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2	Size-Resolved Dust Direct Radiative Effect Efficiency
3	<b>Derived from Satellite Observations</b>
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#### 26 Abstract

27 The role of mineral dust aerosol in global radiative energy budget is often quantified by 28 the dust direct radiative effect (DRE). The dust DRE strongly depends on dust aerosol optical depth (DAOD), therefore, DRE efficiency (DREE=DRE/DAOD) is widely compared across 29 30 different studies to eliminate difference due to the various dust load. Nevertheless, DREE is still influenced by the uncertainties associated with dust particle size distribution (PSD) and optical 31 32 properties. In this study, we derive a global clear-sky size-resolved DREE dataset in both shortwave (SW) and longwave (LW) at top of the atmosphere (TOA) and surface based on satellite 33 34 observations (i.e., satellite-retrieved dust extinction spatial and vertical distributions). In the DREE 35 dataset, dust geometric diameter from 0.1µm to 100 µm is divided into 10 bins and the 36 corresponding monthly mean DREE (with respect to DAOD at 532nm) for each size bin is derived by using the Rapid Radiative Transfer Model (RRTM). Three sets of state-of-the-art dust refractive 37 38 indices (RI) and two sets of dust shape models (sphere vs. spheroid) are adopted to investigate the 39 sensitivity of dust DREE to dust absorption and shape. As a result, the size-resolved dust DREE 40 dataset contains globally distributed monthly mean dust DREE at TOA and surface for each of 10 41 size bins with 5° (longitude)  $\times$  2° (latitude) resolution as well as for each dust RI and shape 42 combination. The size-resolved dust DREE dataset can be used to readily calculate global dust 43 DRE for any DAOD and dust PSD, including the uncertainty in the DRE induced by dust microphysical properties (e.g., dust PSD, RI and shape). By calculating dust DRE based on DAOD 44 climatology retrieved from different satellite sensors and based on different dust PSD, we find that 45 uncertainty in the spatial pattern of DAOD induces more than 10% of the uncertainty in SW dust 46 47 DRE at TOA. The observation-based dust PSD induces around 15%~20% uncertainty in dust DRE 48 at TOA and in the atmosphere. The sensitivity assessments of dust DRE to dust RI and shape

- 49 further suggest that dust non-sphericity induces a negligible effect on dust DRE estimations, while
- 50 dust RI turns out to be the most important factor in determining dust DRE, particularly in SW.
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#### 52

## 53 1 Introduction

54 Mineral dust is an important component of the atmospheric aerosol (Textor et al., 2006; 55 Choobari et al., 2014). They can influence the radiative energy budget of the Earth-Atmosphere 56 system directly through their interaction with both solar and thermal infrared radiation, which is 57 known as the direct radiative effect (DRE) of dust. The DRE of dust consists of two components. 58 In the solar shortwave (SW) spectral region, dust aerosols reflect a fraction of solar radiation back to the space which generally leads to a negative cooling effect at both top of the atmosphere (TOA) 59 60 and surface (Tegen et al., 1996; Myhre et al., 2003). In the longwave (LW) thermal infrared region, 61 dust aerosols trap the thermal radiation emitted from Earth's surface by absorption, which generally leads to a positive warming radiative effect at TOA and surface (Sokolik et al., 1998). 62 In addition to DRE, dust can also influence the radiation and the hydrological cycles indirectly 63 64 through serving as cloud condensation nuclei and ice nuclei and affecting cloud microphysical properties and cloud lifetime, known as indirect effects of dust (Twomey, 1977; Albrecht, 1989). 65 66 The dust DRE depends on many factors including primarily the atmospheric dust content, represented by its optical depth (DAOD), vertical distribution (especially important for LW DRE), 67 68 and particles' physico-chemical properties that are the particle size distribution (PSD), complex refractive index (RI), and shape. Besides dust PSD, RI and shape, the dust DRE also depends on 69 70 the atmospheric composition and structure, notably the atmospheric vertical profile of clouds, 71 water vapor, and temperature, as well as surface properties (Yu et al., 2006). All of these properties 72 vary in space and time and need to be characterized at the best possible spatio-temporal resolution 73 in order to get realistic dust DRE estimates.

74 Among all these factors, DAOD is of first order importance in determining dust DRE since 75 dust DRE is approximately linear with DAOD (Satheesh and Ramanathan, 2000). Many previous 76 studies related to dust DRE are based on DAOD distributions from model simulations. For 77 example, Kok et al. (2017) used four global model simulations to estimate global mean dust DRE 78 efficiency (DREE is defined as DRE/DAOD) and further derive global mean dust DRE. Di Biagio 79 et al. (2020) derived dust DRE based on model-simulated DAOD distributions with global annual 80 mean DAOD constrained by observations. The main advantage of these studies is the availability 81 of continuous and detailed DAOD spatial and temporal variation from model simulations. On the 82 other hand, model-simulated DAOD could be subject to large uncertainties and biases in reproducing DAOD due to parameterizations of various physical processes, therefore need 83 84 observational constraints for evaluation and improvement.

85 Satellite observations are important sources of data for evaluating model simulations, because of their routine sampling on a global scale and over decadal time periods. Previous studies 86 87 have developed sensor-specific methods to distinguish dust aerosol from total aerosol based on the 88 size and shape characteristics of dust particles. Some are based on passive satellite observations 89 such as Moderate Resolution Imaging Spectroradiometer (MODIS, Remer et al. (2005)) and others are based on active observations such as Cloud-Aerosol Lidar with Orthogonal Polarization 90 91 (CALIOP, Winker et al. (2009)). The wide spectral coverage of MODIS measurements allows the 92 retrieval of aerosol particle size information, such as effective radius, fine-mode fraction, aerosol 93 Angstrom exponent, as well as spectral gradient of absorption (Remer et al., 2005; Hsu et al., 2013). 94 Based on the fact that dust aerosols are generally larger in size than other aerosols and have a decreasing absorption from ultraviolet (UV) to the near infrared, the combinations of these 95 96 retrievals provide the basis for dust separation and dust aerosol optical depth (DAOD) retrievals

97	from MODIS (Kaufman et al., 2005; Ginoux et al., 2012; Voss and Evan, 2020; Yu et al., 2009,
98	2019). In addition, some recent studies have also characterized dust distribution through
99	integrating MODIS measurements with other data sources and model simulations, for example,
100	using the DAOD-to-AOD ratio from MERRA-2 (Modern-Era Retrospective analysis for Research
101	and Applications, version 2 ), Gkikas et al. (2021) converted the MODIS AOD retrievals to DAOD.
102	However, passive sensors do not provide the vertical structure of aerosol that is critical for studying
103	aerosol-cloud interactions, LW radiative effects and aerosol influences on the thermal structure of
104	the atmosphere (e.g., Meloni et al., 2005, 2015). By contrast, the active sensor CALIOP can
105	provide the vertical profiles of aerosol extinction and particle properties such as depolarization
106	ratio and color ratio, which have been used for improving DAOD retrievals in thermal infrared
107	(TIR) (Zheng et al., 2022) and evaluating global dust simulations (Yu et al., 2010; Wu et al., 2020).
108	The CALIOP dust identification is mainly based on dust aerosols being non-spherical in shape and
109	their linear depolarization ratio being much larger than spherical aerosols (Sakai et al., 2010).
110	Using CALIOP retrievals, Song et al. (2021) derived a three-dimensional (3D) decadal
111	(2007-2019) global scale dust extinction profile climatology, which provides an observational
112	constraint on both the spatial DAOD pattern and the vertical dust distribution for studying dust
113	DRE and evaluating models. In their study, Song et al. (2021) also compared dust retrievals, in
114	particular DAOD, based on different methods (i.e., CALIOP-based and MODIS-based DAOD
115	retrievals), showed that DAOD often differ significantly between the different products, and further
116	discussed the potential reasons of causing the differences (e.g., instrument calibration errors and
117	errors in discriminating cloud from aerosol, globally uniform dust Lidar Ratio assumption in
118	CALIOP DAOD retrieval and so on). They showed that DAOD derived from CALIOP
119	observations is generally smaller and more concentrated over 'dust belt' regions - extending from

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122 the west coast of north Africa to the Middle East, central Asia, and China - than that derived from 123 MODIS observations. These differences in DAOD in turn lead to different dust DRE estimations, 124 making it difficult to compare different studies to reach meaningful conclusions. Even an 125 agreement of DRE could be a result of the compensation between differences in DAOD and other 126 aforementioned factors, such as dust microphysical properties. Therefore, DRE provides only a 127 weak constraint on model. Instead, a normalized quantity, DRE efficiency (DREE) as the ratio of 128 DRE to DAOD, has been widely used in inter-comparison studies and model evaluations (Di 129 Biagio et al. 2020). Because of the elimination of DAOD, the DREE provides a stronger constraint 130 on dust microphysical properties and their impacts on the dust DRE from different dust source 131 regions (García et al., 2008).

132 In addition to DAOD, dust size is also an important factor in determining dust DRE 133 (Mahowald et al., 2014). Smaller particles are more effective at scattering SW radiation and super-134 micron particles are more effective at absorbing both SW and LW radiation (Tegen and Lacis, 135 1996). Therefore, when other parameters are equal, fine dust would generally have a more negative 136 SW DRE and a less positive LW DRE than coarse dust. Unfortunately, despite its importance, the 137 simulation of dust PSD in the models and satellite retrievals of dust size remain challenging tasks 138 (Ryder et al., 2019). As a result, there is a large uncertainty in our understanding of dust PSD. For 139 example, several recent studies suggested that model simulations tend to underestimate dust size, 140 especially the very coarse dust with diameter in excess of 5 µm (Adebiyi and Kok, 2020). 141 Moreover, dust RI and shape can be important for DRE estimation as well because besides dust 142 PSD they are the other two factors that determine dust spectral optical properties. As such, it is 143 important to investigate the sensitivity of dust DRE to dust PSD, RI and shape. Previous studies 144 suggest that large dust PSD and RI uncertainty leads to a large uncertainty in dust DRE and thereby DREE estimations. For example, Song et al., (2018) shows that the SW DREE of a dust model with a large size and less absorptive RI is very similar to that of a dust model with a smaller size and more absorptive RI, both in the range of satellite derived values in the NE Atlantic region. Not surprisingly, even DREE cannot provide sufficient constraints due to this possible compensation of effects in the dust PSD and RI.

150 The main objective of this study is to derive a global clear-sky size-resolved dust DREE 151 dataset based on satellite observations and demonstrate its usefulness in constraining, comparing, 152 and understanding the dust DRE estimations. As explained below, the size-resolved DREE 153 decomposes the DREE of dust into several size bins and therefore provide a way to take into 154 account the effects of dust PSD explicitly. The sensitivity of dust DRE to dust RI and shape are 155 also assessed in this study. Due to the inhomogeneous spatio-temporal distribution of those 156 aforementioned factors, it is thus important to consider the spatio-temporal variation of dust DREE. 157 Therefore, we organize the DREE dataset at 5° (longitude)  $\times$  2° (latitude) horizontal resolution 158 and at monthly temporal resolution. To the best of our knowledge, this work presents the first such 159 dataset based on retrieved dust properties (i.e., DAOD vertical and horizontal distributions) from 160 satellite observations, although size-resolved DREE from model simulations have been used in 161 previous studies. We will show that our size-resolved DREE can allow users to readily compute 162 the DREE and DRE of dust based on any dust PSD (e.g., from model simulations, satellite 163 retrievals or in-situ measurements). We will also carry out an inter-comparison of the global dust 164 DRE estimations based on different dust PSD and compare the results with previous studies. With 165 these functions, we expect that the size-resolved DREE will be a useful tool for both observational and modeling studies of dust DRE. 166

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167	The rest of the paper is organized as follows. Section 2 provides a description of the data
168	and models used in this study. Section 3 describes the methodology of deriving the size-resolved
169	DREE dataset. In section 4, we describe a methodology of calculating the dust DRE with the size-
170	resolved DREE dataset and its validation. In section 5, we compare the regional and global dust
171	DRE estimations based on different DAOD, dust PSD and compare the results with previous
172	studies. Section 6 provides a summary of the study along with the main conclusions.

#### 173 2 Data and Models

#### 174 2.1 Satellite-based DAOD climatology

175 We use CALIOP-based DAOD climatology and dust vertical distribution derived in Song 176 et al. (2021) to derive a size-resolved dust DREE. The reason for choosing CALIOP-based DAOD 177 climatology is discussed in detail in section 3.2. The CALIOP-based dust climatology dataset contains monthly mean DAOD and dust vertical extinction profile on a 5° (longitude)  $\times$  2° 178 (latitude) spatial resolution grid for the period 2007-2019. The CALIOP-based DAOD and dust 179 180 vertical distribution climatology from 2007 to 2010 are used to derive monthly mean size-resolved 181 dust DREE dataset in this study. The selection of 4 years (2007-2010) for DREE calculations is 182 based on several considerations. Firstly, the multi-year DREE calculations allow us to investigate 183 the effect of interannual variations of atmospheric and surface properties to dust DRE. Secondly, 184 this selection is consistent with Song et al. (2018), making it easier to compare our results with 185 previous work. Thirdly, considering the computational efficiency, we do not extend the calculation 186 to more years.

In addition to CALIOP-based DAOD climatology, we will use the MODIS-based DAOD
 climatology to investigate the sensitivity of dust DRE to DAOD spatial pattern in section 5.2. The
 MODIS-based DAOD climatology achieves global coverage on a 5° (longitude) × 2° (latitude)

190	spatial resolution for the period 2003-2019 by combining the monthly mean Aqua MODIS over-
191	ocean (Yu et al., 2020) and over-land (Pu and Ginoux, 2018) DAOD. In contrast to CALIOP-based
192	DAOD climatology which is based on dust non-sphericity to separate dust aerosol from CALIOP
193	total aerosol observations, MODIS-based DAOD retrieval is mainly based on dust large size to
194	partition DAOD from MODIS total aerosol observations. The two sensor-specific dust partition
195	methods result in different DAOD magnitude and spatial pattern retrievals.

196 Figure 1 shows annual mean DAOD from 2007 to 2010 based on CALIOP and MODIS 197 observations. CALIOP-based and MODIS-based DAOD climatology differ in terms of both 198 magnitude and spatial pattern. MODIS-based DAOD is generally larger than CALIOP-based DAOD. For example, the global  $(60^{\circ}S - 60^{\circ}N)$  4-year mean MODIS-based DAOD is 0.047, while 199 200 CALIOP-based DAOD is 0.032. High DAOD are seen from both CALIOP-based and MODIS-201 based DAOD over the 'dust belt' regions, where large-scale dust activities occur persistently 202 throughout the year. However, the CALIOP-based DAOD is rather low in some other regions that 203 are known to be dusty in certain seasons, such as South America, Australia, and South Africa. In other words, the two satellite-based DAOD spatial pattern differs significantly with CALIOP-204 205 based DAOD more concentrated over 'dust belt' regions.



Figure 1. Global  $(60^{\circ}S - 60^{\circ}N)$  spatial pattern of CALIOP-based and MODIS-based 4-year (2007-2010) mean DAOD (Song et al., 2021) and their difference.

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#### 212 2.2 Dust physical and optical models

213 To study the sensitivity of dust DREE to dust RI and dust shape, we adopt three sets of 214 dust RI (Figure 2) and two dust shapes and compute a total of 6 sets of DREE based on their 215 combinations. The three dust RI sets represent less absorptive, mean absorptive and more 216 absorptive dust aerosols and the two dust shapes include spherical and spheroidal dust shapes (dust 217 shape distribution is shown in Figure 4 (a) in Song et al. 2018). The mean, 10th and 90th percentile 218 of calculated RI for 19 dust samples over 8 regions in Di Biagio et al. (2019) are used to represent 219 mean, less and more absorptive dust in SW. We combine RI of wavelengths from  $0.37 \mu m$  to 220  $0.95\mu m$  measured in Di Biagio et al. (2019) and RI of other wavelengths up to  $3\mu m$  reported in 221 Balkanski et al. (2007) to get full spectral coverage in SW. The mean, minimum and maximum RI 222 of wavelengths beyond  $3\mu m$  measured in Di Biagio et al. (2017) are used to represent mean, less 223 and more absorptive dust in LW. Two dust shapes are used to investigate the effect of dust 224 nonsphericity on dust DRE. One is spherical dust shape, the other one is spheroidal dust shape 225 with dust aspect ratio distribution described by Figure 4 (a) in Song et al. (2018) which is originally 226 from Dubovik et al. (2006). Each combination of dust RI and dust shape is considered as a dust 227 model. As a result, the three dust RI and two dust shapes constitute six dust models in SW and 228 LW, respectively, as shown in Table 1.

229

1.60 Real part of RI (n) 1.55 1.50 1.45 1.40 0.5 1.0 1.5 2.( --01 [k] --01 [k] 0.5 1.0 1.5 2.( Wavelength (µm Deleted:

Deleted: (Figure 4 (a) in Song et al. 2018)

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Figure 2. The SW and LW spectral refractive indices (RI) used in this study<u>obtained from Di Biagio et al. (2017,2019)</u> and Balkanski et al. (2007). The black curves represent the mean RI which indicates the mean absorptive dust. The grey shading represents the upper and lower limits indicating more absorptive and less absorptive dust, respectively.

Table 1. Dust models used in this study. Three dust RI are used in shortwave (SW) and longwave (LW) to represent less, mean, and more absorptive dust, respectively. Two dust shape models are used to represent spherical and spheroidal dust shape. The three dust RI sets and two dust shapes constitute 6 dust models in SW and LW respectively.

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**Deleted:** References for the used datasets are provided in Section 2.2.

SW RI LW RI (Balkanski et al. 2007; Di Biagio et al. 2019) (Di Biagio et al. 2017) 90% Minimum 10% Mean Mean Maxmum MinSWRI MeanSWRI MaxSWRI-MinLWRI-MeanLWRI MaxLWRI-Sphere Sphere MinSWRI-Sphere MeanSWRI-Sphere MaxSWRI-Sphere MinLWRI-Sphere MeanLWRI-Sphere MaxLWRI-Spheroid Spheroid Spheroid Spheroid Spheroid Spheroid Spheroid

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# 241 3 Methodology

#### 242 3.1 Size-resolved dust scattering properties

243 Rapid Radiative Transfer Model (RRTM) (Mlawer et al., 1997) is used to compute both

244 SW and LW radiative fluxes for both clean (i.e., cloud-free and aerosol-free) and dusty

245 atmospheres (i.e., free of clouds and non-dust aerosols). RRTM retains reasonable accuracy in

249 comparison with line-by-line results for single column calculations (Mlawer and Clough, 1998; 250 Mlawer et al., 1997). It divides the solar spectrum into 14 continuous bands ranging from 0.2 to 251 12.2 µm and the thermal infrared (3.08–1000 µm) into 16 bands. We explicitly specify the spectral 252 DAOD, single scattering albedo ( $\omega$ ), and asymmetry parameter (g) of dust aerosols for every band 253 in the RRTM radiative transfer simulations. In contrast to radiative transfer scheme in most global 254 models, which do not account for LW scattering, scattering capability is available through the 255 discrete-ordinate-method radiative transfer (DISORT) in RRTM LW (Stamnes et al., 1988). Four 256 streams are used in DISORT. The Henyey-Greenstein phase function is used and only the first 257 moment of the phase function (i.e., asymmetry parameter) needs to be specified in the RRTM.

258 Dust scattering properties (extinction efficiency  $Qe, \omega$  and g) depend on several factors 259 including dust PSD, RI, and dust shape. To account for the impact of dust PSD, we divide dust 260 diameters into 10 logarithmically spaced size bins. The 10 size bins represent a wide range of dust 261 geometric diameters (i.e., diameter of a sphere with the same volume) ranging from  $0.1 \mu m$  to 262  $100\mu m$ . The geometric diameter (hereafter diameter or D) range of each size bin is listed in Figure 3. For each size bin k, the spectral scattering properties ( $Qe_k^{\lambda}, \omega_k^{\lambda}$  and  $g_k^{\lambda}$ ) are calculated for each 263 264 dust model shown in Table 1 and each spectral band. In the calculations of scattering properties 265  $(Qe_k^{\lambda}, \omega_k^{\lambda} \text{ and } g_k^{\lambda})$ , dust particle number (dN/dD) is assumed to be uniformly distributed within 266 each size bin. We use the Lorenz-Mie theory code of Wiscombe (1980) to compute the spectral 267 optical properties of dust particles in the assumption of sphericity. The spectral optical properties 268 of spheroidal dust particles are derived from the database of Meng et al. (2010). Figure 3 shows  $Qe_k^\lambda, \omega_k^\lambda$  and  $g_k^\lambda$  for MeanSWRI-MeanLWRI-Spheroid dust model. In SW, finer dust has a larger 269 270  $\omega$  and smaller g, implying a more effective SW backscattering of finer dust. As a result, finer dust 271 is expected to have stronger cooling effect (more negative DREE values) at TOA generally. In





279Figure 3. Spectral scattering properties (i.e.,  $Q_e$ : extinction efficiency,  $\omega$ : single scattering property, g: asymmetry280parameter) of each size bin for the MeanSWRI-MeanLWRI-Spheroid dust model. The scattering properties of each281size bin are represented by the corresponding curve indicated in the legend. Each size bin is defined with respect to282dust diameter with unit of micrometers ( $\mu m$ ).

#### 283 3.2 DREE dataset

Based on the dust scattering properties shown in Figure 3 and the procedures summarized in Figure 4, we compute the size-resolved dust DREE for the MeanSWRI-MeanLWRI-Spheroid dust model in SW and LW. In this section, we focus on demonstrating the method of deriving sizeresolved dust DREE for one dust model, but this method is applicable to all six dust models listed in Table 1.

289 First, we use RRTM to simulate monthly mean dust DRE from 2007 to 2010 for each 5° (longitude)  $\times$  2° (latitude) grid with CALIOP-based DAOD<sup>532nm</sup> exceeding 0.01. The 290 291 DAOD<sup>532nm</sup>>=0.01 threshold ensures most dusty regions over the globe are covered (see Figure 292 S1 and Figure S2 in the Supplement) and in the meanwhile balances the computational cost. Dust 293 DRE are calculated for each size bin using the extinction properties of the corresponding size bin 294 shown in Figure 3 (denoted as  $DRE_{k,i,j}$ , hereafter k indicates size bin index and (i, j) indicates 295 longitude-latitude grid index, unless specified otherwise). Note that we do not consider dust RI 296 spatial variation and dust size vertical variation due to the lack of observation-based dust 297 minerology and size estimation on global scale. In  $DRE_{k,i,j}$  calculations, we constrain the monthly 298 mean dust extinction vertical distributions using the CALIOP-based climatological dataset of Song 299 et al. (2021). Worth to mention, our target in this section is DREE<sub>k,i,i</sub> calculations. Considering 300 dust DRE is approximately linear to DAOD (Satheesh and Ramanathan, 2000), the DAOD used 301 in dust DRE calculations will not affect dust DREE results significantly, we simply calculate dust  $DRE_{k,i,j}$  with respect to  $DAOD_{i,j}^{532nm}$  from CALIOP-based DAOD climatology. As a result, 302 303  $DRE_{k,i,j}$  calculated in this section are only intermediate variables used to calculate dust DREE, 304 they do not represent actual DRE contributed by kth size bin. The atmospheric profiles such as 305 water vapor (H<sub>2</sub>O), ozone (O<sub>3</sub>) and temperature ( $T_{atm}$ ) vertical profiles of 72 levels are from 3-

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308	hourly MERRA2 assimilated meteorological fields data (Gelaro et al., 2017). We combine the 1-
309	hourly surface albedo for visible beam from MERRA2 radiation diagnostics with the instantaneous
310	spectral surface albedo from the integrated CALIPSO, Cloud-Sat, CERES, and MODIS merged
311	product (CCCM) (Kato et al., 2011) to get time-dependent spectral surface albedo. Surface
312	temperature is obtained from 1-hourly MERRA2 radiation diagnostics data. The atmospheric and
313	surface properties are all aggregated to monthly mean values at eight UTC times: 0:30, 3:30, 6:30,
314	9:30, 12:30, 15:30, 18:30, 21:30 to obtain monthly-mean diurnal cycle for radiative transfer
315	simulations. Considering DRE <sup>SW</sup> strongly depends on solar zenith angle (SZA), we calculate
316	DRE <sup>SW</sup> for every 1 hour using the corresponding hourly SZA in midmonth day. As a result, every
317	three SZA share the same atmospheric and surface properties in DRE <sup>SW</sup> calculations due to their
318	different temporal resolution.

319 Table 2 List of definitions of variables and their indices.

Variable	Definition				
k	size bin index				
i, j	longitude-latitude grid index				
t	8 UTC times with 3-hour interval (i.e., 0:30, 3:30, 6:30, 9:30, 12:30, 15:30, 18:30, 21:30)				
tt	24 UTC times with 1-hour interval				
day <sup>mm</sup>	The midmonth day of the month				
$\overline{R(t)}, \overline{H_2O(t)}, \overline{O_2(t)},$	3-hourly monthly mean surface albedo and vertical profile of water vapor, ozone, carbon				
$\frac{CO_2(t)}{CO_2(t)}, T_{atm}(t)$	dioxide and atmospheric temperature				
$\zeta_d$	dust properties such as DAOD, dust extinction vertical profile and scattering properties				
$DRE_{SW}^{SW}(tt)$	1-hourly monthly mean DRE <sup>SW</sup> (i.e., monthly mean DRE <sup>SW</sup> at each of 24 UTC times) of				
$1h^{D} \prod_{k,l,j} (00)$	k <sup>th</sup> size bin and (i <sup>th</sup> , j <sup>th</sup> ) grid				
$DRE_{i}^{LW}(t)$	3-hourly monthly mean DRE <sup>LW</sup> (i.e., monthly mean DRE <sup>LW</sup> at each of 8 UTC times) of k <sup>th</sup>				
$3h^{D}M^{D}k_{k,l,j}(t)$	size bin and (i <sup>th</sup> , j <sup>th</sup> ) grid				
$\overline{DRE_{k,i,j}^{SW}}, \overline{DRE_{k,i,j}^{LW}}$	The monthly and diurnally mean dust $DRE^{SW}$ and $DRE^{LW}$ of $k^{th}$ size bin and in $(i^{th}, j^{th})$ grid				
$DREE_{kii}$	The monthly and diurnally mean dust DREE <sup>SW</sup> and DREE <sup>LW</sup> of k <sup>th</sup> size bin and (i <sup>th</sup> , j <sup>th</sup> )				
к,ι,ј	grid				
$DAOD_{i,j}^{532nm}$	The monthly mean dust optical depth at 532nm of (i <sup>th</sup> , j <sup>th</sup> ) grid				

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- 321 The definitions of variables and indices used to derive size-resolved dust DREE dataset are
- 322 summarized in Table 2. Eq. (1) shows the way of deriving 1-hourly monthly mean  $DRE^{SW}$ .

$$\frac{1}{10} REE_{1}^{2}(RC) = DRE_{1}^{2}(RC) \overline{h_{10}}(c_{10}, \overline{h_{10}}(c_{10}, \overline{c_{10}}(c_{10}, \overline{c$$

$$\overline{DRE_{k,i,j}^{SW}} = \frac{\sum_{t} t_{1h} DRE_{k,i,j}^{SW}(tt)}{\sum_{t} tt}$$

Similarly, the 3-hourly monthly mean  $DRE^{LW}(\overline{_{3h}DRE_{k,i,j}^{LW}(t)})$  derived from Eq. (2) is averaged diurnally (over 8 points) to get the monthly and diurnally mean dust  $DRE^{LW}(\overline{DRE_{k,i,j}^{LW}})$  as indicated by Eq. (4).

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$$\overline{DRE_{k,i,j}^{LW}} = \frac{\sum_{t \ 3h} DRE_{k,i,j}^{LW}(t)}{\sum t}$$

$$(4)$$

The method described by Eq. (1) - Eq. (4) will be referred to as the '*conventional*' method of calculating monthly mean dust DRE in Section 4. Based on the monthly mean size-resolved dust  $DRE^{SW}(\overline{DRE_{k,i,j}^{SW}})$  and  $DRE^{LW}(\overline{DRE_{k,i,j}^{LW}})$ , we

derive the monthly mean size-resolved dust DREE ( $\overline{DREE}_{k,i,j}$ ) using Eq. (5) for SW and LW respectively. Note that the monthly mean size-resolved dust DREE ( $\overline{DREE}_{k,i,j}$ ) is calculated by dividing by monthly mean  $DAOD^{532nm}$  since the size-resolved  $\overline{DRE}_{k,i,j}$  was initially derived with respect to monthly mean  $DAOD^{532nm}$ .

$$\overline{DREE_{k,i,j}^{SW \text{ or } LW}} = \frac{\overline{DRE_{k,i,j}^{SW \text{ or } LW}}}{\overline{DAOD_{i}^{532nm}}}$$
(5)

Finally, we average the monthly mean size-resolved dust DREE ( $\overline{DREE}_{k,i,j}$ ) over 4 years to get monthly mean size-resolved dust DREE datasets in addition to the associated interannual standard deviation (std). The std indicates the DREE uncertainty caused by interannual variation of monthly mean atmospheric and surface properties as well as dust vertical distribution. Finally, the dataset developed in this study contains monthly mean size-resolved dust DREE and its associated interannual std at TOA and surface with dimension of 10 bins, 12 months, 90 latitudes,  $\boxed{ \frac{\text{Deleted: } \P}{DRE_{k,i,j}^{SW}} = \frac{\sum t_{1k} DRE_{k,i,j}^{SW}(tt)}{\sum tt} }$ 

(... [1])

(3)

368 72 longitudes for each of six dust models in SW and LW respectively. Figure S1 and Figure S2 in
369 the Supplement demonstrate the global distribution of the monthly mean size-resolved DREE<sup>SW</sup>
370 and DREE<sup>LW</sup> at TOA for June.

371 It is important to note that dust DREE of each grid cell rarely depends on the DAOD 372 because dust DRE is approximately linear with DAOD (Satheesh and Ramanathan, 2000). 373 Therefore, the choice of CALIOP- or MODIS-based DAOD climatology to derive the global 374  $(5^{\circ} \times 2^{\circ})$  size-resolved DREE dataset will not lead to large difference. In other words, the size-375 resolved DREE dataset is rarely related to the robustness of the DAOD used in the derivation 376 process. We select CALIOP-based DAOD to derive the size-resolved dust DREE dataset because 377 that the CALIOP-based dust climatology contains dust vertical distribution, which is especially 378 important for obtaining LW DREE. Nevertheless, using CALIOP-based dust retrieval to derive 379 size-resolved dust DREE dataset has several limitations: (1) The size-resolved dust DREE dataset 380 may miss some regions with tenuous dust layers that below the CALIOP sensitivity. (2) The LW 381 DREE is related to the quality of dust vertical distribution retrieval. By contrast, dust DRE highly 382 depends on DAOD, therefore we will use different DAOD climatological datasets retrieved from 383 different sensors (i.e., CALIOP and MODIS) to investigate global dust DRE in section 5.2. 384 Furthermore, even though dust DREE of each grid cell is rarely related to DAOD, regional or 385 global mean dust DREE will depend on the DAOD spatial distribution (i.e., DAOD 2D distribution) 386 in the region of interest (see details in section 5.2).

Based on the monthly mean size-resolved dust DREE datasets derived above, we further calculate globally annual mean size-resolved dust DREE<sup>SW</sup> and DREE<sup>LW</sup> at TOA and surface for the six dust models (Figure 5). As discussed above, the global mean dust DREEs depends on the DAOD spatial distribution, the global mean dust DREEs shown in Figure 5 is based on CALIOP- ( Deleted: (Satheesh and Ramanathan, 2000)

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393 based DAOD spatial distribution from Song et al. (2021). Generally smaller bins cause stronger 394 cooling in SW and less warming in LW, which is consistent with our discussions in 3.1. This 395 observationally informed globally annual mean size-resolved dust DREE is also consistent with 396 the model-simulated results shown in supplementary Figure S3 in Kok et al. (2017) in terms of the 397 variation trend of DREE with respect to dust size. Moreover, our study explicitly shows the sensitivity of dust DREE to dust RI and dust shape. For example, Figure 5 shows that DREE<sup>SW</sup> is 398 strongly sensitive to dust RI as DREE<sup>SW</sup> of different dust RI is widely separated. Depending on 399 dust RI, DREE<sup>SW</sup> switches from cooling effect (negative value) to warming effect (positive value) 400 at different size bins. More absorptive dust starts to warm the Earth system in SW at smaller dust 401 size, and vice versa. In addition, our results suggest that DREE<sup>SW</sup> is generally not sensitive to dust 402 shape. Specifically, dust shape is not important for DREE<sup>SW</sup> in most size bins, while it is important 403 404 in the fourth size bin (D:  $0.79\mu m \sim 1.58\mu m$ ) with DREE<sup>SW</sup> of spheroidal dust obviously higher (less negative) than spherical dust. In the DREE<sup>LW</sup>, dust shape is almost as important as RI for 405 406 several size bins.



Figure 4. Schematic of the methodology used to derive size-resolved dust DREE dataset. Orange boxes denote dust models used to calculate dust scattering properties. Red boxes denote inputs for RRTM. Green boxes denote outputs from RRTM.



Figure 5. Globally annual mean size-resolved dust DREE in SW (a) and LW (b) for six dust models (six markers).
Horizontal bars indicate the dust diameter range of each size bin. Note: LW DREE is on a logarithm scale; in contrast to global model simulations, we consider dust LW scattering in LW DRE Efficiency calculations.

415 Our size-resolved dust DREE dataset is unique in many aspects: First, our DREE dataset 416 is derived based on CALIOP-based dust 3D distribution. Size-resolved DREE is derived for all 417 grids with CALIOP-based DAOD  $\geq 0.01$ . Second, our size-resolved DREE dataset covers a wide 418 range of dust diameters, specifically, they include dust DREE for ten dust diameter size bins 419 ranging from  $0.1 \mu m$  to 100  $\mu m$ . This is challenging, if not impossible, to obtain from global 420 models because these models generally simulate dust particles with diameter only up to 20  $\mu m$  and 421 coarse dust particles in models deposit quickly and could not be sustained to the remote transport 422 regions (Huneeus et al., 2011; Adebiyi and Kok, 2020) where coarse particles have been observed 423 by in-situ measurements (Weinzierl et al., 2017). As a result, our size-resolved DREE dataset 424 achieves a wide spatial coverage for a large range of dust size. This is critical for investigating 425 impacts of coarse dust and even giant dust particles on dust DRE on both regional and global scales. Third, considering that the dust vertical distribution is important for quantifying DRE<sup>LW</sup>, we 426 constrain dust vertical distribution using CALIOP-based dust retrievals in DREE<sup>LW</sup> computation. 427 Fourth, our size-resolved dust DREE dataset accounts for dust LW scattering in DREE<sup>LW</sup> 428 calculations since scattering capability is available through the DISORT in RRTM LW (Stamnes 429

et al., 1988). Dufresne et al., (2002) suggests that dust LW scattering enhances dust LW warming
effect at TOA by a factor of up to 50%. However, dust LW scattering is generally not considered
in most global models. Therefore, many previous studies artificially account for dust LW scattering
by increasing the radiative perturbation due to LW absorption by a certain fraction. For example,
Kok et al. (2017) accounts for LW scattering by artificially augmenting DRE<sup>LW</sup> by 23% and Di
Biagio et al. (2020) augmented DRE<sup>LW</sup> by 50%.

436 On the other hand, our size-resolved dust DREE dataset has several limitations. First, 437 possible vertical and horizontal variations of dust particle size in each grid box  $(5^{\circ} \times 2^{\circ})$  are not 438 accounted for in our calculation. The entire dust-loading column in each grid box is assumed to 439 have the same dust size distribution. Second, we do not explicitly account for spatial variation of 440 dust RI, in other words, dust RI is assumed to be globally uniform. This uncertainty is assessed 441 through the sensitivity tests of DREE to dust RI using three sets of state-of-the-art dust RI based 442 on laboratory measurement of 19 dust samples all over the world. Third, dust 3D distribution in 443 the DREE calculation is constrained by CALIOP observations. The limits on the sensitivity of 444 CALIOP will affect the 3D distribution of dust in our calculation. Fourth, we account for dust 445 nonsphericity by using spheroidal shape model. This shape can't perfectly represent the highly 446 irregular shape and roughness of real dust. In addition, several studies suggest that dust non-447 sphericity is underestimated by the spheroidal shape model (Huang et al., 2020). The spheroidal 448 shape model assumption thus might produce systematic errors.

Overall, the size-resolved dust DREE dataset is useful in many dust-related studies. First,
with our size-resolved dust DREE dataset, dust DRE could be calculated efficiently for any DAOD
magnitude, DAOD spatial pattern and any dust PSD for any regions or the globe (see details in
Section 4.1). Second, our size-resolved DREE dataset is derived for different RI and different dust

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shapes respectively. As a result, we could estimate dust DRE uncertainty coming from DAOD,
PSD, RI, and shape separately to better understand major uncertainty sources in dust DRE
estimations. Third, our size-resolved DREE dataset could be used to evaluate model simulated
DREE for each size bin.

#### 458 **4** DRE calculation methodology and its validation

#### 459 4.1 DRE calculation based on DREE dataset

With the size-resolved dust DREE dataset derived in section 3.2, DRE of dust with any PSD and DAOD could be computed very efficiently without performing radiative transfer simulations as we do in *conventional* method. This section introduces the methodology of applying the size-resolved DREE dataset to calculate DRE of dust with any PSD and DAOD.

464 DRE of full\_size range of dust can be expressed as the sum of DRE from each size bin 465  $(DRE_k)$ . Dust  $DRE_k$  is approximated to be linearly proportional to DAOD of  $k^{th}$  size bin  $(DAOD_k)$ 466 (Satheesh and Ramanathan, 2000). The similar concept of calculating dust DRE has been used in 467 previous studies e.g., Kok et al. (2017). Eq. (6) shows the process of computing dust DRE using 468 the size-resolved DREE dataset.

$$DRE = \sum_{k} DRE_{k} = \sum_{k} DREE_{k} \times DAOD_{k} = \sum_{k} DREE_{k} \times f_{k} \times DAOD,$$
(6)

where *DRE* represents dust DRE induced by full size range of dust with optical depth of *DAOD*. *f<sub>k</sub>* is the fraction of the DAOD contributed by the *k<sup>th</sup>* size bin.

471 Each variable in Eq. (6) can be obtained or derived from datasets developed in this study 472 and other studies. For example, the size-resolved DREE dataset ( $DREE_{k,i,j}$ ) derived in this study 473 is essential for utilizing this efficient and novel DRE calculation method. DAOD can be obtained 474 from CALIOP-based or MODIS-based DAOD climatological datasets (Song et al., 2021).  $f_k$  can 475 be derived from dust extinction efficiency (*Qe*), the geometric cross-sectional area (A) and dust 476 PSD (dN/dD) based on Eq. (7).

$$f_{k} \equiv \frac{DAOD_{k}}{DAOD} = \frac{\int_{D^{k-}}^{D^{k+}} Qe^{532nm}(D)A(D)\frac{dN}{dD}dD}{\int_{0}^{D^{max}} Qe^{532nm}(D)A(D)\frac{dN}{dD}dD}$$
(7)

Qe is defined according to  $Qe \equiv \frac{\sigma_e}{A}$ , where  $\sigma_e$  is extinction cross section, the geometric 477 cross-sectional area of the particle (A) can be expressed as  $A = \pi r^2$ . Under the assumption of 478 spherical dust particle, r is the radius. Under the assumption of spheroidal dust particle, Vouk 479 (1948) shows that the average projected area of a convex body (e.g., spheroidal particle) is A =480  $\pi r^2$ , where r is the radius of a surface area-equivalent sphere. The average is taken over all 481 482 possible orientations in space, which is consistent with our assumption of randomly oriented dust particles in the atmosphere.  $Qe^{532nm}(D)$  for the six dust models are shown in Figure 6 (a), they 483 484 all converge to 2 as the dust diameter becomes much larger than the wavelength, which is consistent with the principle of geometric optics (van de Hulst, 1957). By contrast,  $Q_e^{550nm}(D)$  of 485 non-spherical dust in Kok et al. (2017) has a much larger value than spherical dust for dust  $D \ge$ 486 487  $1\mu m$  (see their Figure 1(b)). This discrepancy is probably due to the different  $Q_e$  definitions used 488 in the two studies. Kok et al. (2017) defined  $Q_e$  as dust extinction per unit cross section of volume-489 equivalent sphere. Figure 6 (b) shows that  $f_k$  of a specific PSD is not sensitive to dust RI and dust shape, this is also suggested by the similar  $Qe^{532nm}$  v.s. geometric diameter (D) trends of the six 490 491 dust models shown in Figure 6 (a). In contrast,  $f_4$  (i.e.,  $f_k$  for the fourth size bin with D ranging 492 from  $0.79\mu m$  to  $1.58\mu m$ ) is more sensitive to dust shape than other size bins, this is in line with 493 the larger difference in  $Qe^{532nm}$  with shape shown in Figure 6 (a).



Figure 6. (a) Dust extinction efficiency (Qe) at 532nm for six dust models. (b) The colorful bars represent  $f_k$ calculated for six dust models based on a specific dust PSD (dV/dlnD) indicated by black curve. Note,  $f_k$  is not sensitive to different dust models such as dust RI and dust shape.

In summary, the size-resolved dust DREE dataset provides an efficient way to compute DRE for any dust PSD and any DAOD by using Eq. (6) and Eq. (7). To distinguish from the *conventional* method introduced in section 3.2, this method of calculating dust DRE based on sizeresolved DREE dataset is referred to as '*DREE-integration*' method.

#### 502 4.2 Validation of DRE calculation methodology

503 In this section, we select the Sahara Desert (14°N-30°N, 15°W-30°E) to validate the

504 DREE-integration method. We choose MeanSWRI-MeanLWRI-Spheroid dust model and Fennec-

505 Fresh dust PSD (see red curve in Figure 7) measured within 12h of dust uplift in remote Sahara

506 locations by Fennec field campaign to represent microphysical properties of Saharan dust (Ryder

507 et al., 2013a, b). Monthly mean DAOD is from CALIOP-based DAOD climatology.





509 Figure 7. Normalized atmospheric dust volume distribution (dV/dlnD) described in Table 5 (Kok et al., 2017; Ryder et al., 2013a; b; 2018; 2019).
511 Figure 8 shows the comparison of 4-year (2007-2010) monthly mean dust DRE between

the *Conventional* and *DREE-integration* method. In Conventional DRE calculation, dust scattering properties (Qe,  $\omega$  and g) are calculated based on the Fennec-Fresh PSD and then used to calculate monthly mean dust DRE from 2007 to 2010 with RRTM as described in Section 3.2 (Eq. 1 – Eq. 4). While the DREE-integration method is based on the monthly mean size-resolved DREE dataset derived based on 4-year (2007-2010) data as described in Section 4.1 (Eq. 6 – Eq. 7). The excellent

517 agreement in monthly mean dust DRE between two methods validates the *DREE-integration* DRE

518 calculation methodology.







Figure 8. Monthly mean dust DRE<sup>SW</sup> (a) and DRE<sup>LW</sup> (b) comparison between *Conventional* and *DREE-integration* calculation from 2007 to 2010 over Sahara Desert. <u>The DRE Difference line represents the difference between *DREE-integration* and <u>Conventional calculation</u>. Shaded area along *DREE-integration* DRE indicates the one standard deviation caused by the atmospheric and surface variations as well as dust vertical distribution variation within the four years. Orange curves indicate CALIOP-based monthly mean DAOD. The variation of dust DRE match well with DAOD variation.</u>

529 The shaded-area associated with DREE-integration DRE corresponds to the one standard deviation of DREE caused by the 4-year (2007-2010) interannual variation of factors except dust 530 531 microphysical properties such as monthly mean atmospheric and surface properties as well as dust 532 vertical distributions (hereafter those factors is referred to as non-dust-factors for short). The 533 narrow shaded-area along DREE-integration DRE suggests non-dust-factors cause very small 534 uncertainty in dust DRE estimations. However, the small effects of 4-year interannual variation of 535 non-dust-factors may not necessarily be representative due to the limited number of years 536 considered. Section 2.1 discusses in detail for the reason of choosing 2007-2010 to derive size-537 resolved DREE dataset. To check the representative of 4-year interannual variation for non-dust-538 factors, we compare the 4-year (2007-2010) and 10-year (2007-2017) interannual standard 539 deviation (std) of monthly mean non-dust-factors (e.g., surface albedo, surface temperature and 540 dust vertical distribution) in Figure 9. To evaluate the interannual variation of dust vertical distribution, we define dust mean extinction height  $(Z_{\alpha})$  referring to Koffi et al. (2012) as  $Z_{\alpha} =$ 541  $\frac{\sum_{i=1}^{n} \beta_{ext,i} \times Z_{i}}{\sum_{i=1}^{n} \beta_{ext,i}}$ , where  $\beta_{ext,i}$  is the dust extinction coefficient at 532nm at level *i*, and  $Z_{i}$  is the altitude 542

of level *i*. Nevertheless the 10-year std is slightly larger than 4-year std, they are both close to zero and on the same order of magnitude. As such, even though our monthly mean size-resolved DREE dataset is derived from 4-year (2007-2010) data, they could be used to represent DREE and calculate DRE for other years considering the small sensitivity of monthly mean dust DRE to interannual variation of non-dust-factors.



Figure 9. Probability density function (PDF) of 4-year and 10-year interannual standard deviation (std) in monthly
mean (a) surface albedo, (b) surface temperature, and (c) dust mean extinction height. The PSD analyses include
interannual std in 12 months and all 5° (longitude) × 2° (latitude) grid cells over the world and their mean values are
indicated as 'std\_mean' on each figure.

#### 553 5 Regional and global dust DRE based on size-resolved DREE dataset

548

554 After the validation of DREE-integration method in Section 4, we use the DREE-integration 555 method to calculate regional and global dust DRE in this section. There are three main objectives 556 in this section: (1) the most important objective throughout this section is to demonstrate the usefulness of the size-resolved DREE dataset for calculating regional and global dust DRE for any 557 558 given dust PSD; (2) the second objective is to validate the size-resolved DREE dataset by 559 comparing with regional dust DREE reported by field studies based on satellite and ground-based 560 observations (section 5.1); (3) the third objective to assess the sensitivity of dust DRE to DAOD 561 spatial pattern (section 5.2) as well as dust microphysical properties such as dust PSD, RI and 562 shape (section 5.3).

#### 563 5.1 Comparison with observation-based regional dust DREE

Table 3 shows the comparison of our calculations of clear-sky regional mean SW and LW DREE with those reported by field studies based on satellite and ground-based observations. We first calculated regional mean dust DRE using the *DREE-integration* method, then divided by the corresponding regional mean DAOD to get regional mean DREE, and then compared this with observation-based results from previous studies. Comparing DREE allows eliminating differences due to the variation in regional dust loading, optically represented by DAOD.

570 Knowledge of regional dust PSD is necessary for estimating dust DRE regionally. There are 571 several in-situ measurements of dust PSD over Sahara and tropical eastern Atlantic. The state-of-572 the art airborne observations of Saharan dust from the Fennec field campaign (Fennec-Fresh) and 573 transported Saharan dust over tropical eastern Atlantic within Saharan Air Layer (SAL) from both 574 AER-D and Fennec fieldwork campaigns are adopted (Ryder et al., 2013 a, b, 2018, 2019) (see 575 Figure 7). Both campaigns include giant dust particles, measuring up to 100µm diameter for AER-576 D and up to  $300\mu m$  for Fennec. The wide coverage of dust diameter in our size-resolved DREE 577 dataset allows for dust DRE calculations for giant dust up to  $100\mu m$  over both dust source and 578 transported regions where giant particles are observed in those campaigns. This is an advantage of 579 our size-resolved DREE dataset compared to modeled dust DREE, because climate models 580 generally cut off dust diameter at 20 µm and could not sustain coarse dust to remote transport 581 regions due to several missing mechanisms in models (Van Der Does et al., 2018; Drakaki et al., 582 2022; Meng et al., 2022).

583 The Fennec-Fresh dust PSD includes measurements within 12h of dust uplift in remote 584 Sahara locations. It is used to calculate dust DRE for Saharan dust in this section. In reality, dust 585 over the wide Sahara Desert region (15N~30N, 10W~30E) is not all lifted within 12h, so using

586	Fennec-Fresh to represent dust PSD over the wide Sahara Desert could bias dust size coarse, which
587	could partially explain the warm bias in our $DREE^{SW}$ estimation over the Sahara Desert compared
588	to the satellite-based result. Over the tropical Atlantic, both AER-D and Fennec-SAL measured
589	PSD are used to assess the sensitivity of dust DREE to dust PSD. In addition, dust DRE is
590	calculated for three dust RIs to evaluate the sensitivity of dust DREE to dust RI as shown in Table
591	3. Generally, our dust DREE estimations achieve good agreement with observation-based dust
592	DREE. However, there is a significant uncertainty caused by dust RI in DREE, especially for SW.
593	In addition, DRE comparisons between AER-D and Fennec-SAL over the Tropical Atlantic
594	suggests that in-situ measured dust PSD uncertainty leads to a large uncertainty in regional DREE
595	in both SW and LW.
596	Based on the regional DREE study with the state-of-the art RI and PSD, we found DREE <sup>sw</sup>
597	uncertainty could come from both dust RI and dust PSD, while DRE <sup>LW</sup> uncertainty is mainly from

<sup>599</sup> 600 Table 3. Comparison of our DREE estimations for different PSD and RI with Clear-Sky regional SW and LW dust DREE reported by field studies based on satellite and ground-based observations. Specifically, we calculated regional dust DREE for different RI (Min, Mean, Max) and different PSD (AER-D and Fennec-SAL for Tropical Atlantic) and 601 602 603 then compare with observation-based results from previous studies. Note, spheroidal dust shape is assumed in our *DREE-integration* DRE calculations.

Shortwave Spectral Rang	ge						
Region	Season	Level	Satellite-		Tł	nis study	
_			Based				
			DREE <sup>SW</sup>		DREE <sup>SW</sup>		PSD
				Min RI	Mean RI	Max RI	
Sahara Desert (a)	JJA	TOA	0	2.8	16.0	26.6	Fennec-Fresh
(15N~30N, 10W~30E)							
Ilorin <sup>(f)</sup> , Nigeria	Annual	TOA	-15 ~ -35	-28.3	-24.1	-19.9	AER-D
(8.5N, 4.7E)				-23.4	-17.7	-12.9	Fennec-SAL
		Surface	-49 ~ -75	-43.1	-51.7	-59.3	AER-D
				-46.0	-57.1	-66.0	Fennec-SAL
Cape Verde <sup>(f)</sup>	Annual	TOA	-36 ~ -48	-42.3	-38.0	-33.7	AER-D
(16.7N, 22.9W)				-36.6	-30.8	-26.0	Fennec-SAL
		Surface	-68 ~ -90	-59.6	-68.7	-77.7	AER-D
				-61.5	-74.6	-85.3	Fennec-SAL
Tropical Atlantic <sup>(b)</sup>	JJA	TOA	-28	-44.6	-39.9	-35.3	AER-D
$(10N \sim 30N, 20W - 45W)$				-38.4	-32.1	-27.0	Fennec-SAL

<sup>598</sup> dust PSD.

		Surface	-82.1	-61.1	-71.9	-81.7	AER-D
				-64.4	-78.5	-90.0	Fennec-SAL
Tropical Atlantic (c)	JJA	TOA	-35	-41.2	-36.3	-31.5	AER-D
(15N~25N,15W~45W)				-35.1	-28.5	-23.1	Fennec-SAL
		Surface	-65	-57.9	-68.6	-78.1	AER-D
				-61.2	-75.1	-86.3	Fennec-SAL
Longwave Spectral Rang	ge						
Region	Season	Level	Satellite-		This study		
_			Based				
			DREE <sup>LW</sup>		DREELW		PSD
				Min RI	Mean RI	Max RI	
Sahara Desert <sup>(a)</sup>	JJA	TOA	11~26	13.4	11.8	11.4	Fennec-Fresh
(15N~30N, 10W~30E)							
North Africa <sup>(d-e)</sup>	JJA	TOA	15~22	14.4	12.8	12.4	Fennec-Fresh
(15N~35N, 18W~40E)							
Tropical Atlantic (b)	JJA	TOA	10.5	8.2	8.1	8.5	AER-D
(10N~30N, 20W~45W)				13.1	11.8	11.6	Fennec-SAL
Cape Verde <sup>(g)</sup>	Sept	Surface	16	8.0	11.8	15.1	AER-D
(16.7N, 22.9W)				13.0	17.0	19.8	Fennec-SAL
(a) Patadia et al. (2009	9). (b) Son	g et al. (20	18). (c) Li et al.	(2004). (d)	) Zhang and	Christopher	(2003). (e)
Brindle	y and Rus	sell (2009).	(f) Zhou et al.	(2005). (g)	Hansell et a	1. (2010)	

604

#### 605 5.2 Global dust clear-sky DRE based on different DAOD climatology

606 The DAOD is the most important factor in determining dust DRE. As illustrated in Song et 607 al. (2021), the DAOD retrieved from different satellite sensors have a large difference in terms of 608 magnitude and spatial distribution. To evaluate how the current DAOD uncertainty affects dust 609 DRE estimations, the global dust DRE computed based on monthly mean DAOD climatology 610 retrieved from CALIOP observations and MODIS observations are compared in this section. To 611 separate the effect of DAOD from other factors, we use the same dust PSD, RI and shape in the 612 two sets of dust DRE calculations in this section. Specifically, we use the Fennec-Fresh PSD for 613 three major dust source regions (i.e., Sahara (14-30°N, 15°W-30°E), Middle East (10-35°N, 40-614 85°E) and eastern Asia (30-50°N, 75-130°E), they are indicated by three black boxes in Figure 10) 615 and use AER-D PSD for other regions (hereafter Campaign-PSD, see Table 5). The MeanSWRI-616 MeanLWRI-Spheroid dust model described in Table 1 is used to represent dust RI and shape.

617 The two DAOD climatological datasets result in distinct dust DRE spatial pattern as shown 618 in Figure 10, which is consistent with the DAOD spatial patterns shown in Figure 1 suggesting 619 CALIOP DAOD is more concentrated over 'dust belt' regions than MODIS DAOD. The global mean dust DRE<sup>SW</sup>, DRE<sup>LW</sup> and DRE<sup>NET</sup> based on the two DAOD climatology are significantly 620 621 different (Table 4), which is mainly caused by two factors. The first is the difference in DAOD 622 magnitude. The CALIOP-based global mean DAOD is 0.032, while MODIS-based is 0.047. The 623 other factor is the difference in DAOD spatial pattern. After we scale dust DRE to the same global mean DAOD (DAOD = 0.03) to eliminate the effect of DAOD magnitude difference (values in 624 parentheses in Table 4), the DRE<sup>SW</sup> difference reduced from 0.55 W m<sup>-2</sup> (-0.69 vs. -1.24 W m<sup>-2</sup>) 625 to 0.15 W m<sup>-2</sup> (-0.64 vs. -0.79 W m<sup>-2</sup>). Similarly, differences in DRE<sup>LW</sup> and DRE<sup>NET</sup> also reduce 626 627 significantly. It indicates that the global mean DAOD magnitude difference is more important than the subtle difference in spatial pattern. Nevertheless, after scaling to the same global mean DAOD 628 there is still more than 10% difference between the two dust DRE<sup>SW</sup>, with CALIOP-based being 629 630 the more positive one. This is probably because CALIOP-based DAOD is more concentrated over dust sources where dust aerosols induce less negative or even positive DRE<sup>SW</sup> (For example the 631 positive DRE<sup>SW</sup> over the Sahara Desert and Arabia shown in Figure 10), which result in a less 632 negative global mean DRE<sup>SW</sup> than MODIS. 633





Figure 10. Annual mean dust DRE global distribution based on CALIOP-based and MODIS-based DAOD
 climatology. MeanSWRI-MeanLWRI-Spheroid dust model are used to represent dust RI and shape in the calculation.
 Campaign-PSD is used to represent dust PSD, specifically, Fennec-Fresh PSD is used to represent dust PSD over the
 three major dust source regions indicated by three black boxes. AER-D PSD is used to represent dust PSD over other
 regions.

640 Table 4. Globally annual mean DAOD, DRE<sup>SW</sup>, DRE<sup>LW</sup> and DRE<sup>NET</sup> based on CALIOP DAOD and MODIS DAOD 641 climatology. Note, values in the parentheses are for the two DAOD scaled to the same value of 0.03.

	DAOD	DRE <sup>SW</sup> [Wm <sup>-2</sup> ]	DRE <sup>LW</sup> [Wm <sup>-2</sup> ]	DRENET [Wm <sup>-2</sup> ]
CALIOP	0.032 (0.03)	-0.69 (-0.64)	0.25 (0.23)	-0.44 (-0.41)
MODIS	0.047 (0.03)	-1.24 (-0.79)	0.34 (0.22)	-0.90 (-0.57)

642

#### 643 5.3 Global dust clear-sky DRE based on different dust PSD

In the section 5.2, we showed the dust DRE based on the Campaign-PSD. As aforementioned, one of the main advantages of our size-resolved DREE is that it can be combined with different dust PSDs to estimate the dust DRE. To demonstrate this, we calculate another set of dust DRE based on the Kok2017-PSD. Table 5 describes the two dust PSDs used for global dust DRE calculations and their references. Kok2017-PSD is a globally averaged dust PSD and used to represent dust PSD for each dusty grid cell. It is constrained with observations and includes coarse dust particles up to  $20\mu m$ . Although our primary goal here is to demonstrate the capability of our size-resolved DREE, the comparison between the two DRE can also help us understand the impacts of dust PSD uncertainty on the dust DRE estimation. Moreover, we also investigate the sensitivity of DRE to dust RI and dust shape explicitly in this section. The same DAOD climatology (CALIOP-based DAOD climatology) is used for dust DRE calculations to eliminate the impact of dust loading difference.

656 Several recent observation-constrained dust PSDs (e.g., Di Biagio et al., 2020, Adebiyi et 657 al., 2020) suggest that dust size is coarser than Kok2017-PSD. As such, Kok2017-PSD is used to 658 represent the lower limit of the observation-based global dust PSD to investigate the sensitivity of 659 dust DRE to dust PSD. The Campaign-PSD is purely based on aircraft in-situ measurements and 660 the aircraft was extensively equipped to measure giant particles with diameter larger than  $20\mu m$ . 661 We use the dust PSD measured over Sahara (from the Fennec field campaign) to represent dust 662 PSD over three major dust source regions and use dust PSD measured in the Saharan Air Layer 663 over the tropical eastern Atlantic (from AER-D field campaign) to represent dust PSD over dust 664 transport regions. Of course, representing the spatial and temporal variation of global dust PSD 665 with only two PSDs from the field campaigns is only a crude approximation due to the lack of 666 PSD measurements. Dust aerosol over the three wide dust source regions may not be all uplifted 667 within 12 hours as in the Fennec-Fresh measurements, in addition, dust size after long-range transport could be a bit finer than dust PSD measured over tropical eastern Atlantic (Weinzierl et 668 669 al., 2017). Thus, Campaign-PSD likely represents the upper limit of the observation-based global 670 dust PSD for the investigation of sensitivity to dust PSD. By contrast, the climate models miss 671 most of coarse dust (D>5  $\mu m$ ) in the atmosphere (Adebiyi and Kok, 2020), as a result, the purely 672 modeled dust PSD without observational constraints will lead to a substantially different dust DRE.

Deleted: spatially

Deleted: temporally

675 Therefore, the sensitivity test to dust PSD conducted in this study can only represent the

676 uncertainty induced by the current understanding of observation-based dust PSD.

677 Table 5. The two observation-based dust PSDs used in DRE calculations (see Figure 7).

PSD	Description	Reference	
Kok2017-PSD	A globally averaged atmospheric PSD derived from observation constrained globally averaged emitted PSD and model simulated globally averaged dust lifetime. This globally averaged PSD is used to represent dust PSD for each dusty grid cell.	Kok et al. (2017)	Deleted: Kok et al. (2017)
Campaign-PSD	Dust diameter is cutoff at $20\mu m$ (Figure 2a in Kok et al.2017). Fennec-Fresh PSD is used for three major dust source regions (i.e., Sahara (14-30°N, 15°W-30°E), Middle East (10-35°N, 40-85°E) and eastern Asia (30-50°N, 75-130°E)), which are indicated by the three black boxes in Figure 10. AER-D PSD is used for other regions.	Ryder et al. (2013a, b, 2018, 2019)	

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679 We calculated dust DRE of each grid cell  $(DRE_{i,j})$  using DREE-integration method based 680 on the dust PSD described in Table 5. Global mean dust DRE was then calculated by averaging dust  $DRE_{i,j}$  weighted by its surface area. Figure 11 shows the global mean  $DRE^{SW}$ ,  $DRE^{LW}$  and 681 682 DRENET at TOA, surface, and in the atmosphere calculated based on the two sets of PSDs. 683 Obviously, Kok2017-PSD leads to stronger cooling effect in SW and weaker warming effect in 684 LW at TOA compared to Campaign-PSD, which is consistent with the fact that Kok2017-PSD is 685 finer than the Campaign-PSD. In addition, we explicitly include the effects of dust RI and dust 686 shape on DRE in Figure 11. Comparison of uncertainty induced by dust PSD, RI and shape 687 suggests that dust RI uncertainty leads to the largest uncertainty in dust DRE, particularly RI uncertainty induces more than 40% uncertainty in DRE<sup>SW</sup> estimations in the atmosphere (Figure 688 689 12). Dust PSD is also important for quantifying dust DRE, we found that the observation-based 690 dust PSD uncertainty induces around 15%~20% uncertainty in dust DRE at TOA and in the 691 atmosphere. Dust non-sphericity causes a negligible uncertainty in global mean dust DRE, in line 692 with previous studies e.g., Raisanen et al. (2013) and Colarco et al. (2014).



Figure 11. Globally annual mean clear-sky DRE<sup>SW</sup>, DRE<sup>LW</sup> and DRE<sup>NET</sup> at TOA, in the atmosphere and surface calculated based on the two PSDs described in Table 5. The two rows represent dust DRE based on two PSDs. Error bars indicate uncertainty induced by dust RI uncertainty. Different types of bars indicate dust DRE based on different dust shapes. This figure explicitly separates the impacts of different dust microphysical properties on dust DRE. Two values in parenthesis on each plot represent spherical (left) and spheroidal (right) dust DRE corresponding to mean RI.



Figure 12. Comparison of uncertainty induced by dust RI, PSD and shape in DRE<sup>SW</sup>, DRE<sup>LW</sup> and DRE<sup>NET</sup> at TOA (a),
 in the atmosphere (b) and surface (c). The horizontal lines in each plot represent global mean DRE<sup>SW</sup> (blue line in the
 left column), DRE<sup>LW</sup> (red line in the middle column) and DRE<sup>NET</sup> (green line in the right column) averaged over two
 dust PSDs (i.e., Kok2017-PSD and Campaign-PSD) based on MeanRI-Spheroid dust model. The three error bars in
 each column represent DRE uncertainty induced by dust RI (left), dust PSD (middle) and dust shape (right).
 Accordingly, the percentage values on the bottom represent the percentage uncertainty induced by dust RI, PSD and
 shape, respectively.

702

710 It is tempting to compare our global mean dust DRE with results reported in Kok et al. (2017).

711 But it must be noted that the global mean dust DRE shown in Figure 11 is for *clear sky* only, while

712 the global mean dust DRE reported in Kok et al. (2017) is for all sky. The all-sky dust DRE can

713 be separated into contributions from clear-sky and cloudy-sky portions (Myhre et al., 2020):

$$DRE_{all-sky} = (1 - CF) \times DRE_{clear-sky} + CF \times DRE_{cloudy-sky},$$
(8)

714 where CF is cloud fraction, DRE<sub>clear-sky</sub> is dust DRE simulated under the case of removing all 715 clouds, DRE<sub>cloudy-sky</sub> is the dust DRE assuming whole grid is covered by clouds. To compare 716 our global mean dust DRE<sup>SW</sup> based on Kok2017-PSD with the results reported in Kok et al. (2017), we convert our clear-sky  $DRE_{clear-sky}^{SW}$  to  $DRE_{all-sky}^{SW}$  by using MODIS L3 monthly mean cloud 717 fraction. Specifically, we multiply  $DRE_{clear-sky}^{SW}$  by (1-CF) for each grid cell and then calculate 718 global annual mean values. In this process, we neglect the cloudy-sky dust DRE<sup>SW</sup> portion because 719 720 the annual mean cloudy-sky dust DRE<sup>SW</sup> is estimated to be very small, around -0.04 (Zhang et al., 2016). Finally, our estimated global mean  $DRE_{all-sky}^{SW}$  corresponding to DAOD=0.03 is around 721 -0.34 Wm<sup>-2</sup>. Although it is comparable to the -0.48 Wm<sup>-2</sup> from Kok et al. 2017, the following 722 723 differences between the two studies must be kept in mind when interpreting the results. First, the rough conversion from global mean  $DRE_{clear-sky}^{SW}$  to global mean  $DRE_{all-sky}^{SW}$  is subject to the 724 725 approximation of global mean  $DRE_{cloudy-sky} \sim 0$  and the MODIS L3 cloud fraction could be 726 different from modeled cloud fraction used in Kok et al. (2017). Second, the two studies use 727 different dust RI. For example, the imaginary part of RI at 550nm in this study ranges from 0.00061 728 to 0.003, while that in Kok et al. (2017) ranges from 0.0014 as used in GEOS-Chem and GISS 729 model based on Sinyuk et al., (2003) to 0.003 as used in WRF-Chem based on Zhao et al., (2010). 730 Third, in this study Kok2017-PSD is used to represent dust PSD in each dusty grid and applied to 731 our size-resolved dust DREE dataset to calculate global dust DRE. In contrast, the model-732 simulated dust DREE in Kok et al. (2017) has reduced cooling from SW scattering and enhanced 733 warming from SW absorption effects because the short lifetime of coarse dust in models 734 concentrates these particles over bright deserts. Fourth, the two studies use different dust shape 735 models, Kok et al. (2017) accounts for more nonspherical shape model (i.e., tri-axial ellipsoids).

Here we do not compare our global mean  $DRE_{clear-sky}^{LW}$  with  $DRE_{all-sky}^{LW}$  suggested in Kok et al. (2017) because that the lack of knowledge in  $DRE_{cloudy-sky}^{LW}$  prevent us to convert  $DRE_{clear-sky}^{LW}$ to  $DRE_{all-sky}^{LW}$ . Moreover, the two studies use different dust vertical profile, which is critical for DRE<sup>LW</sup> estimations. For instance, dust vertical profile in Kok et al. (2017) is purely based on model simulations, while this study constrains dust vertical profile with CALIOP observations. Considering all these factors, it is hard to tell if the comparison is fair.

#### 742 6 Summary and Conclusion

This study developed a clear-sky size-resolved dust DREE dataset in both SW and LW based on CALIOP-based dust DAOD climatology and dust vertical distributions. The dataset contains global monthly mean dust DREE at TOA and surface with 5° (longitude)  $\times$  2° (latitude) spatial resolution for 10 size bins ranging from  $0.1\mu m$  to  $100\mu m$  diameter, for three state-of-the art dust RI representing more, mean and less absorptive dust, and for two dust shapes representing spherical and spheroidal dust, respectively.

749 The size-resolved DREE dataset allows us to calculate dust DRE of any DAOD climatology and dust PSD efficiently by using the DREE-integration method presented in section 750 751 4.1 without involving radiative transfer simulations. The DREE-integration method is proven to 752 be in great agreement with conventional DRE calculations. With the DREE-integration methodology, we firstly calculated clear-sky regional mean DREE<sup>SW</sup> and DREE<sup>LW</sup> over the Sahara 753 754 Desert and tropical Atlantic. The comparison of our calculations with those reported by field 755 studies based on satellite and ground-based observations shows reasonable agreement. Secondly, 756 we estimated global mean dust DRE with two satellite-based DAOD climatological datasets and 757 two different global dust PSDs. We found that the global mean DAOD magnitude difference 758 between the two DAOD climatological datasets is more important than the subtle difference in 38

759	spatial pattern. Nevertheless, after scaling to the same global mean DAOD there is still more than
760	10% difference between the two dust $DRE^{SW}$ , with CALIOP-based being the more positive one.
761	Moreover, our results explicitly show the uncertainty induced by each dust microphysical property
762	(i.e., dust PSD, RI and shape) separately. When DAOD is constrained: (a) Dust non-sphericity
763	induces negligible effect on dust DRE estimations; (b) The current understanding of observation-
764	based dust PSD induces relatively large uncertainty (15%~20%) in dust DRE at TOA and in the
765	atmosphere (c) Dust RI turns out to be the most important factor in determining dust DRE,
766	particularly in SW. This implies that better understanding of dust mineral composition and RI will
767	significantly improve our understanding in dust DRE in the future.
768	Data availability:
769	The size-resolved dust DREE dataset and the codes to calculate dust DRE for any given
770	dust PSD and DAOD are available at
771	'https://drive.google.com/drive/folders/15_e28Y9JiSWiJnIM_2flEmt2u6i9phEY?usp=sharing'
772	CALIOP- and MODIS-based DAOD climatological datasets are available at
773	<pre>`https://drive.google.com/drive/folders/1aQVupe7govPwR6qmsqUbR4fJQsp1DBCX?usp=shari</pre>
774	ng'
775	
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Supplement of

# Size-Resolved Dust Direct Radiative Effect Efficiency Derived from Satellite Observations

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## 1. Demonstration of monthly-mean size-resolved DREE dataset

Figure S1. Global distribution of <u>four-year (2007-2010) June</u> monthly mean DREE<sup>SW</sup> of MeanSWRI-Spheroid dust model at TOA<sub>v</sub> j obtained from the *size-resolved* DREE dataset. Grey area indicates area without DREE derivations (e.g., DAOD retrieval is not available or *DAOD* <sup>532nm</sup> <=0.01 over the area).<sub>v</sub>



Figure S2. Global distribution of <u>four-year (2007-2010) June</u> monthly-mean DREE<sup>LW</sup> of MeanLWRI-Spheroid dust model at TOA<sub>2</sub> obtained from the *size-resolved* DREE dataset. Grey area indicates area without DREE derivations (e.g., DAOD retrieval is not available or *DAOD*  $^{532nm} \leq 0.01$  over the area).

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