Modeling impacts of dust mineralogy on fast climate response

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- 3 Qianqian Song¹, Paul Ginoux², María Gonçalves Ageitos^{3,4}, Ron L. Miller^{5,6}, Vincenzo Obiso⁵,
- 4 Carlos Pérez García-Pando^{4,7}
- 5 ¹ Atmospheric and Oceanic Sciences Program, Princeton University, Princeton, NJ, USA
- ⁶ ²NOAA Geophysical Fluid Dynamics Laboratory, Princeton, NJ, USA
- 7 ³ Projects and Construction Engineering Department. Universitat Politècnica de Catalunya-Barcelona
- 8 TECH, Terrassa, Spain
- 9 ⁴ Barcelona Supercomputing Center, Barcelona, Spain
- 10 ⁵ NASA Goddard Institute for Space Studies, New York, NY, USA
- ⁶ Department of Applied Physics and Applied Mathematics, Columbia University, New York, NY, USA
- 12⁷ ICREA, Catalan Institution for Research and Advanced Studies, Barcelona, Spain
- 13 Correspondence: Qianqian Song (qs7080@princeton.edu)

14 Abstract

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

45 and photochemistry. On the other hand, lumping together clay minerals (i.e., illite, kaolinite, and 46 smectite), but excluding externally mixed hematite and gypsum, appears to provide both 47 computational efficiency and relative accuracy. Nevertheless, for specific research, it may be 48 necessary to fully resolve mineralogy to achieve accuracy.

49 1 Introduction

50 Soil dust aerosols emitted from erodible land surfaces, hereafter referred to as dust, are the most 51 abundant aerosol component in the atmosphere in terms of dry mass. Dust has significant impacts on 52 the Earth's climate systems (atmosphere, ocean, cryosphere) due to its interaction with terrestrial and 53 solar radiation (Sokolik and Toon, 1999), cloud microphysics (Guo et al., 2021), tropospheric 54 chemistry (Bian and Zender, 2003; Paulot et al., 2016), and oceanic and terrestrial 55 biogeochemistry(Mahowald, 2011; Evans et al., 2019; Dunne et al., 2020). In addition, dust particles 56 deposited on snow and ice decrease surface reflectivity and accelerate snowmelt (Skiles et al., 2018; 57 Réveillet et al., 2022). Dust can influence Earth's radiative energy budget through different pathways; 58 among them: 1) directly by interacting with both solar and terrestrial radiation (i.e., direct radiative 59 effects, hereafter referred to as DRE) 2) by radiatively influencing the thermal dynamical structure of 60 atmosphere and thereby clouds (i.e., semi-direct radiative effect) and 3) indirectly by altering cloud 61 reflectivity (cloud albedo effect) and lifetime (cloud lifetime effect). Unfortunately, the quantitative 62 estimate of dust DRE at the top of atmosphere (TOA) is largely uncertain (Claquin et al., 1998; Miller 63 et al., 2014; Kok et al., 2017; Song et al., 2022). A significant part of this uncertainty has been attributed 64 to neglecting variations in dust mineralogical composition and its evolution during transport (Li et al., 65 2021).

66 The magnitude of dust impacts on the Earth's climate systems depends on its mineralogical 67 composition, as has been shown in multiple studies. In the ShortWave (SW), dust absorption depends 68 on the iron oxides content. Sokolik and Toon (1999) suggested that a small amount of iron oxides 69 internally mixed with less absorptive minerals is able to reverse the sign of DRE_{SW} at TOA from 70 negative (cooling effect) to positive (warming effect). Multiple studies have confirmed the importance 71 of iron oxides to the dust DREsw (Balkanski et al., 2007; Li et al., 2021; Obiso et al., 2023). In 72 LongWave (LW) spectrum, absorption and DRE_{LW} depend on the abundance of quartz, calcite, and 73 clays in coarse and super-coarse modes (Di Biagio et al., 2017; Sokolik and Toon, 1999). As a result, 74 resolving dust mineralogy allows to better understand the impact of dust DRE, such as the fast response 75 of the land surface temperature, as opposed to the slow response of sea surface temperature that will

not be studied here. This fast temperature response will affect precipitation, and atmospheric
circulation (Ming et al., 2010; Persad et al., 2014).

78 In addition, resolving dust mineralogy is also crucial for studying heterogeneous reactions of acid gases 79 with dust aerosols. For example, the uptake of HNO₃, NO_3^- , N_2O_5 on dust particles is suggested to be 80 limited by alkalinity that comprises calcium and magnesium carbonates (Song and Carmichael, 2001; 81 Paulot et al., 2016). These reactions will modify the composition of dust particles and subsequently 82 changing their hygroscopicity, cloud condensation nucleation (CCN), and ice nucleation activities 83 (Kelly et al., 2007), and thereby further affecting precipitation (Rosenfeld et al., 2001). Moreover, 84 heterogeneous reactions with mineral dust could significantly affect tropospheric photochemical 85 oxidation cycles, causing up to 10% reduction in O3 concentrations in dust source regions and nearby 86 (Dentener et al., 1996). Among the different minerals, K-Feldspar appears to dominate ice nucleation, 87 despite being a minor component of aeolian dust (Atkinson et al., 2013; Harrison et al., 2019), although 88 other minerals such as quartz may also contribute (Chatziparaschos et al., 2023). The key factor 89 controlling the production and removal of pollutants and the damages by acid rain is the pH of 90 raindrops, which has been observed to increase due to its dependency on Ca-rich dust (Grider et al., 91 2023).

92 Despite the potential importance of resolving dust mineralogy in various aspects, current climate 93 models tend to use a fixed mineralogy without considering the temporal and spatial variations in 94 dust mineralogical composition. To test the importance of resolving dust mineralogy on the fast 95 climate response (e.g., surface temperature response over land, atmospheric circulation and 96 precipitation response) through its interactions with SW and LW radiation (i.e., through dust DRE), 97 dust mineralogy has been implemented and simulated in the GFDL AM4.0 model (Zhao et al., 98 2018a, b), including its on-line interactions with radiation. Following the pioneering work of 99 Claquin et al. (1999) (C1999), we consider the emission, transport and interactions with radiation and deposition of eight minerals: illite, kaolinite, smectite, hematite, calcite, feldspar, quartz and 100 101 gypsum. Following the recent launch of the Earth Surface Mineral Dust Source Investigation 102 (EMIT) instrument specifically designed to retrieve global distribution of dust mineralogy over 103 dust sources (Green et al., 2020), there have been coordinated efforts to represent dust mineralogy 104 and investigate DRE of mineral-speciated dust in climate models, in particular in Li et al. (2021), 105 Gonçalves Ageitos et al. (2023), and Obiso et al. (2023). However, to the best of our knowledge, 106 there have been no studies investigating the fast climate impact of dust while accounting for its

107 mineral speciation. Our work contributes to these efforts by incorporating dust mineralogy into the 108 GFDL models, and it is distinguished by extending its investigation to the fast climate response of 109 mineral-speciated dust. The impacts of dust mineralogy on other aspects, such as sea surface 110 temperature and slow climate response, heterogeneous reactions, and ice nucleation ability, will 111 be examined in future studies.

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 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.

119 2 Model and Data

120 2.1 Model description

121 We conduct a series of experiments with GFDL AM4.0 (Zhao et al., 2018a, b) over the period 122 2001-2019. These experiments use the AMIP protocol, where sea surface temperature (SST) and 123 sea-ice are imposed based upon average monthly observations (see Gates, 1992 for details). 124 Observed gridded SST and sea-ice concentration boundary conditions are from the reconstructions 125 of Taylor et al., (2000). Historical reconstructions of monthly solar spectral irradiances are from 126 Matthes et al., (2017). For radiation calculations, global monthly mean concentrations of 127 greenhouse gases (GHGs), including nitrous oxide (N_2O), and ozone-depleting substances (ODSs, including CFC-11, CFC-12, CFC-113, and HCFC-22) are specified from Meinshausen et al., 128 129 (2017). The solar irradiances and GHG databases are standard for CMIP6. Longwave (LW) 130 scattering of aerosols is not accounted for in the model.

- 131 In AM4.0, dust emission is calculated interactively following the parameterization of Ginoux et al.
- 132 (2001) with a threshold of wind erosion and global scaling factor of 3.5 $m s^{-1}$ and 0.2 $\mu g s^2 m^{-5}$,
- 133 respectively. Dust size is represented by five bins with diameter ranging from $0.2 \,\mu m$ to $20 \mu m$
- 134 (bin1: $0.2 2 \mu m$, bin2: $2 3.6 \mu m$, bin3: $3.6 6 \mu m$, bin4: $6 12 \mu m$, bin5: $12 20 \mu m$). The

135 corresponding source fractions have been updated from 0.1, 0.225, 0.225, 0.225 and 0.225 to 136 values of 0.04, 0.14, 0.19, 0.49, and 0.14 for the five bins. These updated source functions allocate 137 more fraction to coarser size bins, following the suggested Brittle Fragmentation Theory (BFT) as 138 proposed by Kok et al. (2011). Dust mineral composition in the standard AM4.0 is considered as 139 uniform, with no temporal and spatial variations; in other words, dust Refractive Index (RI) is 140 temporally and spatially homogeneous (case referred to as homogeneous dust hereafter). The dust 141 RI in the standard AM4.0 is taken from Balkanski et al., (2007), assuming a fixed hematite content 142 of 2.7% by volume (HD27), which was calculated for the internal mixture of hematite and five 143 other minerals (calcite, quartz, illite, kaolinite and montmorillonite) using the Maxwell Garnett 144 mixing rule (see details in Balkanski et al., 2007). The decision to fix the hematite content for dust 145 particles at 2.7% was made during the development of the previous GFDL Climate Model CM3 146 (Donner et al., 2011). This decision was prompted by the discovery that dust absorption was 147 unrealistically high (by a factor 3) in CM2 (Delworth et al., 2006) compared to AERONET 148 observations (Balkanski et al., 2007). In CM3, the conjunction of a sharp decrease of black carbon 149 (strong aerosol absorber) with a new emission inventory and the switch to more scattering dust 150 had a negative effect on precipitation bias, and late 20th century warming (see Donner et al., 2011 151 for details). To mitigate this bias, the selection of 2.7% hematite was adopted in CM3, as well as 152 in the subsequent GFDL models. The control run conducted with the homogeneous dust in the 153 standard AM4.0 model is labeled as HD27 as described in Table 2.

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.

157 2.2 Dust mineralogy implementation

158 Claquin et al. (1999) (C1999) is the earliest study providing a soil mineralogy map oriented toward 159 atmospheric and climate modelling. The soil map provides the mineral mass fractions present in 160 the clay and silt size ranges for eight different minerals, namely: illite, smectite, kaolinite, calcite, 161 quartz, feldspars, gypsum, and hematite. In this study, we implement the eight minerals from the 162 soil mineralogy map provided by C1999 in GFDL AM4.0 to resolve dust mineralogy. The soil 163 map is based on soil analyses that are usually done after wet sieving, which disperse mineral 164 aggregates into small particles. This dispersal is particularly relevant for the phyllosilicates, 165 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 166 167 Garcia-Pando et al., 2016). These recent studies also show that the Brittle Fragmentation Theory 168 (BFT; Kok, 2011) represents a practical framework to generate the emitted particle size 169 distribution based on the dispersed soil PSD, which facilitates the utilization of soil mineralogy 170 maps. In our simulations, we employ BFT to reconstruct the mineral aggregates emitted from the 171 original undispersed soils, following the methods described in Goncalves Ageitos et al., 2023. The 172 mass density of the eight minerals, along with a brief description of their importance to Earth's 173 climate, are listed in Table 1. The density of minerals impacts their settling velocity, which is 174 relevant to the removal of particles in the atmosphere. Goethite and hematite are the two major 175 types of iron oxides present in soils. Goethite is less absorptive than hematite and is not resolved 176 in C1999. So, iron oxides are represented by hematite in this study. Hematite has larger density 177 than other minerals, so that hematite deposits more quickly and is not able to be transported to 178 remote regions when not aggregated or internally mixed with lighter clay minerals. Moreover, 179 among the minerals considered here, hematite is the strongest absorber at ultraviolet (UV) and 180 visible wavelengths, while it does not have noticeable absorption at infrared wavelengths (IR) 181 compared to other minerals (Sokolik and Toon, 1999). As such, the correct representation of 182 hematite content in dust aerosols is critical in improving the representation of dust interaction with 183 SW radiation in climate models. All minerals are considered to be externally mixed, except for 184 iron oxides. A large part of the emitted flux of iron oxides is considered to be internally mixed 185 with other minerals, e.g., in the form of accretions in phyllosilicates, in line with observational 186 evidence and previous modeling studies (Kandler et al., 2009; Perlwitz et al., 2015a; Zhang et al., 187 2015; Panta et al., 2023). As suggested by Gonçalves Ageitos et al. (2023), we define two different 188 types of tracers for the iron oxides: one set of tracers carries the mass of the hematite that 189 constitutes small accretions in clay minerals (i.e., internally mixed with clay minerals), are allowed 190 to be up to 5 % of the masses of their host minerals at emission (Perlwitz et al., 2015a; Gonçalves 191 Ageitos et al., 2023). Given the low fractional mass of hematite compared to their host minerals, 192 we assume that these accretions do not change the density of their host particles. These internally 193 mixed accretions form the largest fraction of the emitted hematite. Another smaller set of tracers 194 carries the mass of the remaining fraction of hematite, which is considered to be externally mixed 195 with the other minerals, including the internal mixtures of hematite with clay.

196 In addition to the similar roles of clay minerals in carrying iron oxides, the optical properties of 197 the three clay minerals are very similar, and the optical properties of their external mixture are 198 found to be almost identical to their internal mixture (see Section S1 in the Supplement). This 199 finding suggests the use of a single mineral species to represent all three clay minerals in their 200 interaction with radiation to reduce computational cost. Therefore, the optical properties of one 201 single mineral (clay433) are used to represent the optical properties of all three clay minerals. The 202 clay433 represents a mixed mineral comprising three clay minerals: illite, kaolinite and smectite, 203 with mass fraction of 40%, 30%, and 30%, respectively (see detailed descriptions in 204 Supplementary Section S1). This simplification streamlines the calculations of optical properties 205 for internal mixtures of hematite and the three clay minerals (illite, kaolinite, and smectite), 206 reducing it from an internal mixture of four minerals (hematite, illite, kaolinite and smectite) to an 207 internal mixture of two minerals (hematite and clay433).

208 The optical properties of the internal mixture of hematite and clay433 are calculated using three 209 mixing rules: volume weighted average (VOL-mixing), Maxwell-Garnett mixing rule (MG-210 mixing) and Bruggeman mixing rule (BM-mixing). Generally, VOL-mixing is used for a quasi-211 homogeneous mixture, that is when the components have similar refractive index. For cases 212 involving dominant homogeneous host with small inclusions of contrasting composition, MG-213 mixing is appropriate. BM-mixing is suitable for mixtures that the inclusions virtually occupy the 214 entire volume of the particle, and the host disappears. The detailed discussion regarding the three 215 mixing rules and their applications can be found in Liu and Daum (2008) and Markel (2016). The 216 appropriate selection of mixing rules is important for the determination of the optical properties of 217 the mixtures. Therefore, we incorporate all three mixing rules in this study. These calculations are 218 performed for various volume mixing fractions of hematite with respect to clay433, to construct a 219 lookup table (LUT) for each mixing rule. The optical properties of each mineral as well as the 220 internal mixtures of hematite and clay433 are calculated offline using Mie code with a spherical 221 shape assumption. As all other minerals have similar SW absorption, internal or external mixing 222 does not change their absorption properties. So, we assume all other minerals to be externally 223 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 five size bins. As a result, 45 mineral tracers have been incorporated in AM4.0 to account for dust

227 mineralogy.

228 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

230 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.		
4. Smectite	2570	They are internally mixed with internal hematite.		
5. 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 μm) 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).

231 2.3 AERONET Dust SSA

232 The AERONET Version 3 Level 2.0 Almucantar inversion retrievals (Giles et al., 2019; Sinyuk et 233 al., 2020) from 2000 to 2020 are screened for dust events following the methodology in Goncalves 234 Ageitos et al. (2023) and Obiso et al. (2023). This screening process aims to select dust-dominated 235 events and filter out the AERONET scenes contaminated by other absorbing aerosols. The criteria 236 that are applied to AERONET retrievals to screen dust events are: 1) hourly retrievals from 237 AERONET are considered to represent dust when the fine volume fraction is small (below 15%), 238 2) the SSA increases from 440 nm to 675 nm (a feature that distinguishes dust from other species, 239 see Dubovik et al., 2002), and 3) the mean of the imaginary index at red and near-infrared

- wavelengths (675, 870 and 1020 nm) is lower than 0.0042 (as higher values would indicate the
 presence of absorbing black and brown carbon, following Schuster et al., 2016). We calculate
 AERONET SSA in the visible by averaging AERONET retrieved SSA at two visible wavelengths
- 243 (0.44 μ m and 0.67 μ m) weighted by solar spectrum following Eq. (2).
- 244 **2.4 Laboratory Dust SSA**

245 The lab measured dust SSA at 550 nm is obtained from Di Biagio et al. (2019) (DB-2019 hereafter), 246 in which dust SSA was directly retrieved from scattering and absorption measurements. We 247 acknowledge the limits of laboratory measurements, where the dust samples are not aerosols 248 present in the atmosphere, but instead are reemitted in the lab from soil samples collected from 249 various source regions. Consequently, the laboratory measurements in DB-2019 do not account 250 for dust aerosols transported from other regions to the regions of interest. In addition, in contrast 251 to the modeled dust diameter range of 0.2 μm to 20 μm , DB-2019 measures dust particles with a 252 diameter ranging from 0.2 μm up to 10 μm .

253 2.5 CERES Data

254 To compare modeled fluxes at TOA with observations, we use the Clouds and the Earth's Radiant 255 Energy System (CERES) Energy Balanced and Filled (EBAF) Edition-4.2 data (Loeb et al., 2018). 256 The standard CERES level-3 products provide clear-sky fluxes by averaging all CERES footprints 257 within a region that are completely free of clouds. Therefore, there are many missing regions in 258 monthly mean clear-sky TOA flux maps because completely cloud-free conditions are not always 259 observed at the CERES footprint scale (~20 km at nadir). In contrast to the standard CERES level-260 3 products, CERES EBAF product infers clear-sky fluxes from clear portions of partly cloudy 261 CERES footprints thereby producing a clear-sky TOA flux climatology free of any missing regions 262 (details in Loeb et al., 2018). Starting from CERES EBAF Ed4.1, the product also provides clear-263 sky flux estimates for the total region (i.e., the total CERES footprints) by combining CERES 264 observations and radiative transfer calculations, which represents clear-sky flux with clouds 265 removed from the entire atmospheric column of CERES footprints. These clear-sky fluxes for the 266 total region are defined in a way that is more consistent with how clear-sky fluxes are represented 267 in climate models (for details see CERES EBAF Ed4.1 Data Quality Summary). In this study, 268 the monthly mean TOA 'Clear-Sky Flux Estimate for Total Region' variables in the 269 CERES EBAF Ed4.2 product, the most recent version of the product, are used to compare with 270 modeled monthly mean clear-sky flux at TOA. The comparisons allow us to examine the 271 agreement of modeled clear-sky fluxes from different experiments with observations. The 272 comparison results will be shown in section 5.1.

273 **2.6 CRU TS Data**

274 The CRU TS (Climatic Research Unit gridded Time Series) dataset provides high-resolution $(0.5^{\circ}$

275 latitude $\times 0.5^{\circ}$ longitude) climate dataset over land except Antarctica. The dataset is based on 276 extensive networks of weather stations going back to 1901(Harris et al., 2020). This dataset has 277 been widely used in various research areas since its first release in 2000. The mean 2-meter 278 temperature (TMP) and precipitation rate (PRE) variables from CRU TS v4.07 are used to evaluate 279 our model simulations. The results will be shown in section 5.2 and section 5.3.

280 **3 Experimental Design**

Table 2. List of experiments and their description. Experiments are named based on the type of dust usedor 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.	Balkanski et al. (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)

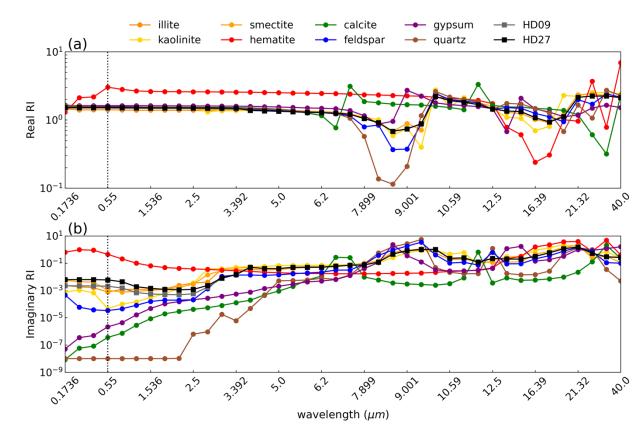
2) BM-LCRH experiment: following BM-LC,	
the tracer of external mixed hematite is	
removed, its mass is combined with internal	
mixed hematite.	
3) BM-LCRHRG experiment: following BM-	
LCRH, gypsum tracer is removed, and its	
mass is proportionally added to all other	
remaining minerals.	

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%.

The other three experiments, namely VOL, MG, and BM, resolve dust mineralogy and activate 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.

296 Each of the experiments ran for 19 years, from 2001 to 2019. We consider the 19-year runs of the 297 experiment as a group of simulations, containing 19 members of one-year simulation. The two 298 control runs (i.e., HD27 and HD09), combined with the three mineral-resolved experiments (i.e., 299 VOL, MG, and BM), form a total of six contrasting pairs. In this study, for each contrasting pair, 300 we define the anomaly as the group mean difference (based on 19-year mean) between mineral-301 resolved experiment and control run. An anomaly is considered statistically significant if the p-302 value, determined by the student's t-test between the two contrasting groups of simulations, is 303 smaller than 0.05.

- **304 4 Optical Properties**
- 305 4.1 Dust optical properties
- 306



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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.

312 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

315 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 that in our calculations, the domain averaged DAOD is always weighted by the area of each grid

325 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 μm, as shown in Eq. (1)

328 and Eq. (2).

$$\overline{DAOD} = \frac{\int_{\lambda_1}^{\lambda_2} DAOD(\lambda) B(\lambda) d\lambda}{\int_{\lambda_1}^{\lambda_2} B(\lambda) d\lambda}$$
Eq. (1)
$$\overline{SSA} = \frac{\int_{\lambda_1}^{\lambda_2} SSA(\lambda) DAOD(\lambda) B(\lambda) d\lambda}{I^{\lambda_2}}$$
Eq. (2)

$$\int_{\lambda 1}^{\lambda 2} B(\lambda) \, DAOD(\lambda) \, d\lambda$$

329 Where $B(\lambda)$ describes the solar radiation energy intensity, which can be calculated by means of

330 the Planck's function $B(T,\lambda)$, 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 (\overline{DAOD}) and single scattering albedo (\overline{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 - 0.625µm) of GFDL AM4.0, while averaged in the UV-VIS band (0.30 - 0.77µm) of GISS ModelE2.1 in Obiso et al. (2023).

Experiments		Emission (Tg/	Load (Tg)	DAOD	<u>SSA</u>
		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)	1600 (1000-3200)	20 (9-26)	0.029 (0.021 - 0.035)	-
CMIP5 ⁽²⁾		2700 (1700-3700)	17 (14-36)	-	-

CMIP6 ⁽³⁾		3472	25	0.029	-
DUSTCOMM ⁽⁴⁾		4700 (3300 - 9000)	26 (22 - 31)	0.028 (0.024 - 0.030)	-
GISS ModelE2.1 ⁽⁵⁾	НОМ	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).

338

339 The lowest SSA of HD27 in Table 3 suggests that HD27 dust, which has been used in the standard 340 AM4.0 model, is the most absorptive among all experiments. The HD09 dust is much less 341 absorptive, attributed to its smaller hematite content, as indicated by the lower imaginary part of 342 RI in the visible range (Figure 1). For the three mineral-resolved experiments, the lower global 343 mean SSA (\overline{SSA}) in VOL suggests that VOL-mixing dust is more absorptive than MG-mixing 344 and BM-mixing dust. This finding is consistent with previous studies that have suggested that 345 VOL-mixing method, when applied to minerals to compute the bulk aerosol optical properties, 346 may artificially enhance absorption relative to scattering and lead to a lower SSA for bulk dust 347 aerosol (Zhang et al., 2015). We can see that the global mean SSA (\overline{SSA}) of HD09 dust is 348 comparable to the values obtained in cases where minerals are resolved (e.g., MG and BM). This 349 implies that, from a global perspective, HD09 dust is as absorptive as mineral-resolved dust (e.g., 350 MG and BM).

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 unchanged across experiments. Besides the subtle difference (~10%) in global mean DAOD across 356 the three experiments as listed in Table 3, the global distribution of DAOD responds differently in 357 HD09 and BM. Compared to HD27, reducing hematite content to HD09 generally decreases 358 DAOD, except over the Sahel region. In contrast, resolving mineralogy as in BM decreases DAOD 359 over the Sahara region while increasing DAOD over the Sahel and Asia regions. The reduction in 360 DAOD over the Sahara region further contributes to the decrease in dust absorption over the region, 361 primarily attributed to the change in dust optical properties, such as the enhancement in dust SSA. 362 The indistinct variation in DAOD across different experiments results from the feedback of dust 363 interactions with radiation (Miller et al., 2004; Pérez et al., 2006; Miller et al., 2014), which is 364 influenced by the distinct scattering properties of dust aerosols in each experiment as shown in 365 Table 3.

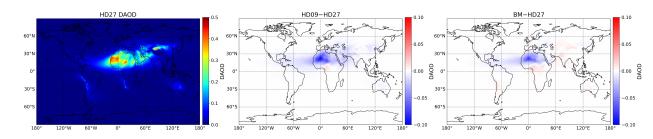


Figure 2. The global distribution of dust optical depth (DAOD) for HD27, and the difference in DAOD between HD09 and BM compared to HD27. The global mean DAOD values (\overline{DAOD}) of each experiment are shown in Table 3.

4.2 Comparison of dust optical properties with observations

366

371 Iron oxides content of dust determines shortwave radiation absorption by dust: the higher amount 372 of iron oxides, the lower the SSA. Following our calculations of dust optical properties in Section 373 4.1, we compared GFDL AM4.0 modeled dust SSA (averaged in the visible band 0.44-0.625 µm) 374 against AERONET SSA retrievals (averaged at two visible wavelength: 0.44 µm and 0.67 µm) in 375 Section 4.2.1 and laboratory measurements of SSA (at 0.55 µm) in Section 4.2.2. The modeled 376 dust SSA is evaluated against observation-based results utilizing the following evaluation metrics: 377 the mean SSA (mSSA) is calculated based on SSA for all locations displayed in Figure 3, the 378 standard deviation (σ), derived from SSA for all locations displayed in Figure 3, is used as an 379 indicator of dust SSA spatial variation; the normalized mean bias (nMB) and normalized root 380 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 381 382 are provided in Section S3 in the Supplement.

383 4.2.1 Comparison with AERONET retrievals

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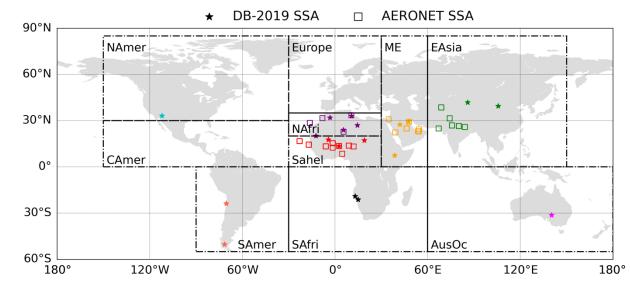


Figure 3. Dust sample locations in Di Biagio et al. (2019) and AERONET stations selected by filtering dust events. AERONET stations for SSA retrievals are in North Africa (NAfri), Sahel, the Middle East (ME) and East Asia (EAsia). Lab measurements by Di Biagio et al. (2017, 2019) expand dust sampling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

389 Figure 3 displays the AERONET stations selected by filtering dust events. The global distribution 390 of modeled dust SSA and AERONET retrieved SSA over the selected AERONET sites are shown 391 in Figure 4. There is a significant decrease in dust absorption from HD27 to HD09 globally due to 392 the reduction in hematite content. HD09 and BM exhibit similar dust absorption on a global scale 393 (e.g., the same global mean \overline{SSA}), but the regional differences are evident. For instance, compared 394 to HD09, resolving mineralogy (e.g., BM) decreases dust absorption over Iceland and Taklamakan 395 regions, while enhances dust absorption over Southern Hemisphere, particularly over Australia. 396 Additionally, there is a shift in dust absorption from the Sahara to the Sahel region after resolving 397 mineralogy.

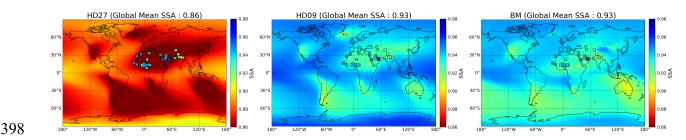
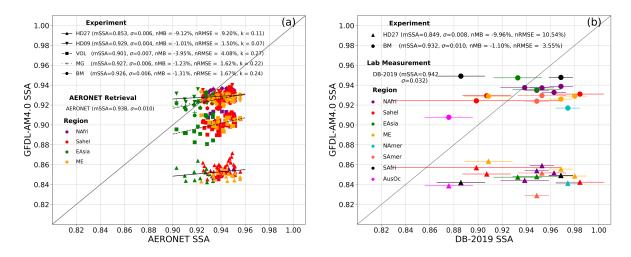


Figure 4. The 19-year (2001-2019) annual mean dust SSA simulated by AM4.0 across the three experiments.

⁴⁰⁰ The squares represent 21-year (2000-2020) annual mean AERONET retrieved dust SSA over the selected 401 AERONET sites.





403 Figure 5. GFDL AM4.0 modeled 19-year (2001-2019) averaged monthly dust SSA (average in 0.44-0.625 404 μm) versus (a) AERONET 21-year (2000-2020) averaged monthly SSA retrievals (average at two visible 405 wavelengths: 0.44 μ m and 0.67 μ m) and (b) laboratory SSA measurements (at 0.55 μ m) of dust particles 406 with diameter ranging from 0.2 μm up to 10 μm obtained by Di Biagio et al. (2019) (DB-2019). The lab 407 measurements were carried out in March 2015, horizontal error bars represent measurement uncertainties. 408 Markers represent different experiments, and colors represent different regions. mSSA in the legend 409 represents the mean SSA averaged over all locations indicated in Figure 2 (squares for AERONET, stars 410 for DB-2019). The standard deviation (σ), normalized mean bias (nMB), normalized root mean square error 411 (nRMSE), and the slope of linear regression (k) are also indicated in the legend.

412 Figure 5a shows GFDL AM4.0 modeled 19-year (2001-2019) averaged dust SSA (average in 0.44-413 0.625 µm) versus AERONET SSA (average at 0.44 µm and 0.67 µm) retrievals. Compared to 414 AERONET SSA retrievals, both HD27 and VOL overestimate dust absorption, as indicated by 415 their relatively low SSA (Figure 5a). HD27 dust is the most absorptive, indicating that the standard 416 AM4.0 dust is overly absorptive. Dust SSA in MG and BM are quite similar (i.e., mSSA: 0.926 417 versus 0.927) and show a better agreement with AERONET measurements ($nMB \approx -1.3\%$ and 418 $nRMSE \approx 1.6\%$), and they exhibit stronger scattering (i.e., higher SSA) than HD27 and VOL. 419 HD09 is almost as scattering as MG and BM, as indicated by the mSSA of 0.929 versus 0.926 and 420 0.927 in Figure 5a, which is consistent with the global mean SSA (\overline{SSA}) results shown in Table 3. 421 Overall, both the fixed mineralogy dust HD09 and mineral-resolved MG and BM dust agree well 422 with AERONET SSA retrievals, while HD27 and VOL are too absorptive. Therefore, we 423 recommend using mixing ratios of MG or BM in calculating optical properties of internal mixture 424 of hematite and clay minerals. Unless otherwise specified, in the following part of the paper, we 425 will refer to mineral-resolved experiments as MG or BM.

426 We further assess the SSA spatial variation (indicated by σ) for each experiment from AM4.0 by 427 comparing it to observation-based results. SSA is generally determined by dust mineralogy, size 428 as well as shape. Various dust mineralogy leads to distinct dust SSA due to the different absorption 429 properties of minerals (Figure 1). Coarser dust generally tends to be more absorptive (i.e., has 430 lower SSA) than finer dust when other factors are the same (Ryder et al., 2018). Spherical dust 431 assumption tends to underestimate dust SSA (Huang et al., 2023). Given the uncertainty in dust 432 shape, we assume dust particles to be spherical in this study, aligning with other model studies 433 (e.g. Gliß et al., 2021). Consequently, in the mineral-resolved experiments of this study, namely 434 VOL, MG, and BM, dust mineralogy and dust size are the two factors affecting the SSA spatial 435 variation.

436 Conversely, in homogeneous dust experiments, specifically HD27 and HD09, SSA variation is 437 solely attributed to variation in dust size, as dust mineralogy remains uniform globally. 438 Interestingly, HD09 demonstrates smaller spatial variation (i.e., lower σ) in SSA compared to 439 HD27 (Figure 5a). To investigate the impact of dust size on SSA for different hematite content 440 (e.g., HD27 and HD09), we perform a simple test in Section S4 of the Supplement. Supplementary 441 Figure S4 and S5 illustrate that the variation of SSA due to the dust particle size is more 442 pronounced with increasing absorption, i.e., from HD09 to HD27. This suggests that enhancement 443 in dust scattering relative to dust absorption (i.e., an increase in SSA) mitigates the sensitivity of 444 SSA to dust size.

445 The conclusions above provide an understanding of the SSA spatial variation (indicated by σ) 446 before (i.e., HD27 and HD09) and after (i.e., VOL, MG, and BM) implementing dust mineralogy. 447 The same σ (0.06) between HD27 and BM can be explained as follows: Because mineral-resolved 448 BM-mixing dust is overall more scattering than HD27 dust, resulting in a reduced sensitivity of 449 SSA to size, therefore, the σ of SSA caused by dust size is reduced in BM relative to HD27. 450 However, the incorporation of dust mineralogy in BM leads to an increase of σ . These contrasting 451 effects compensate for each other, resulting in the same σ . In contrast, HD09 is overall as 452 scattering as BM, as shown in Table 3and Figure 5a, suggesting a similar sensitivity of SSA to 453 size. Therefore, the incorporation of dust mineralogy in BM results in a higher σ compared to 454 HD09. Overall, while the enhancement in σ can be offset by the reduction in σ due to the reduced 455 sensitivity to dust size, resolving dust mineralogy increases σ on its own, consequently enhancing 456 the spatial variation in dust SSA.

Worth to mention, AERONET dust is quite scattering as shown in Figure 5a, therefore its SSA is less sensitive to dust size. The high σ (0.010) of AERONET SSA can be mainly due to spatial 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 (0.004), while implementing dust mineralogy (e.g., MG and BM) retains the agreement with AERONET in mean SSA and, at the same time, increases σ (0.006).

463 Besides the standard deviation (σ), the slopes (k) obtained from the statistics of modeled SSA 464 versus AERONET SSA can also indicate the regional contrast of SSA. The regional contrast of 465 AERONET SSA is well captured by the model when k is one, underestimated when k is lower than 466 one, and overestimated when k is higher than one. As such, the slopes in Figure 5a show that the 467 contrast in SSA from different regions (e.g., North Africa vs. East Asia) observed by AERONET 468 is better captured by mineral-resolved experiments (e.g., VOL, MG and BM with k ranging from 469 0.22 to 0.27) than homogeneous dust experiments (e.g., HD27 and HD09 with k ranging from 0.07 470 to 0.11). However, the modeled regional contrast of SSA in mineral-resolved experiments remains 471 overly underestimated (i.e., k is much lower than one). The significant underestimation of regional 472 SSA contrast (k) and spatial variation (σ) in AM4.0, even after accounting for mineralogy, implies 473 that something important is still missing in models. For instance: 1) the observed regional contrast 474 in iron oxides content may be higher than that in the soil map used in this study, and 2) the model 475 may have underestimated regional contrasts in the dust aerosol size distribution and thus their 476 contribution to SSA, and 3) spatial variation of dust shape, which is not taken into account in the 477 model.

478 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 485 SSA between HD27 used in the standard AM4.0 and DB-2019 (Figure 5b). Consistent with the 486 comparison with AERONET, the comparison with lab measurements suggests that dust 487 representation in the standard AM4.0 (i.e., HD27) is excessively absorptive. The smaller nMB and 488 nRMSE values in BM suggests that SSA of BM agrees better with lab measurements.

489 Moreover, regarding spatial variation (σ), resolving dust mineralogy in BM increases σ from 0.008 490 for HD27 to 0.010 for BM, even though it is still lower than the σ (0.032) in DB-2019 lab 491 measurements. Note that the variation for HD27 results from the high sensitivity of SSA to dust 492 size due to its higher absorption (as discussed in Supplementary Section S4). The inability to 493 reproduce spatial variation observed in the lab measurements is likely attributed to two aspects. 494 The first limitation is the fact that samples of DB-2019 are from soils rather than aeolian dust. 495 Aeolian dust is expected to exhibit greater uniformity in mineralogy than soils because of the 496 atmospheric mixing of dust emitted from various soil sources. The second one is associated with 497 the under-representation of regional contrast in iron oxides content in our model. Observations 498 from the EMIT are therefore essential to constrain soil mineralogy in climate models.

499 **5** Impacts of dust mineralogy on climate

500 Resolving dust mineralogy in climate models affects dust optical properties (as discussed in 501 Section 4) and their spatial and temporal variability, thereby affecting their interactions with 502 shortwave (SW) and longwave (LW) radiation. The variability in dust radiative interactions further 503 induces the fast response of land surface temperature, circulation, and precipitation. To investigate 504 the impacts of resolving dust mineralogy on climate, we need to compare modeled results in 505 mineral-resolved experiments to the baseline homogeneous dust (i.e., non-resolved mineralogy) 506 control run. As a result, the significance of the impacts depends on the selection of the baseline 507 homogeneous dust experiment. In this section, we investigate the impacts of resolving dust 508 mineralogy compared to two baseline homogeneous dust experiments. One baseline experiment is 509 the homogeneous dust used in the standard GFDL AM4.0, in which dust mineralogy is assumed 510 to be temporally and spatially uniform (i.e., non-resolved), with a volume fraction of 2.7% 511 hematite (HD27). The impacts of resolving dust mineralogy, compared to the baseline HD27, can 512 be attributed to two factors: 1) the reduction in dust absorption after resolving dust mineralogy in 513 comparison to HD27 as discussed in Section 4, and 2) spatial and temporal variations in dust 514 scattering properties induced by inhomogeneity in dust mineralogy. The other baseline experiment

- 515 is the homogeneous dust, with a volume fraction of 0.9% hematite (HD09). Given the comparable
- 516 global mean dust scattering properties (e.g., \overline{SSA}) between the control run HD09 and mineral-
- 517 resolved experiments (e.g., MG and BM), the impacts of resolving dust mineralogy, compared to
- 518 baseline HD09, is solely attributed to the inhomogeneity in dust scattering properties induced by
- 519 resolving dust mineralogy.

520 5.1 Impacts on Clear-sky Radiative Fluxes

521 We start our analysis by examining the impacts of resolving dust mineralogy on clear-sky radiative 522 fluxes. By 'clear-sky', we mean that our results do not consider the radiative effects of clouds. We 523 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 524 fluxes with resolved mineralogy, F_2^{\uparrow} is the 19-year mean clear-sky upward radiative fluxes for 525 526 homogeneous dust. Section S5 in the Supplement provides the clear-sky radiative fluxes anomalies 527 at TOA and surface (SFC) induced by resolving dust mineralogy over the global scale, we see 528 much more significant anomalies over the North Africa than other regions, which makes senses 529 because that dust aerosol is the most dominant aerosol species in this area. The changes in dust 530 aerosol optical properties have a greater potential to lead to significant impacts on radiation and 531 climate over the region than in the others. Therefore, this section focuses on the North Africa 532 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 533 534 specifically analyze the results for the June-July-August (JJA) season when dust loading is at its 535 highest and the West African Monsoon is the strongest.

536 5.1.1 Impacts on Clear-sky Radiative Fluxes relative to HD27

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 relative to HD27, it's worth recapping that the effects of resolving mineralogy relative to HD27 can be attributed to two factors: the reduction in dust absorption and the variation in dust scattering properties induced by the mineralogical inhomogeneity.

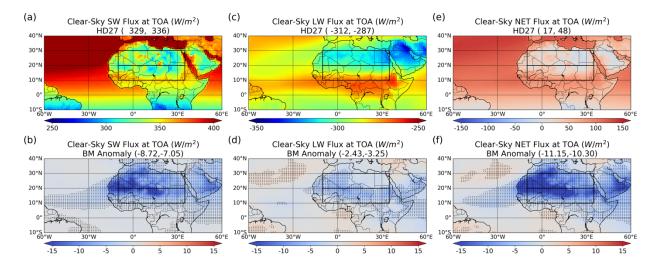
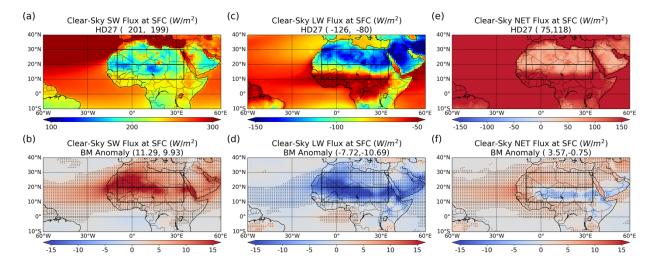




Figure 6. Seasonal mean JJA climatology (2001-2019) clear-sky SW (1st column), LW (2nd column) and Net (3rd column) radiative flux at TOA for the HD27 control run (1st row) and their anomalies resulting from resolving dust mineralogy in Bruggeman-mixing experiment (2nd row). Downward direction is defined 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.

548 The first row in Figure 6 illustrates the modeled clear-sky shortwave (SW), longwave (LW) and 549 net (NET: the combination of SW and LW) radiative flux at TOA from the HD27 control run. 550 Relative to HD27, mineral-resolved dust (e.g., BM-mixing dust) generally reflects more SW 551 radiation back to space and induces negative SW flux anomalies at TOA (Figure 6a, b; Positive: 552 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 553 554 combining both SW and LW, resolving mineralogy turns out to induce substantial decrease in NET 555 flux at TOA, with a more than 50% negative anomaly over the Sahara and around a 20% negative

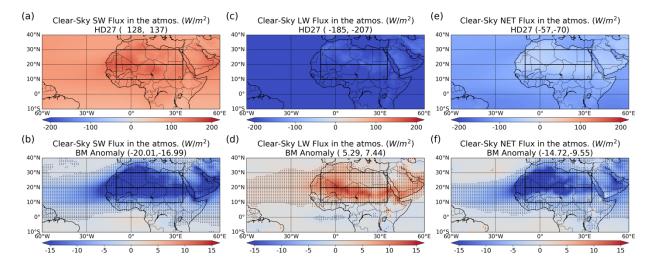
- anomaly over the Sahel (see values in parentheses in Figure 6e, f). Therefore, less NET radiation
- 557 reaches the Earth at TOA in the mineral-resolved dust cases due to their lower absorptivity.





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

560 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: 561 562 downward). In the LW, the cooling of the mineral-resolved dust layer, due to its low absorption, 563 results in less LW radiation being emitted toward Earth's surface. This reduction in the downward 564 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 565 566 approximately canceled out by the negative anomalies in LW radiation (Figure 7). As a result, a 567 similar amount of radiation reaches the Earth's surface in both HD27 and mineral-resolved cases. 568 Despite less NET radiation entering the Earth at TOA in mineral-resolved cases, the similar 569 amount of NET radiation reaching the Earth's surface indicates that less NET radiation is absorbed 570 in the atmosphere in mineral-resolved cases.



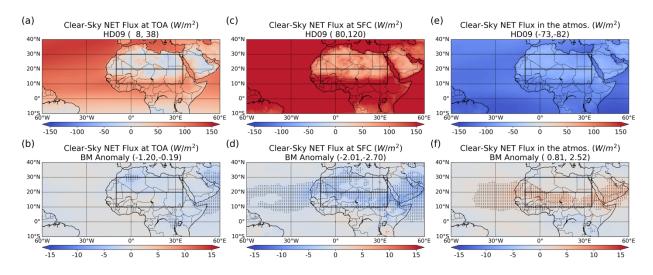
571

- 572 Figure 8. As in the Figure 6, but for the radiative flux absorbed in the atmosphere.
- 573 In Figure 8, the negative SW flux anomalies are partially offset by positive LW flux anomalies,

574 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

- 576 parentheses in Figure 8b, d, f).
- 577 5.1.2 Impacts on Clear-sky Radiative Fluxes relative to HD09
- 578 Before comparing mineral-resolved experiments with HD09 control run, where the homogeneous 579 dust is as absorptive as mineral-resolved dust (e.g., MG and BM) from a global perspective, to 580 understand their impacts on clear-sky radiative fluxes relative to HD09, it's worth recapping that 581 the effects of resolving mineralogy relative to HD09 are primarily attributed to the variation in 582 dust scattering properties induced by the mineralogical inhomogeneity.



⁵⁸³

Figure 9. As in the Figure 6, but for HD09 control run. In addition, SW and LW flux anomalies are not shown here. Clear-sky net flux at TOA (1st column), at surface (2nd column), and in the atmosphere (3rd column) are shown in this figure.

587 **Error! Reference source not found.** Figure 9 shows the clear-sky fluxes anomalies with respect t 588 o HD09 over North Africa, the anomalies over the global scale are shown in Figure S7, S9, S11 589 and S13 in the Supplement.

- 590 In contrast to the anomalies with respect to HD27 control run, resolving dust mineralogy does not
- 591 cause substantial anomalies (< 5%) in clear-sky fluxes with respect to HD09 control run. This can
- 592 be attributed to their similarity in dust scattering properties from a global mean perspective,
- 593 particularly SSA as shown in Figure 5. The comparable effects of HD09 and mineral-resolved dust

594 on radiation suggest that resolving dust mineralogy does not have significant impacts on clear-sky 595 fluxes when homogeneous dust is as scattering as mineral-resolved dust aerosols on a global scale. 596 However, the equivalence between HD09 and mineral-resolved dust in terms of their interactions 597 with radiation may be related to the three limitations in the current model simulations: 1) Soil 598 mineralogy: The limited soil mineralogy database fails to adequately capture the regional variation 599 of iron content (or SSA) within the region; 2) Dust emission based on Ginoux et al. (2001) uses a 600 continuous function of topography, which does not take into account geomorphological 601 characteristics of the surface to differentiate soil properties of dust sources as done by others 602 (Zender et al., 2003; Bullard et al., 2011); 3) Dust transport: Excessive numerical diffusion may 603 occur when solving advection equation (Ginoux, 2003). Given all those limitations of our model 604 simulations, this finding may differ with improved representation of dust sources and transport. 605 Such improvement may come from spaceborne soil mineralogy dataset (e.g., EMIT) that may 606 capture accurately the regional contrasts in iron oxides content.

607 5.1.3 Compare Clear-sky Radiative Fluxes with CERES Observations

608 Furthermore, we conduct a comparison of modeled SW upward, LW upward, NET downward flux 609 at TOA with observation-based results from CERES EBAF Ed4.2 product (see Table 4). The 610 difference between modeled flux and CERES observations are listed in parentheses within the title 611 of each figure in Table 4. Compared to HD27, the more scattering HD09 and mineral-resolved BM 612 achieve much better agreement with CERES observations in clear-sky flux (i.e., SWup, LWup and 613 NETdn) at TOA. This is evident in the smaller values of HD09 – CERES (e.g., NETdn: 1.6 for the 614 Sahara and 2.4 for the Sahel) and BM – CERES (e.g., NETdn: 0.4 for the Sahara and 2.1 for the 615 Sahel) compared to HD07 - CERES (e.g., NETdn: 11.3 for the Sahara and 12.4 for the Sahel), as 616 shown by the values in parentheses in Table 4. Between HD09 and BM, BM tends to agree slightly 617 better with CERES.

Table 4. Comparison of modeled clear-sky SW upward (SWup, 1st row), LW upward (LWup, 2nd row) and 618 NET downward (NETdn, 3rd row) fluxes at TOA with CERES observation-based results over 2001-2019 619 620 JJA. The 1st column shows the clear-sky flux estimates at TOA from CERES EBAF Ed4.2 product, which 621 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 622 column) experiments from CERES observations. The two values in parentheses represent domain average 623 624 for the Sahara and Sahel regions as indicated in figures in Section 5.1.2. Specifically, the first column 625 (CERES) is domain averaged flux, while the second (HD27 – CERES), third (HD09 – CERES), and fourth 626 (BM - CERES) columns are domain averaged flux differences between model and CERES observation-627 based results.

	CERES	HD27 – CERES	HD09 – CERES	BM – CERES
Clr SWup flux at TOA (W/m ²)	(135, 113)	(-9.3, -9.4)	(-1.1, -2.2)	(-0.6, -2.3)
Clr LWup flux at TOA (W/m ²)	(314, 291)	(-1.4, -3.1)	(-0.6, -0.5)	(0.2, -0.1)
Clr NETdn flux at TOA (W/m ²)	(6, 36)	(11.3, 12.4)	(1.6, 2.4)	(0.4, 2.1)

629 5.2 Impacts on land temperature

630 Here we explore the impacts on the temperature vertical profile and near-land surface temperature 631 relative to HD27 and HD09, respectively. Compared to the HD27 control run, lower absorption of 632 radiation in the atmosphere by mineral-resolved dust aerosols results in statistically significant 633 negative temperature anomalies in the atmosphere ranging from 800 mb up to 500 mb where dust 634 aerosols are mainly located (Figure 10). In contrast, there is no statistically significant temperature 635 anomaly for mineral-resolved dust cases compared to HD09, as illustrated by the red curves in 636 Figure 10 This finding aligns with the insubstantial anomalies (<5%) in clear-sky NET radiative 637 fluxes discussed in Section 5.1.2. In the subsequent part of the section, we will delve into 638 comparing mineral-resolved experiment (using BM as an example) with the HD27 control run. 639 This comparison will help us further understand the impact of dust aerosols with distinct 640 absorption on land temperature.

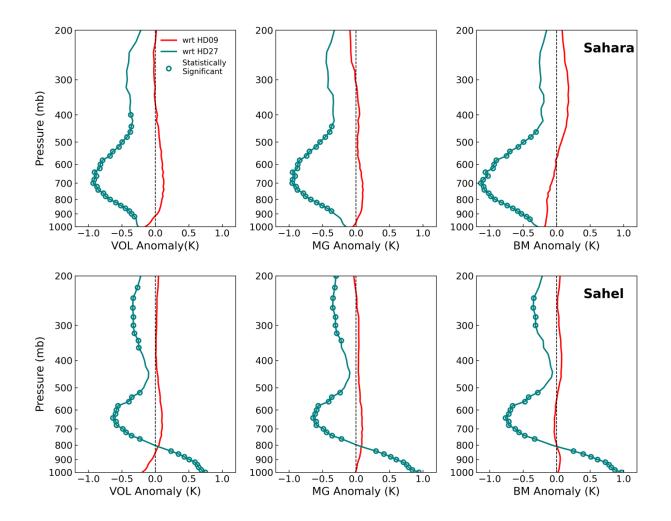


Figure 10. Vertical profile of temperature anomaly induced by resolving dust mineralogy for the Sahara (1st row) and the Sahel (2nd row) regions in the three mineral-resolved experiments (i.e., VOL, MG, BM). Green lines represent temperature anomalies with respect to HD27 control run. Red lines are temperature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

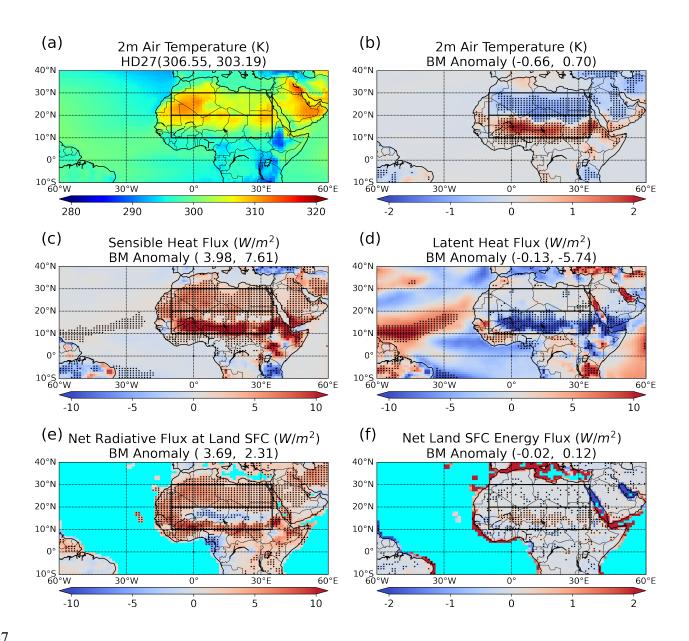


Figure 11. Air temperature at 2-meter from HD27 control run (a), anomaly (b) induced by implementing 648 649 Bruggeman-mixing minerals in BM experiment, surface sensible heat flux (c), latent heat flux (d), net 650 radiative flux (e), net energy flux (f) anomalies between BM and HD27; Upward flux is positive in (c) and 651 (d), while downward flux is positive in (e) and (f). Net energy flux (f) is the subtraction of (c), (d), and 652 downward ground flux from (e). Note that ground flux is not shown in the figure considering its relatively 653 small magnitude, but it is included in the land surface net energy flux calculations in subplot (f). The dotted 654 area denotes anomalies that are statistically significant. The two values in parentheses within the title of 655 each figure are domain average for the Sahara and Sahel regions.

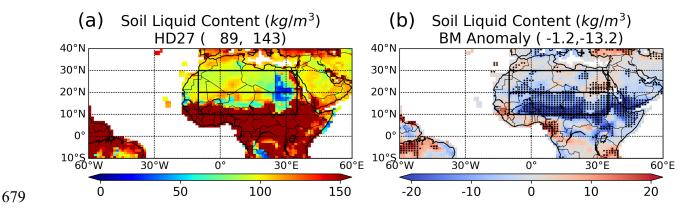
656 Figure 11a shows air temperature at 2-meter from HD27 control run over the Northern Africa.

657 Near the land surface, more scattering mineral-resolved dust induces a temperature decrease (i.e.,

658 negative temperature anomaly –0.66 K) over the Sahara and a temperature increase (i.e., positive

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.

Radiative flux perturbation over land is quickly equivalented by balancing surface radiative fluxes 661 662 with sensible heat flux, latent heat flux and ground flux (i.e., downward heat flux into the ground), 663 which results in nearly zero net energy flux at land surface as shown in Figure 11f. Precisely, the 664 radiative flux anomaly comprises two contributions: one is the instantaneous radiative forcing (IRF) 665 caused by the change in dust mineralogy in the atmosphere, and the other one is the associated 666 radiative feedbacks. For simplicity, we will not partition the radiative flux anomaly in our 667 discussion here. Over the Sahel region, the positive net radiative flux anomaly at land surface is 668 balanced out by the increased sensible heat flux and the decreased latent heat flux as well as ground 669 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 670 671 the Sahel in BM case (Figure 11d) is due to the depletion of soil moisture (and therefore 672 evaporation) in the region as shown in Figure 12. The depletion of soil moisture is caused by the 673 decrease in moisture carried by onshore winds over the Sahel and the decrease in precipitation 674 over the same region, as will be discussed in section 5.3. Therefore, a large enhancement of sensible heat flux (~7.6 W/m^2) is needed (Figure 11c) not only to compensate for the depletion 675 in latent heat flux (~5.7 W/m^2 in d), but also to balance out the increased net radiative flux (~2.3 676 W/m^2 in Figure 11e). As a result, higher land surface temperature with anomaly around 0.7 K is 677 678 needed in the region to achieve the required sensible heat flux enhancement.



680 Figure 12. Soil liquid content in HD27 control run (a) and anomaly resulting from implementing 681 Bruggeman-mixing minerals in the BM experiment (b). The two values in parentheses within the title of 682 each figure are domain average for the Sahara and Sahel regions.

683 Over the Sahara region, latent heat flux does not change from HD27 case to BM case, therefore, 684 the increased net radiative flux (~3.69 W/m^2) in BM compared to HD27 is mainly balanced out by the enhanced sensible heat flux (~3.98 W/m^2) which requires a larger temperature gradient 685 686 between surface and atmosphere. However, there is a very strong negative temperature anomaly 687 (around -1K) in the atmosphere near 700 hPa due to less dust absorption in BM as we discussed 688 in Figure 10. The strong negative temperature anomaly in the lower atmosphere effectively 689 increases the vertical temperature gradient. As such, it is not necessary for the land surface 690 temperature to increase; in fact, it may need to decrease by approximately 0.66 K to achieve the 691 desired enhancement in sensible heat flux and reach equilibrium.

692 Additionally, to assess the effectiveness of various dust scattering properties (e.g., HD27, HD09, 693 and BM) in matching observations of near-surface temperature, we compare the modeled near 694 surface temperature (T_{2m}) with CRU TS observations, which is described in Section 2.6, over the 695 Sahara and Sahel regions (Table 5). Considering the relatively large inter-model spread of regional 696 surface air temperature, we compare the Sahara-Sahel regional contrast in surface air temperature 697 to the CRU rather than comparing their absolute values. Table 5 shows that HD09 and BM improve 698 the agreement with CRU in Sahara-Sahel temperature contrast compared to HD27, and BM 699 exhibits the closest agreement with CRU.

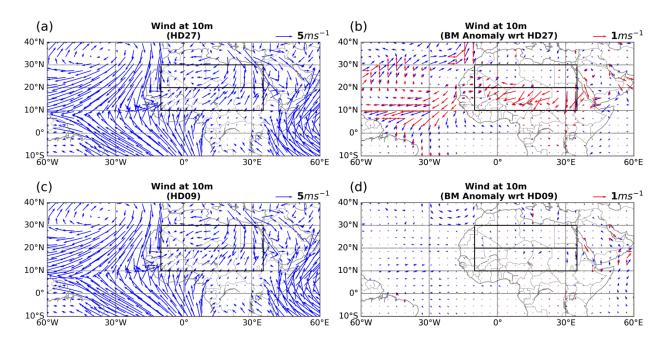
Table 5. The 19-year (2001~2019) JJA mean 2-meter Air Temperature (T_{2m} , unit: K) and their standard deviation over the 19 years from CRU observations and modeled experiments over the Sahara and Sahel regions. The 'Contrast' row indicates the T_{2m} regional contrast between the Sahara and the Sahel.

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

703

704 **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 (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.



710

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.

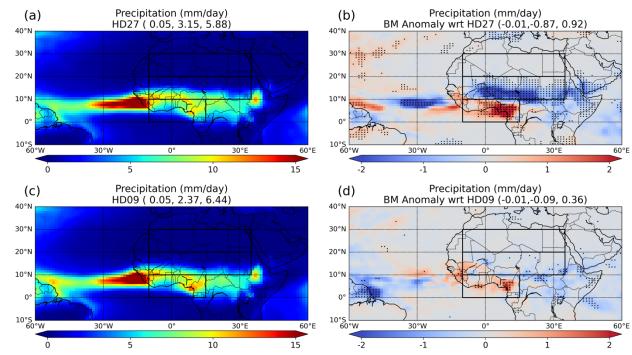
Global precipitation is higher by 0.017 mm/day for the BM mineral-speciated case compared to HD27 experiment. This is consistent with the lower SW absorption in the former, given the global compensation between latent heating associated with precipitation and net radiative cooling (e.g., Allen and Ingram, 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,

thereby enhancing upward motion. The rising warm air spawns a large-scale onshore flow,

727 carrying the low-level moist air from the Atlantic to the Sahel, thus enhancing precipitation over 728 this region (Balkanski et al., 2021). The more scattering mineral-resolved BM and MG dust absorb 729 less radiation and cause less warming of the atmosphere (Section 4.1), reducing adiabatic cooling 730 through ascent and Sahel precipitation. The suppressed ascent in BM compared to HD27 is 731 associated with a reduction both in the wind divergence aloft and in convergence at the surface 732 (Figure 13b). The reduction in convergence results in northeast wind anomalies at 10-meter over 733 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 734 735 Sahel, is consistent with the reduction of ascent and precipitation over this region (Figure 14a, b).



⁰ ⁵ ¹⁰ ¹⁵ ⁻² ⁻¹ ⁰ ¹ ²
⁷³⁶ 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 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
in reduced moist static energy and buoyancy, limiting convection to the coastal region.

These changes in precipitation have non-negligible effects on soil moisture content in North Africa due to its moisture-starved environment. The decrease in precipitation over the Sahel in BM leads to a reduction in soil moisture content. Conversely, the increase of precipitation over the GC leads to increases of soil moisture (Figure 12). The change in soil moisture content further affects the partitioning of surface energy fluxes and the efficiency of the latent heat flux, thereby affecting land surface temperature, as illustrated by Figure 11.

So far, we have been focusing on discussing the impacts of resolving dust mineralogy on winds and precipitation relative to the HD27 control run. The large discrepancy in optical properties between HD27 and mineral-resolved dust allows us to better understand how distinct dust absorption impacts our climate through its distinct radiative effects.

As discussed in section 4.1, HD09 dust is nearly as scattering as mineral-resolved dust but exhibits smaller regional variability. Section 5.1.2 shows that resolving dust mineralogy does not lead to statistically significant anomalies on radiation relative to HD09. Consistently, there are no further statistically significant impacts on winds (Figure 13c, d) and precipitation (Figure 13c, d).

763 To investigate the effectiveness of various dust scattering properties (e.g., HD27, HD09, BM) in 764 matching observations of precipitation rate, we compare the modeled precipitation with CRU TS 765 observations over the Sahara, Sahel and GC regions (Table 6). The greater difference between 766 HD09, BM and CRU (i.e., HD09 - CRU and BM - CRU) indicate that more scattering HD09 and 767 BM lead to a larger discrepancy between the modeled precipitation and CRU observations. In 768 contrast, Balkanski et al. (2021) describes the same balance of increased dust absorption and Sahel 769 precipitation but find improved agreement with Global Precipitation Climatology Project (GPCP) 770 data by assuming homogeneous dust containing 3% iron oxides by volume. Contrasts between that 771 study and ours result from differences between the GPCP and CRU data sets, contrasts in dust absorption (related to contrasts in the dust size distribution or assumed index of refraction), non-772 773 dust model biases in precipitation or differences between the slow response computed by 774 Balkanski et al. (2021) and the fast response that we calculate. The fast and slow response even

- exhibit differences in the sign of the calculated precipitation anomaly within some regions of the
- 776 WAM (Miller and Tegen, 1998; Jordan et al., 2018).

777 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

780 between HD27 control run and CRU observations, along with the corresponding std of this 19-year

781 difference. Similar for HD09 – CRU and BM – CRU.

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

782

783 6 Potential for reducing mineral tracers

784 Thus far in this study, we have been using 45 mineral tracers in mineral-resolved experiments (i.e., 785 VOL, MG, and BM). However, it is important to investigate the potential of reducing the number 786 of mineral tracers in climate models to lower computational costs. In this section, we take BM as 787 a reference for providing the best comparisons with CRU temperature and CERES flux 788 observations, and conduct an experiment named BM-RT to assess the possibility of reducing 789 mineral tracers in BM. The BM-RT experiment consists of three sub-experiments, namely, BM-790 LC, BM-LCRH, and BM-LCRHRG. In each of the three sub-experiments, the number of mineral 791 tracers is progressively reduced, allowing for an examination of the relative impacts of different 792 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). Therefore, in the first sub-experiment BM-LC (where 'LC' represents 'Lump Clay minerals'), we lump together the three clay minerals as one mineral species 'clay433'.

800 Based upon the C1999 soil mineral composition that we use, externally mixed hematite is mainly 801 concentrated over the Sahel region (Ginoux et al. 2023, in preparation) and cannot be transported 802 to remote regions due to its high mass density. Obiso et al. (2023) shows that visible extinction 803 due to externally mixed hematite is negligible compared to other mineral components including 804 hematite internally mixed with other minerals. Thus, we further remove external hematite tracers 805 in the second sub-experiment BM-LCRH (where 'RH' indicates 'Remove externally mixed 806 Hematite'). The mass fraction of external hematite is combined with internal hematite to ensure 807 that the total mineral fraction at emission remains equal to one. In this sub-experiment, the number 808 of mineral tracers is reduced from 35 in BM-LC to 30 in BM-LCRH.

Since there are no known specific impacts of gypsum on climate, we conducted the third subexperiment, BM-LCRHRG ('RG' indicates 'Remove Gypsum'), where gypsum was removed. The mass fraction previously attributed to gypsum at emission, which is very low at the global scale, was proportionally redistributed among all other minerals. The number of mineral tracers is finally reduced from 30 in BM-LCRH to 25 in BM-LCRHRG.

We analyze the 19-year (2001-2019) time series of total dust mineral emission before and after reducing mineral tracers in Figure 15. We observe subtle differences in total mineral emission between experiments, which arises from the feedback of mineral radiative interactions. However, these differences are numerically small, and Student's t-test suggests that the time series of the four experiments are not statistically different. Additionally, the globally averaged DAOD and SSA of each sub-experiment remains highly similar to those of the reference experiment BM, as listed in Table 3.

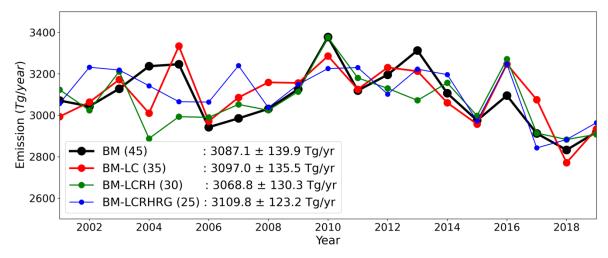


Figure 15. Time series of total dust mineral emission from 2001 to 2019 before and after reducing the number of mineral tracers. The legend displays the following information: 'Experiment name (number of mineral tracers): 19-year averaged total mineral emission \pm 19-year standard deviation of total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

826 Given the highly similar optical properties of minerals before and after reducing mineral tracers, 827 we further investigate their impact on climate. Firstly, we examine the clear-sky flux anomaly of 828 each of the three sub-experiments at TOA and surface relative to the reference experiment BM, as 829 shown in Supplementary Figure S14 for TOA and Supplementary Figure S15 for surface. We only 830 observe a few statistically significant (p-value < 0.05) anomalies over the North Africa, suggesting 831 that the reduction of mineral tracers in the three sub-experiments has a weak impact on radiation. 832 Furthermore, we investigate the anomaly in temperature profile, surface winds, and precipitation 833 of each of the three sub-experiments relative to the reference experiment BM. The results are 834 presented in Figure S16-S18 in the Supplement. No statistically significant (p-value < 0.05) 835 temperature anomalies (Figure S16) and surface wind anomalies (Figure S17) is observed. Only a 836 few statistically significant anomalies in precipitation are observed in the Supplementary Figure 837 S18. These results suggest that the reduction of mineral tracers in the three sub-experiments has a

838 weak impact on climate.

821

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.

844 7 Conclusions

We simulate the distribution of dust mineralogy (i.e., illite, kaolinite, smectite, hematite, calcite, feldspar, quartz, and gypsum) and activate their interaction with radiation in the GFDL AM4.0 model. Our investigation focuses on the radiative impacts of resolving dust mineralogy on Earth's atmosphere and its fast response of land temperature, surface winds and precipitation.

849 We set up two baseline homogeneous dust control runs: HD27 and HD09, in which dust 850 mineralogy is considered as temporally and spatially uniform, the former following the standard 851 configuration for the dust optical properties in GFDL AM4.0 and the latter including a more 852 scattering dust. Three experiments with resolved mineralogy are also conducted: VOL, MG, and 853 BG, using three different mixing rules for the internal mixture between hematite and clay minerals 854 (i.e., volume weighted mean, Maxwell Garnett, and Bruggeman). The comparison of dust 855 absorption properties (e.g., SSA) with observation-based results suggests that the homogeneous 856 dust used in the standard GFDL AM4.0 (i.e., HD27) is overly absorptive, Maxwell Garnett and 857 Bruggeman mixing rules are more appropriate than volume weighted mixing rule in calculating 858 optical properties of internal mixtures of hematite and clays. Compared to HD27, the homogeneous 859 dust with reduced hematite content (HD09) and mineral-resolved dust (i.e., MG and BM) exhibit 860 much better agreement with AERONET retrievals and laboratory measurements in terms of dust 861 absorption properties (i.e., SSA). Additionally, resolving dust mineralogy enhances regional 862 variability in dust SSA compared to homogenous dust, further improves the agreement with 863 AERONET, even though it remains lower than the observed variability.

864 The two homogeneous dust control runs, HD27 and HD09, with distinct dust absorption properties, 865 allow us to investigate the impact of dust mineralogy on Earth's radiation and fast climate response 866 relative to distinct baseline homogeneous dust. In comparison to HD27, resolving mineralogy 867 reduces dust absorption. During JJA, the reduced dust absorption results in a reduction of over 50% 868 in NET downward radiation across the Sahara and approximately 20% over the Sahel at TOA. 869 Additionally, there is a reduction of around 25% in the atmospheric absorption of radiation over 870 the Shahara and around 10% over the Sahel in the atmosphere. The reduced surface absorption of 871 radiation by mineral-resolved dust leads to a temperature decrease of 0.66 K at the land surface 872 across the Sahara and an increase of 0.7 K over the Sahel. The reduced NET downward radiation 873 at TOA, attributed to the less absorption of radiation by mineral-resolved dust, suppresses ascent 874 and weakens the monsoon inflow from the Gulf of Guinea. This brings less moisture to the Sahel, 875 which combined with decreased ascent induces a reduction of precipitation. On the other hand, 876 compared to HD09, resolving dust mineralogy results in dust absorption comparable to that of 877 HD09 on a global scale. However, when resolving mineralogy, there is an increase in spatial 878 variation of dust absorption. Additionally, we observe a noticeable change in global distribution 879 of dust absorption, with more dust absorption distributed in the Southern Hemisphere and lower 880 dust absorption over Iceland and Taklamakan regions. Nevertheless, the higher spatial variation in dust absorption does not lead to statistically significant changes in any of the climate aspects 881 882 mentioned above. The models with reduced absorption (HD09 and fully resolved mineralogy) 883 improve the comparison with observations of CERES fluxes and CRU land surface temperature. 884 We see a slightly better agreement with observations for fully resolved mineralogy than HD09 885 however it is not statistically significant. As such, when using fixed mineralogical composition, 886 we recommend using a 0.9% hematite content in volume, which represents the lowest of the three 887 hematite mixings considered by Balkanski et al. (2007).

888 Historically, climate models have relied on fixed refractive index to consider dust radiative forcing 889 starting (IPCC, 2001) with strongly absorptive value based on dust samples in Sahara (Patterson 890 et al., 1977) to more scattering values after dust absorption could be inferred from satellite and 891 surface observations (e.g., Sinyuk et al., 2003; Balkanski et al., 2007). With the launch of EMIT 892 in July 2022 and the expected delivery of a high-resolution map of soil mineralogy in source areas, 893 dust interactions with radiation in climate models will be calculated directly from the simulated 894 mineralogical composition (Li et al., 2021). Still, the additional burden of simulating a dozen 895 minerals may be too prohibitive for large ensemble climate models simulations. In such cases, our 896 analysis suggests the use of a fixed value providing similar radiative effects as the comprehensive 897 representation of minerals. However, our recommendation is directed toward the GFDL AM4.0 898 model with all its uncertainties related to mineral distribution, emission sources, and aerosol 899 transport. Moreover, incorporating dust mineralogy in models is likely to be important in other 900 aspects, such as cloud properties, ocean biogeochemistry, air quality and photochemistry. For 901 studies with resolved mineralogy, we show that the number of mineral tracers can be reduced from 902 45 to 25 without losing the quality of comparison with observations of CERES fluxes and CRU 903 surface temperature. Such reduction can be achieved by lumping together clay minerals, removing 904 external hematite and gypsum. For specific research such as biogeochemistry, it may be necessary905 to fully resolve mineralogy to achieve accuracy.

906 This study has some limitations. First, the soil mineralogy map from C1999 is based on extensive 907 extrapolation and limited observations. In terms of the need to improve knowledge of soil 908 mineralogy in dust source regions, the launch in July 2022 of the EMIT instrument operating from 909 the International Space Station will provide mineral identifications of dust sources using 910 hyperspectral measurements (Green et al., 2020). The EMIT soil map measurements will improve 911 resolving dust mineralogy in climate models and advance our understanding of dust's effects in 912 the Earth system. Second, hematite and goethite are the most common iron oxides present in soils. 913 However, goethite is not considered in this study because not included in the used soil mineralogy 914 map. Previous studies suggest that goethite is generally more abundant than hematite, but it is less 915 absorptive than hematite in the visible spectrum (Formenti et al., 2014). Therefore, the abundance 916 of iron oxides may be underestimated in this study, which may lead to underestimation of dust 917 absorption in the SW. A more recent database by Journet et al. (2014) (J2014) includes the 918 distribution of goethite but it shares many limitations as C1999 (e.g., extensive extrapolation) and 919 has other major disadvantages, such as numbers of missing soil fractions of some minerals at some 920 locations. Third, the refractive index of hematite used in our study is close to the upper range of 921 the values available in literature (Zhang et al., 2015). Hence, the last two limitations, 922 underestimation of iron oxide content and overestimation of absorption by hematite, may have 923 compensating effects.

924 This study, by prescribing SST, calculates only the fast response to the dust DRE, without 925 including the slow response by the sea surface temperature. This avoids the need to spin-up the 926 model for decades before reaching new equilibrium but may overestimate the eventual response, 927 as shown by Miller and Tegen (1998) and Balkanski et al. (2021). This complicates model 928 evaluation because the observations include the slow response to dust. Variables like precipitation 929 are especially sensitive to the inclusion of the slow response because prescribed SST experiments 930 omit the surface energy balance over the ocean. Thus, the surface DRE beneath the aerosol layer, 931 which is generally negative, is not fully balanced by a fast reduction of evaporation (Miller et al., 932 2004a). The addition of the surface balance in the slow response can reverse the sign of the fast 933 precipitation anomaly (Miller and Tegen, 1998; Jordan et al., 2018). In this study, the increase of 934 dust scattering (e.g., through consideration of dust mineral composition) generally reduces model

- 935 biases for all variables except precipitation. Future works may include satellite-based inventory of
- 936 soil mineralogy using fully coupled Earth's system components.

937 8 Competing interests

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

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