved experiment and control run. An anomaly is considered statistically significant if the p-

value, determined by the student's t-test between the two contrasting groups of simulations, is

smaller than 0.05.

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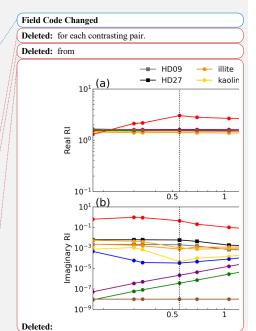
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4 Optical Properties

4.1 Dust optical properties



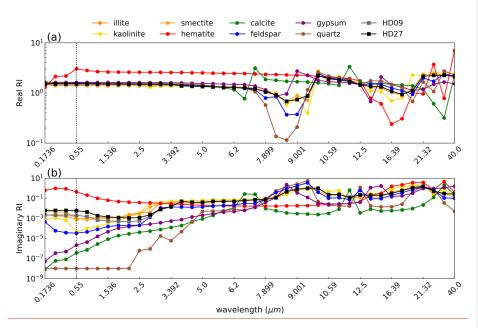


Figure 1. Real part and imaginary part of complex refractive indices (RI) of two homogeneous dust (e.g., HD27 and HD09) and eight minerals (Scanza et al., 2015). The dotted lines represent the real part (a) and imaginary part (b) of dust or minerals for the corresponding wavelength. The black and grey dotted lines are for HD27 and HD09, respectively. The colorful dotted lines are for the eight minerals.

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we use the refractive indices (RI) of each mineral from Scanza et al. (2015) and the RI of HD27 and HD09 from Balkanski et al. (2007) to calculate the LUT of optical properties. The spectral RI of each mineral and homogeneous dust (e.g., HD27 and HD09) are shown in Figure 1. The HD09 dust has lower imaginary part of RI at 550 nm than HD27 dust, indicating its lower absorption in the visible band due to a reduced content of hematite.

After calculating LUT of optical properties (details in Section 2.2), we incorporate the interaction of minerals with radiation into GFDL AM4.0. The modeled emission, load, deposition, and lifetime for each mineral are provided in Table S1 in the Supplement. Table 3 provides global total dust emission, load, globally averaged dust aerosol optical depth (DAOD) and SSA for each experiment listed in Table 2 and their comparisons with previous studies. DAOD and SSA from AM4.0 simulations are averaged in the visible band (0.44 - 0.625µm) of GFDL AM4.0. Unless otherwise specified, DAOD and SSA in this study refer to the average in the visible band. Note

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Deleted: Figure 1 shows the refractive indices (RI) of each mineral (Scanza et al., 2015) as well as the RI of two homogeneous dust mixtures HD27 and HD09 (Balkanski et al., 2007). The HD09 dust has lower imaginary part of RI at 550 nm than HD27 dust, indicating its lower absorption in the visible band due to a reduced content of hematite. ¶ With the optical properties of each mineral component calculated...

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Deleted: . DAOD and SSA from AM4.0 simulations are averaged in the visible band (0.44-0.625µm) of GFDL AM4.0. Note that in our calculations, the domain averaged DAOD is always weighted by the area of each grid cell. The domain averaged SSA is always weighted by the area and DAOD of each grid cell. Additionally, the spectral-averaged DAOD and SSA are always weighted by solar spectrum, which peaks at around 0.50 µm. The emission, load, deposition, and lifetime for each mineral are provided in Table S1 in the Supplement. The subtle difference in DAOD and large variation in dust SSA across experiments suggests that resolving dust mineralogy or changing hematite content in homogeneous dust does not affect visible extinction of dust aerosols (i.e., DAOD). However, it significantly affects the relative strength of dust absorption versus scattering. The variation in total dust emission and DAOD across different experiments results from the feedback of dust interactions with radiation (Miller et al., 2004

that in our calculations, the domain averaged DAOD is always weighted by the area of each grid cell. The domain averaged SSA is always weighted by the area and DAOD of each grid cell. Additionally, the spectrally averaged DAOD and SSA are always weighted by the TOA solar radiation intensity at the corresponding wavelengths, peaking around $0.50 \, \mu m$, as shown in Eq. (1) and Eq. (2).

$$DAOD = \frac{\int_{\lambda_1}^{\lambda_2} DAOD(\lambda) B(\lambda) d\lambda}{\int_{\lambda_1}^{\lambda_2} B(\lambda) d\lambda}$$
 Eq. (1)

$$SSA = \frac{\int_{\lambda_1}^{\lambda_2} SSA(\lambda) DAOD(\lambda) B(\lambda) d\lambda}{\int_{\lambda_1}^{\lambda_2} B(\lambda) DAOD(\lambda) d\lambda}$$
 Eq. (2)

Where $B(\lambda)$ describes the solar radiation energy intensity, which can be calculated by means of the Planck's function B(T, λ), using the temperature of the Sun (T = 5800 K).

Table 3. 19-year (2001-2019) averaged global dust emission, load, globally averaged visible band dust optical depth (*pAoD*) and single scattering albedo (*sSA*) for each experiment in this study. We use each grid-cell surface area as a weight for the global DAOD average. We use each grid-cell surface area times DAOD in each grid-cell as a weight for the global SSA average. In addition, we include the results from previous studies for the purpose of comparison. Note, the modeled DAOD and SSA in this study are averaged in the visible band (0.44 ε 0.625μm) of GFDL AM4.0, while averaged in the UV-VIS band (0.30 ε 0.77μm) of GISS ModelE2.1 in Obiso et al. (2023).

Experiments		Emission (Tg/	Load (Tg)	DAOD	ŞSA
		yr)			
	HD27	3354	23.6	0.022	0.86
	HD09	3119	21.5	0.020	0.93
	VOL	3154	21.6	0.022	0.91
	MG	3083	21.1	0.021	0.93
	BM	3087	21.1	0.021	0.93
BM-RT	BM-LC	3097	21.1	0.021	0.930
	BM-LCRH	3069	20.9	0.021	0.930
	BM-LCRHRG	3110	21.4	0.021	0.928
AeroCom (1	1)	(1000 2200)	20	0,029	<u>-</u>
<u>CMIP5⁽²⁾</u>		(1000-3200) 2700 (1700-3700)	(9-26) 17 (14-36)	(0 <u>.021</u> - 0 <u>.035</u>) =	=

Moved down [1]:; Pérez et al., 2006; Miller et al., 2014), which is influenced by the distinct scattering properties of dust aerosols in each experiment

Moved down [2]: in VOL suggests that VOL-mixing dust is more absorptive than MG-mixing and BM-mixing dust. This finding is consistent with previous studies that have suggested that VOL-mixing method, when applied to minerals to compute the bulk aerosol optical properties, may artificially enhance absorption relative to scattering and lead to a lower SSA for bulk dust aerosol (Zhang et al., 2015).

Deleted: The HD27 dust used in the standard AM4.0 model is the most absorptive among all experiments. The HD09 dust is much less absorptive due to its smaller hematite content, thereby resulting in a lower imaginary part of RI in the visible range (Figure 1). We can see that the SSA of HD09 dust is very similar to the values obtained in cases where minerals are resolved (i.e., MG and BM). For the three mineral-resolved experiments, the lower global mean SSA...

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CMIP6 ⁽³⁾		<u>3472</u>	<u>25</u>	0.029	Ξ
DUSTCOMM ⁽⁴⁾		<u>4700</u> (3300 - 9000)	<u>26</u> (22 - 31)	<u>0.028</u> (0.024 - 0.030)	=
GISS ModelE2.1 (5) HOM		4031	31.3	0.020	0.917
	EXT	4152	32.4	0.020	0.936
	INT	4284	33.7	0.021	0.942

(1) Results are from AeroCom Phase I, which were taken from Table 3 in Huneeus et al. (2011), and the 1 standard error range was obtained by eliminating the two highest and lowest values.

(2) Results are from Table 3 in Wu et al. (2020)

(3) Results from Zhao et al. (2022)

(4) Results are from Table 3 in Kok et al. (2021)

(5) Results from Obiso et al. (2023).

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AM4.0 model, is the most absorptive among all experiments. The HD09 dust is much less absorptive, attributed to its smaller hematite content, as indicated by the lower imaginary part of RI in the visible range (Figure 1). For the three mineral-resolved experiments, the lower global mean SSA (SSA) in VOL suggests that VOL-mixing dust is more absorptive than MG-mixing and BM-mixing dust. This finding is consistent with previous studies that have suggested that VOL-mixing method, when applied to minerals to compute the bulk aerosol optical properties, may artificially enhance absorption relative to scattering and lead to a lower SSA for bulk dust aerosol (Zhang et al., 2015). We can see that the global mean SSA (SSA) of HD09 dust is comparable to the values obtained in cases where minerals are resolved (e.g., MG and BM). This implies that, from a global perspective, HD09 dust is as absorptive as mineral-resolved dust (e.g.,

The lowest SSA of HD27 in Table 3 suggests that HD27 dust, which has been used in the standard

MG and BM). 519 520

In addition to the globally averaged dust properties listed in Table 3, we illustrate the distribution

of global dust mass across 5 size bins (Figure S3 in the Supplement) and the global distribution of DAOD (Figure 2) for the three experiments: before (e.g., HM27 and HD09) and after (e.g., BM)

resolving mineralogy. The global dust mass distribution across the 5 size bins remains largely

523 unchanged across experiments. Besides the subtle difference (~10%) in global mean DAOD across Deleted: 2

Deleted: (1) Results based on 15 global aerosol models within the AeroCom project (Huneeus et al., 2011).

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the three experiments as listed in Table 3, the global distribution of DAOD responds differently in HD09 and BM. Compared to HD27, reducing hematite content to HD09 generally decreases DAOD, except over the Sahel region. In contrast, resolving mineralogy as in BM decreases DAOD over the Sahara region while increasing DAOD over the Sahel and Asia regions. The reduction in DAOD over the Sahara region further contributes to the decrease in dust absorption over the region, primarily attributed to the change in dust optical properties, such as the enhancement in dust SSA. The indistinct variation in DAOD across different experiments results from the feedback of dust interactions with radiation (Miller et al., 2004; Pérez et al., 2006; Miller et al., 2014), which is

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Table 3.

influenced by the distinct scattering properties of dust aerosols in each experiment as shown in

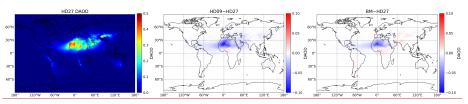


Figure 2. The global distribution of dust optical depth (DAOD) for HD27, and the difference in DAOD between HD09 and BM compared to HD27. The global mean DAOD values (DAOD) of each experiment are shown in Table 3.

4.2 Comparison of dust optical properties with observations

Iron oxides content of dust determines shortwave radiation absorption by dust: the higher amount of iron oxides, the lower the SSA. Following our calculations of dust optical properties in Section 4.1, we compared GFDL AM4.0 modeled dust SSA (averaged in the visible band 0.44-0.625 µm) against AERONET SSA retrievals (averaged at two visible wavelength: 0.44 μm and 0.67 μm) in Section 4.2.1 and laboratory measurements of SSA (at 0.55 µm) in Section 4.2.2. The modeled dust SSA is evaluated against observation-based results utilizing the following evaluation metrics: the mean SSA (mSSA) is calculated based on SSA for all locations displayed in Figure 3, the standard deviation (σ), derived from SSA for all locations displayed in Figure 3, is used as an indicator of dust SSA spatial variation; the normalized mean bias (nMB) and normalized root mean square error (nRMSE) are utilized to assess the mean bias and root mean square error, respectively, of modeled SSA in comparison to observed SSA. Definitions of nMB and nRMSE are provided in Section \$3 in the Supplement.

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4.2.1 Comparison with AERONET retrievals

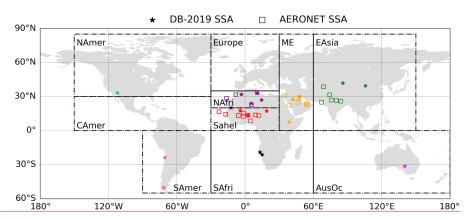
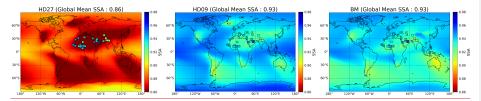
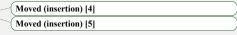


Figure 3, Dust sample locations in Di Biagio et al. (2019) and AERONET stations selected by filtering dust events. AERONET stations for SSA retrievals are in North Africa (NAfri), Sahel, the Middle East (ME) and East Asia (EAsia). Lab measurements by Di Biagio et al. (2017, 2019) expand dust sampling to include soils from North America (NAmer), South America (SAmer), South Africa (SAfri), Australia (AusOc).

Figure 3 displays the AERONET stations selected by filtering dust events. The global distribution of modeled dust SSA and AERONET retrieved SSA over the selected AERONET sites are shown in Figure 4. There is a significant decrease in dust absorption from HD27 to HD09 globally due to the reduction in hematite content. HD09 and BM exhibit similar dust absorption on a global scale (e.g., the same global mean SSA), but the regional differences are evident. For instance, compared to HD09, resolving mineralogy (e.g., BM) decreases dust absorption over Iceland and Taklamakan regions, while enhances dust absorption over Southern Hemisphere, particularly over Australia. Additionally, there is a shift in dust absorption from the Sahara to the Sahel region after resolving mineralogy.



<u>Figure 4.</u> The 19-year (2001-2019) annual mean dust SSA simulated by AM4.0 across the three experiments. The squares represent 21-year (2000-2020) annual mean AERONET retrieved dust SSA over the selected AERONET sites.



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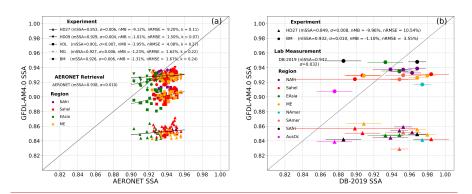


Figure 5, GFDL AM4.0 modeled 19-year (2001-2019) averaged monthly dust SSA (average in 0.44-0.625 μm) versus (a) AERONET 21-year (2000-2020) averaged monthly SSA retrievals (average at two visible wavelengths: $0.44 \mu m$ and $0.67 \mu m$) and (b) laboratory SSA measurements (at $0.55 \mu m$) of dust particles with diameter ranging from $0.2 \mu m$ μp to $10 \mu m$ obtained by Di Biagio et al. (2019) (DB-2019). The lab measurements were carried out in March 2015, horizontal error bars represent measurement uncertainties. Markers represent different experiments, and colors represent different regions. mSSA in the legend represents the mean SSA averaged over all locations indicated in Figure 2 (squares for AERONET, stars for DB-2019). The standard deviation (σ), normalized mean bias (nMB), normalized root mean square error (nRMSE), and the slope of linear regression (k) are also indicated in the legend.

Figure 5a shows GFDL AM4.0 modeled 19-year (2001-2019) averaged dust SSA (average in 0.44-0.625 μm) versus AERONET SSA (average at 0.44 μm and 0.67 μm) retrievals. Compared to AERONET SSA retrievals, both HD27 and VOL overestimate dust absorption, as indicated by their relatively low SSA (Figure 5a). HD27 dust is the most absorptive, indicating that the standard AM4.0 dust is overly absorptive. Dust SSA in MG and BM are quite similar (i.e., mSSA: 0.926 versus 0.927) and show a better agreement with AERONET measurements ($nMB \approx -1.3\%$ and $nRMSE \approx 1.6\%$), and they exhibit stronger scattering (i.e., higher SSA) than HD27 and VOL. HD09 is almost as scattering as MG and BM, as indicated by the mSSA of 0.929 versus 0.926 and 0.927 in Figure 5a, which is consistent with the global mean SSA (SSA) results shown in Table 3. Overall, both the fixed mineralogy dust HD09 and mineral-resolved MG and BM dust agree well with AERONET SSA retrievals, while HD27 and VOL are too absorptive. Therefore, we recommend using mixing ratios of MG or BM in calculating optical properties of internal mixture of hematite and clay minerals. Unless otherwise specified, in the following part of the paper, we will refer to mineral-resolved experiments as MG or BM.

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611 We further assess the SSA spatial variation (indicated by σ) for each experiment from AM4.0 by 612 comparing it to observation-based results. SSA is generally determined by dust mineralogy, size Deleted: various 613 as well as shape. Various dust mineralogy leads to distinct dust SSA due to the different absorption 614 properties of minerals (Figure 1). Coarser dust generally tends to be more absorptive (i.e., has 615 lower SSA) than finer dust when other factors are the same (Ryder et al., 2018). Spherical dust 616 assumption tends to underestimate dust SSA (Huang et al., 2023). Given the uncertainty in dust 617 shape, we assume dust particles to be spherical in this study, aligning with other model studies Deleted: (e.g. Gliß et al., 2021) 618 (e.g. Gliß et al., 2021). Consequently, in the mineral-resolved experiments of this study, namely 619 VOL, MG, and BM, dust mineralogy and dust size are the two factors affecting the SSA spatial 620 variation. 621 Conversely, in homogeneous dust experiments, specifically HD27 and HD09, SSA variation is 622 solely attributed to variation in dust size, as dust mineralogy remains uniform globally. 623 Interestingly, HD09 demonstrates smaller spatial variation (i.e., lower σ) in SSA compared to 624 HD27 (Figure 5a). To investigate the impact of dust size on SSA for different hematite content Deleted: Figure 3a). 625 (e.g., HD27 and HD09), we perform a simple test in Section \$\frac{\$54}{2}\$ of the Supplement. Supplementary Deleted: S3 626 Figure \$4 and \$5 illustrate that the variation of SSA due to the dust particle size is more Deleted: S3 illustrates 627 pronounced with increasing absorption, i.e., from HD09 to HD27. This suggests that enhancement 628 in dust scattering relative to dust absorption (i.e., an increase in SSA) mitigates the sensitivity of 629 SSA to dust size. 630 The conclusions above provide an understanding of the SSA spatial variation (indicated by σ) 631 before (i.e., HD27 and HD09) and after (i.e., VOL, MG, and BM) implementing dust mineralogy. 632 The same σ (0.06) between HD27 and BM can be explained as follows: Because mineral-resolved 633 BM-mixing dust is overall more scattering than HD27 dust, resulting in a reduced sensitivity of 634 SSA to size, therefore, the σ of SSA caused by dust size is reduced in BM relative to HD27. 635 However, the incorporation of dust mineralogy in BM leads to an increase of σ . These contrasting 636 effects compensate for each other, resulting in the same σ . In contrast, HD09 is overall as 637 scattering as BM, as shown in Table 3 and Figure 5a, suggesting a similar sensitivity of SSA to Deleted: Table 3 Deleted: Figure 3 638 size. Therefore, the incorporation of dust mineralogy in BM results in a higher σ compared to Deleted: in BM relative 639 HD09, Overall, while the enhancement in σ can be offset by the reduction in σ due to the reduced Deleted: is attributed to

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650 sensitivity to dust size, resolving dust mineralogy increases σ on its own, consequently enhancing 651 the spatial variation in dust SSA. 652 Worth to mention, AERONET dust is quite scattering as shown in Figure 5a, therefore its SSA is 653 less sensitive to dust size. The high σ (0.010) of AERONET SSA can be mainly due to spatial 654 variations in dust mineral composition. Reducing dust hematite content (HD09) leads to a better agreement in mean SSA with AERONET (i.e., more scattering dust) but results in very low σ 655 656 (0.004), while implementing dust mineralogy (e.g., MG and BM) retains the agreement with 657 AERONET in mean SSA and, at the same time, increases σ (0.006). 658 Besides the standard deviation (σ) , the slopes (k) obtained from the statistics of modeled SSA 659 versus AERONET SSA can also indicate the regional contrast of SSA. The regional contrast of 660 AERONET SSA is well captured by the model when k is one, underestimated when k is lower than 661 one, and overestimated when k is higher than one. As such, the slopes in Figure 5a show that the contrast in SSA from different regions (e.g., North Africa vs. East Asia) observed by AERONET 662 663 is better captured by mineral-resolved experiments (e.g., VOL, MG and BM with k ranging from 664 0.22 to 0.27) than homogeneous dust experiments (e.g., HD27 and HD09 with k ranging from 0.07 665 to 0.11). However, the modeled regional contrast of SSA in mineral-resolved experiments remains 666 overly underestimated (i.e., k is much lower than one). The significant underestimation of regional SSA contrast (k) and spatial variation (σ) in AM4.0 even after accounting for mineralogy, implies 667 668 that something important is still missing in models. For instance: 1) the observed regional contrast 669 in iron oxides content may be higher than that in the soil map used in this study, and 2) the model 670 may have underestimated regional contrasts in the dust aerosol size distribution and thus their 671 contribution to SSA, and 3) spatial variation of dust shape, which is not taken into account in the 672 model. 673 4.2.2 Comparison with laboratory measurements

We further compare the GFDL AM4.0 modeled dust SSA (average in 0.44-0.625 µm) with DB-

2019 laboratory measurements of SSA at 0.55μm (Figure 5b). Figure 3 shows the locations where

dust samples were collected for the lab measurements. Considering that MG and BM are very

similar in terms of dust absorption and agree the best with AERONET SSA, we select BM as a

representative to compare with DB-2019 SSA. Moreover, to evaluate how dust absorption is

represented in the standard AM4.0 relative to lab measurements, we also show the comparison of

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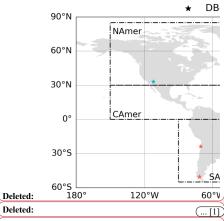
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Moved up [5]: Dust sample locations in Di Biagio et al. (2019) and AERONET stations selected by filtering dust events. AERONET stations for SSA retrievals are in North Africa (NAfri), Sahel, the Middle East (ME) and East Asia (EAsia). Lab measurements by Di Biagio et al. (2017, 2019) expand dust sampling to include soils from North America (NAmer), South America (SAmer), South Africa (SAfri), Australia (AusOc).

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734 SSA between HD27 used in the standard AM4.0 and DB-2019 (Figure 5b). Consistent with the

comparison with AERONET, the comparison with lab measurements suggests that dust

736 representation in the standard AM4.0 (i.e., HD27) is excessively absorptive. The smaller nMB and

737 nRMSE values in BM suggests that SSA of BM agrees better with lab measurements.

738 Moreover, regarding spatial variation (σ), resolving dust mineralogy in BM increases σ from 0.008

739 for HD27 to 0.010 for BM, even though it is still lower than the σ (0.032) in DB-2019 lab

measurements. Note that the variation for HD27 results from the high sensitivity of SSA to dust

741 size due to its higher absorption (as discussed in Supplementary Section \$4). The inability to

742 reproduce spatial variation observed in the lab measurements is likely attributed to two aspects.

743 The first limitation is the fact that samples of DB-2019 are from soils rather than aeolian dust.

744 Aeolian dust is expected to exhibit greater uniformity in mineralogy than soils because of the

atmospheric mixing of dust emitted from various soil sources. The second one is associated with

746 the under-representation of regional contrast in iron oxides content in our model. Observations

747 from the EMIT are therefore essential to constrain soil mineralogy in climate models.

5 Impacts of dust mineralogy on climate

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749 Resolving dust mineralogy in climate models affects dust optical properties (as discussed in

750 Section 4) and their spatial and temporal variability, thereby affecting their interactions with

shortwave (SW) and longwave (LW) radiation. The variability in dust radiative interactions further

752 induces the fast response of land surface temperature, circulation, and precipitation. To investigate

753 the impacts of resolving dust mineralogy on climate, we need to compare modeled results in

mineral-resolved experiments to the baseline homogeneous dust (i.e., non-resolved mineralogy)

control run. As a result, the significance of the impacts depends on the selection of the baseline

homogeneous dust experiment. In this section, we investigate the impacts of resolving dust

mineralogy compared to two baseline homogeneous dust experiments. One baseline experiment is

758 the homogeneous dust used in the standard GFDL AM4.0 in which dust mineralogy is assumed

to be temporally and spatially uniform, (i.e., non-resolved), with a volume fraction of 2.7%

hematite (HD27). The impacts of resolving dust mineralogy compared to the baseline HD27, can

be attributed to two factors: 1) the reduction in dust absorption after resolving dust mineralogy in

comparison to HD27 as discussed in Section 4, and 2) spatial and temporal variations in dust

scattering properties induced by inhomogeneity in dust mineralogy. The other baseline experiment

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Deleted: . To understand the impacts of dust minerals on climate, we proceed in two steps. Firstly

Deleted: on radiation and climate relative to the HD27 control run. The HD27 control run represents

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Deleted: the standard AM4.0 model. Secondly, we go beyond the representation of dust aerosols

Deleted: the standard AM4.0 model by replacing our homogeneous dust in

Deleted: control run with more scattering homogeneous dust (i.e., HD09) to assess the impacts of resolving

Deleted: of dust mineralogy on climate relative to the

782 is the homogeneous dust, with a volume fraction of 0.9% hematite (HD09). Given the comparable

783 global mean dust scattering properties (e.g., SSA) between the control run HD09 and mineral-

784 resolved experiments (e.g., MG and BM), the impacts of resolving dust mineralogy, compared to

785 baseline HD09, is solely attributed to the inhomogeneity in dust scattering properties induced by

786 resolving dust mineralogy.

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5.1 Impacts on Clear-sky Radiative Fluxes

788 We start our analysis by examining the impacts of resolving dust mineralogy on clear-sky radiative

789 fluxes. By 'clear-sky', we mean that our results do not consider the radiative effects of clouds. We

use anomalies (ΔF) to evaluate the impacts of resolving mineralogy on clear-sky radiative fluxes,

which is defined as, $\Delta F = F_1^{\uparrow} - F_2^{\uparrow}$, where F_1^{\uparrow} is the 19-year mean clear-sky upward radiative 791

792 fluxes with resolved mineralogy, F_2^{\uparrow} is the 19-year mean clear-sky upward radiative fluxes for

793 homogeneous dust. Section S5 in the Supplement provides the clear-sky radiative fluxes anomalies

794 at TOA and surface (SFC) induced by resolving dust mineralogy over the global scale, we see

795 much more significant anomalies over the North Africa than other regions, which makes senses

because that dust aerosol is the most dominant aerosol species in this area. The changes in dust

aerosol optical properties have a greater potential to lead to significant impacts on radiation and

798 climate over the region than in the others. Therefore, this section focuses on the North Africa

799 region, where the Sahara Desert, the largest dust source in the world, is located. The Sahara (20°N-

30°N, 10°W-35°E) and the Sahel (10°N-20°N, 10°W-35°E) regions are studied separately. We

specifically analyze the results for the June-July-August (JJA) season when dust loading is at its

802 highest and the West African Monsoon is the strongest.

803 5.1.1 Impacts on Clear-sky Radiative Fluxes relative to HD27

804 Before comparing mineral-resolved experiments (e.g., BM) with HD27 control run, the dust has

been used in the standard GFDL AM4.0, to understand their impacts on clear-sky radiative fluxes

806 relative to HD27, it's worth recapping that the effects of resolving mineralogy relative to HD27

807 can be attributed to two factors: the reduction in dust absorption and the variation in dust scattering

808 properties induced by the mineralogical inhomogeneity. Deleted: experiment. As shown in Table 3

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Deleted: is much more scattering than HD27 and approximately as

Deleted: as the mineral-resolved dust (i.e., MG-mixing, and BM-mixing dust) in terms of globally averaged SSA. Therefore, this step sheds light on the impacts of resolving dust mineralogy on climate given similar globally averaged scattering properties with and without resolved

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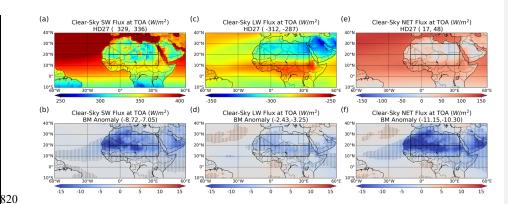


Figure 6. Seasonal mean JJA climatology (2001-2019) clear-sky SW (1st column), LW (2nd column) and Net (3nd column) radiative flux at TOA for the HD27 control run (1st row) and their anomalies resulting from resolving dust mineralogy in Bruggeman-mixing experiment (2nd row). Downward direction is defined as positive. The dotted area denotes anomalies that are statistically significant. The two values in parentheses within the title of each figure are domain average for the Sahara and Sahel regions.

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The first row in Figure 6 illustrates the modeled clear-sky shortwave (SW), longwave (LW) and net (NET: the combination of SW and LW) radiative flux at TOA from the HD27 control run. Relative to HD27, mineral-resolved dust (e.g., BM-mixing dust) generally reflects more SW radiation back to space and induces negative SW flux anomalies at TOA (Figure 6a, b; Positive: downward). Relative to the HD27 control run, the LW flux anomaly at TOA resulting from resolving mineralogy is less substantial compared to SW flux anomaly (Figure 6c, d). After combining both SW and LW, resolving mineralogy turns out to induce substantial decrease in NET flux at TOA, with a more than 50% negative anomaly over the Sahara and around a 20% negative anomaly over the Sahel (see values in parentheses in Figure 6e, f). Therefore, less NET radiation reaches the Earth at TOA in the mineral-resolved dust cases due to their lower absorptivity.

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Deleted: ., VOL-mixing, MG-mixing, and

Deleted: Figure 4 a-d; Positive: downward). In addition, MG-mixing and BM-mixing dust induce stronger reflection anomalies than VOL-mixing dust. The results are consistent with the fact that mineral-resolved dust is less absorptive than HD27 dust in the visible spectrum, and MG-mixing and BM-mixing dust are even less absorptive than VOL-mixing dust, as discussed in Section 4.

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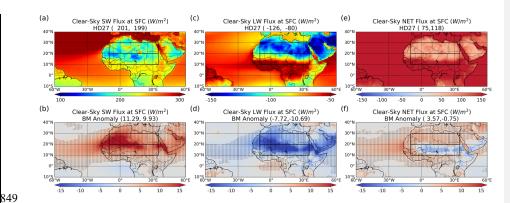


Figure 7. As in Figure 6, but for the surface.

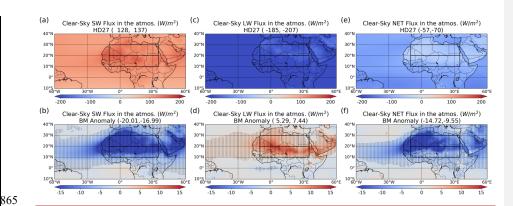
 At the surface (SFC) in Figure 7, the enhanced scattering of mineral-resolved dust scatters more SW radiation toward Earth's surface, leading to a positive SW flux anomaly at SFC (Positive: downward). In the LW, the cooling of the mineral-resolved dust layer, due to its low absorption, results in less LW radiation being emitted toward Earth's surface, This reduction in the downward LW emission outweighs the change in the upward LW emission from the Earth's surface, thereby causing a negative LW flux anomaly at SFC. The positive anomalies in SW radiation are approximately canceled out by the negative anomalies in LW radiation (Figure 7). As a result, a similar amount of radiation reaches the Earth's surface in both HD27 and mineral-resolved cases. Despite less NET radiation entering the Earth at TOA in mineral-resolved cases, the similar amount of NET radiation reaching the Earth's surface indicates that less NET radiation is absorbed in the atmosphere in mineral-resolved cases.

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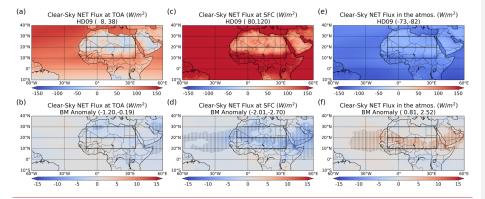


<u>Figure 8. As in the Figure 6, but for the radiative flux absorbed in the atmosphere.</u>

<u>In Figure 8</u>, the negative SW flux anomalies are partially offset by positive LW flux anomalies, resulting in negative NET flux anomalies in the atmosphere. These anomalies amount to approximately a 25% reduction over the Sahara and 10% reduction over the Sahel (see values in parentheses in <u>Figure 8b, d, f</u>).

5.1.2 Impacts on Clear-sky Radiative Fluxes relative to HD09

Before comparing mineral-resolved experiments with HD09 control run, where the homogeneous dust is as absorptive as mineral-resolved dust (e.g., MG and BM) from a global perspective, to understand their impacts on clear-sky radiative fluxes relative to HD09, it's worth recapping that the effects of resolving mineralogy relative to HD09 are primarily attributed to the variation in dust scattering properties induced by the mineralogical inhomogeneity.



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880 Figure 9. As in the Figure 6, but for HD09 control run, In addition, SW and LW flux anomalies are not Moved (insertion) [14] 881 shown here. Clear-sky net flux at TOA (1st column), at surface (2nd column), and in the atmosphere (3 Moved (insertion) [15] 882 column) are shown in this figure. Deleted: Figure 7 883 Figure 9 shows the clear-sky fluxes anomalies with respect to HD09 over North Africa, the 884 anomalies over the global scale are shown in Figure \$7, \$9, \$11 and \$13 in the Supplement. Deleted: S5, Deleted: and 885 In contrast to the anomalies with respect to HD27 control run, resolving dust mineralogy does not cause substantial anomalies (< 5%) in clear-sky fluxes with respect to HD09 control run. This can 886 887 be attributed to their similarity in dust scattering properties from a global mean perspective, 888 particularly SSA as shown in Figure 5. The comparable effects of HD09 and mineral-resolved dust Deleted: Figure 3 889 on radiation suggest that resolving dust mineralogy does not have significant impacts on clear-sky Deleted: regionally averaged 890 fluxes when homogeneous dust is as scattering as mineral-resolved dust aerosols on a global scale. Deleted: . The 891 However, the equivalence between HD09 and mineral-resolved dust in terms of their interactions 892 with radiation may be related to the three limitations in the current model simulations: 1) Soil 893 mineralogy: The limited soil mineralogy database fails to adequately capture the regional variation 894 of iron content (or SSA) within the region; 2) Dust emission based on Ginoux et al. (2001) uses a 895 continuous function of topography, which does not take into account geomorphological 896 characteristics of the surface to differentiate soil properties of dust sources as done by others 897 (Zender et al., 2003; Bullard et al., 2011); 3) Dust transport: Excessive numerical diffusion may 898 occur when solving advection equation (Ginoux, 2003). Given all those limitations of our model 899 simulations, this finding may differ with improved representation of dust sources and transport. 900 Such improvement may come from spaceborne soil mineralogy dataset (e.g., EMIT) that may 901 capture accurately the regional contrasts in iron oxides content. 902 5.1.3 Compare Clear-sky Radiative Fluxes with CERES Observations 903 Furthermore, we conduct a comparison of modeled SW upward, LW upward, NET downward flux 904 at TOA with observation-based results from CERES EBAF Ed4.2 product (see Table 4). The Deleted: Figure 8) 905 difference between modeled flux and CERES observations are listed in parentheses within the title 906 of each figure in Table 4. Compared to HD27, the more scattering HD09 and mineral-resolved BM Deleted: Figure 8 achieve much better agreement with CERES observations in clear-sky flux (i.e., SWup, LWup and 907 908 NETdn) at TOA. This is evident in the smaller values of HD09 - CERES (e.g., NETdn: 1.6 for the 909 Sahara and 2.4 for the Sahel) and BM - CERES (e.g., NETdn: 0.4 for the Sahara and 2.1 for the

Sahel) compared to HD07 - CERES (e.g., NETdn: 11.3 for the Sahara and 12.4 for the Sahel), as

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shown by the values in parentheses in <u>Table 4</u>. Between HD09 and BM, BM tends to agree slightly

920 better with CERES.

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Table 4 Comparison of modeled clear-sky SW upward (SWup, 1st row), LW upward (LWup, 2nd row) and NET downward (NETdn, 3rd row) fluxes at TOA with CERES observation-based results over 2001-2019 JJA. The 1st column shows the clear-sky flux estimates at TOA from CERES_EBAF_Ed4.2 product, which represents clear-sky flux with clouds removed from the atmospheric column. The following columns show the difference of modeled clear-sky flux at TOA in HD27 (2nd column), HD09 (3rd column) and BM (4th column) experiments from CERES observations. The two values in parentheses represent domain average for the Sahara and Sahel regions as indicated in figures in Section 5.1.2. Specifically, the first column (CERES) is domain averaged flux, while the second (HD27 – CERES), third (HD09 – CERES), and fourth (BM – CERES) columns are domain averaged flux differences between model and CERES observation-based results.

	CERES	HD27 – CERES	HD09 – CERES	BM – CERES
Clr SWup flux at TOA (W/m²)	(135, 113)	(-9.3, -9.4)	(-1.1, -2.2)	(-0.6, -2.3)
Clr LWup flux at TOA (W/m²)	(314, 291)	(-1.4, -3.1)	(-0.6, -0.5)	(0.2, -0.1)
Clr NETdn flux at TOA (W/m²)	(6, 36)	(11.3, 12.4)	(1.6, 2.4)	(0.4, 2.1)

5.2 Impacts on land temperature

Here we explore the impacts on the temperature vertical profile and near-land surface temperature, relative to HD27 and HD09, respectively. Compared to the HD27 control run, lower absorption of radiation in the atmosphere by mineral-resolved dust aerosols results in statistically significant negative temperature anomalies in the atmosphere ranging from 800 mb up to 500 mb where dust aerosols are mainly located (Figure 10). In contrast, there is no statistically significant temperature anomaly for mineral-resolved dust cases compared to HD09, as illustrated by the red curves in Figure 10 This finding aligns with the insubstantial anomalies (<5%) in clear-sky NET radiative fluxes discussed in Section 5.1.2. In the subsequent part of the section, we will delve into comparing mineral-resolved experiment (using BM as an example) with the HD27 control run. This comparison will help us further understand the impact of dust aerosols with distinct absorption on land temperature.

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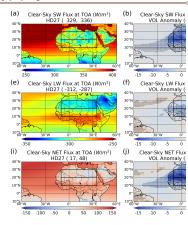
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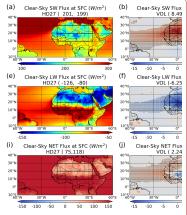
Moved up [15]: In addition, SW and LW flux anomalies are not shown here.

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Deleted:relative to HD27 and HD09, respectively. Compared to the HD27 control run, lower absorption of radiation in the atmosphere by mineral-resolved dust

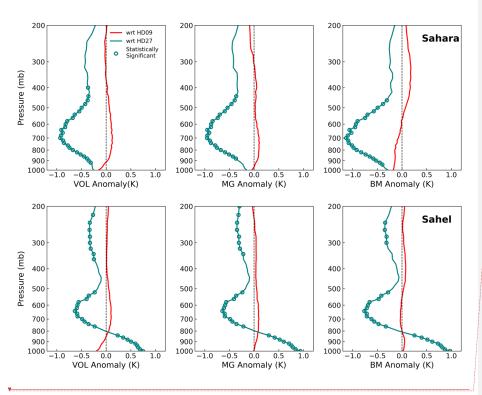


Figure 10 Vertical profile of temperature anomaly induced by resolving dust mineralogy for the Sahara (1st row) and the Sahel (2nd row) regions in the three mineral-resolved experiments (i.e., VOL, MG, BM). Green lines represent temperature anomalies with respect to HD27 control run. Red lines are temperature anomalies with respect to HD09 control run. The circles represent statistically significant temperature anomaly.

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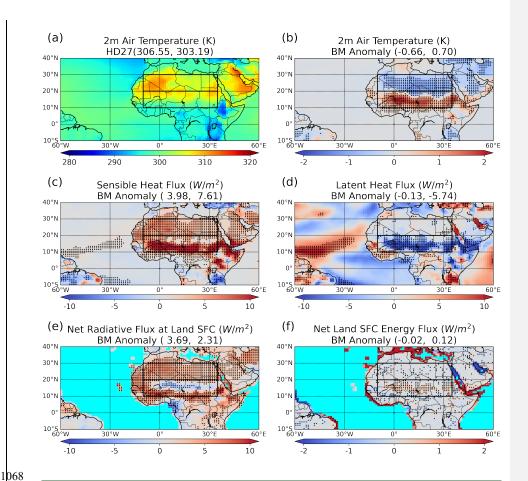


Figure 11, Air temperature at 2-meter from HD27 control run (a), anomaly (b) induced by implementing Bruggeman-mixing minerals in BM experiment, surface sensible heat flux (c), latent heat flux (d), net radiative flux (e), net energy flux (f) anomalies between BM and HD27; Upward flux is positive in (c) and (d), while downward flux is positive in (e) and (f). Net energy flux (f) is the subtraction of (c), (d), and downward ground flux from (e). Note that ground flux is not shown in the figure considering its relatively small magnitude, but it is included in the land surface net energy flux calculations in subplot (f). The dotted area denotes anomalies that are statistically significant. The two values in parentheses within the title of each figure are domain average for the Sahara and Sahel regions.

Figure 11a shows air temperature at 2-meter from HD27 control run over the Northern Africa. Near the land surface, more scattering mineral-resolved dust induces a temperature decrease (i.e., negative temperature anomaly _0.66 K) over the Sahara and a temperature increase (i.e., positive

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temperature anomaly 0.70 K) over the Sahel as shown Figure 11b. To understand this phenomenon, we further analyze the surface energy budget in Figure 11c-f.

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Radiative flux perturbation over land is quickly equivalented by balancing surface radiative fluxes with sensible heat flux, latent heat flux and ground flux (i.e., downward heat flux into the ground), which results in nearly zero net energy flux at land surface as shown in Figure 11f. Precisely, the radiative flux anomaly comprises two contributions: one is the instantaneous radiative forcing (IRF) caused by the change in dust mineralogy in the atmosphere, and the other one is the associated radiative feedbacks. For simplicity, we will not partition the radiative flux anomaly in our discussion here. Over the Sahel region, the positive net radiative flux anomaly at land surface is balanced out by the increased sensible heat flux and the decreased latent heat flux as well as ground flux. Note that the ground flux is generally small in magnitude and not shown in Figure 11, but we include it in calculating net surface energy flux in Figure 11f. The decrease of latent heat flux over the Sahel in BM case (Figure 11d) is due to the depletion of soil moisture (and therefore evaporation) in the region as shown in Figure 12. The depletion of soil moisture is caused by the decrease in moisture carried by onshore winds over the Sahel and the decrease in precipitation over the same region, as will be discussed in section 5.3. Therefore, a large enhancement of sensible heat flux ($\sim 7.6 W/m^2$) is needed (Figure 11c) not only to compensate for the depletion in latent heat flux ($\sim 5.7 W/m^2$ in d), but also to balance out the increased net radiative flux (~ 2.3 W/m^2 in Figure 11e). As a result, higher land surface temperature with anomaly around 0.7 K is needed in the region to achieve the required sensible heat flux enhancement.

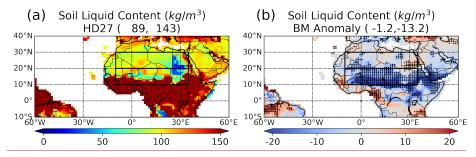


Figure 12, Soil liquid content in HD27 control run (a) and anomaly resulting from implementing Bruggeman-mixing minerals in the BM experiment (b). The two values in parentheses within the title of each figure are domain average for the Sahara and Sahel regions.

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Over the Sahara region, latent heat flux does not change from HD27 case to BM case, therefore, the increased net radiative flux ($\sim 3.69 \ W/m^2$) in BM compared to HD27 is mainly balanced out by the enhanced sensible heat flux ($\sim 3.98 \ W/m^2$) which requires a larger temperature gradient between surface and atmosphere. However, there is a very strong negative temperature anomaly (around -1K) in the atmosphere near 700 hPa due to less dust absorption in BM as we discussed in Figure 10. The strong negative temperature anomaly in the lower atmosphere effectively increases the vertical temperature gradient. As such, it is not necessary for the land surface temperature to increase; in fact, it may need to decrease by approximately $0.66 \ K$ to achieve the desired enhancement in sensible heat flux and reach equilibrium.

Additionally, to assess the effectiveness of various dust scattering properties (e.g., HD27, HD09, and BM) in matching observations of near-surface temperature, we compare the modeled near surface temperature (T_{2m}) with CRU TS observations, which is described in Section 2.6, over the Sahara and Sahel regions (Table 5). Considering the relatively large inter-model spread of regional surface air temperature, we compare the Sahara-Sahel regional contrast in surface air temperature to the CRU rather than comparing their absolute values. Table 5 shows that HD09 and BM improve the agreement with CRU in Sahara-Sahel temperature contrast compared to HD27, and BM exhibits the closest agreement with CRU.

Table 5. The 19-year (2001~2019) JJA mean 2-meter Air Temperature (T_{2m} , unit: K) and their standard deviation over the 19 years from CRU observations and modeled experiments over the Sahara and Sahel regions. The 'Contrast' row indicates the T_{2m} regional contrast between the Sahara and the Sahel.

Region	CRU (K)	HD27 (K)	HD09 (K)	ВМ (К)
Sahara	305.8 ± 0.18	306.55 ± 0.52	306.2 ± 0.68	305.89 ± 0.61
Sahel	304.1 ± 0.32	303.19 ± 0.46	303.87 ± 0.51	303.89 ± 0.59
Contrast	1.7 ± 0.5	3.36 ± 0.98	2.3 ± 1.19	2.0 ± 1.2

5.3 Impacts on winds and precipitation

To understand the fast circulation and hydrological response resulting from resolving dust mineralogy, we examine surface wind speed anomalies (Figure 13) and precipitation anomalies

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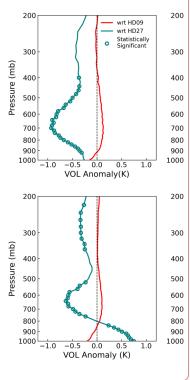
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(Moved up [14]: Figure 9.

Moved up [16]: Vertical profile of temperature anomaly induced by resolving dust mineralogy for the Sahara (1st row) and the Sahel (2nd row) regions in the three mineral-resolved experiments (i.e., VOL, MG, BM). Green lines represent temperature anomalies with respect to HD27

Moved up [17]: Air temperature at 2-meter from HD27 control run (a), anomaly (b) induced by implementing Bruggeman-mixing minerals in BM experiment, surface sensible heat flux (c), latent heat flux (d), net radiative flux (e), net energy flux (f) anomalies between BM and HD27;

Moved up [19]: Soil liquid content in HD27 control run (a) and anomaly resulting from implementing Bruggeman-mixing minerals in the BM experiment (b). The two values in parentheses within the title of each figure are domain average for the Sahara and Sahel regions.



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(Figure 14) induced by mineral-resolved dust. We compare mineral-resolved experiments (using BM as an example) with HD27 and HD09, respectively, to investigate the effects of resolving dust mineralogy.

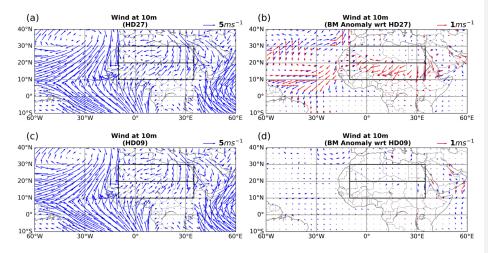


Figure 13 Surface wind at 10-meter from HD27 (a) and HD09 (c) control runs and their anomalies (b) and (d) resulting from implementing Bruggeman-mixing minerals in the BM experiment. Statistically significant wind anomalies are highlighted by red arrows.

Global precipitation is higher by 0.017 mm/day for the BM mineral-speciated case compared to HD27 experiment. This is consistent with the lower SW absorption in the former, given the global compensation between latent heating associated with precipitation and net radiative cooling (e.g., Allen and Ingram, 2002; Samset, 2022). (Global radiative cooling is also compensated, although to a smaller extent, by the sensible heat flux.)

However, within the Sahel, precipitation for BM is reduced compared to HD27 (Figure 14b), with weaker onshore flow during the summer monsoon (Figure 13b), displacing West African precipitation toward the Guinea coast. This reduction is consistent with several previous calculations of the fast response calculated with fixed SST (e.g., Stephens et al., 2004; Miller et al., 2004b; Lau et al., 2009; Jin et al., 2016; Jordan et al., 2018).

Dust absorbs radiation and redistributes heating from the surface to within the dust layer (Miller and Tegen, 1999; Strong et al., 2015). The heating of the air warms the lower to middle troposphere,

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thereby enhancing upward motion. The rising warm air spawns a large-scale onshore flow, carrying the low-level moist air from the Atlantic to the Sahel, thus enhancing precipitation over this region (Balkanski et al., 2021). The more scattering mineral-resolved BM and MG dust absorb less radiation and cause less warming of the atmosphere (Section 4.1), reducing adiabatic cooling through ascent and Sahel precipitation. The suppressed ascent in BM compared to HD27 is associated with a reduction both in the wind divergence aloft and in convergence at the surface (Figure 13b). The reduction in convergence results in northeast wind anomalies at 10-meter over the Sahel (Figure 13b), which are opposite in direction to the southwest onshore winds of the West African Monsoon (Figure 13a). The inhibition of onshore winds, bringing less moisture to the Sahel, is consistent with the reduction of ascent and precipitation over this region (Figure 14a, b).

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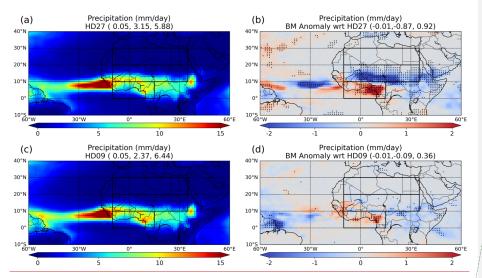


Figure 14, 19-year (2001-2019) JJA mean precipitation from HD27 (a) and HD09 (c) control runs and anomalies resulting from implementing Bruggeman-mixing minerals with respect to HD27 (b) and with respect to HD09 (d). The three values in the parenthesis are domain averaged values for the Sahara, Sahel, and GC regions.

Besides the Sahel, there is a statistically significant positive anomaly (0.92 mm/day) of precipitation over the region to the south of the Sahel in BM relative to HD27 (Figure 14b). We will call this region, the Guinea Coast (GC: 0-10°N, 10°W-15°E) region. One possible reason for the increase of precipitation over the GC is that the region is located to the south of the Saharan dust layer, where the suppression of ascent over the Sahel in BM suppresses the subsidence in the

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Deleted: Figure 12) and precipitation anomalies (Figure 13) induced by mineral-resolved dust. Based on the elevated heat pump (EHP) mechanism of Lau et al., (2009), more absorptive dust absorbs radiation and redistributes heating from the surface to within the dust layer (Miller and Tegen, 1998; Strong et al., 2015). Atmospheric heating leads to ascent thus enhancing precipitation. In this sense, more scattering mineral-resolved dust aerosols absorb less radiation and cause less warming of the atmosphere, suppressing ascent motion and reducing precipitation. As discussed in the Section 4.1, mineral-resolved dust (i.e., VOL, MG, and BM) is more scattering than HD27. Relative to HD27 case, mineral-resolved dust causes less warming of the atmosphere due to less absorption and suppresses ascent. The suppressed ascent is associated with a reduction both in the wind divergence aloft and in convergence at the surface. The reduction in convergence results in northeast wind anomalies at 10-meter over the Sahel (Figure 12a, b), which are opposite in direction to the southwest onshore winds of the West African Monsoon. The inhibition of onshore winds. bringing less moisture to the Sahel, combined with the suppression of ascent motion work together to reduces precipitation over this region (Figure 13a, b).

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1266 1267	GC region and, therefore, enhances the precipitation (Guo et al. 2021). Alternatively, the moist onshore flow that is weakened in BM is subject to greater dilution by the dry desert air, resulting		
1268	in reduced moist static energy and buoyancy, limiting convection to the coastal region.		
1269	These changes in precipitation, have non-negligible effects on soil moisture content in North Africa		Deleted: pattern
1270	due to its moisture-starved environment. The decrease in precipitation over the Sahel in BM leads		
1271	to a reduction in soil moisture content. Conversely, the increase of precipitation over the GC leads		
1272	to increases of soil moisture (Figure 12). The change in soil moisture content further affects the	(Deleted: content (Figure 11).
1273	partitioning of surface energy fluxes and the efficiency of the latent heat flux, thereby affecting		
1274	land surface temperature, as illustrated by Figure 11.		Field Code Changed
1275	So far, we have been focusing on discussing the impacts of resolving dust mineralogy on winds		
1276	and precipitation relative to the HD27 control run. The large discrepancy in optical properties		
1277	between HD27 and mineral-resolved dust allows us to better understand how <u>distinct</u> dust		
1278	<u>absorption impacts</u> our climate through <u>its</u> distinct radiative effects.	(Deleted: aerosols with varying optical properties impact
1279	As discussed in section 4.1, HD09 dust is nearly as scattering as mineral-resolved dust but exhibits		Deleted: their
1280	smaller regional variability. Section 5.1.2 shows that resolving dust mineralogy does not lead to		Deleted: 5.1
1281	statistically significant anomalies on radiation relative to HD09. Consistently, there are no further		
1282	statistically significant impacts on winds (Figure 13c, d) and precipitation (Figure 13c, d).		Deleted: Figure 12
1283	To investigate the effectiveness of various dust scattering properties (e.g., HD27, HD09, BM) in	>	Field Code Changed
1284	matching observations of precipitation rate, we compare the modeled precipitation with CRU TS		Deleted: Furthermore, to
1285	observations over the Sahara, Sahel and GC regions (Table 6). The greater difference between		
1286	HD09, BM and CRU (i.e., HD09 – CRU and BM – CRU) indicate that more scattering HD09 and		
1287	BM lead to a larger discrepancy between the modeled precipitation and CRU observations. Jn		Deleted: This discrepancy may reflect
1288	contrast, Balkanski et al. (2021) describes the same balance of increased dust absorption and Sahel		-
1289	precipitation but find improved agreement with Global Precipitation Climatology Project (GPCP)		
1290	data by assuming homogeneous dust containing 3% iron oxides by volume. Contrasts between that		
1291	study and ours result from differences between the GPCP and CRU data sets, contrasts in dust		
1292	absorption (related to contrasts in the dust size distribution or assumed index of refraction), non-		
1293	dust model biases in precipitation or differences between the slow response computed by	-/ >	Deleted: model Deleted: ,
1294	Balkanski et al. (2021) and the fast response that we calculate. The fast and slow response even	> >	Deleted: the absence of
		1/6	Deleted: climate
		,(i	Deleted: , whose inclusion can change

1308 exhibit differences in the sign of the calculated precipitation anomaly within some regions of the

WAM (Miller and Tegen, 1998; Jordan et al., 2018).

Table 6. Comparison of modeled precipitation rate (PRE, unit: mm/day) with observations from CRU TS dataset over 2001-2019 JJA. CRU column represents 19-year (2001-2019) JJA mean PRE over the region as well as 19-year standard deviation (std). HD27 - CRU column shows the 19-year mean PRE difference between HD27 control run and CRU observations, along with the corresponding std of this 19-year

difference. Similar for HD09 – CRU and BM – CR
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Comparison Region	CRU (mm/day)	HD27 – CRU (mm/day)	HD09 – CRU (mm/day)	BM – CRU (mm/day)
Sahara	0.08 ± 0.013	-0.03 ± 0.03	-0.03 ± 0.07	-0.04 ± 0.05
Sahel	2.99 ± 0.27	0.16 ± 0.56	-0.62 ± 0.43	-0.71 ± 0.41
Guinea Coast	6.16 ± 0.49	-0.28 ± 0.90	0.28 ± 1.02	0.64 ± 0.83

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6 Potential for reducing mineral tracers

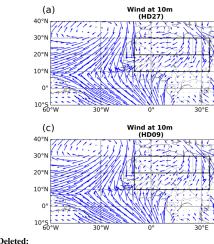
Thus far in this study, we have been using 45 mineral tracers in mineral-resolved experiments (i.e., VOL, MG, and BM). However, it is important to investigate the potential of reducing the number of mineral tracers in climate models to lower computational costs. In this section, we take BM as a reference for providing the best comparisons with CRU temperature and CERES flux observations, and conduct an experiment named BM-RT to assess the possibility of reducing mineral tracers in BM. The BM-RT experiment consists of three sub-experiments, namely, BM-LC, BM-LCRH, and BM-LCRHRG. In each of the three sub-experiments, the number of mineral tracers is progressively reduced, allowing for an examination of the relative impacts of different minerals on climate compared to the reference BM.

As discussed in section 4.1, the three clay minerals (i.e., illite, kaolinite, smectite) exhibit similar optical properties and perform similar functions in climate by hosting hematite. Hence, they can be combined in their interaction with radiation without significant impacts on climate. In addition, by lumping the three clay minerals together, the number of mineral tracers can be reduced from 45 in BM (nine types of minerals × five size bins) to 35 (seven types of minerals × five size bins).

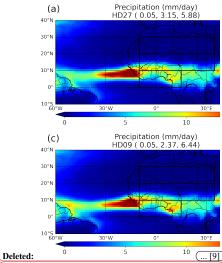
Moved up [18]: Figure 12.

Moved up [20]: Surface wind at 10-meter from HD27 (a) and HD09 (c) control runs and their anomalies (b) and (d) resulting from implementing Bruggeman-mixing minerals in the BM experiment. Statistically significant wind anomalies are highlighted by red arrows.

Moved up [21]: 19-year (2001-2019) JJA mean precipitation from HD27 (a) and HD09 (c) control runs and anomalies resulting from implementing Bruggeman-mixing minerals with respect to HD27 (b) and with respect to HD09 (d). The three values in the parenthesis are domain averaged values for the Sahara, Sahel, and GC regions.



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Deleted: Note, for anomalies, we only average the dotted area, in other words, we only calculate the domain average of statistically significant anomalies.

1350	Therefore, in the first sub-experiment BM-LC (where 'LC' represents 'Lump Clay minerals'), we	
1351	lump together the three clay minerals as one mineral species 'clay433'.	
1352	Based upon the C1999 soil mineral composition that we use, externally mixed hematite is mainly	Deleted: Externally
1353	concentrated over the Sahel region (Ginoux et al. 2023, in preparation) and cannot be transported	
1354	to remote regions due to its high mass density. Obiso et al. (2023) shows that visible extinction	
1355	due to externally mixed hematite is negligible compared to other mineral components including	Deleted: external
1356	hematite internally mixed with other minerals. Thus, we further remove external hematite tracers	Deleted: hematite and
1357	in the second sub-experiment BM-LCRH (where 'RH' indicates 'Remove externally mixed	
1358	Hematite'). The mass fraction of external hematite is combined with internal hematite to ensure	
1359	that the total mineral fraction at emission remains equal to one. In this sub-experiment, the number	
1360	of mineral tracers is reduced from 35 in BM-LC to 30 in BM-LCRH.	
1361	Since there are no known specific impacts of gypsum on climate, we conducted the third sub-	
1362	experiment, BM-LCRHRG ('RG' indicates 'Remove Gypsum'), where gypsum was removed. The	
1363	mass fraction previously attributed to gypsum at emission, which is very low at the global scale,	
1364	was proportionally redistributed among all other minerals. The number of mineral tracers is finally	
1365	reduced from 30 in BM-LCRH to 25 in BM-LCRHRG.	
1366	We analyze the 19-year (2001-2019) time series of total dust mineral emission before and after	
1367	reducing mineral tracers in Figure 15. We observe subtle differences in total mineral emission	
1368	between experiments, which arises from the feedback of mineral radiative interactions. However,	
1369	these differences are numerically small, and Student's t-test suggests that the time series of the	
1370	four experiments are not statistically different. Additionally, the globally averaged DAOD and	
1371	SSA of each sub-experiment remains highly similar to those of the reference experiment BM, as	
1372	listed in Table 3.	Field Code Changed

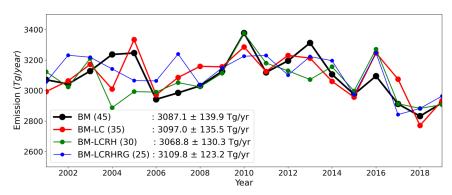


Figure 15. Time series of total dust mineral emission from 2001 to 2019 before and after reducing the number of mineral tracers. The legend displays the following information: 'Experiment name (number of mineral tracers): 19-year averaged total mineral emission \pm 19-year standard deviation of total mineral emission'.

Given the highly similar optical properties of minerals before and after reducing mineral tracers, we further investigate their impact on climate. Firstly, we examine the clear-sky flux anomaly of each of the three sub-experiments at TOA and surface relative to the reference experiment BM, as shown in Supplementary Figure $\underline{\$14}$ for TOA and Supplementary Figure $\underline{\$15}$ for surface. We only observe a few statistically significant (p-value < 0.05) anomalies over the North Africa, suggesting that the reduction of mineral tracers in the three sub-experiments has a weak impact on radiation.

Furthermore, we investigate the anomaly in temperature profile, surface winds, and precipitation of each of the three sub-experiments relative to the reference experiment BM. The results are presented in Figure \$16-\$18 in the Supplement. No statistically significant (p-value < 0.05) temperature anomalies (Figure \$16) and surface wind anomalies (Figure \$17) is observed. Only a few statistically significant anomalies in precipitation are observed in the Supplementary Figure \$18. These results suggest that the reduction of mineral tracers in the three sub-experiments has a weak impact on climate.

The results from the BM-RT experiment suggest combining clay minerals and excluding the externally mixed hematite and gypsum to simulate dust impact on radiation. This does not preclude similar conclusion for other impacts of dust on the Earth's climate systems. The removal of mineral tracers reduces the number of mineral tracers from 45 to 25, effectively lowering computational costs without causing statistically significant impacts on simulating climate.

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1406 7 **Conclusions** 1407 We simulate the distribution of dust mineralogy (i.e., illite, kaolinite, smectite, hematite, calcite, 1408 feldspar, quartz, and gypsum) and activate their interaction with radiation in the GFDL AM4.0 1409 model. Our investigation focuses on the radiative impacts of resolving dust mineralogy on Earth's 1410 atmosphere and its fast response of land temperature, surface winds and precipitation. 1411 We set up two baseline homogeneous dust control runs: HD27 and HD09, in which dust 1412 mineralogy is considered as temporally and spatially uniform, the former following the standard 1413 configuration for the dust optical properties in GFDL AM4.0 and the latter including a more 1414 scattering dust. Three experiments with resolved mineralogy are also conducted: VOL, MG, and 1415 BG, using three different mixing rules for the internal mixture between hematite and clay minerals 1416 (i.e., volume weighted mean, Maxwell Garnett, and Bruggeman). The comparison of dust 1417 absorption properties (e.g., SSA) with observation-based results suggests that the homogeneous 1418 dust used in the standard GFDL AM4.0 (i.e., HD27) is overly absorptive, Maxwell Garnett and Deleted: 1419 Bruggeman mixing rules are more appropriate than volume weighted mixing rule in calculating 1420 optical properties of internal mixtures of hematite and clays. Compared to HD27, the homogeneous 1421 dust with reduced hematite content (HD09) and mineral-resolved dust (i.e., MG and BM) exhibit 1422 much better agreement with AERONET retrievals and laboratory measurements in terms of dust 1423 absorption properties (i.e., SSA). Additionally, resolving dust mineralogy enhances regional Deleted: mineral-resolved 1424 variability in dust SSA compared to homogenous dust, further improves the agreement with 1425 AERONET, even though it remains lower than the observed variability. 1426 The two homogeneous dust control runs, HD27 and HD09, with distinct dust absorption properties, 1427 allow us to investigate the impact of dust mineralogy on Earth's radiation and fast climate response. Deleted: . During JJA, compared 1428 relative to distinct baseline homogeneous dust. In comparison to HD27, resolving mineralogy Deleted: dust 1429 reduces dust absorption. During JJA, the reduced dust absorption results in a reduction of over 50% 1430 in NET downward radiation across the Sahara and approximately 20% over the Sahel at TOA. 1431 Additionally, there is a reduction of around 25% in the atmospheric absorption of radiation over 1432 the Shahara and around 10% over the Sahel in the atmosphere. The reduced surface absorption of 1433 radiation by mineral-resolved dust leads to a temperature decrease of 0.66 K at the land surface Deleted: 39K 1434 across the Sahara and an increase of 0.7 K over the Sahel. The reduced NET downward radiation Deleted: 6K 1435 at TOA, attributed to the less absorption of radiation by mineral-resolved dust, suppresses ascent

1442 and weakens the monsoon inflow from the Gulf of Guinea. This brings less moisture to the Sahel, 1443 which combined with decreased ascent induces a reduction of precipitation. On the other hand, 1444 compared to HD09, resolving dust mineralogy results in dust absorption comparable to that of 1445 HD09 on a global scale. However, when resolving mineralogy, there is an increase in spatial 1446 variation of dust absorption. Additionally, we observe a noticeable change in global distribution 1447 of dust absorption, with more dust absorption distributed in the Southern Hemisphere and lower 1448 dust absorption over Iceland and Taklamakan regions. Nevertheless, the higher spatial variation in 1449 dust absorption does not lead to statistically significant changes in any of the climate aspects 1450 mentioned above. The models with reduced absorption (HD09 and fully resolved mineralogy) 1451 improve the comparison with observations of CERES fluxes and CRU land surface temperature. 1452 We see a slightly better agreement with observations for fully resolved mineralogy than HD09 1453 however it is not statistically significant. As such, when using fixed mineralogical composition, 1454 we recommend using a 0.9% hematite content in volume, which represents the lowest of the three 1455 hematite mixings considered by Balkanski et al. (2007). Historically, climate models have relied on fixed refractive index to consider dust radiative forcing 1456 1457 starting (IPCC, 2001) with strongly absorptive value based on dust samples in Sahara (Patterson 1458 et al., 1977) to more scattering values after dust absorption could be inferred from satellite and 1459 surface observations (e.g., Sinyuk et al., 2003; Balkanski et al., 2007). With the launch of EMIT 1460 in July 2022 and the expected delivery of a high-resolution map of soil mineralogy in source areas, 1461 dust interactions with radiation in climate models will be calculated directly from the simulated 1462 mineralogical composition (Li et al., 2021). Still, the additional burden of simulating a dozen 1463 minerals may be too prohibitive for large ensemble climate models simulations. In such cases, our 1464 analysis suggests the use of a fixed value providing similar radiative effects as the comprehensive 1465 representation of minerals. However, our recommendation is directed toward the GFDL AM4.0 1466 model with all its uncertainties related to mineral distribution, emission sources, and aerosol 1467 transport. Moreover, incorporating dust mineralogy in models is likely to be important in other 1468 aspects, such as cloud properties, ocean biogeochemistry, air quality and photochemistry. For 1469 studies with resolved mineralogy, we show that the number of mineral tracers can be reduced from 1470 45 to 25 without losing the quality of comparison with observations of CERES fluxes and CRU

surface temperature. Such reduction can be achieved by lumping together clay minerals, removing

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external hematite and gypsum. For specific research such as biogeochemistry, it may be necessary to fully resolve mineralogy to achieve accuracy.

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1504 1505 This study has some limitations. First, the soil mineralogy map from C1999 is based on extensive extrapolation and limited observations. In terms of the need to improve knowledge of soil mineralogy in dust source regions, the launch in July 2022 of the EMIT instrument operating from the International Space Station will provide mineral identifications of dust sources using hyperspectral measurements (Green et al., 2020). The EMIT soil map measurements will improve resolving dust mineralogy in climate models and advance our understanding of dust's effects in the Earth system. Second, hematite and goethite are the most common iron oxides present in soils. However, goethite is not considered in this study because not included in the used soil mineralogy map. Previous studies suggest that goethite is generally more abundant than hematite, but it is less absorptive than hematite in the visible spectrum (Formenti et al., 2014). Therefore, the abundance of iron oxides may be underestimated in this study, which may lead to underestimation of dust absorption in the SW. A more recent database by Journet et al. (2014) (J2014) includes the distribution of goethite but it shares many limitations as C1999 (e.g., extensive extrapolation) and has other major disadvantages, such as numbers of missing soil fractions of some minerals at some locations. Third, the refractive index of hematite used in our study is close to the upper range of the values available in literature (Zhang et al., 2015). Hence, the last two limitations, underestimation of iron oxide content and overestimation of absorption by hematite, may have compensating effects.

This study, by prescribing SST, calculates only the fast response to the dust DRE, without including the slow response by the sea surface temperature. This avoids the need to spin-up the model for decades before reaching new equilibrium but may overestimate the eventual response, as shown by Miller and Tegen (1998) and Balkanski et al. (2021). This complicates model evaluation because the observations include the slow response to dust. Variables like precipitation are especially sensitive to the inclusion of the slow response because prescribed SST experiments omit the surface energy balance over the ocean. Thus, the surface DRE beneath the aerosol layer, which is generally negative, is not fully balanced by a fast reduction of evaporation (Miller et al., 2004a). The addition of the surface balance in the slow response can reverse the sign of the fast precipitation anomaly (Miller and Tegen, 1998; Jordan et al., 2018). In this study, the increase of dust scattering (e.g., through consideration of dust mineral composition) generally reduces model

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1508 biases for all variables except precipitation. <u>Future works may include satellite-based inventory of</u>

1509 <u>soil mineralogy using fully coupled Earth's system components.</u>

8 Competing interests

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1511 The contact author has declared that none of the authors has any competing interests.

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