Modeling Large Dust Aerosols in the Community Earth System Model

2 Version 2 (CESM2)

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Key Points:

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- A new model is developed to simulate dust aerosols for a wide distribution of sizes with observational constraints and improved mineralogy
- Including particles larger than 10 microns in diameter greatly alters simulated dust budgets while not degrading finer dust representation
- Challenges remain, especially for representing the physical mechanisms that transport super coarse dust in Earth system models

Abstract

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Dust aerosols have a wide size distribution from less than 1.0 nm to over 100 µm and dominate the Earth's atmospheric aerosol mass. However, most Earth system models inadequately represent dust aerosols larger than 10 µm in diameter, limiting the accuracy of dust cycle and climatic impact simulations. Here, we introduce a new modeling framework that captures the observed full-size distribution of dust aerosols, incorporating recent advances into a mineralresolved version of the Community Earth System Model and addressing known issues in previous versions. Comprehensive evaluation against diverse observations of bulk dust and component minerals demonstrates that the model reproduces the observed dust cycle across sizes. Incorporating the previously unrepresented fractions substantially alters dust budget estimates, highlighting potential changes in simulated climatic impacts and underscoring the importance of comprehensive size-resolved dust modeling. Despite these advancements, uncertainties persist. Future developments should address misrepresented physical processes that hinder accurate modeling of the transport of dust aerosols larger than 10 µm in diameter, whose gravitational settling velocity must be reduced by 60% in the new model to match observed size distribution. Expanding observational datasets covering the full-size distribution is also essential to better constrain the dust cycle and improve the representation of dust optical properties and climatic effects.

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Plain Language Summary

56 Mineral dust constitutes the largest mass of airborne particles on Earth. These particles span a 57 wide range of sizes from tiny to giant, but not all sizes play an equally important role. As particles that both scatter and absorb solar and thermal radiation while delivering vital nutrients 58 to ecosystems, mineral dust plays a critical role in various Earth system processes and in 59 60 projections of future climate changes. However, most Earth system models (ESMs), including 61 the Community ESM Version 2 (CESM2), either neglect or underrepresent particles large than 10 microns in diameter, limiting our understanding of dust's influence on climate. Furthermore, 62 the CESM2 dust module, including its mineral-resolved version used in this study, lacks several 63 key physical processes known to affect dust emission and transport. This study enhances the 64 representation of dust aerosols in the mineral-resolved version of CESM2 by extending the upper

- size limit of simulated dust particles to 70 microns at emission and adding omitted physical
 processes for dust emission based on the latest scientific findings. The updated model is
 evaluated against a wide range of observations under present-day climate conditions.

 1 Introduction
 Mineral dust, the most abundant aerosol by mass (Gliß et al., 2021), has diverse climatic impacts
 (Kok et al., 2023) through direct interactions with radiation, indirectly modifying cloud
 properties, and by depositing micronutrients (e.g., iron or phosphorus) to marine and terrestrial
- impacting human health (Goudie, 2014; Maki et al., 2022; Pérez García-Pando et al., 2014).

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79 models capable of reproducing the spatiotemporal life cycle and physicochemical properties of

Accurately quantifying these impacts at large (e.g., continental and global) scales requires

dust aerosols, which are sensitive to dust size (Mahowald et al., 2014). Such capability, however,

ecosystems (Jickells et al., 2005; Mahowald, 2011; Mahowald et al., 2010, 2018; Mccutcheon et

al., 2021; Meskhidze et al., 2005). Dust influences weather and climate, especially near source

regions, and acts as an air pollutant causing transportation hazards (Bhattachan et al., 2019) and

- remains highly limited, partially due to a lack of high-quality observations (Mahowald et al.,
- 82 2024) with sufficient coverage to constrain key factors, such as dust size (Baker & Jickells,
- 83 2006; Liao & Seinfeld, 1998) and mineral composition (Sokolik & Toon, 1999), which remain
- especially uncertain (Adebiyi & Kok, 2020; Li et al., 2021; Mahowald et al., 2014).
- Dust aerosols, typically represented by multiple modes or bins in Earth system models (ESMs),
- have a broad size range from just above 0.0 up to hundreds of microns (Adebiyi et al., 2023).
- Like many other ESMs, various versions of the Community ESM (CESM) have focused on
- simulating dust particles smaller than $10 \mu m$ in geometric diameter (referred to as diameter
- herein unless stated otherwise; fine: smaller than 1.0 μ m, fine/coarse: 1.0 5.0 μ m, and medium
- $\,$ coarse: 5.0 $10~\mu m)$ (Zender et al., 2003), even though dust particles between 10 and $20~\mu m$
- (super coarse), and even larger than 20 μm (super coarse/giant), has been observed during
- transport from North Africa to Barbados (Prospero et al., 1970) or East Asia to North Pacific
- 93 (Betzer et al., 1988). Particles larger than 10 μm in diameter, especially those larger than 20 μm ,
- were assumed to be rare and received less attention, because (1) most of them were thought to be
- rapidly removed from the atmosphere due to gravitational settling and because they are also

more likely to be removed through below-cloud scavenging (Jones et al., 2022) because of larger 96 size; (2) they were considered less relevant to climate; (3) conventional dry-deposition 97 mechanisms (Petroff & Zhang, 2010; Zender, et al., 2003; Zhang et al., 2001) underestimate their 98 lifetimes and long-range transport (Drakaki et al., 2022; Ginoux, 2003; Ratcliffe et al., 2024); 99 and (4) air quality measurements typically focus on dust particles with diameters smaller than 2.5 100 101 μ m (particular matter: PM_{2.5}) and 10 μ m (PM₁₀) in diameter (Mahowald et al., 2025). The conventional assumption of a 10 µm cutoff diameter in the dust size distribution was 102 103 recognized as insufficient for dust aerosol modeling in a limited number of early studies over 104 two decades ago (Betzer et al., 1988; Mahowald et al., 2005). Such limitation gains more attention following recent campaign measurements using in situ and remote sensing instruments 105 (Van Der Does et al., 2016; Ryder et al., 2013, 2019; Ryder et al., 2018; Weinzierl et al., 2017; 106 Weinzierl et al., 2009). These measurements indicate that dust aerosols with diameters larger 107 108 than 10 µm and up to 40 µm are observed in downwind regions up to 2000 km from North Africa (van der Does et al., 2018). A synthesis of model and observational data highlights that 109 110 dust particles in the 10 - 20 µm diameter range greatly contribute to regional deposition in areas such as the Southern Ocean (Adebiyi et al., 2023). 111 112 Most current ESMs, including CESM with either Bulk Aerosol Module (BAM) (Adebiyi & Kok, 2020) or Modal Aerosol Module (MAM) (Li et al., 2022; Liu et al., 2016), underestimate the 113 abundance of dust particles larger than 5.0 µm in diameter (Adebiyi & Kok, 2020), and even 114 more so for particles larger than 10 µm involved in long-range transport (Ratcliffe et al., 2024), 115 116 compared to observations. This underrepresentation can potentially bias previous estimates of dust aerosol budgets (e.g., smaller cutoff sizes may result in lower emission mass fluxes, or 117 attribute too much dust to the fine fraction) and assessments of dust impacts on Earth-climate-118 human interactions (Kok et al., 2023). 119 To date, only a few studies have attempted to simulate the full-size distribution of dust particles 120 121 in either regional (Drakaki et al., 2022) or global ESMs (Ginoux, 2003; Ratcliffe et al., 2024; Wang et al., 2024), and even fewer in CESM (Meng et al., 2022), although some other ESMs 122 have a dust cutoff size larger than 10 µm in diameter (Gonçalves Ageitos et al., 2023; Klose et 123 al., 2021; Obiso et al., 2024; Perlwitz et al., 2015a). Most CESM-based dust modeling studies 124 125 have not adequately included particles larger than 10 µm in diameter (Albani et al., 2014; Li et

126	al., 2022; Meng et al., 2022). This is because they used older model versions or focused on
127	specific objectives that addressed only limited aspects of dust cycle modeling. For example,
128	Meng et al. (2022) did not focus on model developments within CESM (version 1) but rather on
129	achieving an emitted size distribution consistent with observed dust fluxes. Consequently, their
130	study did not extend model-data comparisons to other critical dust properties, such as surface
131	concentrations, optical depth, and deposition, all of which warrant further investigation. Ke et al.
132	(2022) extended the maximum dust diameter to 20 μm but did not account for coarser aerosols,
133	which remain abundant and are observed downwind of North Africa. The size distribution
134	simulated by their model also requires evaluation against observations, which are unfortunately
135	lacking, before a reliable assessment of the contribution of dust particles between 10 and 20 μm
136	in diameter can be made. There, thus, remains a need for the development of a more
137	comprehensive and precise representation of the dust cycle, which adequately incorporates the
138	dynamics of giant dust particles, along with measurements for evaluating such a model.
139	The primary objective of this study is to advance the mineral-resolved CESM2 (Li et al., 2022)
140	to comprehensively represent dust aerosols across a wide size range, up to 70 µm in geometric
141	diameter at emission (slightly above the diameter boundary between sand and dust particles at
142	$62.5\;\mu\text{m}).$ To achieve this, we develop a ten-mode MAM (MAM10) and constrain the simulated
143	dust size distribution with observations. We also incorporate recent developments into this more
144	comprehensive framework to simulate the climatic impacts of dust aerosols.
145	Specifically, this study introduces several key developments:
146	(1) The new MAM10: We develop this new aerosol module to include dust particles larger than
147	$20~\mu m$ and up to $70~\mu m$ in geometric diameter upon emission within the mineral-resolved
148	Community Atmosphere Model Version 6 (CAM6) framework (Li et al., 2022) as an extension
149	to the CAM5-MAM9 (Ke et al., 2022), which lacks mineral speciation. The mixing state of dust
150	aerosols with non-dust aerosols, such as sea salt, has been revised as well. The simulated dust
151	size distribution is constrained by comprehensive measurements including the super coarse and
152	giant dust fractions (Formenti & Di Biagio, 2024).
153	(2) Improved dust emission scheme: We improve the dust emission scheme in the mineral-
154	resolved CESM2 by adding drag partitioning by non-erodible elements (e.g., rocks and

155	vegetation) and intermittency due to the coarse temporal resolution of the model relative to the
156	timescale of surface wind oscillations (Leung et al., 2023).
157	(3) Updated soil mineralogy atlas: We incorporate a new atlas from the Earth surface Mineral
158	dust source InvesTigation (EMIT) project (Green et al., 2020) to characterize mineralogy in
159	current and potential future erodible soils. Prior to the EMIT mission, dust source composition
160	was estimated from two highly uncertain global soil atlases based on limited mineral
161	measurements and, heavily interpolated over the non-agricultural regions that are the key dust
162	source areas (Claquin et al., 1999; Journet et al., 2014) (pre-EMIT soil mineral atlases). The new
163	EMIT soil mineral atlas produced the first global-scale mineral atlas of dust source regions based
164	upon direct measurements. This EMIT dataset has been shown to significantly reduce
165	uncertainties in dust mineral composition and, consequently, in the simulated dust shortwave
166	direct radiative effect for both the present day and projections for the late 21st century, relative to
167	simulations using pre-EMIT soil atlases (Li et al., 2025).
168	(4) Updated optics for iron oxides: We modify the optics for iron oxides using observationally
169	constrained values, which have been shown to capture both the laboratory-measured dust aerosol
170	optics and their relationship with iron oxide content (Li et al., 2024).
171	This paper evaluates the performance of the new mineral-resolved CESM2 in simulating key
172	features of the observed dust cycle. We also quantify the simulated dust budget, including the
173	contribution of newly incorporated super coarse and giant dust particles. Our analysis focuses on
174	the updated model, with comparisons made to the previous version (Li et al., 2022), without
175	attributing improvements to specific model developments.
176	2 General Model Overview and Treatments in Previous Versions
177	We utilize the National Center for Atmospheric Research CESM version 2.1.0 (CESM2.1.0).
178	This model comprises multiple components, including the atmosphere component (CAM6) and
179	the land component (Community Land Model version 4.5; CLM4.5), crucial for representing
180	Earth's processes in climate projections.
181	Parameterizations for eolian processes responsible for dust emission are primarily implemented
182	in CLM4.5, which communicates with CAM6 through a coupler, transmitting vertical emission

fluxes. CAM6 then redistributes the emission flux into different aerosol modes when employing 183 a MAM, and simulates the transport, deposition, and radiative impacts of dust aerosols. 184 185 2.1 Modal Aerosol Module CAM6 by default utilizes a four-mode MAM (MAM4) (Liu et al., 2016) to represent a total of 186 six aerosol species: Aitken (A2: soil dust, sulfate, sea salt, and secondary organic matter), 187 accumulation (A1: sulfate, secondary organic matter, primary organic matter, black carbon, sea 188 salt, and soil dust), coarse (A3: soil dust, sea salt, and sulfate), and a primary carbon mode (A4: 189 primary organic matter and black carbon). These species, represented by separate tracers, are 190 191 assumed to be internally mixed within the same mode and externally mixed between different modes. Due to the varying physical properties of species (e.g., hygroscopicity, optical properties, 192 and density), this aerosol mixing state is essential for accurately modeling the full aerosol cycle 193 and diagnosing optical properties, which influence radiative effect estimates. In addition, the 194 195 total number concentration of aerosol species in a mode is also advected as a separate tracer, along with the mass mixing ratio of each species (Liu et al., 2016). 196 197 The size distribution of each mode assumes a log-normal shape, with the mode parameters listed in Table 1. These parameters are prescribed in the user namelist file, including the globally 198 199 constant geometric standard deviation, the initialized geometric mean diameter for online predictions, and the range used to constrain the simulated geometric mean diameter. It is worth 200 noting that a previous study (Li et al., 2022) adjusted the coarse mode size distribution 201 parameters (geometric standard deviation, initialized geometric mean diameter, and its range) to 202 those used in CAM5, leading to large changes in the modeled dust aerosol cycle and improving 203 the representation of the dust size distribution. This adjustment is retained here to better 204 represent the dust aerosol size distribution in CAM6-MAM4. 205 206 207 208 209 210

Table 1. Dust Mode Parameters in MAM4.

	Dust Modes	A1	A2	A3	
Dust Modes		(Aitken)	(Accumulation)	(Coarse)	
Emitted Geometric Diameter Limits (μm) for Mass-to-Number Flux Conversions		0.010 - 0.10		1.0 - 10	
Geom	etric Standard Deviation	1.6	1.8	1.8	
Initialized Dry Geometric Mean Diameter (µm) for the Number Distribution		0.026	0.11	2.0	
Low and Upper Bounds for Simulated Dry Geometric Mean Diameter (µm; Number Distribution)		0.0087 - 0.052	0.054 - 0.44	1.0 - 4.0	
Emitted	Fractional Contribution to Total Dust	0.0010	0.011	0.99	
mass fractions	From Clay	1.0	1.0	0.69	
	From Silt	0.0	0.0	0.31	

Processes that determine the removal rate of aerosols from the atmosphere include dry and wet deposition. The dry deposition (e.g., gravitational settling and turbulent removal) rate, calculated in the model via parameterizations, partially depends on the aerosol size (i.e., the predicted geometric mean diameter and geometric standard deviation for each mode). The model we take as default here uses a revised dry deposition scheme (Petroff & Zhang, 2010), compared to that used in the officially released version (Zhang et al., 2001). This revised scheme, as previously shown, helps mitigate the overestimated dry deposition velocity of fine aerosols, such as black carbon (Wu et al., 2018) and dust in the fine modes (Aitken plus accumulation) (Li et al., 2022), as tested in CAM5 and CAM6 (officially released versions), respectively.

Wet deposition depends on the relative location of aerosols to clouds. It consists of in-cloud and below-cloud scavenging, both of which are applied separately to convective and stratiform precipitation. The in-cloud scavenging uses cloud properties (e.g., cloud water, precipitation production profiles) to compute aerosol loss rates, scaled by solubility factors that represent the aerosol fraction incorporated into cloud water. Stratiform cloud-borne aerosols have a solubility factor of 1.0 for all activated aerosols, while interstitial aerosols are unaffected. Convective in-

cloud scavenging applies solubility and activation factors specific to aerosol mode (0.40 for dust) 227 and hygroscopicity (0.068), with lower activation values for less hygroscopic modes. 228 The below-cloud scavenging removes interstitial aerosols through impaction and diffusion, with 229 230 removal rates dependent on solubility factor, scavenging coefficient, and precipitation rate (Neale et al., 2010). CAM6-MAM4 calculates size-dependent scavenging coefficients (Easter et 231 al., 2004) and applies a solubility factor of 0.10, which effectively serves as a tunable parameter. 232 Stratiform cloud-borne aerosols are not subject to the below-cloud scavenging. However, 233 234 aerosols scavenged at higher altitudes can be resuspended at lower atmospheric levels. In CAM6, the fraction of resuspended aerosol is set equal to the fraction of evaporated precipitation. 235 A particle's hygroscopic growth or shrinkage depends on its hygroscopicity, dry radius, density, 236 ambient relative humidity, and temperature, following the κ-Köhler theory (Ghan and Zaveri, 237 2007). This process determines the particle's final wet size and volume at equilibrium under 238 239 ambient conditions. The volume-averaging mixing rule is then applied to predict the bandaveraged (see Li et al., 2021 for band information) complex refractive index of the wet aerosol 240 mixture, based on the relative abundance and refractive index of each component (using its dry-241 state value for aerosols), including water (Ghan and Zaveri, 2007). To parameterize the optical 242 properties of the aerosol mixture, such as single-scattering albedo across different bands, the 243 model uses a look-up table containing Chebyshev polynomial coefficients (see Section 3.1.3 for 244 details on how we generate it) and an interpolation method based on the band-averaged complex 245 refractive index of the wet aerosol mixture (Ghan and Zaveri, 2007). 246 2.2 Default Dust Modeling Without Mineral Speciation 247 The default dust emission in the officially released CESM2 relies on the dust entrainment and 248 deposition model (DEAD) (Zender et al., 2003) in CLM4.5, which simulates the dust 249 mobilization based on the mechanism proposed by Marticorena and Bergametti (1995). This 250 emission scheme was previously updated to a more physically based one (Kok, et al., 2014), 251 shifting emissions to topographic depressions, where abundant erodible sediment accumulates, 252

without using a time-invariant empirical source function such as previously used (Ginoux et al., 253 2001; Mahowald et al., 2006; Zender et al., 2003). 254 Dust emission is initiated when the near-surface friction velocity exceeds the fluid soil threshold 255 256 friction velocity for saltation, the process by which wind lifts sand particles (100 µm) and propels them into ballistic trajectories, ejecting smaller dust aerosol-sized particles into the air 257 upon collision (Kok et al., 2012). This soil threshold friction velocity is determined primarily by 258 near-surface meteorological fields and top 1.0 - 2.0 cm soil properties such as moisture content, 259 260 grain size distribution, and land cover (e.g., vegetation) (e.g., Shao, 2008). Vegetation influences dust emissions via the leaf area index (Mahowald et al., 2006) in CLM4.5. When the leaf area 261 index exceeds 0.30 m² m⁻², vegetation is assumed to fully block the transfer of wind momentum 262 to the surface soil, preventing dust emission. Below this threshold, the fraction of a grid cell that 263 can emit dust aerosols increases linearly with decreasing leaf area index. The suppression of dust 264 emissions due to soil moisture is activated when the near-surface soil gravimetric water content 265 exceeds a threshold determined by the clay content of the soil. This relationship is represented in 266 the land model using a semi-empirical formulation (Fécan et al., 1998) and the top 10 cm soil 267 gravimetric water content. Observational studies linking wind threshold behavior to soil 268 gravimetric water content typically focus on the top 1.0 - 2.0 cm of soil, which dries rapidly 269 following precipitation. In contrast, the uppermost soil layer in the model spans 0.0 - 10 cm and 270 retains moisture longer, introducing inertia not representative of surface conditions and thus 271 uncertainty in the dust emission modeling (Darmenova et al., 2009), which we do not address in 272 this study. 273 274 The size distribution of emitted total dust aerosols follows the brittle fragmentation theory (Kok, 2011a), which allocates 0.10, 1.0, and 98.9% of dust mass into Aitken, accumulation, and coarse 275 modes, respectively, regardless of the friction velocity at the time of emission (Kok, 2011b). 276 277 After emission, dust aerosols are transported, interacting with radiation and clouds, and removed from the atmosphere in CAM6. 278 Evaluations using observations, including in situ measurements and remote sensing retrievals, 279 suggest that compared to the default emission scheme in the officially released CAMs (Zender, 280 Bian, et al., 2003), the more physically based emission scheme, along with the updated size 281

distribution of emitted total dust aerosols, maintains or improves accuracy in modeling the dust 282 climatology in previous CAM versions (Kok, Albani, et al., 2014; Li et al., 2022). 283 2.3 Mineral-Resolved Dust Modeling 284 The dust model (Section 2.2) was modified to categorize the single dust tracer into eight separate 285 mineral tracers (Li et al., 2021; Scanza et al., 2015), enabling detailed optical property 286 calculations based on dust mineral composition. The eight minerals include illite, kaolinite, 287 montmorillonite (smectite), iron oxides (hematite + goethite), quartz, calcite, feldspar, and 288 gypsum. They represent the most common clay- (soil grain diameter below 2.0 um) and silt-289 sized (diameter between 2.0 - 63 µm) soil categories (Claquin et al., 1999). 290 291 The distribution of the eight minerals in the clay- and/or silt-sized categories was derived offline 292 from a global soil atlas, based on the mean mineralogy table of Claquin et al. (1999), where mineralogy is inferred from a global atlas of soil type. Additional modifications by Scanza et al. 293 (2015) and Li et al. (2021) include normalizing the mineral fractions within each clay- or silt-294 sized category for 28 soil types. An equal and opposite adjustment to the illite percentage ensures 295 consistent hematite amounts in both clay- and silt-sized categories. A nearest-neighbor algorithm 296 was then applied to fill the grid cells in the land model for dust emission. The conversion of soil 297 298 mineralogy to dust aerosol mineralogy follows the brittle fragmentation theory (Kok, 2011a), as previously described (Scanza et al., 2015). 299 300 The mass distribution of each mineral across the three emission modes matches that of the model without mineral speciation (Section 2.2). Each mineral, treated as a separate tracer, undergoes 301 the same processes as dust in the released model version, including advection, deposition, and 302 aerosol microphysics (e.g., coagulation). 303 304 All component minerals are assumed to be internally mixed within a mode. Mineral densities are set to values used in a previous study (Li et al., 2021). The scavenging efficiency and 305 hygroscopicity of these minerals are consistent with those prescribed for dust without speciation, 306 as described in Section 2.1. The complex refractive indices for all component minerals, except 307 308 iron oxides, are based on Scanza et al. (2015). For iron oxides, an observationally constrained refractive index is applied, which improved the accuracy in predicting optical properties of dust 309

aerosols using the volume-averaging mixing rule, compared to other publicly available indices 310 (Li et al., 2024). 311 3 Updates to Dust Representations in the Model 312 We introduced two key enhancements to the mineral-resolved CAM6: additional modes via the 313 new MAM10 to simulate super coarse and giant dust particles (Section 3.1) and improved 314 physics to model dust emission (Section 3.2). The development of MAM10 in CAM6 builds 315 upon the previous MAM9 and MAM7 frameworks in CAM5 (Ke et al., 2022; Liu et al., 2012) 316 without mineral speciation, while the advanced dust emission physics are based on those 317 implemented in CAM6 (Leung et al., 2024), which likewise has no mineral speciation. The first 318 modification primarily involves changes to the atmosphere component, and the second focuses 319 on the land component. 320 3.1 A New Modal Aerosol Module 321 MAM10 represents dust aerosols in five distinct transport modes (A5, A7, A8, A9, and A10), as 322 summarized in Table 2. Each of these modes contains two internally mixed species: dust and 323 324 sulfate. While we do not detail sulfate representations, we note that sulfate has multiple sources (Liu et al., 2012), and that sulfate in the coarse modes originates from the condensation of 325 sulfuric acid gas produced by the oxidation of sulfur dioxide, which is either directly emitted or 326 formed via the oxidation of dimethyl sulfide. Sea salt is represented in A1, A2, A4, and A6, 327 328 separate from dust modes, similar to MAM7 and MAM9 (Ke et al., 2022; Liu et al., 2012). Other non-dust aerosol species, such as particulate organic matter and black carbon in the non-dust 329 accumulation mode (A2) aged from the primary carbon mode (A3), are also simulated in 330 MAM10 using the same mode settings as in MAM9. 331 It is important to note that simulations of the non-dust species in MAM10 are expected to vary 332 333 from MAM4 due to different size settings and mixing states of dust aerosols with the other species (e.g., sea salt). These distinctions could affect the simulated dust aerosols by modifying 334 dust deposition rates. For example, the internal mixing of sea salt with dust aerosols can increase 335 the hygroscopicity of the mixture, thereby enhancing the wet removal rate of dust aerosol. The 336 simulation of non-dust aerosol species, their climatic impacts, and their influence on dust aerosol 337

simulations fall outside the scope of this study.

Table 2. Dust Mode Parameters in MAM10.

Dust modes		A5 (Fine)	A7 (Fine/Coarse)	A8 (Medium Coarse)	A9 (Super Coarse)	A10 (Super Coarse/Giant)
Dust Emission Size Cut-offs (μm)		0.10 - 1.0	1.0 - 5.0	5.0 - 10	10 - 20	20 - 70
Geometric Standard Deviation (GSD)		1.8	1.7	1.5	1.5	1.5
Initialized Dry Geometric Mean Diameter (GMD; µm) (Number Distribution)		0.56	2.0	5.5	8.6	14
Low and Upper Bounds for Simulated Dry GMD (number distribution; µm) ^a		0.36 - 0.76	1.7 - 2.4	4.0 - 6.3	8.3 - 10	10 - 18
Emitted Mass	Fractional Contribution to Total Dust	0.0028	0.14	0.19	0.24	0.43
Fractions	From Clay	1.0	$0.97 (0.57)^{b}$	0.87 (0.57) ^b	0.87	0.87
	From Silt	0.0	0.030 (0.43) ^b	0.13 (0.43) ^b	0.13	0.13

Note. ^a Lower and upper bounds are applied to constrain the simulated geometric mean diameter of each dust mode. ^b Values outside parentheses are derived using the brittle fragmentation theory for non-hematite minerals, while values in parentheses correspond to hematite and have been refined.

As in previous MAM versions, each mode is described by three parameters: the geometric standard deviation, geometric mean diameter, and volume or mass concentration, detailed in Tables 1 and 2 and the following subsections.

3.1.1 Refinement of Dust Size Distribution

The mode parameters (Table 2), along with adjustments to the dry settling velocity of dust aerosols (Table 3), were fine-tuned to match the observed size distribution (Formenti & Di

350	Biagio, 2024). The optimized dust aerosol size distribution from Adebiyi and Kok (2020),
351	extended to include the giant dust mode (emission size range in geometric diameter: 20 - $70~\mu m$).
352	supports using a constant standard deviation of 2.0 across all modes. However, initial tests with
353	this value failed to match observations near North African dust sources, whereas a subsequent
354	test, reducing the standard deviation to 1.5 for the three largest dust modes (A8, A9, and A10;
355	Table 2), achieved a better model performance (see Section 7.2.3).
356	The rationale for adopting a geometric standard deviation of 1.5, rather than 2.0, which provides
357	the best fit to the observed size distribution using five lognormal modes, likely reflects persistent
358	uncertainties in dust modeling and model-observation comparisons. For example, modeling the
359	evolution of dust size remains inherently challenging. We applied the obtained geometric
360	standard deviation uniformly in all the calculations requiring this parameter, including
361	diagnostics of dust optical properties. The assumption behind using a constant geometric
362	standard deviation across different modes globally is that the width of the size distribution
363	remains unchanged between freshly emitted and aged dust aerosols. This approach may not fully
364	capture variations in the geometric standard deviation, as atmospheric processes (e.g., size-
365	dependent cloud interactions and deposition) can modify the size distribution during transport. A
366	more accurate solution would be desirable as the model development advances with additional
367	measurements of both freshly emitted and aged dust aerosols.
368	While the mode parameters for dust between 0.40 and 10 µm are relatively well constrained by
369	observations, those for particles outside this range are more uncertain (Formenti & Di Biagio,
370	2024). This reduced confidence for the submicron accumulation mode stems from several
371	factors, including contamination by non-dust aerosols (e.g., biomass burning, especially for
372	particles smaller than $0.50\ \mu m$ in diameter) and assumptions about dust shape that limit the
373	accuracy of the measurements used for size distribution optimization (Dubovik et al., 2000). For
374	the giant dust mode, the uncertainty is mainly due to the scarcity of measurements and
375	instrument limitations. Although AErosol RObotic NETwork (AERONET) retrievals are
376	abundant and capture aerosol mass up to $30\ \mu m$ in diameter, they are sensitive to assumptions
377	about large particle sizes.
378	Due to challenges in representing the dust size distribution, the model requires a prescribed range
379	of the geometric median diameter for each mode to reasonably simulate it. Sensitivity tests

without these bounds show that the last four largest dust modes (A7, A8, A9, and A10) tend to merge into a single broad mode. The geometric mean diameters of the fine/coarse and medium-coarse modes are sensitive to the prescribed upper bounds, while the super coarse and giant modes are more influenced by the lower bounds. Limited observational data to constrain the simulated diameters for the three largest dust modes (A8, A9, and A10), combined with the use of a globally constant geometric standard deviation for each mode and the absence of mechanisms to transport dust particles larger than 5.0 µm in diameter, prevents this study from fully capturing the observed dust size dynamics.

3.1.2 Tuning the Gravitational Settling Velocity for Large Dust Particles

The Stokes settling velocity overestimates the settling velocity of particles larger than 20 μm in diameter by more than 10%, primarily because the particle Reynolds number (Re) approaches or exceeds 1.0, the upper limit for which Stokes' law is valid (Stokes, 1851). The magnitude of this error becomes more pronounced with increasing dust particle size. For example, particles of 200 μm in diameter can have a settling velocity bias of up to 60%. To address this issue, a time-invariant Stokes correction factor was applied in CAMs to coarse particles up to a maximum aerosol diameter of 1000 μm (Zender et al., 2003), although previous versions of CAM did not simulate aerosol particles of such large sizes. Despite this correction, large dust particles still appear to settle too quickly compared to the real atmosphere, limiting the model's ability to match measured dust size distributions (see discussions in Section 8 for the proposed reasons so far).

Due to misrepresented or absent mechanisms in the model's dust-related parameterizations, an adjustment, as applied in previous studies (Drakaki et al., 2022; Meng et al., 2022), is used to better simulate coarse dust aerosols. Specifically, we reduce the settling velocity for the three largest dust modes (A8, A9, and A10) by 50, 75, and 85%, respectively (Table 3), to counteract gravitational settling, thereby narrowing the differences in dust size distribution between the model and observations. While more formal alternative methods may exist (see discussions in Section 8), this approach offers a practical compromise to address the overestimation of coarse dust particle settling velocity in the models until particle transport mechanisms are better understood and incorporated, a critical task beyond the scope of this study.

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Mode version	MAM4	MAM10		
Emission Scheme	Kok et al. (2014); linear dependency on modeled LAI and the threshold LAI is set as 0.30 m ² /m ² above which no emission occurs	Leung et al. (2023; 2024) with MERRA2 LAI and increased threshold LAI to 1.0 m ² /m ²		
Size Distribution	As in Li et al. (2022); also shown in Table 1	See Table 2		
Dust Tuning Parameter (dust_emis_fact: Inversely Linked to Dust Emission Rate)	3.6	0.35		
Dust Optical Depth in the Visible Band (0.44 - 0.63 µm)	0.030	0.031		
Percentage Reduction to Gravitational Settling Velocity of Dust Aerosol	0.0	50, 75, 85 for A8, A9, and A10, respectively		
Simulation Period	2007 - 2011 (one additional year for model spin-up)	2007 - 2011 (one additional year for model spin-up)		
Look-Up Table for Aerosol Optic Diagnostics	As in Li et al. (2022)	Regenerated to accommodate with new dust modes (Sect. 3.1.3)		
Soil Atlases	Mineral fractions derived from Claquin et al. (1999) and normalized to unity	Derived from EMIT following ATBD (see Section 3.4)		

Note. Abbreviations: MAM - Modal Aerosol Module; LAI - Leaf Area Index; MERRA2 -

Modern-Era Retrospective analysis for Research and Applications Version 2; EMIT - Earth

surface Mineral dust source InvesTigation; ATBD - Algorithm Theoretical Basis Document.

Changes in the gravitational settling velocity of coarse mode aerosols can also influence their

wet deposition processes, which we do not quantify separately from the total dust budget

estimation, including dry deposition. This interaction arises for two main reasons. First,

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modifying the dry deposition velocity in a coarse mode can alter simulated dust burdens, which directly affect the wet scavenging rate as a first-order function of dust burden, thereby impacting dust aerosol removal via wet deposition. Second, due to the mode's width, dust number or mass concentrations in one mode can contribute to the total dust mass within the size range represented by neighboring modes. This contrasts with the binned approach, where size bins have fixed boundaries and no overlap between modes. 3.1.3 Parameterization of Aerosol Optical Properties CAM6 uses parameterized aerosol optical properties to calculate aerosol radiative effects while minimizing computational costs. In this study, we use the approach outlined by Ghan and Zaveri (2007), generating a coefficient look-up table for MAM10. Sensitivity tests by Ghan and Zaveri (2007) suggest that this method reproduces aerosol optical properties calculated by the Mie Theory with errors generally less than 20%, and under most conditions, within 30% of the Mie solution, except when their absolute values are small. The look-up table, applied to all aerosol modes and species, provides coefficients for total aerosol specific absorption (σ_{abs}), extinction (absorption plus scattering, σ_{ext}), and the asymmetry parameter (g) across 14 shortwave bands, as well as specific absorption for 16 longwave bands. We derive the coefficients by fitting Chebyshev polynomials to mass-weighted (m_w) specific optical properties (i.e., $\log(\sigma_{sca}/m_w)$, σ_{abs}/m_w , and g) based on Mie simulations with seven real (n_r) and ten imaginary (n_w) components of refractive indices that bracket the probable absorption range of aerosols. These components are equally spaced between 1.3 - 1.6 and 0.0 - 0.79, respectively. A brief introduction to the parameterization of aerosol optical properties using the Mie Theory is provided in Text S1 in Supporting Information, with detailed descriptions available in Ghan and Zaveri (2007). It is important to note that the Mie Theory cannot be applied to particles with non-spherical shapes for optical property calculations. However, dust aerosols are typically non-spherical (Kalashnikova & Sokolik, 2004) and exhibit complex morphologies (Falkovich et al., 2001), which increase their extinction efficiency (Huang et al., 2023). To address this limitation, we apply shape corrections by increasing the mass extinction efficiency by 16% for fine dust aerosols (A5 and A7) and by 28% for coarser dust aerosols (A8, A9, and A10), based on

previous calculations (Kok et al., 2017) and an extension of the dust emission cutoff geometric 445 diameter to 70 µm. This adjustment is applied only to shortwave bands, as the dust model is 446 tuned to the estimated global visible-band dust optical depth of 0.030 (Ridley et al., 2016), 447 although shape effects may similarly enhance the longwave extinction efficiency (Huang et al., 448 2023). 449 450 3.2 Amendments to Dust Emission Physics Despite the improvements achieved (Kok, et al., 2014), the physically based scheme previously 451 incorporated into the mineral-resolved CESM2-CAM6 (Li et al., 2022) (described in Section 2.2) 452 still lacks key physics necessary for accurately predicting dust emission strength and location. 453 This limitation led to the development of a more scientifically comprehensive scheme (Leung et 454 al., 2023) designed for integration into ESMs (Leung et al., 2024). Here, we incorporate this 455 scheme into our mineral-resolved dust model. 456 457 The additional physics included in the emission process now accounts for two critical factors. First, intermittency from unresolved instantaneous wind fluctuations, such as turbulence, which 458 occur on timescales (around one minute or less) smaller than the model's 30-minute time step 459 within the near-surface boundary layer. Second, drag partitioning due to surface roughness 460 461 elements, like vegetation and rocks, that partially absorb wind momentum. This absorption reduces the downward momentum flux available to uplift soil dust particles. 462 We briefly summarize the key elements below. Texts S2 and S3 in Supporting Information 463 provide additional details on the parameterizations of emission intermittency and drag 464 partitioning due to non-erodible elements, respectively. For further information, see Leung et al. 465 (2023).466 Moreover, a global unconsolidated sediments map (GUM) (Börker et al., 2018) is applied as a 467 surface erodibility constraint in the updated dust emission model. This implementation 468 suppresses dust emission from consolidated surfaces, allowing emission only from 469 unconsolidated regions (e.g., loose sands, silts, and dust beds) and spectrally identified by EMIT 470 as potential dust sources. 471

3.2.1 Emission Intermittency

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In ESMs with prognostic dust, accounting for intermittency could be crucial (Comola et al., 2019) to avoid considerable biases in dust emission simulations (Dupont et al., 2013). Saltation intermittency is due to the highly turbulent surface winds that frequently sweep across the dust emission threshold friction velocity, causing multiple sudden initiations and terminations of dust emission on a timescale of minutes. Theoretical studies indicate that the saltation intermittency effect is further complicated by the fact that there are two threshold friction velocities for dust emission: the fluid threshold required to initiate saltation, and the impact threshold required to sustain saltation after it has been initiated (Bagnold, 1941; Kok et al., 2012). In most schemes, dust emission begins when friction velocity exceeds the fluid threshold but ceases when friction velocity falls below this threshold. However, in practice, emission persists even when friction velocity decreases to a value between the two thresholds, ceasing only when it drops below the impact threshold. This saltation intermittency effect could be important under two conditions where accounting for intermittency can improve dust emission simulations (Kok et al., 2012). First, when the model surface wind speed is below the fluid erosion threshold, turbulent fluctuations can still initiate saltation and dust emission, which is then sustained until the wind drops below the impact threshold. Second, soil cohesion, while not affecting the impact erosion threshold, considerably increases the fluid erosion threshold (Comola et al., 2019). These conditions typically occur in marginal source regions (e.g., semi-arid areas) due to higher soil moisture than arid regions. Consequently, incorporating the intermittency effect tends to increase dust emissions over these marginal source regions more than over arid regions (Leung et al., 2023). To include the intermittency effect, a substepping scheme was developed to parameterize the effects of instantaneous wind fluctuations on initiating and ceasing dust emission (Comola et al., 2019). A saltation intermittency factor, ranging between 0.0 and 1.0, is introduced in the updated dust emissions scheme (Leung et al., 2023), representing the fraction of time during which saltation and dust emissions are active within a model time step (see Text S2 in Supporting Information). The updated dust emission is then modeled as the product of this intermittency

factor and the vertical dust emission calculated by the previous version of the scheme for each 500 model time step. 501 3.2.2 Drag Partitioning Due to Non-Erodible Elements 502 503 A surface roughness correction factor is applied to the modeled friction velocity to account for drag partitioning effects from rocks or vegetation (Leung et al., 2023), which were either absent 504 or not well represented in CESM versions before that study. The effects of these two elements 505 are calculated separately and then combined using a weighted mean based on the fractional area 506 of each grid cell dominated by rocks or vegetation (see Text S3 in Supporting Information). 507 The drag partitioning due to rocks is quantified using a semi-empirical equation by Marticorena 508 509 and Bergametti (1995), with aeolian roughness length provided by satellite retrievals of the 510 backscatter coefficient at 5.3 GHz (Prigent et al., 2005). To separate the roughness map for rocks from vegetation, the minimum value of the monthly aeolian roughness length in a climatological 511 year was used. This map is applied to grid cells where the vegetation area index (the sum of the 512 one-sided leaf area index and stem area index) is less than 1.0 m² m⁻². The parameterization thus 513 accounts only for photosynthetic vegetation, excluding other types such as plant debris and 514 senescent leaves. 515 The drag partitioning due to vegetation is parameterized following Pierre et al. (2014), building 516 on Okin (2008), by using a gamma distribution to represent the gaps between plants. This 517 518 parameterization is straightforward to implement in ESMs, as drag partitioning can be approximated using only the leaf area index. In the simulation with the updated model here, the 519 leaf area index is taken from the Modern-Era Retrospective analysis for Research and 520 Applications Version 2 (MERRA2) (Gelaro et al., 2017). 521 3.3 Conversion of Soil Mineralogy to Aerosol Mineralogy 522 The distribution of emitted minerals across five dust aerosol modes is determined using the 523 524 extended brittle fragmentation theory (Meng et al., 2022) and the method proposed by Scanza et al. (2015). The original brittle fragmentation theory predicts that the emitted particle size 525 distribution follows a power law for dust particle diameters (D_d) between 2.0 and 10 µm, with 526 emission outside this range determined by the size-crack propagation length (λ). In contrast to 527

Scanza et al. (2015), our calculation of the emitted mineral distribution employs the bulk dust 528 distribution derived by Meng et al. (2022), which accounts for the dependence of the size-crack 529 propagation length on the diameter of soil aggregates (diameter D_{agg}), in addition to the fully 530 dispersed soil size distribution used in Kok (2011a) and Scanza et al. (2015). This new approach 531 yields a mineral size distribution consistent with the bulk dust distribution used in our model. 532 However, the dispersed soil size distribution is treated as a free parameter, determined by fitting 533 the emitted dust size distribution to observations. The resulting soil particle distribution is 534 skewed toward finer sizes, potentially overrepresenting the clay-sized fraction relative to the 535 parent soil. In contrast, minerals more evenly distributed across both clay- and silt-sized 536 fractions, such as iron oxides, are less sensitive to the assumed soil size distribution. 537

Following Meng et al. (2022), Equation 1 expresses the size-crack propagation length which is proportional to the size of the fragmented soil dust aggregate:

$$\lambda = f_{\lambda} D_{agg}, \tag{1}$$

where f_{λ} , the ratio of the size-crack propagation length to the dust aggregate size, is set to 0.15.

The normalized emitted mass (M_d) particle size distribution accounting for soil aggregation is given by Equation 2:

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$$\frac{dM_{d}}{dlnD_{d}} = \frac{D_{d}}{C_{m}} \left[1 + erf\left(\frac{ln (D_{d}/\overline{D_{s}})}{\sqrt{2}ln (\sigma_{s})}\right) \right] \int_{0}^{\infty} exp\left(-\frac{D_{d}}{f_{\lambda}D_{agg}}\right)^{3} P_{agg}(D_{agg}) dD_{agg}, \tag{2}$$

where erf is the error function, and C_m is a normalized constant ensuring unit integral of Equation 1 over the size range of 0.10 - $70~\mu m$. The size distribution of fully dispersed soil is approximated by a lognormal distribution with a volume median diameter $\overline{D_s} = 1.13 \pm 0.58~\mu m$ and a geometric standard deviation $\sigma_s = 1.9 \pm 0.25$. The size distribution of fragmented soil dust aggregates is also modeled as a log normal distribution with a volume median diameter $\overline{D_{agg}} = 1.27 \pm 47~\mu m$ and a geometric standard deviation $\sigma_s = \sim 3.0 \pm 1.0$:

$$P_{agg}(D_{agg}) = \left[\frac{1}{D_{agg}ln(\sigma_{agg})\sqrt{2\pi}}\right] exp\left[-\frac{\left(ln(D_{agg}/\overline{D_{agg}})\right)^{2}}{2\left(ln(\sigma_{agg})\right)^{2}}\right]. \tag{3}$$

The emitted mass fraction of mineral i in different dust modes, arising from clay-sized (M_{c,i}) or other-sized (M_{o,i}) soil category, is computed by integrating either Equation 4 for the accumulation dust mode or Equation 5 for larger dust modes. The integration bounds correspond to the mode cutoff sizes listed in Table 2:

$$\frac{dM_{d,i}}{d\ln D_d} = M_{c,i} \frac{dM_d}{d\ln D_d} \text{ for } D_d \le 2 \text{ } \mu\text{m}, \tag{4}$$

$$\begin{split} \frac{dM_{d,i}}{dlnD_{d}} &= M_{c,i} \frac{dM_{d}}{dlnD_{d}} + M_{o,i} \frac{D_{d}}{C_{m}} \left[erf \left(\frac{ln \left(D_{d} / \overline{D_{s}} \right)}{\sqrt{2}ln \left(\sigma_{s} \right)} \right) \right. \\ &\left. - erf \left(\frac{ln \left(2 / \overline{D_{s}} \right)}{\sqrt{2}ln \left(\sigma_{s} \right)} \right) \right] \int\limits_{0}^{\infty} exp \left(- \frac{D_{d}}{f_{\lambda} D_{agg}} \right)^{3} P_{agg} \left(D_{agg} \right) dD_{agg}, \text{ for 2 } \mu m < D_{d} \\ &\leq 70 \ \mu m. \end{split}$$
 (5)

3.4 The EMIT Soil Mineralogy Atlas

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EMIT detects ten soil minerals (illite, kaolinite, smectite, hematite, goethite, calcite, gypsum, chlorite, dolomite, and vermiculite) using their spectral signatures with an advanced shortwave and near-infrared imaging spectrometer (Thompson et al., 2024) from its vantage point on the International Space Station (Green et al., 2020). Mineral identification is conducted at the instrument's native spatial resolution of 60 meters, employing the Tetracorder linear feature-matching algorithm (Clark et al., 2024) to interpret surface reflectance. Mass fractions are subsequently estimated using a Hapke-like radiative transfer model (Hapke, 1981), which accounts for scattering and reflection within approximately the top centimeter of soil (Clark & Roush, 1984; Clark et al., 2024), the primary source region for dust emission.

To derive size-resolved mineral mass fractions in emitted aerosols, it is necessary to estimate the distribution of minerals across soil particle sizes. We partition EMIT-derived mineral

565	abundances into clay ($\leq 2.0 \mu\text{m}$) and silt (2.0 - $50 \mu\text{m}$) fractions based on established empirical
566	relationships informed by soil texture data, Food and Agriculture Organization (FAO) soil
567	classification maps, and auxiliary constraints, including observed iron oxide concentrations
568	(Perlwitz et al., 2015a) and AERONET-retrieved dust single-scattering albedo (Obiso et al.,
569	2024). Iron oxides are assumed to be uniformly distributed across particle sizes. The resulting
570	mineral mass fractions are allocated to different dust modes following methodologies established
571	in prior work (Gonçalves Ageitos et al., 2023; Perlwitz et al., 2015a) and subsequently applied in
572	our model simulation.
573	A comprehensive description of the methodology used to generate the mineral atlases is
574	available in previously published documentation (Brodrick, Clark, et al., 2023; Clark et al., 2024;
575	Okin et al., 2023). The publicly released EMIT soil mineral atlases are archived at National
576	Aeronautics and Space Administration (NASA)'s Distributed Active Archive Center (DAAC)
577	(Brodrick, Okin, et al., 2023).
578	EMIT soil mineralogy used in this study is derived using Tetracorder expert system 5.27c1
579	(Clark, 2024; Clark et al., 2024), which has known deficiencies, including misidentifying some
580	nano-hematite-nano-goethite mixtures as jarosite. Thus, the total hematite abundance could be
581	under-reported by the EMIT delivery used for this study. The EMIT team is working on a new
582	reflectance model, and this known deficiency has been mitigated in newer versions (Tetracorder
583	expert system 5.27e1 and later) and the EMIT team will recalculate the global mineral starting
584	later in 2025.
585	The EMIT retrievals are made across latitudes between 60° S and 60° N. We mask areas where
586	there is solid rock (see Section 3.2). High-latitude dust sources exist (Groot Zwaaftink et al.,
587	2016; Meinander et al., 2022). Although their contribution to the global mean annual dust direct
588	radiative effect is minor due to small dust loadings, limited vertical extent, low surface
589	temperatures, and reduced solar irradiance at high latitudes, they can considerably impact
590	regional climates by interacting with clouds (Shi et al., 2021), glaciers, and land or ocean
591	ecosystems (Bullard, 2017). To account for these sources and fill gaps in areas where the EMIT
592	soil atlas provides no data, soil mineralogy from Claquin et al. (1999) is applied.

For our dust model, chlorite, vermiculite, and kaolinite are grouped and collectively transported as kaolinite, using the physical properties of kaolinite. Initial tests indicate that different combinations (e.g., combining chlorite, vermiculite, and illite, and using the physical properties of illite) have a negligible impact on dust direct radiative effect calculations, given the smaller optic differences among them compared to their differences with iron oxides, particularly at the visible band. Similarly, hematite and goethite are combined and treated as iron oxides due to the limited availability and questionable accuracy of refractive indices for goethite (Warren et al., 2019). Constrained refractive indices, which accurately capture the optical properties of dust aerosols in the shortwave spectral range with the combined iron oxides (Li et al., 2024), are applied to the total iron oxide component.

4 Model Configuration

CAM6 with either aerosol model is configured with sea surface temperature prescribed from measurements, with the atmosphere coupled to active land and sea ice models, and to data ocean and slab glacier models. Two simulations (Table 3), both tuned to have almost identical global dust optical depth (0.030) in the visible band, are conducted to quantify the modeled dust cycle, allowing for a thorough evaluation of the new and old dust models and inter-model comparisons. Therefore, one simulation utilizes the new model with all implemented changes introduced in Section 3, while the other uses the old model introduced in Section 2 with observationally constrained optics for simulated iron oxide aerosols. Both simulations are performed at a horizontal spatial resolution of 1.25° × 0.9° (longitude by latitude) with 56 vertical layers up to 2.0 hPa with offline meteorological fields (air temperature, horizontal wind, and relative humidity) taken from the MERRA2 reanalysis data for the period 2006 - 2011 (only the last five-year results are used for data analysis). Anthropogenic emissions are from the Coupled Model Intercomparison Project Phase 6 (CMIP6) inventory for the year 2000 (Eyring et al., 2016).

Observational Data for Model Performance Evaluation

5.1 Observations of Dust Aerosol Size Distribution

- We utilize previously compiled and harmonized measurements of size distributions (Formenti &
- Di Biagio, 2024) for freshly emitted dust (residence time less than 1.0 day), as well as dust

following medium (1.0 - 4.0 days), and long (more than 4.0 days) range transport. This 621 classification includes 12, 36, and 10 locations for short-, medium-, and long-range transported 622 dust, respectively. 623 624 The compilation comprises 58 datasets collected from literature published over the past 50 years. These datasets were gathered at various locations using different measurement techniques, 625 626 including microscopy, Coulter multisizers, optical particle counters, optical array probes, and mobility particle sizers. They feature diverse operational size definitions (e.g., optical, projected-627 628 area, aerodynamic, and geometric), sampling concentrations and size information (e.g., resolutions and ranges). Such variability complicates accurate and comparable analysis of dust 629 size distributions across sources and transport stages, posing challenges for model evaluation. To 630 address these inconsistencies, Formenti and Di Biagio harmonized the data at four levels (Levels 631 632 0, 1, 2a, and 2b). Detailed documentation of the dataset is provided by Formenti and Di Biagio (2024).633 This study uses their Level 2b data to evaluate modeled dust size distributions. This dataset is 634 derived through three key steps: (1) interpolating original data from publications over a common 635 size range with equi-logarithmically spaced diameters; (2) renormalizing the interpolated data so 636 that the integral equals unity over a defined diameter range, enabling direct comparison of size 637 distribution shapes rather than absolute values; and (3) harmonizing observed particle diameters 638 by converting them to a common equivalent geometric diameter that accounts for dust 639 asphericity. These procedures ensure a robust and consistent dataset that accounts for inherent 640 differences in dust size distribution measurements, facilitating a better understanding of dust 641 642 particle size dynamics across different transport stages and a consistent comparison of modeled dust size distribution. 643 5.2 Dust Optical Depth and Deposition Flux 644 To evaluate our simulated dust optical depth in the visible band, we compared it against 645 AERONET retrievals (aerosol optical depth Version 2 Direct Sun Algorithm, Level 2.0) at the 646 wavelength of 0.55 µm that were postprocessed by Albani et al. (2014). The postprocessing 647 648 includes rigorous data quality control procedures and station selection criteria based on the dominance of dust optical depth in the reported total aerosol optical depth (Albani et al., 2014). 649

The dust deposition flux data utilized in this study were originally compiled by Albani et al. 650 (2014) from various sources (Ginoux et al., 2001; Lawrence & Neff, 2009; Mahowald et al., 651 2009; Tegen et al., 2002) for present-day climate conditions. Reported mass fractions of dust 652 particles smaller than 10 µm in diameter are used to estimate the total dust deposition flux, 653 including contributions from coarser particles, for comparison with the total flux simulated by 654 both MAM versions. 655 **5.3 Elemental Aluminum Concentration** 656 Elemental aluminum, often found in mineral dust aerosols associated with aluminosilicate, is 657 widely used to identify mineral aerosols within an aerosol population (Formenti et al., 2001; 658 Measures & Vink, 2000; Prospero et al., 2001). We compare elemental aluminum particulate 659 matter derived from simulated minerals with aerodynamic diameters of up to 2.5 µm (PM_{2.5}) and 660 10 μm (PM₁₀) to newly compiled data from Mahowald et al. (2024). This dataset synthesizes a 661 wide range of mass measurements for PM_{2.5} and PM₁₀ aerosol particles from the literature and 662 public health databases. 663 The cutoff size at 2.5 µm or 10 µm is established according to aerodynamic diameter guidelines, 664 which inform inlet design in reference methods. These PM_{2.5} and PM₁₀ values correspond to 665 PM_{1.6} and PM_{6.9}, respectively, in geometric space, which defines the aerosol sizes in our model. 666 We considered this difference in size concept when preparing the model results for comparison, 667 following Reid et al. (2003), using a dynamic shape factor of approximately 1.5. 668 669 **5.4 Mineral Aerosol** We assess modeled mineral fractions in surface concentrations and deposition fluxes using data 670 previously compiled at 154 locations (Perlwitz et al., 2015a, 2015b), with dust primarily 671 transported from North Africa, the Middle East, and Asia. This dataset includes observations 672 from individual points and averages along ship tracks, with sampling periods ranging from single 673 days to several years, spanning the late 1960s to the 2000s. For details on sample numbers, 674 standard deviations, sampling periods, size ranges, locations, and vessel tracks, see Perlwitz et al. 675 (2015) and references therein. Although most studies in the compilation aimed to measure 676 mineral fractions from dust aerosols (e.g., by extracting dust with polycarbonate or quartz 677

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microfibers, cellulose filters, or other filters, or by selecting dust-heavy episodes), various sources of error still exist, which may affect the interpretation of the model-data comparison. To evaluate the model's ability to simulate dust mineral composition, we compare model results with observed minerals filtered by Gonçalves Ageitos et al. (2023) to exclude potential anthropogenic contamination by aerosols from industrial activities and combustion processes (Chen et al., 2012; Luo et al., 2008). Observed minerals are mapped to model components, recalibrating mass fractions and standard deviations to account for unrepresented species (e.g., grouping all iron oxides as hematite). The interannual variability of mineral fractions is considered low, enabling comparison between observed values and modeled multi-annual means (Perlwitz et al., 2015a). Model outputs are matched to observations in time (monthly climatology), space, and size by collocating resolved size distributions at corresponding locations and times. For MAM4, only observations with particle sizes smaller than 10 µm in diameter are retained, while broader size ranges are used for MAM10 following Gonçalves Ageitos et al. (2023). Here, we additionally include model-observation comparisons for dust particles larger than 20 µm. Text S4 in Supporting Information describes in detail how the mineral fraction dataset is used. EMIT provides total mineral abundances in soils but does not resolve their size distribution. It also does not directly retrieve quartz and feldspar. The first version of the EMIT-derived soil map for use in mineral-resolved models, including this study, incorporates pre-EMIT soil mineral atlases and empirical relationships that rely on soil texture data to describe the partitioning of quartz and feldspar, as well as the size distribution of calcite. In order to minimize the impact of these assumptions on model-observation comparisons, we group quartz, feldspar, and calcite simulated by MAM10 into a single category for comparison with observations. 6 Statistical Metrics and Limitations in Model-Observation Comparison We assess model performance in simulating dust mineralogy by comparing modeled and observed mineral composition (e.g., ratios of minerals both modeled and observed) and size distribution at corresponding locations and times, using monthly climatology. The modeled dust size distribution is evaluated using monthly climatology, whereas comparisons involving elemental aluminum, dust optical depth, and total dust deposition flux are based on annual

means, except when assessing their seasonal cycles, which are evaluated on a monthly basis. For
most model-observation pairs, we compute (normalized) mean bias, (normalized) root mean
square error, or Pearson correlation coefficients in space or time. Statistical significance of the
correlations is tested at the 95% confidence level.
Although model outputs are selected to align as closely as possible in both space and time with
available measurements, several potential limitations in the model-observation comparison
remain and may affect the interpretation of results, as previously summarized (Li et al., 2022).
These limitations include the following aspects:
First, temporal mismatch. Observational campaigns often span only a few days to weeks, while
model results are reported as monthly means. Although we compare monthly climatology, this
discrepancy may introduce uncertainties, particularly given the episodic nature of dust events.
The short duration of measurements may not reliably represent climatological conditions, and
this mismatch is a common issue across all model-observation comparisons presented in this
study. We do not quantify the uncertainty introduced by this limitation.
Second, differences in spatial representativeness. Model outputs reflect averages over grid cells
that can span hundreds of kilometers, whereas observational data, such as those from ground
stations or aircraft, capture dust properties at much finer spatial scales. This difference in spatial
sampling volumes can lead to discrepancies, particularly in regions with strong spatial gradients
in dust concentration.
Third, potential bias in data site selection. Some AERONET site selections are based on modeled
aerosol fields, which may not accurately reflect actual optical depth conditions for both dust and
non-dust aerosols. This introduces potential bias into the validation process.
Fourth, observational uncertainties. Observational datasets themselves are subject to errors. For
example, AERONET aerosol optical depth retrievals may be affected by cloud-screening
algorithms (Levy et al., 2010), and postprocessing procedures applied to dust size distribution
data from different field campaigns (Section 5.1) can introduce additional uncertainties
(Formenti & Di Biagio, 2024).
Fifth, inconsistencies in mineral types between the model and observations, as well as

differences in the number of observation points used for MAM4 and MAM10. Modeled mineral fractions are collocated with observations in time (monthly climatology), space (nearest model grid cell), size, and mineral composition. For the compositional comparison, only minerals present in both the model and observations at a given site are considered, with uncommon minerals excluded rather than dropping the site entirely. This approach maximizes the number of usable observational points per mineral, as not all sites report all modeled minerals. However, it introduces discrepancies in relative mass fractions at sites where the set of modeled minerals differs, for example, between pre-EMIT (Claquin et al., 1999) and EMIT-based simulations, where chlorite, vermiculite, and kaolinite are grouped and collectively simulated as kaolinite. Additionally, the distinct size ranges represented by MAM4 and MAM10 result in different subsets of valid observational data for each model. As a result, the statistical metrics reflect not only differences in model performance for comparable size ranges but also artifacts introduced by these inconsistencies, complicating direct comparisons between MAM4 and MAM10 in reproducing observed mineral mass fractions.

7 Results

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7.1 Simulated Global Annual Dust Budget Climatology

- With a globally averaged dust optical depth of approximately 0.030 in the visible band (0.44 -
- 752 0.63 µm) following the dust model tuning (Table 3), the inclusion of dust particles larger than
- 10 μm in diameter increases the simulated annual global dust budget. During the 2007 2011
- period, the MAM10 model simulates higher dust emission, deposition fluxes (9500 versus 2900
- Tg year⁻¹), and loadings (40 versus 24 Tg) compared to MAM4. The inclusion of super coarse
- and giant dust particles enhances dry deposition more than wet deposition, increasing the dry-to-
- vet deposition ratio. This disparity primarily results from the stronger gravitational settling of
- 758 coarser dust particles, whose deposition predominantly occurs in arid regions with low
- precipitation (see Section 7.3). Table 4 provides further details on the distribution of simulated
- 760 global dust quantities across different size modes.
- The disproportionately large deposition (or emission) relative to loadings in MAM10, compared
- to MAM4, is primarily due to the short lifetime of super coarse (1.1 days) and super coarse/giant
- dust particles (0.33 days) (i.e., due to differences in particle size distribution). This shorter

764	lifetime of large particles, combined with the reduced sensitivity of visible-band dust optical
765	depth to large particles because of their low mass extinction efficiency (e.g., A5: 2.0 m ² g ⁻¹
766	versus A10: 0.041 m ² g ⁻¹ ; and MAM10: 0.40 m ² g ⁻¹ versus MAM4: 1.3 m ² g ⁻¹), and dust tuning
767	toward a global mean visible-band dust optical depth of 0.030 (Tables 3 and 4) collectively
768	explains why MAM10, despite simulating substantial increases in the dust loading and emission,
769	maintains a global mean visible-band dust optical depth comparable to MAM4.
770	Particularly, our diagnosed dust quantities for a cutoff size of 10 μm in diameter, based on the
771	simulated size distribution of aged dust, suggest that the higher dust loadings in MAM10
772	compared to MAM4 primarily result from dust particles between 2.0 and 10 μm . Dust particles
773	larger than 10 μm account for 10 Tg (25% of total dust loading), while dust smaller than 10 μm
774	accounts for 30 Tg (75% of total dust loading) in MAM10, which is 25% higher than in MAM4.
775	The high loading within the 2.0 - $10~\mu m$ geometric diameter range, which may partially explain
776	the overestimated surface elemental aluminum with aerodynamic diameters between 2.5 and 10
777	μm (see Section 7.2.1), primarily results from the reduced simulated settling velocity of medium-
778	coarse dust particles in MAM10 (Table 3). This high loading in the 2.0 - $10~\mu m$ geometric
779	diameter range, accompanied by a slight compensatory reduction in absolute dust mass for
780	particles smaller than $2.0~\mu m$, mainly reflects the reduced mass extinction efficiency of dust
781	aerosol in this size range in MAM10. The reduced efficiency, in turn, arises from the model's
782	size distribution parameters for both emitted and transported dust, which are shifted toward
783	coarser particles, thereby lowering visible-band extinction relative to MAM4. As a result,
784	MAM10 requires a higher dust emission rate (implemented through a lower dust emission tuning
785	factor, which is inversely related to the emission rate; Table 3) to maintain the global mean
786	visible-band dust optical depth at approximately 0.030.
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Table 4. Simulated Global Annual Mean Dust Quantities (years: 2007 - 2011).

Mode Information	Total; < 10; < 20 μm	A5 (Fine)	A7 (Fine/Coarse)	A8 (Medium Coarse)	A9 (Super Coarse)	A10 (Super Coarse/Giant)
Emission/Deposition (Tg year ⁻¹)	9500; 3100 (33% of total); 5400 (57% of total)	100	1400	1600	2300	4100
Loadings (Tg)	40; 30 (75% of total); 36 (90% of total)	1.9	14	13	7.0	3.5
Lifetime (days)	1.5	11	3.6	2.9	1.1	0.32
Optical Depth in the Visible Band (0.44 - 0.63 µm)	0.031	0.0073	0.016	0.0072	0.0012	0.00028
Visible-Band Mass Extinction Efficiency (m ² g ⁻¹)	0.40	2.0	0.58	0.28	0.087	0.041

Note. In the second column from the left, numbers in parentheses, if present, indicate the global percentage contribution of dust larger than 10 or 20 μ m in diameter to size-integrated dust budgets.

We also compare the simulated dust quantities with the inverse modeling results of Kok et al. (2021) for dust diameters smaller than 20 μ m. MAM10 simulates a global mean dust emission of 5400 Tg year⁻¹ for particles smaller than 20 μ m in diameter, which falls within the broad range of 3400 - 9100 Tg year⁻¹ estimated by Kok et al. (2021). However, the simulated dust loading (36 Tg) is 20% higher than the upper range of 22 - 30 Tg reported by Kok et al. (2021). This indicates that MAM10 probably overestimates the amount of dust particles smaller than 20 μ m in diameter, despite its simulated size distribution comparing well with observations (see Section 7.2.3). To match the dust mass within that size range, tuning the global dust optical depth in the visible band from 0.031 to 0.025, which still falls within the observational range of 0.025 - 0.035 (Ridley et al., 2016), might be a good practice if the mass extinction efficiency and dust size distribution remain unchanged.

We did not apply the adjustment in the global dust optical depth for the present study for two primary reasons. First, simulated dust mass is not the focus but rather one of several dust-related quantities we aim to represent reasonably. Second, this adjustment reflects a pragmatic compromise, as the dust optical depth is more directly constrained by observations than dust loading, which is inferred indirectly. As the model-observation agreement on the dust size distribution can still be improved, another possible approach is to further refine the dust size distribution and gravitational settling velocity, which might help reduce the likely overestimation of dust mass compared to Kok et al. (2021). Moreover, the probable underestimation of the impact of dust particle asphericity on mass extinction efficiency may partially explain the overprediction of dust particles smaller than 20 µm by MAM10 relative to Kok et al. (2021).

7.2 Model-Observation Comparison

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7.2.1 Annual Mean Dust Cycle Climatology

while maintaining the MAM4 model's accuracy for dust particles smaller than 10 µm in diameter (Figure 1). The incorporation of super coarse and giant dust particles, along with substantially improved dust emission physics and size distribution (see Section 7.2.3), provides limited improvement in capturing the overall climatology of dust optical depth and deposition fluxes. Across different regions, the combined changes to CAM6 result in varying degrees of accuracy in reproducing the observed dust cycle compared to the earlier version. Simulated dust deposition aligns more closely with observations in some regions (e.g., the South Pacific) but shows larger discrepancies in others (e.g., North Indian). At high latitudes, while suffering from limited data for comparison, both new and old models likely systematically overestimate dust deposition fluxes. This overestimation, which exceeds a factor of two, does not occur in low-tomiddle latitude regions. Given the low dust emissions at high latitudes, particularly in the Southern Hemisphere, this discrepancy suggests that the new model, like MAM4, may transport dust from low-latitude source regions likely too efficiently to high latitudes. Consequently, the underestimation of surface dust concentrations and overestimation of deposition observed at several locations, such as near El Djouf and the Antarctic that are present in MAM4 (Li et al.,

The new MAM10 reproduces observed dust cycle within an order of magnitude at most sites

2022) and earlier versions of CAM (BAM in CAM4 and MAM3 in CAM5) (Albani et al., 2014), are still evident in MAM10. This phenomenon remains particularly evident at sites like King George in the Antarctic (62° S, 58° W) compared to that in MAM4 (Li et al., 2022), where previous studies using observations combined with multiple model ensembles, including an earlier version of CAM4, have reported similar findings (Kok, et al., 2021).

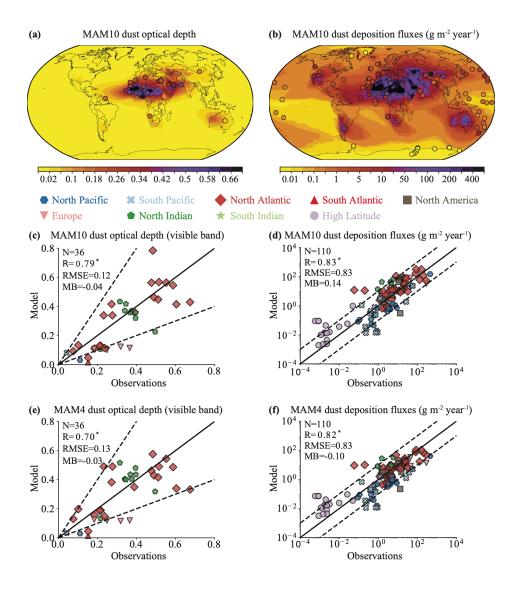


Figure 1. Comparison of simulated total dust quantities with observational data (total number: N) previously compiled (Albani et al., 2014). Panels (a), (c), and (e) show dust optical depth in the 0.44 - 0.63 μm band, while panels (b), (d), and (f) present deposition fluxes. The deposition fluxes represent values across all size ranges. Black solid lines in scatter plots indicate the one-one line, while dashed lines show a factor of 2.0 (panels c and e) or 10 (panels d and f)

847	difference. Model (MAM10: panels a - d; MAM4: panels e and f) performance metrics include
848	Pearson's correlation (R; "*" indicates significance at the 95% confidence level), root mean
849	square error (RMSE), and mean bias (MB), calculated in the logarithmic space for deposition
850	fluxes and in the normal space for dust optical depth.
851	The new model achieves considerable improvements in representing the spatial variation of
852	surface elemental aluminum with aerodynamic diameters smaller than 2.5 μm (Figure 2).
853	However, it almost systematically overestimates that for aerodynamic diameters smaller than 10
854	μm (Figure 3), more specifically between 2.5 and 10 μm , indicating a probable overestimation of
855	emissions for particles in that size range in the model. The cause of this overestimation is
856	unclear. If the model-data comparison for diameters smaller than 2.5 μm and both data are
857	reliable, a probable explanation could be insufficient vertical transport of dust likely due to the
858	process split issue (Wan et al., 2024) in the model (dust dry deposition is applied immediately
859	after emission and then turbulence mixing), which tends to cause dust to unduly accumulate in
860	the bottom model layer. The likely overestimation of dust mass between 2.0 and 10 μm in
861	geometric diameter, as inferred in Section 7.1 compared to Kok et al. (2021), may also partially
862	explain this overestimation.

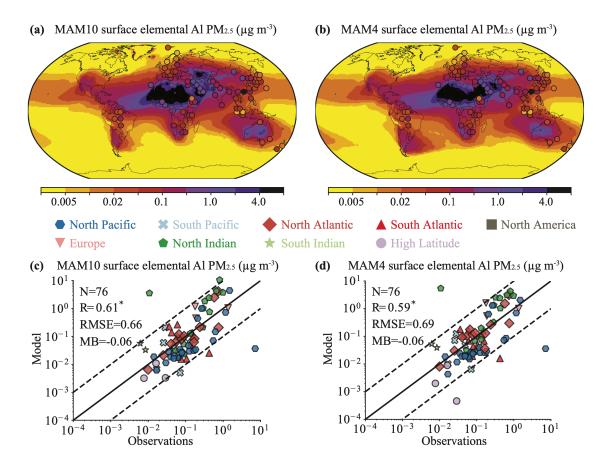


Figure 2. Derived surface elemental aluminum (Al) concentrations based on simulated minerals compared to observations (total number: N) previously compiled (Mahowald et al., 2024) with aerodynamic diameters smaller than 2.5 μm (particular matter: PM_{2.5}). Panels (a) and (c) correspond to MAM10, and panels (b) and (d) to MAM4. Observational data were remapped onto a 4° × 4° regular grid, reducing the dataset to 76 points for better visualization and modeldata comparison. Model performance metrics include Pearson's correlation (R; "*" indicates significance at the 95% confidence level), root mean square error (RMSE), and mean bias (MB), calculated in the logarithmic space.

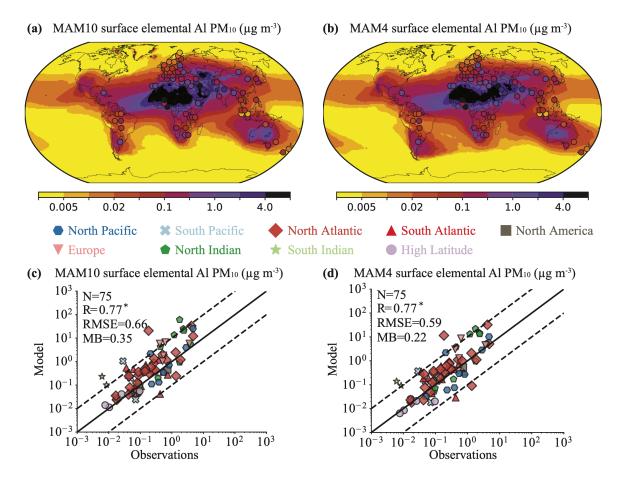


Figure 3. Derived surface elemental aluminum (Al) concentrations based on simulated minerals compared to observations (total number: N) previously compiled (Mahowald et al., 2024). Same as Figure 2 but with aerodynamic diameters smaller than 10 μm (particular matter: PM₁₀).

The persistent discrepancies between modeling and observations highlight uncertainty in modeling the dust cycle and in model-data comparisons, influenced by multiple factors summarized in Section 6 and by a previous study (Li et al., 2022), for which we still lack sufficient information to address. Here, we highlight two additional factors beyond.

First, a known yet unquantifiable error source stems from sparse station-based measurements available for model-data comparison, particularly in dust source regions. This limitation biases dust aerosol emission modeling, likely leading to overestimated emissions over North Africa (Li et al., 2022), which remains evident in MAM10, due to the global tuning to achieve a mean dust optical depth of approximately 0.030. Thus, regional refinement of dust emission might be necessary.

Second, the available measurements themselves are subject to uncertainty, such as those related to deposition (Mahowald et al., 2005) and surface elemental aluminum concentrations (Mahowald et al., 2025). Post-processing procedures applied to reported measurements by original studies likely introduce further uncertainties, as noted for dust size distribution harmonization (Formenti & Di Biagio, 2024).

7.2.2 Seasonality of Dust Aerosol

Given the importance of simulating dust seasonality, which interacts with other Earth system components like ocean and land biogeochemistry, we assess the model's ability to reproduce dust seasonality in Figure 4 using previously compiled climatologically monthly mean data for AERONET dust optical depth at the 0.55 µm and surface concentrations (Albani et al., 2014).

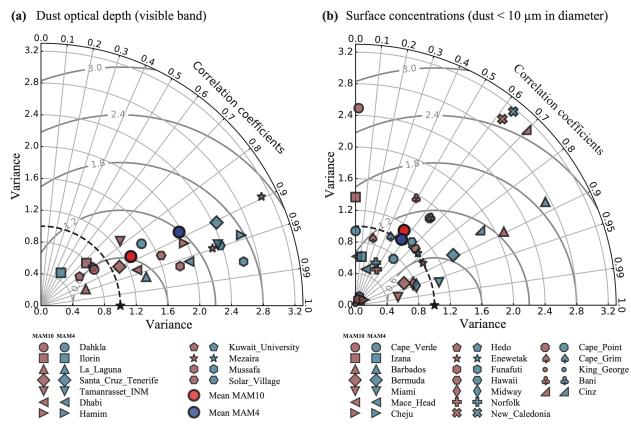


Figure 4. Model-data comparison of dust seasonality, defined as the monthly mean climatology. Panel (a) shows results obtained for dust optical depth, while panel (b) presents surface dust concentrations with the geometric diameter smaller than 10 μm. Seasonal data compiled by Albani et al. (2014) were used to evaluate the model's performance.

MAM10 simulates different seasonal cycle amplitudes of dust optical depth in the visible band (Figure 4a) and surface concentrations (Figure 4b) from MAM4, although the temporal correlations are similar between the two models, better for dust optical depth than for the surface concentrations. Overall, the new model slightly improves the simulated seasonal amplitude of the visible-band dust optical depth (Figure 4a), while slightly degrading that for the surface concentrations (Figure 4b). This improvement is primarily attributed to the inclusion of drag partitioning in the updated dust emission scheme (Leung et al., 2023), which reduces dust emission during peak summer while having a minimal effect in winter (Leung et al., 2024). The different performance on the dust optical depth in the visible band and surface concentrations is not paradoxical, as the observations were made at different locations. The two variables also differ: dust optical depth is a column-integrated variable, whereas surface concentration represents only the bottom model level and depends on the accuracy of simulated vertical dust transport. Because of this difference, another possible explanation for the different behavior in the dust optical depth versus surface concentration is that the model's vertical mixing of dust aerosols is likely insufficient, as discussed in Section 7.2.1, and has a seasonal cycle that is also not well simulated. Determining the "true" cause requires further investigation, which is beyond the scope of this study.

7.2.3 Dust Size Distribution

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Monthly dust size distributions are calculated offline using simulated monthly mass mixing ratios and number concentrations of different aerosol species in each dust mode, the prescribed geometric standard deviation, and the air temperature and pressure near the atmospheric levels where dust size measurements were taken. Using the calculated monthly values at locations near the measurement sites, the climatological dust size distribution is then derived and compared with observations.

Three things are worth noting before interpreting the model-observation comparisons. First, there is a non-linear relationship between the dust size distribution and the variables used in the calculations. As a result, using monthly modeled values to compute the monthly dust size distribution may introduce uncertainties, compared to performing the calculations at each model time step and then averaging over a month. Second, the size distribution measurements for dust

930	between 0.40 and 10 μm are more reliable due to the higher data volume within this range
931	(Formenti & Di Biagio, 2024) compared to outside this range, where measuring dust becomes
932	more challenging. Third, in addition to uncertainties introduced during postprocessing (Formenti
933	& Di Biagio, 2024), this dataset and the model-data comparison may face similar shortcomings
934	discussed for the other datasets (Section 6).
935	Despite any shortcomings in the observations and model-observation comparison, the size
936	settings and reduced settling velocities applied to the last three dust modes in MAM10 provide a
937	significantly better representation of observed dust size distributions compared to MAM4, which
938	simulates almost no dust particles larger than 10 μm (Figure 5). The simulated dust size
939	distribution in MAM10 is within observational uncertainties across most size ranges, except for
940	medium-range transported dust coarser than 50 $\mu\text{m},$ which the new mode underestimates (though
941	not significantly), and particles smaller than 0.40 $\mu m,$ which both models overestimate. MAM10
942	simulates a clear size dynamic, where the two largest dust modes gradually disappear during
943	transport, while the fine modes remain largely unchanged, which is consistent with observations
944	(Formenti & Di Biagio, 2024). In contrast, MAM4 shows little change in its single coarse mode
945	across varying distances from the source regions.

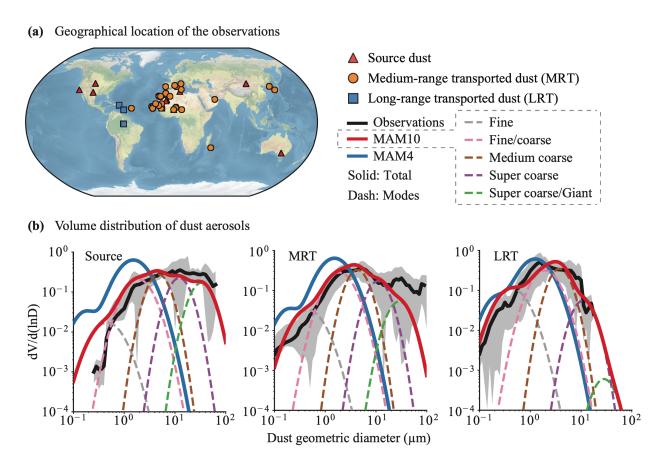


Figure 5. Modeled dust volume size distributions compared to observations previously compiled (Formenti & Di Biagio, 2024). Panel (a) indicates observation locations for the size-resolved data, while panel (b) compares modeled and observed size distribution for dust categorized by transport distance from source regions for MAM10 (red solid lines and dash lines for individual dust modes) and MAM4 (blue solid lines). Model results corresponding to the atmospheric levels, locations, and months of observations were used. Both modeled and observed data are normalized for dust particles across the full-size ranges. The five dust modes correspond to distinct size ranges of emitted particles: Fine $(0.10 - 1.0 \, \mu m)$, fine/coarse $(1.0 - 5.0 \, \mu m)$, medium coarse $(5.0 - 10 \, \mu m)$, super coarse $(10 - 20 \, \mu m)$, and super coarse/giant $(20 - 70 \, \mu m)$.

The new model's performance is improved by incorporating new dust modes that represent super coarse and giant dust and applying reductions in their gravitational settling velocity: 85% for the giant dust mode and 75% for the super coarse dust mode. We also reduce 50% of the simulated gravitational settling velocity for the medium-coarse dust mode. These reductions are generally comparable to the approximately 45 - 60% reductions for particles $10 - 30 \mu m$ in the Georgia Institute of Technology Goddard model (Ginoux, 2003), though lower than the 80 and 90%

reductions applied in Weather Research and Forecasting (WRF) (Drakaki et al., 2022) and 962 CAM-BAM (Meng et al., 2022), respectively. This reduction amplitude difference could reflect 963 structural (e.g., model resolution) and parametric (e.g., drag coefficients) variations across 964 models. 965 In MAM10, giant dust particles still likely settle too quickly during medium-range transport, 966 967 while observations suggest invariant dynamics for dust above 10 µm, or dust fails to be sufficiently uplifted to reach the measurement altitude. A greater reduction in settling velocity is 968 avoided to prevent overestimating the fraction of dust coarser than 20 µm over source regions. It 969 is worth noting again the uncertainties in the observations and the model-observation comparison 970 971 approach, which may complicate the interpretation of the results. For example, the unknown processes (see Section 8) may systematically reduce the settling velocity of super coarse and 972 973 giant dust particles only under specific atmospheric conditions that occur during the field campaigns and do not occur elsewhere. Because modeled climatological mean dust size 974 distributions are compared with measurements from field campaigns that typically last only a 975 few days (Formenti & Di Biagio, 2024) and are therefore more representative of specific events 976 977 than the models' climatological means, this time mismatch could probably introduce uncertainties in the model-observation comparison, particularly for medium- and long-range 978 979 transported dust. 980 7.2.4 Mineral Aerosol As described in Section 3, the new MAM10 integrates several recent advancements, including: 981 (1) a revised dust aerosol size distribution, (2) an extended brittle fragmentation theory that 982 enables the emission of super coarse and giant dust particles, (3) an improved EMIT-derived soil 983 mineral atlas, and (4) modified iron oxide content in the three largest dust size modes through 984 updates to the EMIT soil mineral atlas. This study does not attempt a systematic comparison 985 between simulations using the EMIT and pre-EMIT soil mineral atlases, which will be the focus 986 of a forthcoming publication. Nor do we isolate the contribution of each individual development. 987 Instead, our focus is on evaluating the overall performance of the new model, incorporating these 988

combined developments, in reproducing observed mineral fractions in surface concentrations,

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dry deposition, and total deposition.

Simulations conducted with and without these modifications exhibit similar spatial distributions 991 of mineral mass fractions. However, the combined developments increase the abundance of 992 993 kaolinite relative to the other minerals (Figure S1 in Supporting Information), primarily at the expense of smectite (Figure S1 in Supporting Information) and quartz (Figure S2 in Supporting 994 Information), as well as iron oxides (Figure 6) and gypsum (Figure S3 in Supporting 995 Information), both at the global scale and across the majority of grid cells. 996 A direct comparison between the new and old models using observed mineral mass fractions is 997 998 not feasible, as the metrics presented in the paired subpanels of Figures 7 and S4 in Supporting Information correspond to different particle size categories and numbers of observation points. 999 1000 Despite this limitation, the new model successfully simulates mineral fractions in dust aerosol particles larger than 10 or 20 µm in diameter, which are not represented in the old model. The 1001 1002 extension of mineral size representation alone marks a substantial advancement of the new model over the old one. The existing discrepancies between the modeled and observed minerals 1003 1004 do not necessarily reflect limitations in the EMIT surface mineralogy retrievals (see Section 8).

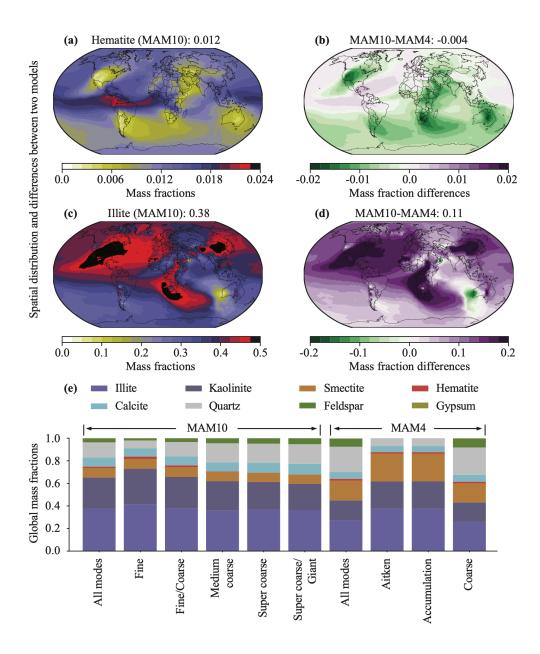


Figure 6. Simulated mineral mass fractions. Panels (a) - (d) show the spatial distribution of simulated hematite and illite, as well as their differences between MAM10 and MAM4. Panel (e) presents the global mean mass fractions of simulated minerals by MAM10 and MAM4 in different size ranges.

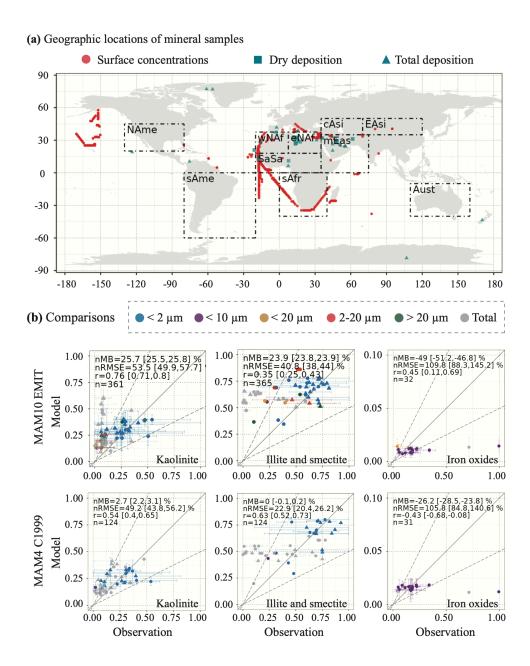


Figure 7. Mineral sample locations and model-simulated mineral fraction in surface concentrations or depositions compared to previously compiled observations (total number: n) (Perlwitz et al., 2015) using the approach of Gonçalves Ageitos et al. (2023). (a) Mineral sample locations categorized by geophysical zones, obtained from surface concentrations (red circles), dry deposition (green squares), or total deposition (green triangles). (b) Model-data comparisons for kaolinite, smectite, and iron oxides across different size (geometric diameter) segments: top panels for MAM10 with the EMIT soil atlas and bottom panels for MAM4 with the Claquin et al. (1999) soil atlas (C1999). Model performance is evaluated using normalized mean bias

(nMB), normalized root mean square error (nRMSE), and size and spatial correlation (r), based 1019 1020 on different sample sizes (n: number of observational samples used in each plot). See Figure S4 1021 in Supporting Information for the other minerals. 1022 Overall, the new model shows improved capability but still faces challenges in reproducing both the observed magnitude and spatial variability of mineral mass fractions (Figures 7 and S4 in 1023 1024 Supporting Information). It captures the mass fraction of kaolinite, the combined fraction of illite 1025 and smectite (Figure 7), and the combined fraction of quartz, feldspar, and calcite (Figure S4 in 1026 Supporting Information) within a factor of two at many sites and across multiple size categories. Furthermore, it reproduces the observed spatial distribution of several minerals, including iron 1027 1028 oxides with a moderate correlation of 0.45 with measured mass fractions (Figure 7). 1029 Limited observations at sparse locations also indicate size-dependent variations in aerosol mineral composition. For example, the mass fraction of iron oxides tends to decrease with 1030 1031 increasing particle size (Kandler et al., 2009; Panta et al., 2023). This trend is captured, to some extent, by the combined modifications (Figure 6). MAM10 simulates highly uncertain mass 1032 fractions of iron oxides in the last two dust modes, due to the lack of robust observational 1033 constraints on the abundance of iron oxides in giant dust particles. While some studies report 1034 almost zero iron oxides in giant dust particles (Kandler et al., 2009; Panta et al., 2023), this 1035 reflects an underestimation by electron microscopy, which identifies iron oxides in isolated 1036 particles but not in inclusions, rather than their actual absence. 1037 7.3 Improved Representation of Size-Resolved Dust Quantities 1038 1039 Dust aerosols of different sizes/modes exhibit varying physical properties, meaning the modeled dust quantities and their distribution among modes are sensitive to changes in size and mode 1040 number. Figure 8a addresses how the new model improves the relative distribution of dust 1041 quantities, including the optical depth, deposition, loadings, or PM_{2.5} globally, compared to 1042 MAM4. 1043 In MAM4, a single coarse mode constitutes 80% (Figure 8a) of the total dust optical depth in the 1044 visible band centered at 0.53 µm, which fails to capture the diverse scattering and absorption 1045 behaviors of dust particles of different sizes. The increased number of dust modes in MAM10 1046

addresses this limitation by distributing 70% of the global dust optical depth across the first two smallest dust modes (fine and fine/coarse), with the fine/coarse mode being the primary contributor (50%; Figure 8a). Among the medium coarse, super coarse, and super coarse/giant dust modes, the dust optical depth is more evenly distributed, with each mode contributing minimally. This reduced contribution primarily arises from lower mass or volume extinction efficiencies and the preferential removal of coarse and giant dust particles by size-dependent aerosol removal mechanisms, such as gravitational settling.

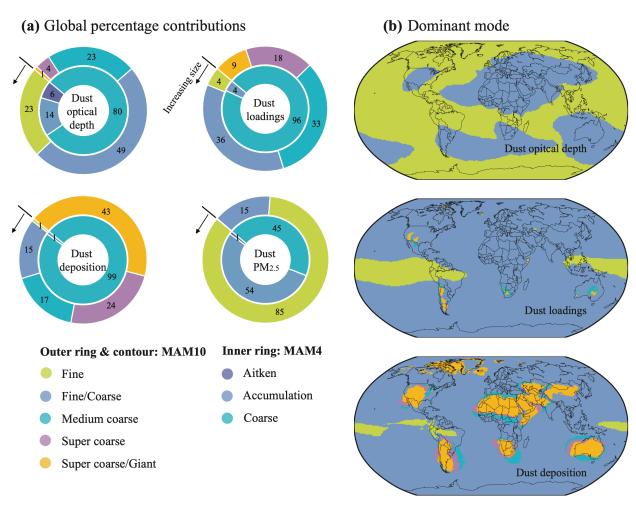


Figure 8. Simulated percentage contribution of individual dust modes to various dust quantities (panel a; MAM10: outer ring; MAM4: inner ring) and spatial distribution of the dominant dust mode in MAM10 (panel b). The dominant mode is the dust mode that contributes the most to a given dust quantity among the five (three) dust modes in MAM10(4). The dust quantities include: (1) visible-band dust optical depth (top-left rings in panel a and top map in panel b), (2) column-integrated dust mass (top-right rings in panel a and middle map in panel b), (3) total (dry

1061	+ wet) dust deposition (bottom-left rings in panel a and bottom map in panel b), and (4) surface
1062	particular matter concentrations (bottom-right rings in panel a) with aerodynamic diameters
1063	smaller than 2.5 μm (PM _{2.5}). The five dust modes in MAM10 cover size ranges of 0.10 - 1.0
1064	(fine), 1.0 - 5.0 (fine/coarse), 5.0 - 10 (medium coarse), 10 - 20 (super coarse), and 20 - 70 μm
1065	(super coarse/giant), respectively, for emitted particles. In comparison, the three dust modes,
1066	Aitken, accumulation, and coarse, in MAM4 cover size ranges of 0.010 - 0.10, 0.10 - 1.0, 1.0 -
1067	10 μm, respectively.
1068	When tuning the model to match the well-constrained global dust optical depth of approximately
1069	0.030 in the visible band, we adjust the total emitted dust mass to match this total dust optical
1070	depth value. Because the mass extinction efficiency of fine dust peaks at visible wavelengths and
1071	a substantial portion of dust mass resides in the accumulation/fine and fine/coarse modes,
1072	visible-band dust optical depth provides a relatively strong constraint on dust particles within
1073	these size ranges. In contrast, this constraint weakens for coarser particles, whose contribution to
1074	visible-band dust optical depth is limited, while the observational global dust optical depth is
1075	available in the visible band. As a result, tuning based solely on visible-band dust optical depth
1076	offers limited leverage on super coarse and giant dust emissions. To overcome this limitation,
1077	alternative constraints, such as satellite-derived infrared-band dust optical depth, which is more
1078	sensitive to particles larger than $10 \mu m$, or retrievals of dust effective diameter, are promising.
1079	However, their utility is currently limited by sparse observational coverage and the reliance on a
1080	single coarse mode in the retrieval process (Zheng et al., 2022).
1081	In terms of deposition, contributions from differently sized dust modes vary substantially (Figure
1082	8a). In MAM4, the single coarse mode used to simulate dust in the 1.0 - 10 μm size range
1083	accounts for 99% of total dust deposition. In contrast, MAM10 reduces this contribution to 30%
1084	by simulating dust in this size range using the fine/coarse and medium-coarse modes. Super
1085	coarse/Giant dust dominates total deposition in MAM10, contributing 40% to the global
1086	deposition budget, while fine-mode dust contributes 1.0%, similar to that in MAM4.
1087	The improved representation of dust through additional modes is also evident in the modeled
1088	PM _{2.5} at the bottom model level. In MAM10, PM _{2.5} is represented exclusively by fine dust
1089	modes (fine and fine/coarse), and the contributions of the three largest dust modes to PM _{2.5} are

1090	negligible, while the single coarse mode in MAM4 contributes about 45% of PM _{2.5} .
1091	Consequently, in MAM4 any attempt to increase coarse-mode aerosols also increased PM _{2.5}
1092	aerosols, which limited the ability of the model to separately simulate fine and coarse mode
1093	aerosols.
1094	Spatially, the distinct physical properties of different dust modes and the distribution of dust
1095	mass or number fluxes over them highlight their distinct roles in influencing the dust optical
1096	depth in the visible band, loadings, and deposition across regions (Figure 8b). Specifically,
1097	Figure 8b suggests the following three key findings.
1098	First, the fine-coarse mode along with the accumulation mode dominates visible-band dust
1099	optical depth everywhere: in the tropics along the Intertropical Convergence Zone (ITCZ), and in
1100	high-latitude regions controlled by the accumulation mode, and the other regions dominated by
1101	the fine coarse mode. However, coarser dust modes (larger than $5.0~\mu m$ in diameter) can
1102	contribute over 30% near or over source regions (Figure S5 in Supporting Information).
1103	Therefore, an inappropriate representation of dust aerosols larger than $5.0~\mu m$ in diameter may
1104	lead to the misestimation of finer dust emission, given the tuning process used to achieve a
1105	global dust optical depth of 0.030 in the visible band. For example, neglecting the contribution of
1106	coarser modes to the global dust optical depth could result in excessive emissions of finer dust
1107	from source regions to compensate for the missing contribution from coarser particles.
1108	Second, the fine/coarse mode dominates the dust loadings in most grid cells (Figure 8b) due to
1109	its longer atmospheric lifetime compared to coarser modes and its higher emission rates relative
1110	to dust in the accumulation mode.
1111	Third, the two smallest dust modes (fine and fine/coarse) also dominate dust deposition in
1112	remote regions and along the ITCZ, whereas the giant dust mode's dominant influence remains
1113	confined to areas near dust sources due to its large size and short atmospheric lifetime. This
1114	spatial pattern (Figure 8b) underscores the critical role of the two smallest dust modes in
1115	transporting nutrients such as iron and phosphorus to ecosystems like the Amazon rainforest and
1116	the central Pacific, surpassing the contributions of other modes. The two smallest dust modes
1117	actually dominate most dust quantities, except for total global deposition, which is primarily
1118	simulated by the super coarse/giant dust mode.

7.4 Role of Dust Particles Larger Than 10 Microns in Diameter

1119

Our model results suggest that dust particles larger than 10 µm in diameter are an important 1120 component of dust aerosols and should be included in models to estimate global and regional 1121 1122 dust budgets, consistent with findings from previous studies using alternative models (Adebiyi et al., 2023). These particles contribute 70% of the total deposition flux and 30% of the total 1123 1124 atmospheric loading in MAM10 (Figure 8a), which is slightly lower than a previous estimate of 35% (Adebiyi et al., 2023). While most of these particles (diameter larger than 10 µm) remain 1125 near dust source regions, such as the Sahara and the Sahel, a considerable amount of dust mass 1126 can be transported over long distances, as observed in various measurements (Prospero et al., 1127 1128 1970; Ryder et al., 2013; Ryder et al., 2018, 2019; Weinzierl et al., 2017; Weinzierl et al., 2009) and previously simulated (Adebiyi et al., 2023). Although these transported particles contribute 1129 1130 far less to the total (size-integrated) dust budget than the global percentages shown in Table 4, they can still deposit onto remote ecosystems, including ocean basins, the Amazon Rainforest, 1131 Greenland, Antarctica, and the Tibetan Plateau, where they supply essential micro- and 1132 macronutrients. 1133 Our calculations reveal a substantial increase in dust deposition across different regions when 1134 super coarse and giant dust particles are included, which is also revealed by a previous study 1135 (Adebiyi et al., 2023), along with the improved representation of particles in the 5.0 - 10 µm size 1136 range (Figure 9). Therefore, the inclusion of such super coarse and giant particles highlights the 1137 potential for a great upward revision of previously estimated dust budgets (Kok et al., 2023) and 1138 probably their climatic impacts, particularly through altering the land and ocean 1139 1140 biogeochemistry, despite a disproportionally low impact of this fraction of dust on the direct 1141 radiative effect relative to their contribution to the dust budget, which will be quantified in our future papers. 1142 Given the limited availability of high-quality observations, the new model likely underestimates 1143 dust deposition in the Southern Ocean, with values an order of magnitude lower than previous 1144 estimates (Kok et al., 2023), even without accounting for dust coarser than 20 µm (2.0 versus 25 1145 Tg year-1). The separation of sea salt from dust aerosols was intended to mitigate the excessive 1146 wet removal of dust observed in MAM4, particularly in regions downwind of South American 1147

sources, such as the Patagonian Desert. The effect, however, is weaker than anticipated.

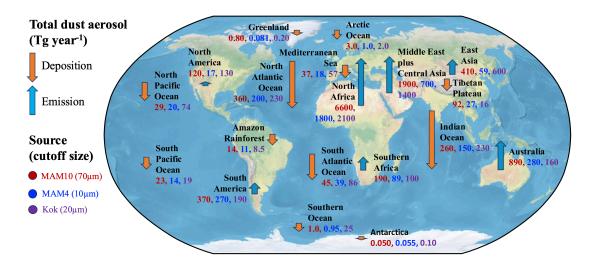


Figure 9. Regional dust emission and deposition budgets (Tg year⁻¹). Upward blue arrows represent dust emissions, while downward orange arrows indicate total (dry + wet) dust deposition. Numbers in red, blue, and purple correspond to results from MAM10 (dust geometric diameter smaller than 70 μm), MAM4 (dust geometric diameter smaller than 10 μm), and the best estimates (geometric diameter smaller than 20 μm) from Kok et al. (2023), which integrate observations with multi-model simulations. The marine geophysical definitions follow the shapefiles available at https://marineregions.org/, while the continental definitions follow Python's built-in geophysical files.

Although the global contribution of dust particles larger than 10 μ m in diameter to the total dust optical depth in the visible band is small, their regional impact can be considerable. For example, campaign measurements over the Sahara Desert indicate a 40% contribution to the total dust optical depth at 0.55 μ m (Ryder et al., 2018), which is similarly simulated here (Figure S5 in Supporting Information) and overall consistent with previous simulations (Adebiyi et al., 2023). This suggests that, although excluding dust larger than 10 μ m in diameter may not considerably affect the global visible-band dust optical depth, it could bias estimates of the finer dust emission toward higher values, especially near source regions such as in the Sahara Desert, to compensate for the missing contribution of dust larger than 10 μ m in diameter, given that the model is tuned to a global visible-band dust optical depth of 0.030. Since fine-sized dust has a longer atmospheric lifetime and tends to scatter solar radiation more than it absorbs, this bias could

result in three key outcomes: (1) increased transport of dust away from source regions; (2) 1169 1170 enhanced cooling effects everywhere; and (3) overestimation of PM_{2.5} levels over source regions, 1171 such as Sahel and Sahara deserts. 1172 **8 Further Discussion and Conclusions** 1173 This study addresses two previously unresolved issues in CESM2: (1) the absence of dust particles larger than 10 µm in diameter, whose substantial presence is indicated by observations, 1174 1175 and (2) the underrepresentation of dust mass and particle number between 1.0 - 10 µm, which relies on a single mode that unrealistically dominates most dust quantities. These goals are 1176 1177 achieved through the addition of modes representing larger particle diameters within the new mineral-resolved aerosol framework, MAM10. This new model also simulates more 1178 1179 comprehensive dust emission physics, building upon previous work (Ke et al., 2022; Leung et al., 2024; Li et al., 2022; Liu et al., 2012), and uses a new EMIT atlas of surface soil mineralogy 1180 1181 (Brodrick, Green, et al., 2023; Green et al., 2020) that is based upon a considerably larger number of measurements in dust source regions compared to pre-EMIT soil atlases (Claquin et 1182 al., 1999; Journet et al., 2014). A surface erodibility constraint is also imposed using the GUM 1183 dataset (Börker et al., 2018). 1184 1185 The model results compared to observations at varying distances from sources (Formenti & Di Biagio, 2024) indicate a reasonable representation of dust size distribution (Figure 5), including 1186 particles larger than 10 and up to 70 µm at emission. This improvement is mainly due to the 1187 introduction of new modes and a reduction in the modeled gravitational settling velocity of dust 1188 particles in the last three dust modes (corresponding to the largest diameters), consistent with 1189 1190 previous studies (Drakaki et al., 2022; Ginoux, 2003; Meng et al., 2022), whereas the impact of 1191 the updated mass extinction efficiency associated with EMIT-derived iron oxides is negligible. With such improvement, the updated model maintains the overall dust cycle accuracy of MAM4 1192 1193 for particles under 10 or 2.5 µm when compared with observations for dust optical depth at 1194 visible wavelengths (Figure 1), surface elemental aluminum concentrations (a proxy for dust aerosols) (Figures 2 and 3), and total dust deposition (Figure 1). 1195 The new MAM10 also more realistically simulates the seasonal variation of visible-band dust 1196 1197 optical depth (Figure 4) and the distribution of dust across size ranges (Figure 8), addressing a key limitation in MAM4. In MAM4, a realistic PM_{2.5} results in an unrealistic mass of dust at 1198

1199	larger diameters due to the influence of a single coarse mode representing both PM _{2.5} and coarse
1200	particles. By introducing additional modes at larger diameters, MAM10 allows the coarser dust
1201	fraction to vary independently of the fine fraction (accumulation mode), which now more
1202	appropriately dominates PM _{2.5} .
1203	Although our results regarding dust larger than 10 µm in diameter are subject to large uncertainty
1204	(as discussed below), they emphasize the importance of simulating a fuller dust size distribution
1205	than what was typically done in previous studies (Ginoux et al., 2001; Li et al., 2022; Zender,
1206	Bian, et al., 2003) for accurately representing the dust cycle and its climatic effects. Dust
1207	particles coarser than 10 or 20 μm need to be included in both regional and global models for
1208	two reasons.
1209	First, they substantially influence the simulated dust mass (Table 4 and Figure 9) (Adebiyi et al.,
1210	2023) (Table 4). Including these particles will revise previous estimates of dust source and
1211	deposition budgets, and dust longwave direct radiative effect upward (Albani et al., 2014; Kok,
1212	et al., 2021; Kok et al., 2023; Li et al., 2022).
1213	What is more important is that, even though do not proportionally affect the visible-band dust
1214	optical depth as to the dust deposition flux, dust particles coarser than 10 or 20 μm in diameter
1215	still can constitute a considerable fraction of the visible-band dust optical depth regionally
1216	(Figure S5 in Supporting Information). The common practice of tuning models to match a global
1217	visible-band dust optical depth target (e.g., 0.030) while omitting dust larger than 10 μm in
1218	diameter can introduce compensating biases, particularly in dust source regions. Specifically,
1219	models may overestimate the emissions of fine dust to reproduce the observed dust optical depth
1220	in the visible band, leading to an undue enhancement of long-range dust transport, surface
1221	cooling, and PM _{2.5} concentrations over regions like the Sahel and Sahara. These biases highlight
1222	the importance of explicitly representing dust larger than $10~\mu m$ in diameter, not only to refine
1223	dust source and sink estimates but also to improve assessments of dust-climate impacts through
1224	dust-radiation interactions (Kok et al., 2017) and effects on land and ocean biogeochemistry
1225	(Jickells et al., 2005; Mahowald, 2011; Mahowald et al., 2010, 2018; Mccutcheon et al., 2021;
1226	Meskhidze et al., 2005).
1227	Regarding the dust mineral composition, while a systematic, clean comparison of simulated
1228	mineralogy using EMIT and pre-EMIT soil mineral atlases is reserved for a forthcoming study,

1229	this study highlights the effects of key model developments on simulated mineral fractions and
1230	their consistency with observations. Specifically, the new MAM10 extends the capabilities of
1231	MAM4 by enabling the simulation of minerals in dust aerosol particles larger than 10 or 20 μm
1232	in diameter, although challenges remain in reproducing observed mineral mass fractions across
1233	different size categories (Figures 7 and S4 in Supporting Information). This improvement,
1234	compared to previous studies, provides insights into the climatic impacts of dust, which remain
1235	highly uncertain, especially when considering surface soil mineralogy (Li et al., 2021) and dust
1236	size distribution (Kok et al., 2017; Mahowald et al., 2014). The revised assessment of these
1237	impacts, along with more detailed mineral evaluations, will be discussed in our forthcoming
1238	paper(s).
1239	The remaining discrepancies between modeled and observed dust mineral aerosols may stem
1240	from the known deficiencies in the EMIT soil mineral atlas (e.g., misidentification of certain
1241	nano-hematite-nano-goethite mixtures as jarosite) and from the representations of iron oxides in
1242	the model (e.g., omission of nano-iron oxides). Furthermore, the EMIT instrument cannot
1243	directly detect quartz and feldspar, as these minerals lack diagnostic VSWIR (Visible to
1244	Shortwave Infrared) absorption features. Instead, their presence is inferred in part from existing
1245	soil mineralogy atlases, which carry substantial uncertainties in representing their abundances in
1246	clay- and silt-sized categories. All these limitations, whose significance may depend on particle
1247	size, also hinder models from accurately capturing the size dependence of key mineral
1248	components (e.g., iron oxides and K-feldspar), and, consequently, from reliably quantifying
1249	dust's climatic impacts. Additional sources of error include inaccuracies in dust size distribution,
1250	emission, transport, and deposition processes; observational uncertainties (including the limited
1251	availability of mineralogical composition measurements for dust aerosols); and the model-
1252	observation comparison methodology. Therefore, these discrepancies do not necessarily provide
1253	a complete reflection of the accuracy of the soil mineralogical characterization in the EMIT atlas.
1254	Despite the advancements, several challenges remain that hinder the model's ability to accurately
1255	simulate dust cycles with a physical basis, particularly in reproducing dust size distributions.
1256	First, there is a lack of precise measurements of the size distribution of mineral dust in the size
1257	range larger than 20 μm in diameter, especially in source regions (Formenti & Di Biagio, 2024),
1258	limiting the ability to constrain emission and atmospheric abundance of dust (mineral) aerosols

1259	in this size range (primarily applied to quartz, feldspar and possibly carbonates. Iron oxides and
1260	phyllosilicates are typically small, less than a couple microns). This scarcity stems from various
1261	factors, including a historical underestimation of the importance of larger dust particles,
1262	instrumental challenges (e.g., low sampling efficiencies of instrument inlets) for airborne
1263	measurements (Adebiyi et al., 2023), and the rarity of field campaigns equipped to capture a full
1264	dust size distribution, given that instruments, such as light optical aerosol counters (Ryder et al.,
1265	2013, 2019; Ryder et al., 2018; Weinzierl et al., 2017; Weinzierl et al., 2009) or multiple aerosol
1266	spectrometers, only detect subsets of the dust size distribution. Consequently, our global
1267	estimates of total dust and the contributions of particles larger than 10 or 20 μm in diameter are
1268	highly uncertain, which are sensitive to the size distribution representation in the model.
1269	A second challenge involves simulating dust particles larger than $5.0~\mu m$ in diameter, which may
1270	behave differently from finer particles in terms of transport, deposition, and radiative effects. An
1271	accurate representation of their transport and deposition could be crucial for quantifying their
1272	regional and global impacts. While reducing the gravitational settling velocity of large dust
1273	particles, particularly at geometric diameters larger than 20 µm, can achieve certain
1274	improvements like those shown here (Figure 5) and in previous studies (Drakaki et al., 2022;
1275	Ginoux, 2003; Meng et al., 2022), the physical mechanisms for slowing descent of these particles
1276	remain unclear (van der Does et al., 2018), as earlier studies also noticed (Betzer et al., 1988;
1277	Mahowald et al., 2005).
1278	Recent findings suggest potential mechanisms for maintaining particles aloft, such as topography
1279	(Heisel et al., 2021; Rosenberg et al., 2014), electric forces on charged particles (van der Does et
1280	al., 2018; Toth et al., 2020), underrepresented uplifts by convection events (combining the other
1281	possible mechanisms or multiple uplifts occurring within a long-lived system) (van der Does et
1282	al., 2018) or turbulence (radiation- or shear-induced) in the free troposphere (Rodakoviski et al.,
1283	2023), horizontal and vertical spatial resolutions of models (Feng et al., 2022) or aerosol splitting
1284	processes in models (Wan et al., 2024), and variations in particle shape and orientation (Bagheri
1285	& Bonadonna, 2016; Huang et al., 2020; Mallios et al., 2020). Testing these mechanisms is
1286	beyond the scope of this study but deserves future investigations. Here, we only approximate the
1287	effects by adjusting the gravitational settling velocity for dust particles represented by the last
1288	three dust modes corresponding to the largest diameters. However, this adjustment is applied
1289	uniformly across regions, ignoring potential spatial variability. Probably in part because of the

1290	uniform adjustment, the model remains unable to reproduce the slow removal rate of giant dust
1291	particles during source-to-medium-range transport (Figure 5).
1292	The third challenge is on the accurate representation of the physical (e.g., optical) properties of
1293	dust particles larger than 20 μm in diameter based on their mineral composition, which affects
1294	dust climatic impacts. Translating soil composition into aerosol mineralogy is particularly
1295	challenging for iron oxides and quartz in giant dust aerosol particles due to the scarcity of
1296	measurements and the need for advanced techniques. Dust aerosol particles larger than 20 μm in
1297	diameter are difficult to sample and analyze compared to particles smaller than 10 μm in
1298	diameter. Uncertainties stemming from the representation of atmospheric processes, such as
1299	chemical reactions and cloud processing (Langmann, 2013; Longo et al., 2016; Usher et al.,
1300	2003), are likely limited, given the low number concentration and short lifetime of dust particles
1301	larger than 20 μm.
1302	Fourth, several factors influencing the modeled dust size distribution, such as the prescribed
1303	geometric standard deviation as a global constant and settling velocity for particles larger than
1304	5.0 µm in diameter, remain poorly constrained. Empirical tuning is used in this study to match
1305	observed size distributions. While observationally derived geometric standard deviations can be
1306	prescribed, missing or inaccurate physics, such as mechanisms for maintaining particles aloft, or
1307	other unknown errors may have hindered achieving the intended outcomes. Therefore, we
1308	adjusted dust parameters to match the observed size distribution. However, there is still room for
1309	further refinement of either dust mode parameters or the gravitational settling velocity (Figure 5),
1310	which might lead to considerably different results.
1311	Additional approaches to further constrain super coarse and giant dust particles are to match
1312	observed dust optical depth at longwave wavelengths or effective diameter measurements
1313	(Zheng et al., 2022). However, uncertainties in the dust optical depth retrievals at longwave
1314	wavelengths could make this challenging. Sparse observations of effective diameters—limited
1315	by assumptions such as mono-modal particle size distributions, the near absence of reliable
1316	observations over land (Zheng et al., 2022), and uncertainties in the emitting temperature of dust
1317	layers, partly due to substantial variability in the vertical structure of dust plumes from one event
1318	to another (Li & Sokolik, 2018)—further complicate this method. From these perspectives,
1319	future constraints on effective diameter retrievals could potentially reduce uncertainties in

1320	modeling the large (especially diameters larger than $20~\mu m$) dust particle cycle as well, in
1321	addition to those in longwave dust radiative effect estimates. These approaches are deferred to
1322	future investigations.
1323	Fifth, reproducing the dust cycle across different environments and measurements remains
1324	challenging. The complex interactions of factors like surface winds, vegetation, and soil moisture
1325	dynamics (Kok et al., 2012; Shao, 2008) govern dust emission, transport, and deposition.
1326	Uncertainties in model parameterization that affect these processes further complicate
1327	predictions for different dust phases. As a result, like MAM4 (Li et al., 2022), the new model
1328	cannot match the observed dust deposition, visible-band dust optical depth, and surface
1329	aluminum elemental PM_{10} concentrations simultaneously (Figures 1 and 3), which has been
1330	recognized for more than a decade (Huneeus et al., 2011).
1331	Sixth, MAM10 does not include the effect of preexisting ice crystals in cirrus considered in
1332	CAM5 (Shi et al., 2015) for dust of all size ranges, which is deferred for future studies.
1333	Excluding this mechanism could lead to an overestimation of simulated ice nuclei concentrations
1334	as preexisting ice crystals may inhibit homogeneous freezing. In addition, dust-cloud interactions
1335	(DeMott et al., 2003; Karydis et al., 2011) are not included in this model for particles with
1336	diameters larger than 20 µm. However, their impact is likely minimal, as cloud effects scale with
1337	aerosol number concentrations (Andreae & Rosenfeld, 2008), which are very low for dust larger
1338	than 20 µm in diameter.
1339	Lastly, MAM10 does not account for additional constituents except sulfate that may be internally
1340	mixed with dust aerosols, such as soot and primary biological particles (e.g., microorganisms and
1341	biological material fragments) (Kellogg & Griffin, 2006; Shinn et al., 2000), which exhibit
1342	regional contrasts. The mixing of these species with dust aerosols is excluded from the MAM10
1343	framework presented here. Below, we outline the specific reasons, using sea salt as an example;
1344	however, future studies may need to investigate the impact of an alternative treatment, in which
1345	dust is internally mixed with these species, on simulations of the dust cycle and its climatic
1346	effects.
1347	The external mixing of dust with the other species except sulfate is inherited from previous
1348	aerosol model versions, MAM7 and MAM9, for simplicity. Here, we advance the model to

1349	simulate a fuller and more accurate size distribution of dust aerosols. While MAM4 simulates
1350	dust as a mixture with other components, such as sea salt, the mechanisms driving the mixing of
1351	dust particles and sea salt remain insufficiently detailed (Andreae et al., 1986; Zhang et al.,
1352	2005). The mixing state of dust and sea salt also depends on the region. Observations show
1353	infrequent internal mixing of African dust with sea salt, particularly during peak dust outbreak
1354	seasons when the largest transport occurs across the Atlantic Ocean (Kandler et al., 2007; Reid et
1355	al., 2003) and the dust plume is elevated. In these scenarios, African dust particles are typically
1356	transported above the marine boundary layer (Karyampudi et al., 1999) and dust layers remain
1357	distinct from it (Reid et al., 2002), resulting in frequently low sea salt mass fractions (less than
1358	10%) in dust plumes (Chiapello et al., 1999). In contrast, East Asian dust transported eastward
1359	shows higher degrees of internal mixing with soluble salts (Niimura et al., 1998), including sea
1360	salt, ranging from 18 to 42% by volume (Osada, 2013). Over 60% of dust particles collected at
1361	Japanese islands, after several hours of transport through marine air, were found internally mixed
1362	with sea salt (Okada et al., 1990; Niimura et al., 1998; Zhang et al., 2003b). Based upon these
1363	findings, the separation of dust and sea salt in MAM10 may be not a good assumption for East
1364	Asian dust aerosols, potentially biasing estimates of their wet removal rates and climatic effects.
1365	Moreover, observations on the mixing state of dust with sea salt, especially in the Southern
1366	Ocean, are far more limited. For the mixing of primary biological particles with dust aerosols,
1367	some attempts exist (Adachi et al., 2020; Brodsky et al., 2023), but the results remain
1368	preliminary.
1369	Beyond addressing current limitations, the new model developed in this study allows a more
1370	accurate and complete representation of the dust climatic impacts than previous CAM versions.
1371	It complements existing models (Di Biagio et al., 2020, Ke et al., 2022, Wang et al., 2024) in
1372	quantifying the role of super coarse and giant dust particles in dust-radiation interactions and
1373	their influence on modeling trace metal deposition, which is critical for terrestrial and ocean
1374	biogeochemistry (Brodsky et al., 2023; Lu et al., 2024; Mahowald et al., 2018; Wong et al.,
1375	2021).
1376	The modeling framework developed here not only improves the representation of large dust
1377	particles but could also be used to simulate other aerosols with broad size distributions, such as
1378	volcanic ash, whose size often exceeds 10 μm in diameter (Brown et al., 2012).

Recognizing all the unresolved issues, it is imperative for future research endeavors to 1379 encompass a more comprehensive analysis, incorporating a broader range of species present in 1380 1381 dust aerosols and a more advanced, physically based modeling approach to simulate large-size dust aerosols with observational constraints. This refinement of the modeling approaches in the 1382 future will help improve accuracy in regional and global climate assessments, enhancing our 1383 1384 understanding of aerosol composition and their roles in the Earth-Human-Climate system. Acknowledgments 1385 L.L., N.M.M., R.L.M., and R.N.C. received support from the NASA EMIT project. EMIT is 1386 supported by the NASA Earth Venture Instrument program under the Earth Science Division of 1387 1388 the Science Mission Directorate. L.L. and N.M.M. also acknowledge assistance from the 1389 Department of Energy (DOE) under grant DE-SC0021302, the high-performance computing resources from Derecho provided by National Center for Atmospheric Research (NCAR)'s 1390 1391 Computational and Information Systems Laboratory (CISL), sponsored by the National Science Foundation (NSF), and a NASA Interdisciplinary Science (IDS) project under grant 1392 80NSSC20K1674. C.P.G.P., M.G.A., and V.O. acknowledge funding by the European Research 1393 Council under the Horizon 2020 research and innovation program through the ERC Consolidator 1394 1395 Grant FRAGMENT under grant 773051, the AXA Research Fund through the AXA Chair on Sand and Dust Storms at BSC, and the EuropeanUnion's Horizon 2020 research and innovation 1396 1397 program under grant agreement No 821205 (FORCeS). C.P.G.P and V.O. also acknowledge Grant PID2022-140365OB-I and M.G.A. acknowledges Grant PID2022-139362OB-I00, both 1398 funded by MICIU/AEI/10.13039/501100011033 and by ERDF, EU. R.L.M. also received 1399 support from the NASA Modeling, Analysis, and Prediction Program under grant 1400 NNG14HH42I. J.F.K. acknowledges support from the NSF Directorate for Geosciences under 1401 1402 grants 1856389 and 2151093. P.F. and C.D.B. received support from the DustClim project, 1403 which is part of ERA4CS, an ERA-NET project initiated by JPI Climate and funded by FORMAS (SE), DLR (DE), BMWFW (AT), IFD (DK), MINECO (ES), and ANR (FR) with co-1404 1405 funding from the European Union's Horizon program under grant 690462. P.F. and C.D.B. also received support from the French national program LEFE/INSU (Les Enveloppes Fluides et 1406 1407 l'Environnement/Institut National des Sciences de l'Univers) and by the OSU-EFLUVE (Observatoire des Sciences de l'Univers-Enveloppes Fluides de la Ville à l'Exobiologie) through 1408

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1420	Author Contributions
1421	L.L. conceived the study, developed the MAM10 code in CAM6, performed the model
1422	simulations, conducted the formal analysis, generated the figures, and wrote the initial
1423	manuscript draft. N.M.M. provided the elemental data and supervised the work. X.L. and Z.K.
1424	supplied the MAM9 code in CAM5. M.G.A. conducted the mineral evaluation. D.M.L. and
1425	J.F.K. supplied the new dust emission scheme code. M.G.A., C.P.G.P., R.L.M., V.O., P.G.,
1426	P.G.B., D.R.T., R.N.C., G.S.O., R.O.G., and B.Z. contributed to the development of the EMIT
1427	soil mineral atlas. S.A. provided the post-processed AERONET data, and A.A.A. derived the
1428	geometric standard deviations for the five dust modes in MAM10, which L.L. subsequently
1429	retuned to improve model performance. All authors reviewed and provided comments on the
1430	original manuscript draft.
1431	Code and Data Availability Statement
1432	The core component of the CAM6-MAM4 and observations (AERONET dust aerosol optical
1433	depth, surface concentrations, and deposition fluxes) are available in a publicly accessible
1434	repository at https://doi.org/10.5281/zenodo.6989502 and are documented in detail by Li et al.
1435	(2022). The MAM10 code will be available in a publicly accessible repository. The EMIT soil
1436	mineralogy data is publicly available through the NASA LP DAAC, subject to their data use
1437	policy. The elemental aluminum data is detailed by Mahowald et al. (2024) and archived with
1438	the link of https://zenodo.org/records/11391232 . Detailed documentation of the observed mineral

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