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2		Size-Resolved Dust Direct Radiative Effect Efficiency
3		Derived from Satellite Observations
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24 Abstract

25 The role of mineral dust aerosol in global radiative energy budget is often quantified by the dust direct radiative effect (DRE). The dust DRE strongly depends on dust aerosol optical 26 27 depth (DAOD), therefore, DRE efficiency (DREE=DRE/DAOD) is widely compared across 28 different studies to eliminate difference due to the various dust load. Nevertheless, DREE is still 29 influenced by the uncertainties associated with dust particle size distribution (PSD) and optical 30 properties. In this study, we derive a global clear-sky size-resolved DREE dataset in both 31 shortwave (SW) and longwave (LW) at top of the atmosphere (TOA) and surface based on satellite 32 observations (i.e., satellite-retrieved dust extinction spatial and vertical distributions). In the DREE 33 dataset, dust geometric diameter from 0.1µm to 100 µm is divided into 10 bins and the 34 corresponding monthly mean DREE (with respect to DAOD at 532nm) for each size bin is derived 35 by using the Rapid Radiative Transfer Model (RRTM). Three sets of state-of-the-art dust refractive 36 indices (RI) and two sets of dust shape models (sphere vs. spheroid) are adopted to investigate the 37 sensitivity of dust DREE to dust absorption and shape. As a result, the size-resolved dust DREE 38 dataset contains globally distributed monthly mean dust DREE at TOA and surface for each of 10 39 size bins with 5° (longitude) \times 2° (latitude) resolution as well as for each dust RI and shape 40 combination. The size-resolved dust DREE dataset can be used to readily calculate global dust 41 DRE for any DAOD and dust PSD, including the uncertainty in the DRE induced by dust 42 microphysical properties (e.g., dust PSD, RI and shape). By calculating dust DRE based on DAOD 43 climatology retrieved from different satellite sensors and based on different dust PSD, we find that 44 uncertainty in the spatial pattern of DAOD induces more than 10% of the uncertainty in SW dust 45 DRE at TOA. The observation-based dust PSD induces around 15% ~20% uncertainty in dust DRE 46 at TOA and in the atmosphere. The sensitivity assessments of dust DRE to dust RI and shape





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

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51 **1 Introduction**

52 Mineral dust is an important component of the atmospheric aerosol (Textor et al., 2006; 53 Choobari et al., 2014). They can influence the radiative energy budget of the Earth-Atmosphere system directly through their interaction with both solar and thermal infrared radiation, which is 54 known as the direct radiative effect (DRE) of dust. The DRE of dust consists of two components. 55 56 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) 57 and surface (Tegen et al., 1996; Myhre et al., 2003). In the longwave (LW) thermal infrared region, 58 dust aerosols trap the thermal radiation emitted from Earth's surface by absorption, which 59 60 generally leads to a positive warming radiative effect at TOA and surface (Sokolik et al., 1998). 61 In addition to DRE, dust can also influence the radiation and the hydrological cycles indirectly 62 through serving as cloud condensation nuclei and ice nuclei and affecting cloud microphysical 63 properties and cloud lifetime, known as indirect effects of dust (Twomey, 1977; Albrecht, 1989). The dust DRE depends on many factors including primarily the atmospheric dust content, 64 represented by its optical depth (DAOD), vertical distribution (especially important for LW DRE), 65 66 and particles' physico-chemical properties that are the particle size distribution (PSD), complex 67 refractive index (RI), and shape. Besides dust PSD, RI and shape, the dust DRE also depends on 68 the atmospheric composition and structure, notably the atmospheric vertical profile of clouds, 69 water vapor, and temperature, as well as surface properties (Yu et al., 2006). All of these properties vary in space and time and need to be characterized at the best possible spatio-temporal resolution 70 71 in order to get realistic dust DRE estimates.





72 Among all these factors, DAOD is of first order importance in determining dust DRE since 73 dust DRE is approximately linear with DAOD (Satheesh and Ramanathan, 2000). Many previous 74 studies related to dust DRE are based on DAOD distributions from model simulations. For example, Kok et al. (2017) used four global model simulations to estimate global mean dust DRE 75 76 efficiency (DREE is defined as DRE/DAOD) and further derive global mean dust DRE. Di Biagio 77 et al. (2020) derived dust DRE based on model-simulated DAOD distributions with global annual 78 mean DAOD constrained by observations. The main advantage of these studies is the availability 79 of continuous and detailed DAOD spatial and temporal variation from model simulations. On the 80 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 81 82 observational constraints for evaluation and improvement.

83 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 84 85 have developed sensor-specific methods to distinguish dust aerosol from total aerosol based on the 86 size and shape characteristics of dust particles. Some are based on passive satellite observations 87 such as Moderate Resolution Imaging Spectroradiometer (MODIS, Remer et al. (2005)) and others 88 are based on active observations such as Cloud-Aerosol Lidar with Orthogonal Polarization 89 (CALIOP, Winker et al. (2009)). The wide spectral coverage of MODIS measurements allows the 90 retrieval of aerosol particle size information, such as effective radius, fine-mode fraction, aerosol Angstrom exponent, as well as spectral gradient of absorption (Remer et al., 2005; Hsu et al., 2013). 91 92 Based on the fact that dust aerosols are generally larger in size than other aerosols and have a 93 decreasing absorption from ultraviolet (UV) to the near infrared, the combinations of these 94 retrievals provide the basis for dust separation and dust aerosol optical depth (DAOD) retrievals





from MODIS (Kaufman et al., 2005; Ginoux et al., 2012; Voss and Evan, 2020; Yu et al., 2009, 95 96 2019). In addition, some recent studies have also characterized dust distribution through 97 integrating MODIS measurements with other data sources and model simulations, for example, 98 using the DAOD-to-AOD ratio from MERRA-2 (Modern-Era Retrospective analysis for Research 99 and Applications, version 2), Gkikas et al. (2021) converted the MODIS AOD retrievals to DAOD. 100 However, passive sensors do not provide the vertical structure of aerosol that is critical for studying 101 aerosol-cloud interactions, LW radiative effects and aerosol influences on the thermal structure of 102 the atmosphere (e.g., Meloni et al., 2005, 2015). By contrast, the active sensor CALIOP can provide the vertical profiles of aerosol extinction and particle properties such as depolarization 103 104 ratio and color ratio, which have been used for improving DAOD retrievals in thermal infrared 105 (TIR) (Zheng et al., 2022) and evaluating global dust simulations (Yu et al., 2010; Wu et al., 2020). 106 The CALIOP dust identification is mainly based on dust aerosols being non-spherical in shape and 107 their linear depolarization ratio being much larger than spherical aerosols (Sakai et al., 2010).

108 Using CALIOP retrievals, Song et al. (2021) derived a three-dimensional (3D) decadal (2007-2019) global scale dust extinction profile climatology, which provides an observational 109 110 constraint on both the spatial DAOD pattern and the vertical dust distribution for studying dust 111 DRE and evaluating models. In their study, Song et al. (2021) also compared dust retrievals, in 112 particular DAOD, based on different methods and showed that DAOD often differ significantly 113 between the different products. For example, they showed that DAOD derived from CALIOP 114 observations is generally smaller and more concentrated over 'dust belt' regions - extending from 115 the west coast of north Africa to the Middle East, central Asia, and China - than that derived from 116 MODIS observations. These differences in DAOD in turn lead to different dust DRE estimations, 117 making it difficult to compare different studies to reach meaningful conclusions. Even an





agreement of DRE could be a result of the compensation between differences in DAOD and other aforementioned factors, such as dust microphysical properties. Therefore, DRE provides only a weak constraint on model. Instead, a normalized quantity, DRE efficiency (DREE) as the ratio of DRE to DAOD, has been widely used in inter-comparison studies and model evaluations (Di Biagio et al. 2020). Because of the elimination of DAOD, the DREE provides a stronger constraint on dust microphysical properties and their impacts on the dust DRE from different dust source regions (García et al., 2008).

125 In addition to DAOD, dust size is also an important factor in determining dust DRE 126 (Mahowald et al., 2014). Smaller particles are more effective at scattering SW radiation and super-127 micron particles are more effective at absorbing both SW and LW radiation (Tegen and Lacis, 128 1996). Therefore, when other parameters are equal, fine dust would generally have a more negative 129 SW DRE and a less positive LW DRE than coarse dust. Unfortunately, despite its importance, the 130 simulation of dust PSD in the models and satellite retrievals of dust size remain challenging tasks 131 (Ryder et al., 2019). As a result, there is a large uncertainty in our understanding of dust PSD. For 132 example, several recent studies suggested that model simulations tend to underestimate dust size, 133 especially the very coarse dust with diameter in excess of 5 µm (Adebiyi and Kok, 2020). 134 Moreover, dust RI and shape can be important for DRE estimation as well because besides dust 135 PSD they are the other two factors that determine dust spectral optical properties. As such, it is 136 important to investigate the sensitivity of dust DRE to dust PSD, RI and shape. Previous studies 137 suggest that large dust PSD and RI uncertainty leads to a large uncertainty in dust DRE and thereby 138 DREE estimations. For example, Song et al., (2018) shows that the SW DREE of a dust model 139 with a large size and less absorptive RI is very similar to that of a dust model with a smaller size 140 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

142 of effects in the dust PSD and RI.

143 The main objective of this study is to derive a global clear-sky size-resolved dust DREE 144 dataset based on satellite observations and demonstrate its usefulness in constraining, comparing, 145 and understanding the dust DRE estimations. As explained below, the size-resolved DREE 146 decomposes the DREE of dust into several size bins and therefore provide a way to take into 147 account the effects of dust PSD explicitly. The sensitivity of dust DRE to dust RI and shape are 148 also assessed in this study. Due to the inhomogeneous spatio-temporal distribution of those 149 aforementioned factors, it is thus important to consider the spatio-temporal variation of dust DREE. 150 Therefore, we organize the DREE dataset at 5° (longitude) \times 2° (latitude) horizontal resolution 151 and at monthly temporal resolution. To the best of our knowledge, this work presents the first such 152 dataset based on retrieved dust properties (i.e., DAOD vertical and horizontal distributions) from 153 satellite observations, although size-resolved DREE from model simulations have been used in 154 previous studies. We will show that our size-resolved DREE can allow users to readily compute 155 the DREE and DRE of dust based on any dust PSD (e.g., from model simulations, satellite 156 retrievals or in-situ measurements). We will also carry out an inter-comparison of the global dust 157 DRE estimations based on different dust PSD and compare the results with previous studies. With these functions, we expect that the size-resolved DREE will be a useful tool for both observational 158 159 and modeling studies of dust DRE.

160 The rest of the paper is organized as follows. Section 2 provides a description of the data 161 and models used in this study. Section 3 describes the methodology of deriving the size-resolved 162 DREE dataset. In section 4, we describe a methodology of calculating the dust DRE with the size-163 resolved DREE dataset and its validation. In section 5, we compare the regional and global dust





164 DRE estimations based on different DAOD, dust PSD and compare the results with previous

165 studies. Section 6 provides a summary of the study along with the main conclusions.

166 2 Data and Models

167 2.1 Satellite-based DAOD climatology

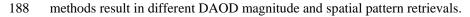
168 We use CALIOP-based DAOD climatology and dust vertical distribution derived in Song 169 et al. (2021) to derive a size-resolved dust DREE. The reason for choosing CALIOP-based DAOD 170 climatology is discussed in detail in section 3.2. The CALIOP-based dust climatology dataset 171 contains monthly mean DAOD and dust vertical extinction profile on a 5° (longitude) \times 2° 172 (latitude) spatial resolution grid for the period 2007-2019. The CALIOP-based DAOD and dust 173 vertical distribution climatology from 2007 to 2010 are used to derive monthly mean size-resolved 174 dust DREE dataset in this study. The selection of 4 years (2007-2010) for DREE calculations is 175 based on several considerations. Firstly, the multi-year DREE calculations allow us to investigate 176 the effect of interannual variations of atmospheric and surface properties to dust DRE. Secondly, this selection is consistent with Song et al. (2018), making it easier to compare our results with 177 178 previous work. Thirdly, considering the computational efficiency, we do not extend the calculation 179 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) spatial resolution for the period 2003-2019 by combining the monthly mean Aqua MODIS overocean (Yu et al., 2020) and over-land (Pu and Ginoux, 2018) DAOD. In contrast to CALIOP-based DAOD climatology which is based on dust non-sphericity to separate dust aerosol from CALIOP total aerosol observations, MODIS-based DAOD retrieval is mainly based on dust large size to

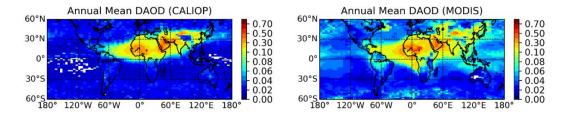




187 partition DAOD from MODIS total aerosol observations. The two sensor-specific dust partition



189 Figure 1 shows annual mean DAOD from 2007 to 2010 based on CALIOP and MODIS 190 observations. CALIOP-based and MODIS-based DAOD climatology differ in terms of both 191 magnitude and spatial pattern. MODIS-based DAOD is generally larger than CALIOP-based 192 DAOD. For example, the global $(60^{\circ}S - 60^{\circ}N)$ 4-year mean MODIS-based DAOD is 0.047, while CALIOP-based DAOD is 0.032. High DAOD are seen from both CALIOP-based and MODIS-193 194 based DAOD over the 'dust belt' regions, where large-scale dust activities occur persistently 195 throughout the year. However, the CALIOP-based DAOD is rather low in some other regions that are known to be dusty in certain seasons, such as southwestern United States, South America, 196 197 Australia, and South Africa. In other words, the two satellite-based DAOD spatial pattern differs 198 significantly with CALIOP-based DAOD more concentrated over 'dust belt' regions.



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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).

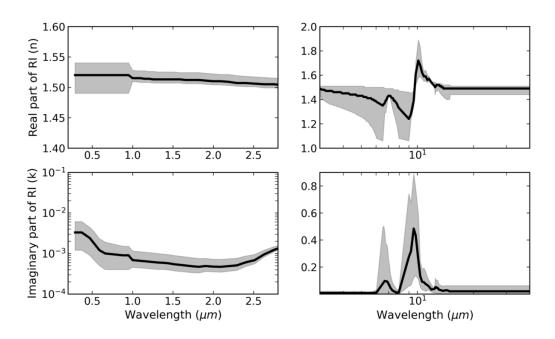
202 2.2 Dust physical and optical models

To study the sensitivity of dust DREE to dust RI and dust shape, we adopt three sets of dust RI (Figure 2) and two dust shapes (Figure 4 (a) in Song et al. 2018) and compute a total of 6 sets of DREE based on their combinations. The three dust RI sets represent less absorptive, mean absorptive and more absorptive dust aerosols and the two dust shapes include spherical and spheroidal dust shapes. The mean, 10th and 90th percentile of calculated RI for 19 dust samples





208 over 8 regions in Di Biagio et al. (2019) are used to represent mean, less and more absorptive dust 209 in SW. We combine RI of wavelengths from $0.37\mu m$ to $0.95\mu m$ measured in Di Biagio et al. 210 (2019) and RI of other wavelengths up to $3\mu m$ reported in Balkanski et al. (2007) to get full 211 spectral coverage in SW. The mean, minimum and maximum RI of wavelengths beyond $3\mu m$ 212 measured in Di Biagio et al. (2017) are used to represent mean, less and more absorptive dust in 213 LW. Two dust shapes are used to investigate the effect of dust nonsphericity on dust DRE. One is 214 spherical dust shape, the other one is spheroidal dust shape with dust aspect ratio distribution 215 described by Figure 4 (a) in Song et al. (2018) which is originally from Dubovik et al. (2006). 216 Each combination of dust RI and dust shape is considered as a dust model. As a result, the three 217 dust RI and two dust shapes constitute six dust models in SW and LW, respectively, as shown in 218 Table 1.



219

Figure 2. The SW and LW spectral refractive indices (RI) used in this study. 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. References for the used datasets are provided in Section 2.2.





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

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

226

227 **3 Methodology**

228 3.1 Size-resolved dust scattering properties

229 Rapid Radiative Transfer Model (RRTM) (Mlawer et al., 1997) is used to compute both 230 SW and LW radiative fluxes for both clean (i.e., cloud-free and aerosol-free) and dusty atmospheres (i.e., free of clouds and non-dust aerosols). RRTM retains reasonable accuracy in 231 232 comparison with line-by-line results for single column calculations (Mlawer and Clough, 1998; 233 Mlawer et al., 1997). It divides the solar spectrum into 14 continuous bands ranging from 0.2 to 234 $12.2 \,\mu$ m and the thermal infrared ($3.08-1000 \,\mu$ m) into 16 bands. We explicitly specify the spectral 235 DAOD, single scattering albedo (ω), and asymmetry parameter (g) of dust aerosols for every band 236 in the RRTM radiative transfer simulations. In contrast to radiative transfer scheme in most global 237 models, which do not account for LW scattering, scattering capability is available through the discrete-ordinate-method radiative transfer (DISORT) in RRTM LW (Stamnes et al., 1988). 238

Dust scattering properties (extinction efficiency Qe, ω and g) depend on several factors including dust PSD, RI, and dust shape. To account for the impact of dust PSD, we divide dust diameters into 10 logarithmically spaced size bins. The 10 size bins represent a wide range of dust geometric diameters (i.e., diameter of a sphere with the same volume) ranging from $0.1\mu m$ to $100\mu m$. The geometric diameter (hereafter diameter or *D*) range of each size bin is listed in Figure

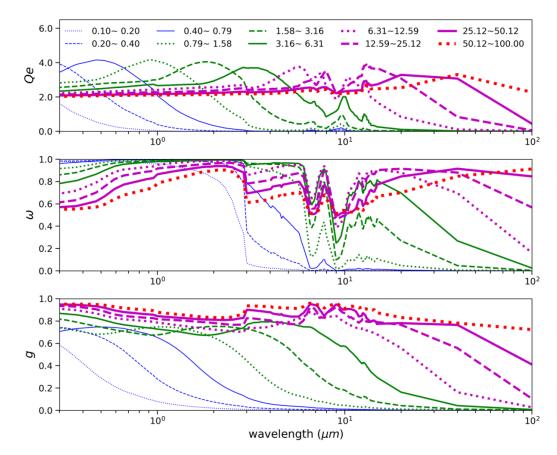




244	3. For each size bin k, the spectral scattering properties ($Qe_k^{\lambda}, \omega_k^{\lambda}$ and g_k^{λ}) are calculated for each
245	dust model shown in Table 1 and each spectral band. In the calculations of scattering properties
246	$(Qe_k^{\lambda}, \omega_k^{\lambda} \text{ and } g_k^{\lambda})$, dust particle number (dN/dD) is assumed to be uniformly distributed within
247	each size bin. We use the Lorenz-Mie theory code of Wiscombe (1980) to compute the spectral
248	optical properties of dust particles in the assumption of sphericity. The spectral optical properties
249	of spheroidal dust particles are derived from the database of Meng et al. (2010). Figure 3 shows
250	Qe_k^{λ} , ω_k^{λ} and g_k^{λ} for MeanSWRI-MeanLWRI-Spheroid dust model. In SW, finer dust has a larger
251	ω and smaller g, implying a more effective SW backscattering of finer dust. As a result, finer dust
252	is expected to have stronger cooling effect (more negative DREE values) at TOA generally. In
253	LW, $\frac{Qe_k^{10\mu m}}{Qe_k^{532nm}}$ is generally enhanced as dust size increases, which implies that coarser dust has larger
254	extinction in LW (optically represented by $DAOD^{10\mu m}$) than finer dust when $DAOD^{532nm}$ is
255	constrained by CALIOP retrieval. As a result, larger $DAOD^{10\mu m}$ will enhance the LW warming
256	(more positive LW DREE) at TOA of coarser size bins. On the other hand, the increased ω and g
257	of the coarser size bins indicates stronger forward scattering, which reduces the enhancement in
258	LW warming induced by larger $DAOD^{10\mu m}$.







259

Figure 3. Spectral scattering properties of each size bin for the MeanSWRI-MeanLWRI-Spheroid dust model. The scattering properties of each size bin are represented by the corresponding curve indicated in the legend. Each size bin is defined with respect to dust diameter with unit of micrometers (μm).

263 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. First, we use RRTM to simulate monthly mean dust DRE from 2007 to 2010 for each 5°

270 (longitude) \times 2° (latitude) grid with CALIOP-based $DAOD^{532nm}$ exceeding 0.01. The





271	$DAOD^{532nm} >= 0.01$ threshold ensures most dusty regions over the globe are covered (see Figure
272	S1 and Figure S2 in the Supplement) and in the meanwhile balances the computational cost. Dust
273	DRE are calculated for each size bin using the extinction properties of the corresponding size bin
274	shown in Figure 3 (denoted as $DRE_{k,i,j}$, hereafter k indicates size bin index and (i,j) indicates
275	longitude-latitude grid index, unless specified otherwise). Note that we do not consider dust RI
276	spatial variation and dust size vertical variation due to the lack of observation-based dust
277	minerology and size estimation on global scale. In $DRE_{k,i,j}$ calculations, we constrain the monthly
278	mean dust extinction vertical distributions using the CALIOP-based climatological dataset of Song
279	et al. (2021). Dust $DRE_{k,i,j}$ is calculated with respect to $DAOD_{i,j}^{532nm}$ from CALIOP-based DAOD
280	climatology. The atmospheric profiles such as water vapor (H2O), ozone (O3) and temperature
281	(T_{atm}) vertical profiles of 72 levels are from 3-hourly MERRA2 assimilated meteorological fields
282	data (Gelaro et al., 2017). We combine the 1-hourly surface albedo for visible beam from
283	MERRA2 radiation diagnostics with the instantaneous spectral surface albedo from the integrated
284	CALIPSO, Cloud-Sat, CERES, and MODIS merged product (CCCM) (Kato et al., 2011) to get
285	time-dependent spectral surface albedo. Surface temperature is obtained from 1-hourly MERRA2
286	radiation diagnostics data. The atmospheric and surface properties are all aggregated to monthly
287	mean values at eight UTC times: 0:30, 3:30, 6:30, 9:30, 12:30, 15:30, 18:30, 21:30 to obtain
288	monthly-mean diurnal cycle for radiative transfer simulations. Considering DRE ^{SW} strongly
289	depends on solar zenith angle (SZA), we calculate DRE ^{SW} for every 1 hour using the corresponding
290	hourly SZA in midmonth day. As a result, every three SZA share the same atmospheric and surface
291	properties in DRE ^{SW} calculations due to their different temporal resolution.

292 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 ^{mm}	The midmonth day of the month
$\overline{\frac{R(t)}{CO_2(t)}, \overline{T_{atm}(t)}},$	3-hourly monthly mean surface albedo and vertical profile of water vapor, ozone, carbon dioxide and atmospheric temperature
ζ_d	dust properties such as DAOD, dust extinction vertical profile and scattering properties
$\overline{{}_{1h}DRE^{SW}_{k,i,j}(tt)}$	1-hourly monthly mean DRE ^{SW} (i.e., monthly mean DRE ^{SW} at each of 24 UTC times) of k^{th} size bin and (i th , j th) grid
$\overline{_{3h}DRE_{k,i,j}^{LW}(t)}$	3-hourly monthly mean DRE ^{LW} (i.e., monthly mean DRE ^{LW} at each of 8 UTC times) of k^{th} size bin and (i^{th}, j^{th}) 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
$DREE_{k,i,j}$	The monthly and diurnally mean dust DREE ^{SW} and DREE ^{LW} of k^{th} size bin and (i^{th}, j^{th}) grid
$\overline{DAOD}_{i,j}^{532nm}$	The monthly mean dust optical depth at 532nm of (i^{th}, j^{th}) grid

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

$$\overline{{}_{1h}DRE_{k,i,j}^{SW}(tt)} = DRE_{k,i,j}^{SW}(\overline{R(t)}, \overline{H_2O(t)}, \overline{O_3(t)}, \overline{CO_2(t)}, \zeta_d, SZA(day^{mm}, tt)),$$
(1)

296 where 't' indicates 8 UTC times with 3-hour interval. 'tt' indicates 24 UTC times with 1-hour interval. ' day^{mm} ' indicates the midmonth day of the month, and ' $\overline{R(t)}$, $\overline{H_2O(t)}$, $\overline{O_3(t)}$, $\overline{CO_2(t)}$ ' 297 298 represent 3-hourly monthly mean surface albedo and vertical profile of water vapor, ozone, carbon 299 dioxide, respectively. The temporal resolution inconsistency of SZA as well as atmospheric and 300 surface properties requires every three SZA share the same atmospheric and surface properties in the calculations. ζ_d represents dust properties such as DAOD, dust extinction vertical profile and 301 302 scattering properties which are independent of UTC time in our calculations. Dust extinction 303 vertical profile is interpolated to the 72 levels in consistency with vertical profiles of water vapor, ozone and temperature from MERRA2. 304

305 Eq. (2) shows the way of deriving 3-hourly monthly mean DRE^{LW}. Surface emissivity ('*E*') 306 is obtained from Huang et al. (2016), which contains monthly mean spectral surface emissivity 307 with 0.5-degree spatial resolution. $\overline{T_{atm}(t)}$ represents 3-hourly monthly mean vertical profile of





atmospheric temperature. With the aid of the 3-hourly monthly mean atmospheric properties,
 monthly mean DRE^{LW} is calculated for every 3 hours.

$$\overline{_{3h}DRE_{k,i,j}^{LW}(t)} = DRE_{k,i,j}^{LW}(\overline{E}, \overline{H_2O(t)}, \overline{O_3(t)}, \overline{CO_2(t)}, \overline{T_{atm}(t)}, \zeta_d)$$
(2)

Then the 1-hourly monthly mean dust $DRE^{SW}(\overline{{}_{1h}DRE^{SW}_{k,i,j}(tt)})$ derived from Eq. (1) is averaged diurnally (over 24 points) to get the monthly and diurnally mean dust $DRE^{SW}(\overline{DRE^{SW}_{k,i,j}})$ as indicated by Eq. (3). Similarly, the 3-hourly monthly mean $DRE^{LW}(\overline{{}_{3h}DRE^{LW}_{k,i,j}(t)})$ derived from Eq. (2) is averaged diurnally (over 8 points) to get the monthly and diurnally mean dust DRE^{LW} $(\overline{DRE^{LW}_{k,i,j}})$ as indicated by Eq. (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.

$$\overline{DRE_{k,i,j}^{SW}} = \frac{\sum_{tt} \overline{\sum_{lh} DRE_{k,i,j}^{SW}(tt)}}{\sum_{tt} t}$$
(3)

$$\overline{DRE_{k,i,j}^{LW}} = \frac{\sum_{t} \overline{{}_{3h} DRE_{k,i,j}^{LW}(t)}}{\sum_{t} t}$$
(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 $DAOD^{532nm}$.

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



343



324 of monthly mean atmospheric and surface properties as well as dust vertical distribution. Finally, 325 the dataset developed in this study contains monthly mean size-resolved dust DREE and its 326 associated interannual std at TOA and surface with dimension of 10 bins, 12 months, 90 latitudes, 72 longitudes for each of six dust models in SW and LW respectively. Figure S1 and Figure S2 in 327 the Supplement demonstrate the global distribution of the monthly mean size-resolved DREE^{SW} 328 and DREE^{LW} at TOA for June. 329 330 It is important to note that dust DREE of each grid cell rarely depends on the DAOD 331 because dust DRE is approximately linear with DAOD (Satheesh and Ramanathan, 2000). 332 Therefore, the choose of CALIOP- or MODIS-based DAOD climatology to derive the global 333 $(5^{\circ} \times 2^{\circ})$ size-resolved DREE dataset will not lead to large difference. In other words, the size-334 resolved DREE dataset is rarely related to the robustness of the DAOD used in the derivation 335 process. We select CALIOP-based DAOD to derive the size-resolved dust DREE dataset because 336 that the CALIOP-based dust climatology contains dust vertical distribution, which is especially 337 important for obtaining LW DREE. Nevertheless, using CALIOP-based dust retrieval to derive 338 size-resolved dust DREE dataset has several limitations: (1) The size-resolved dust DREE dataset 339 may miss some regions with tenuous dust layers that below the CALIOP sensitivity. (2) The LW 340 DREE is related to the quality of dust vertical distribution retrieval. By contrast, dust DRE highly depends on DAOD, therefore we will use different DAOD climatological datasets retrieved from 341 342 different sensors (i.e., CALIOP and MODIS) to investigate global dust DRE in section 5.2.

344 global mean dust DREE will depend on the DAOD spatial distribution (i.e., DAOD 2D distribution)

Furthermore, even though dust DREE of each grid cell is rarely related to DAOD, regional or

global mean dust DKEE will depend on the DAOD spatial distribution (i.e., DAOD 2D distribution

in the region of interest (see details in section 5.2).

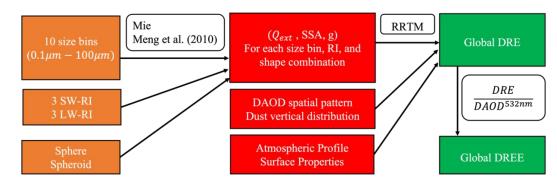




346 Based on the monthly mean size-resolved dust DREE datasets derived above, we further calculate globally annual mean size-resolved dust DREE^{SW} and DREE^{LW} at TOA and surface for 347 348 the six dust models (Figure 5). As discussed above, the global mean dust DREEs depends on the 349 DAOD spatial distribution, the global mean dust DREEs shown in Figure 5 is based on CALIOP-350 based DAOD spatial distribution from Song et al. (2021). Generally smaller bins cause stronger 351 cooling in SW and less warming in LW, which is consistent with our discussions in 3.1. This 352 observationally informed globally annual mean size-resolved dust DREE is also consistent with 353 the model-simulated results shown in supplementary Figure S3 in Kok et al. (2017) in terms of the 354 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^{SW} is 355 strongly sensitive to dust RI as DREE^{SW} of different dust RI is widely separated. Depending on 356 dust RI, DREE^{SW} switches from cooling effect (negative value) to warming effect (positive value) 357 358 at different size bins. More absorptive dust starts to warm the Earth system in SW at smaller dust size, and vice versa. In addition, our results suggest that DREE^{SW} is generally not sensitive to dust 359 shape. Specifically, dust shape is not important for DREE^{SW} in most size bins, while it is important 360 361 in the fourth size bin (D: $0.79\mu m \sim 1.58\mu m$) with DREE^{SW} of spheroidal dust obviously higher (less negative) than spherical dust. In the DREE^{LW}, dust shape is almost as important as RI for 362 363 several size bins.

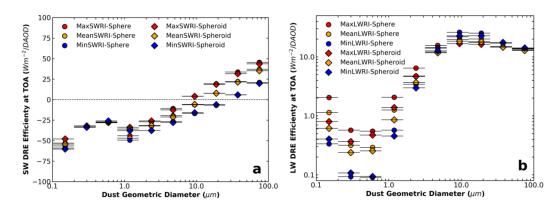






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Figure 4. Schematic of the methodology used to derive size-resolved dust DREE dataset. Orange boxes denote dust
 models used to calculate dust scattering properties. Red boxes denote inputs for RRTM. Green boxes denote outputs
 from RRTM.



368

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.

372 Our size-resolved dust DREE dataset is unique in many aspects: First, our DREE dataset 373 is derived based on CALIOP-based dust 3D distribution. Size-resolved DREE is derived for all 374 grids with CALIOP-based DAOD ≥ 0.01 . Second, our size-resolved DREE dataset covers a wide 375 range of dust diameters, specifically, they include dust DREE for ten dust diameter size bins 376 ranging from $0.1 \mu m$ to 100 μm . This is challenging, if not impossible, to obtain from global 377 models because these models generally simulate dust particles with diameter only up to 20 μm and 378 coarse dust particles in models deposit quickly and could not be sustained to the remote transport 379 regions (Huneeus et al., 2011; Adebiyi and Kok, 2020) where coarse particles have been observed





380 by in-situ measurements (Weinzierl et al., 2017). As a result, our size-resolved DREE dataset 381 achieves a wide spatial coverage for a large range of dust size. This is critical for investigating 382 impacts of coarse dust and even giant dust particles on dust DRE on both regional and global scales. 383 Third, considering that the dust vertical distribution is important for quantifying DRE^{LW}, we constrain dust vertical distribution using CALIOP-based dust retrievals in DREE^{LW} computation. 384 385 Fourth, our size-resolved dust DREE dataset accounts for dust LW scattering in DREE^{LW} 386 calculations since scattering capability is available through the DISORT in RRTM LW (Stamnes 387 et al., 1988). Dufresne et al., (2002) suggests that dust LW scattering enhances dust LW warming 388 effect at TOA by a factor of up to 50%. However, dust LW scattering is generally not considered 389 in most global models. Therefore, many previous studies artificially account for dust LW scattering 390 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^{LW} by 23% and Di 391 392 Biagio et al. (2020) augmented DRE^{LW} by 50%.

393 On the other hand, our size-resolved dust DREE dataset has several limitations. First, possible vertical variations in dust particle size are not accounted for in our calculation. The entire 394 395 dust-loading column is assumed to have the same dust size distribution. Second, we do not 396 explicitly account for spatial variation of dust RI, in other words, dust RI is assumed to be globally 397 uniform. This uncertainty is assessed through the sensitivity tests of DREE to dust RI using three 398 sets of state-of-the-art dust RI based on laboratory measurement of 19 dust samples all over the 399 world. Third, dust 3D distribution in the DREE calculation is constrained by CALIOP observations. 400 The limits on the sensitivity of CALIOP will affect the 3D distribution of dust in our calculation. 401 Fourth, we account for dust nonsphericity by using spheroidal shape model. This shape can't 402 perfectly represent the highly irregular shape and roughness of real dust. In addition, several





- 403 studies suggest that dust non-sphericity is underestimated by the spheroidal shape model (Huang
- 404 et al., 2020). The spheroidal shape model assumption thus might produce systematic errors.
- 405 Overall, the size-resolved dust DREE dataset is useful in many dust-related studies. First, 406 with our size-resolved dust DREE dataset, dust DRE could be calculated efficiently for any DAOD 407 magnitude, DAOD spatial pattern and any dust PSD for any regions or the globe (see details in 408 Section 4.1). Second, our size-resolved DREE dataset is derived for different RI and different dust 409 shapes respectively. As a result, we could estimate dust DRE uncertainty coming from DAOD, 410 PSD, RI, and shape separately to better understand major uncertainty sources in dust DRE 411 estimations. Third, our size-resolved DREE dataset could be used to evaluate model simulated 412 DREE for each size bin.

413 **4 DRE calculation methodology and its validation**

414 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.

419 DRE of full size range of dust can be expressed as the sum of DRE from each size bin 420 (DRE_k) . Dust DRE_k is approximated to be linearly proportional to DAOD of k^{th} size bin $(DAOD_k)$ 421 (Satheesh and Ramanathan, 2000). The similar concept of calculating dust DRE has been used in 422 previous studies e.g., Kok et al. (2017). Eq. (6) shows the process of computing dust DRE using 423 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)





424 where *DRE* represents dust DRE induced by full size range of dust with optical depth of *DAOD*.

425 f_k is the fraction of the DAOD contributed by the k^{th} size bin.

Each variable in Eq. (6) can be obtained or derived from datasets developed in this study and other studies. For example, the size-resolved DREE dataset ($DREE_{k,i,j}$) derived in this study is essential for utilizing this efficient and novel DRE calculation method. DAOD can be obtained from CALIOP-based or MODIS-based DAOD climatological datasets (Song et al., 2021). f_k can be derived from dust extinction efficiency (Qe), the geometric cross-sectional area (A) and dust 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 $Qe \equiv \frac{\sigma_e}{4}$, where σ_e is extinction cross section, the geometric 432 cross-sectional area of the particle (A) can be expressed as $A = \pi r^2$. Under the assumption of 433 spherical dust particle, r is the radius. Under the assumption of spheroidal dust particle, Vouk 434 (1948) shows that the average projected area of a convex body (e.g., spheroidal particle) is A =435 436 πr^2 , where r is the radius of a surface area-equivalent sphere. The average is taken over all 437 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 438 439 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 440 441 non-spherical dust in Kok et al. (2017) has a much larger value than spherical dust for dust $D \ge D$ 442 $1\mu m$ (see their Figure 1(b)). This discrepancy is probably due to the different Q_e definitions used 443 in the two studies. Kok et al. (2017) defined Q_e as dust extinction per unit cross section of volume-444 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
- 446 dust models shown in Figure 6 (a). In contrast, f_4 (i.e., f_k for the fourth size bin with D ranging
- 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
- 448 the larger difference in Qe^{532nm} with shape shown in Figure 6 (a).

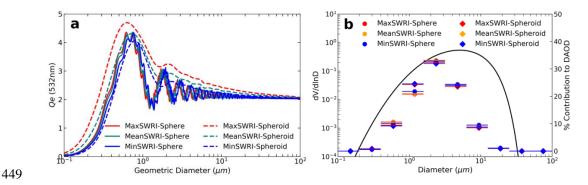


Figure 6. (a) Dust extinction efficiency (Qe) at 532nm for six dust models. (b) The colorful bars represent f_k calculated for six dust models based on a specific dust PSD (dV/dlnD) indicated by black curve. Note, f_k is not sensitive to different dust models such as dust RI and dust shape.

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.

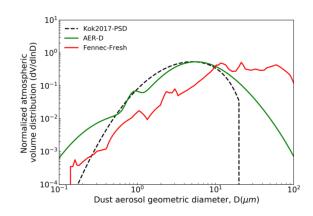
457 4.2 Validation of DRE calculation methodology

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

- 459 DREE-integration method. We choose MeanSWRI-MeanLWRI-Spheroid dust model and Fennec-
- 460 Fresh dust PSD (see red curve in Figure 7) measured within 12h of dust uplift in remote Sahara
- 461 locations by Fennec field campaign to represent microphysical properties of Saharan dust (Ryder
- 462 et al., 2013a, b). Monthly mean DAOD is from CALIOP-based DAOD climatology.





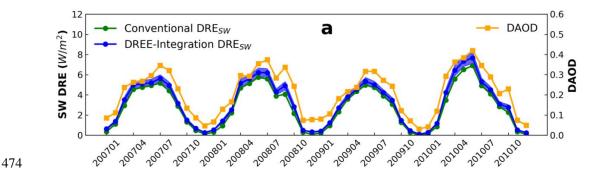




464 Figure 7. Normalized atmospheric dust volume distribution (dV/dlnD) described in Table 5 (Kok et al., 2017; Ryder 465 et al., 2013a; b; 2018; 2019).

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

473 calculation methodology.







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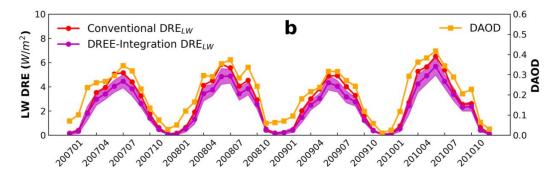


Figure 8. Monthly mean dust DRE^{SW} (a) and DRE^{LW} (b) comparison between *Conventional* and *DREE-integration*calculation from 2007 to 2010 over Sahara Desert. Shaded area along *DREE-integration* DRE indicates the one
standard deviation caused by the atmospheric and surface variations as well as 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.

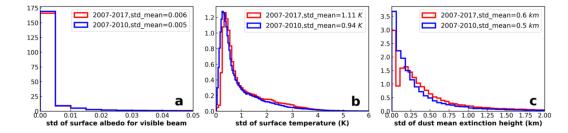
481 The shaded-area associated with DREE-integration DRE corresponds to the one standard 482 deviation of DREE caused by the 4-year (2007-2010) interannual variation of factors except dust 483 microphysical properties such as monthly mean atmospheric and surface properties as well as dust 484 vertical distributions (hereafter those factors is referred to as non-dust-factors for short). The 485 narrow shaded-area along DREE-integration DRE suggests non-dust-factors cause very small 486 uncertainty in dust DRE estimations. However, the small effects of 4-year interannual variation of 487 non-dust-factors may not necessarily be representative due to the limited number of years 488 considered. Section 2.1 discusses in detail for the reason of choosing 2007-2010 to derive size-489 resolved DREE dataset. To check the representative of 4-year interannual variation for non-dust-490 factors, we compare the 4-year (2007-2010) and 10-year (2007-2017) interannual standard 491 deviation (std) of monthly mean non-dust-factors (e.g., surface albedo, surface temperature and 492 dust vertical distribution) in Figure 9. To evaluate the interannual variation of dust vertical 493 distribution, we define dust mean extinction height (Z_{α}) referring to Koffi et al. (2012) as Z_{α} = $\frac{\sum_{i=1}^{n}\beta_{ext,i} \times Z_i}{\sum_{i=1}^{n}\beta_{ext,i}}$, where $\beta_{ext,i}$ is the dust extinction coefficient at 532nm at level *i*, and Z_i is the altitude 494 495 of level *i*. Nevertheless the 10-year std is slightly larger than 4-year std, they are both close to zero



500



- 496 and on the same order of magnitude. As such, even though our monthly mean size-resolved DREE
- 497 dataset is derived from 4-year (2007-2010) data, they could be used to represent DREE and
- 498 calculate DRE for other years considering the small sensitivity of monthly mean dust DRE to
- 499 interannual variation of non-dust-factors.



501 Figure 9. Probability density function (PDF) of 4-year and 10-year interannual standard deviation (std) in monthly 502 mean (**a**) surface albedo, (**b**) surface temperature, and (**c**) dust mean extinction height. The PSD analyses include 503 interannual std in 12 months and all 5° (longitude) \times 2° (latitude) grid cells over the world and their mean values are 504 indicated as 'std_mean' on each figure.

505 **5** Regional and global dust DRE based on size-resolved DREE dataset

506 After the validation of DREE-integration method in Section 4, we use the DREE-integration 507 method to calculate regional and global dust DRE in this section. There are three main objectives in this section: (1) the most important objective throughout this section is to demonstrate the 508 509 usefulness of the size-resolved DREE dataset for calculating regional and global dust DRE for any 510 given dust PSD; (2) the second objective is to validate the size-resolved DREE dataset by 511 comparing with regional dust DREE reported by field studies based on satellite and ground-based 512 observations (section 5.1); (3) the third objective to assess the sensitivity of dust DRE to DAOD 513 spatial pattern (section 5.2) as well as dust microphysical properties such as dust PSD, RI and 514 shape (section 5.3).

515 5.1 Comparison with observation-based regional dust DREE

516 Table 3 shows the comparison of our calculations of clear-sky regional mean SW and LW

517 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.
- 522 Knowledge of regional dust PSD is necessary for estimating dust DRE regionally. There are 523 several in-situ measurements of dust PSD over Sahara and tropical eastern Atlantic. The state-of-524 the art airborne observations of Saharan dust from the Fennec field campaign (Fennec-Fresh) and 525 transported Saharan dust over tropical eastern Atlantic within Saharan Air Layer (SAL) from both 526 AER-D and Fennec fieldwork campaigns are adopted (Ryder et al., 2013 a, b, 2018, 2019) (see 527 Figure 7). Both campaigns include giant dust particles, measuring up to $100\mu m$ diameter for AER-D and up to $300\mu m$ for Fennec. The wide coverage of dust diameter in our size-resolved DREE 528 529 dataset allows for dust DRE calculations for giant dust up to $100 \mu m$ over both dust source and 530 transported regions where giant particles are observed in those campaigns. This is an advantage of 531 our size-resolved DREE dataset compared to modeled dust DREE, because climate models 532 generally cut off dust diameter at 20 μm and could not sustain coarse dust to remote transport 533 regions due to several missing mechanisms in models (Van Der Does et al., 2018; Drakaki et al., 534 2022; Meng et al., 2022).

The Fennec-Fresh dust PSD includes measurements within 12h of dust uplift in remote Sahara locations. It is used to calculate dust DRE for Saharan dust in this section. In reality, dust over the wide Sahara Desert region (15N~30N, 10W~30E) is not all lifted within 12h, so using Fennec-Fresh to represent dust PSD over the wide Sahara Desert could bias dust size coarse, which could partially explain the warm bias in our DREE^{SW} estimation over the Sahara Desert compared to the satellite-based result. Over the tropical Atlantic, both AER-D and Fennec-SAL measured





- 541 PSD are used to assess the sensitivity of dust DREE to dust PSD. In addition, dust DRE is
- 542 calculated for three dust RIs to evaluate the sensitivity of dust DREE to dust RI as shown in Table
- 543 3. Generally, our dust DREE estimations achieve good agreement with observation-based dust
- 544 DREE. However, there is a significant uncertainty caused by dust RI in DREE, especially for SW.
- 545 In addition, DRE comparisons between AER-D and Fennec-SAL over the Tropical Atlantic
- 546 suggests that in-situ measured dust PSD uncertainty leads to a large uncertainty in regional DREE
- 547 in both SW and LW.
- 548 Based on the regional DREE study with the state-of-the art RI and PSD, we found DREE^{SW}
- 549 uncertainty could come from both dust RI and dust PSD, while DRE^{LW} uncertainty is mainly from
- 550 dust PSD.

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 then compare with observation-based results from previous studies. Note, spheroidal dust shape is assumed in our DREE-integration DRE calculations.

Shortwave Spectral Ran	ge						
Region	Season	Level	Satellite- Based DREE ^{SW}	This study			
					DREE ^{SW}		PSD
				Min RI	Mean RI	Max RI	
Sahara Desert ^(a) (15N~30N, 10W~30E)	JJA	TOA	0	2.8	16.0	26.6	Fennec-Fresh
Ilorin ^(f) , 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 ^(f)	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 ^(b)	JJA	TOA	-28	-44.6	-39.9	-35.3	AER-D
(10N~30N, 20W-45W)				-38.4	-32.1	-27.0	Fennec-SAL
		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- Based		This study		
			DREE ^{LW}		DREELW		PSD
				Min RI	Mean RI	Max RI	
Sahara Desert ^(a) (15N~30N, 10W~30E)	JJA	TOA	11~26	13.4	11.8	11.4	Fennec-Fresh
North Africa ^(d-e) (15N~35N, 18W~40E)	JJA	TOA	15~22	14.4	12.8	12.4	Fennec-Fresh
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 ^(g)	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 Brindle	· · · ·	Č (8). (c) Li et al. (f) Zhou et al.		U	1	(2003). (e)

556

557 5.2 Global dust clear-sky DRE based on different DAOD climatology

558 The DAOD is the most important factor in determining dust DRE. As illustrated in Song et 559 al. (2021), the DAOD retrieved from different satellite sensors have a large difference in terms of magnitude and spatial distribution. To evaluate how the current DAOD uncertainty affects dust 560 561 DRE estimations, the global dust DRE computed based on monthly mean DAOD climatology 562 retrieved from CALIOP observations and MODIS observations are compared in this section. To 563 separate the effect of DAOD from other factors, we use the same dust PSD, RI and shape in the 564 two sets of dust DRE calculations in this section. Specifically, we use the Fennec-Fresh PSD for 565 three major dust source regions (i.e., Sahara (14-30°N, 15°W-30°E), Middle East (10-35°N, 40-566 85°E) and eastern Asia (30-50°N, 75-130°E), they are indicated by three black boxes in Figure 10) 567 and use AER-D PSD for other regions (hereafter Campaign-PSD, see Table 5). The MeanSWRI-568 MeanLWRI-Spheroid dust model described in Table 1 is used to represent dust RI and shape.

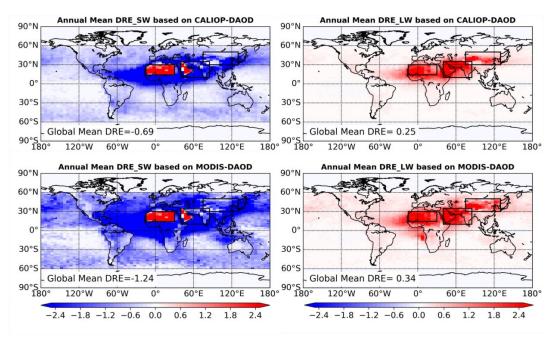
The two DAOD climatological datasets result in distinct dust DRE spatial pattern as shown in Figure 10, which is consistent with the DAOD spatial patterns shown in Figure 1 suggesting CALIOP DAOD is more concentrated over 'dust belt' regions than MODIS DAOD. The global mean dust DRE^{SW}, DRE^{LW} and DRE^{NET} based on the two DAOD climatology are significantly



586



573	different (Table 4), which is mainly caused by two factors. The first is the difference in DAOD
574	magnitude. The CALIOP-based global mean DAOD is 0.032, while MODIS-based is 0.047. The
575	other factor is the difference in DAOD spatial pattern. After we scale dust DRE to the same global
576	mean DAOD ($\overline{DAOD} = 0.03$) to eliminate the effect of DAOD magnitude difference (values in
577	parentheses in Table 4), the DRE ^{SW} difference reduced from 0.55 W m ⁻² (-0.69 vs1.24 W m ⁻²)
578	to 0.15 W m ⁻² (-0.64 vs0.79 W m ⁻²). Similarly, differences in DRE ^{LW} and DRE ^{NET} also reduce
579	significantly. It indicates that the global mean DAOD magnitude difference is more important than
580	the subtle difference in spatial pattern. Nevertheless, after scaling to the same global mean DAOD
581	there is still more than 10% difference between the two dust DRE ^{SW} , with CALIOP-based being
582	the more positive one. This is probably because CALIOP-based DAOD is more concentrated over
583	dust sources where dust aerosols induce less negative or even positive DRE ^{SW} (For example the
584	positive DRE ^{SW} over the Sahara Desert and Arabia shown in Figure 10), which result in a less
585	negative global mean DRE ^{SW} than MODIS.







- 587 Figure 10. Annual mean dust DRE global distribution based on CALIOP-based and MODIS-based DAOD 588 climatology. MeanSWRI-MeanLWRI-Spheroid dust model are used to represent dust RI and shape in the calculation. 589 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
- 591 regions.
- Table 4. Globally annual mean DAOD, DRE^{SW}, DRE^{LW} and DRE^{NET} based on CALIOP DAOD and MODIS DAOD
 climatology. Note, values in the parentheses are for the two DAOD scaled to the same value of 0.03.

	DAOD	DRE ^{SW} [Wm ⁻²]	$\overline{DRE^{LW}}$ [Wm ⁻²]	DRENET [Wm ⁻²]
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)

594

595 5.3 Global dust clear-sky DRE based on different dust PSD

596 In the section 5.2, we showed the dust DRE based on the Campaign-PSD. As aforementioned, 597 one of the main advantages of our size-resolved DREE is that it can be combined with different 598 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 599 600 calculations and their references. Kok2017-PSD is a globally averaged dust PSD and used to 601 represent dust PSD for each dusty grid cell. It is constrained with observations and includes coarse 602 dust particles up to $20\mu m$. Although our primary goal here is to demonstrate the capability of our 603 size-resolved DREE, the comparison between the two DRE can also help us understand the 604 impacts of dust PSD uncertainty on the dust DRE estimation. Moreover, we also investigate the 605 sensitivity of DRE to dust RI and dust shape explicitly in this section. The same DAOD 606 climatology (CALIOP-based DAOD climatology) is used for dust DRE calculations to eliminate 607 the impact of dust loading difference.

Several recent observation-constrained dust PSDs (e.g., Di Biagio et al., 2020, Adebiyi et al., 2020) suggest that dust size is coarser than Kok2017-PSD. As such, Kok2017-PSD is used to represent the lower limit of the observation-based global dust PSD to investigate the sensitivity of dust DRE to dust PSD. The Campaign-PSD is purely based on aircraft in-situ measurements and





612	the aircraft was extensively equipped to measure giant particles with diameter larger than $20\mu m$.
613	We use the dust PSD measured over Sahara (from the Fennec field campaign) to represent dust
614	PSD over three major dust source regions and use dust PSD measured in the Saharan Air Layer
615	over the tropical eastern Atlantic (from AER-D field campaign) to represent dust PSD over dust
616	transport regions. Of course, representing the spatially and temporally variation of global dust PSD
617	with only two PSDs from the field campaigns is only a crude approximation due to the lack of
618	PSD measurements. Dust aerosol over the three wide dust source regions may not be all uplifted
619	within 12 hours as in the Fennec-Fresh measurements, in addition, dust size after long-range
620	transport could be a bit finer than dust PSD measured over tropical eastern Atlantic (Weinzierl et
621	al., 2017). Thus, Campaign-PSD likely represents the upper limit of the observation-based global
622	dust PSD for the investigation of sensitivity to dust PSD. By contrast, the climate models miss
623	most of coarse dust (D>5 μ m) in the atmosphere (Adebiyi and Kok, 2020), as a result, the purely
624	modeled dust PSD without observational constraints will lead to a substantially different dust DRE.
625	Therefore, the sensitivity test to dust PSD conducted in this study can only represent the
626	uncertainty induced by the current understanding of observation-based dust PSD.

627 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	Kok et al. (2017)
	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.	
	Dust diameter is cutoff at $20\mu m$ (Figure 2a in Kok et al.2017).	
Campaign-PSD	Fennec-Fresh PSD is used for three major dust source regions (i.e.,	Ryder et al. (2013a,
	Sahara (14-30°N, 15°W-30°E), Middle East (10-35°N, 40-85°E) and	b, 2018, 2019)
	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.	

628

629

We calculated dust DRE of each grid cell $(DRE_{i,j})$ using DREE-integration method based 630 on the dust PSD described in Table 5. Global mean dust DRE was then calculated by averaging





dust $DRE_{i,i}$ weighted by its surface area. Figure 11 shows the global mean DRE^{SW} , DRE^{LW} and 631 632 DRE^{NET} at TOA, surface, and in the atmosphere calculated based on the two sets of PSDs. 633 Obviously, Kok2017-PSD leads to stronger cooling effect in SW and weaker warming effect in 634 LW at TOA compared to Campaign-PSD, which is consistent with the fact that Kok2017-PSD is 635 finer than the Campaign-PSD. In addition, we explicitly include the effects of dust RI and dust 636 shape on DRE in Figure 11. Comparison of uncertainty induced by dust PSD, RI and shape 637 suggests that dust RI uncertainty leads to the largest uncertainty in dust DRE, particularly RI uncertainty induces more than 40% uncertainty in DRE^{SW} estimations in the atmosphere (Figure 638 639 12). Dust PSD is also important for quantifying dust DRE, we found that the observation-based 640 dust PSD uncertainty induces around 15%~20% uncertainty in dust DRE at TOA and in the 641 atmosphere. Dust non-sphericity causes a negligible uncertainty in global mean dust DRE, in line 642 with previous studies e.g., Raisanen et al. (2013) and Colarco et al. (2014).

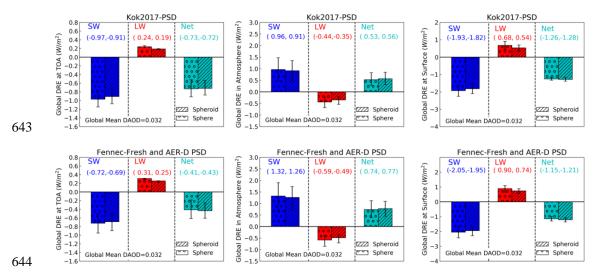


Figure 11. Globally annual mean clear-sky DRE^{SW}, DRE^{LW} and DRE^{NET} at TOA, in the atmosphere and surface calculated based on the two PSDs described in Table 5. The two rows represent dust DRE based on two PSDs. Error bars indicate uncertainty induced by dust RI uncertainty. Different types of 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.





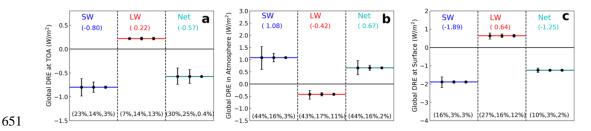


Figure 12. Comparison of uncertainty induced by dust RI, PSD and shape in DRE^{SW}, DRE^{LW} and DRE^{NET} at TOA (a), in the atmosphere (b) and surface (c). The horizontal lines in each plot represent global mean DRE^{SW} (blue line in the left column), DRE^{LW} (red line in the middle column) and DRE^{NET} (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.

It is tempting to compare our global mean dust DRE with results reported in Kok et al. (2017). But it must be noted that the global mean dust DRE shown in Figure 11 is for *clear sky* only, while the global mean dust DRE reported in Kok et al. (2017) is for *all sky*. The all-sky dust DRE can

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)

663 where CF is cloud fraction, $DRE_{clear-sky}$ is dust DRE simulated under the case of removing all clouds, $DRE_{cloudy-sky}$ is the dust DRE assuming whole grid is covered by clouds. To compare 664 our global mean dust DRE^{SW} based on Kok2017-PSD with the results reported in Kok et al. (2017), 665 we convert our clear-sky $DRE_{clear-sky}^{SW}$ to $DRE_{all-sky}^{SW}$ by using MODIS L3 monthly mean cloud 666 fraction. Specifically, we multiply $DRE_{clear-sky}^{SW}$ by (1-CF) for each grid cell and then calculate 667 global annual mean values. In this process, we neglect the cloudy-sky dust DRE^{SW} portion because 668 the annual mean cloudy-sky dust DRE^{SW} is estimated to be very small, around -0.04 (Zhang et 669 al., 2016). Finally, our estimated global mean $DRE_{all-skv}^{SW}$ corresponding to DAOD=0.03 is around 670 671 -0.34 Wm⁻². Although it is comparable to the -0.48 Wm⁻² from Kok et al. 2017, the following 672 differences between the two studies must be kept in mind when interpreting the results. First, the





rough conversion from global mean $DRE_{clear-skv}^{SW}$ to global mean $DRE_{all-skv}^{SW}$ is subject to the 673 674 approximation of global mean $DRE_{cloudy-sky} \sim 0$ and the MODIS L3 cloud fraction could be 675 different from modeled cloud fraction used in Kok et al. (2017). Second, the two studies use 676 different dust RI. Third, in this study Kok2017-PSD is used to represent dust PSD in each dusty grid and applied to our size-resolved dust DREE dataset to calculate global dust DRE. In contrast, 677 678 the model-simulated dust DREE in Kok et al. (2017) has reduced cooling from SW scattering and 679 enhanced warming from SW absorption effects because the short lifetime of coarse dust in models 680 concentrates these particles over bright deserts. Fourth, the two studies use different dust shape 681 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. 682 (2017) because that the lack of knowledge in $DRE_{cloudy-sky}^{LW}$ prevent us to convert $DRE_{clear-sky}^{LW}$ 683 to $DRE_{all-sky}^{LW}$. Moreover, the two studies use different dust vertical profile, which is critical for 684 685 DRE^{LW} estimations. For instance, dust vertical profile in Kok et al. (2017) is purely based on model 686 simulations, while this study constrains dust vertical profile with CALIOP observations. 687 Considering all these factors, it is hard to tell if the comparison is fair.

688

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) × 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.





695 The size-resolved DREE dataset allows us to calculate dust DRE of any DAOD 696 climatology and dust PSD efficiently by using the DREE-integration method presented in section 697 4.1 without involving radiative transfer simulations. The DREE-integration method is proven to 698 be in great agreement with conventional DRE calculations. With the DREE-integration methodology, we firstly calculated clear-sky regional mean DREE^{SW} and DREE^{LW} over the Sahara 699 700 Desert and tropical Atlantic. The comparison of our calculations with those reported by field 701 studies based on satellite and ground-based observations shows reasonable agreement. Secondly, 702 we estimated global mean dust DRE with two satellite-based DAOD climatological datasets and 703 two different global dust PSDs. We found that the global mean DAOD magnitude difference 704 between the two DAOD climatological datasets is more important than the subtle difference in 705 spatial pattern. Nevertheless, after scaling to the same global mean DAOD there is still more than 10% difference between the two dust DRE^{SW}, with CALIOP-based being the more positive one. 706 707 Moreover, our results explicitly show the uncertainty induced by each dust microphysical property 708 (i.e., dust PSD, RI and shape) separately. When DAOD is constrained: (a) Dust non-sphericity 709 induces negligible effect on dust DRE estimations; (b) The current understanding of observation-710 based dust PSD induces relatively large uncertainty (15%~20%) in dust DRE at TOA and in the 711 atmosphere (c) Dust RI turns out to be the most important factor in determining dust DRE, 712 particularly in SW. This implies that better understanding of dust mineral composition and RI will 713 significantly improve our understanding in dust DRE in the future.

714 *Data availability:*

The size-resolved dust DREE dataset and the codes to calculate dust DRE for any given
dust PSD and DAOD are available at

717 'https://drive.google.com/drive/folders/15_e28Y9JiSWiJnIM_2flEmt2u6i9phEY?usp=sharing'





- 718 CALIOP- and MODIS-based DAOD climatological datasets are available at
- $719 \quad `https://drive.google.com/drive/folders/1aQVupe7govPwR6qmsqUbR4fJQsp1DBCX?usp=shariing the standard sta$
- 720 ng'
- 721
- 722 Acknowledgement:

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