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2	Net Radiative Effects of Dust in Tropical North Atlantic Based on
3	Integrated Satellite Observations and In Situ Measurements
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27 Abstract

28 In this study, we integrate the recent in situ measurements with satellite retrievals of dust 29 physical and radiative properties to quantify the dust direct radiative effects on the shortwave (SW) 30 and longwave (LW) radiation (denoted as DREsw and DRE<sub>Lw</sub>, respectively) in the tropical North 31 Atlantic during summer months from 2007 to 2010. Through linear regression of CERES 32 measured top-of-atmosphere (TOA) flux versus satellite aerosol optical depth (AOD) retrievals, we estimate the instantaneous DREsw efficiency at the TOA to be  $-49.7\pm7.1$  W/m<sup>2</sup>/AOD and -33 34  $36.5 \pm 4.8 \text{ W/m}^2$ /AOD based on AOD from MODIS and CALIOP, respectively. We then perform 35 various sensitivity studies based on recent measurements of dust particle size distribution (PSD), refractive index, and particle shape distribution to determine how the dust microphysical and 36 37 optical properties affect DRE estimates and its agreement with abovementioned satellite-derived 38 DREs. Our analysis shows that a good agreement with the observation-based estimates of 39 instantaneous DREsw and DRELW can be achieved through a combination of recently observed 40 PSD with substantial presence of coarse particles, a less absorptive SW refractive index, and 41 spheroid shapes. Based on this optimal combination of dust physical properties we further estimate 42 the diurnal mean dust DRE<sub>sW</sub> in the region of  $-10 \text{ W/m}^2$  at TOA and  $-26 \text{ W/m}^2$  at surface, 43 respectively, of which ~30% is canceled out by the positive DRE<sub>LW</sub>. This yields a net DRE of about  $-6.9 \text{ W/m}^2$  and  $-18.3 \text{ W/m}^2$  at TOA and surface, respectively. Our study suggests that the 44 45 LW flux contains useful information of dust particle size, which could be used together with SW 46 observation to achieve more holistic understanding of the dust radiative effect.

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#### 49 **1. Introduction**

50 Mineral dust is the most abundant atmospheric aerosol component in terms of dry mass 51 [Choobari et al., 2014, Textor et al., 2006]. The Sahara is the largest source of atmospheric dust 52 aerosols, with an estimated emission of 670 Mt yr<sup>-1</sup> [Rajot et al., 2008, Washington et al., 2003]. 53 African dust from Sahara is regularly lifted by strong near-surface winds and transported 54 westwards within the Saharan Air Layer (SAL) over to the tropical North Atlantic during northern 55 summer [Cuesta et al., 2009, Karyampudi et al., 1999]. During the transport, dust aerosols can 56 scatter and absorb both shortwave solar (referred to as "SW") and longwave thermal infrared 57 (referred to as "LW") radiation, and thereby influence Earth's energy budget [McCormick et al., 58 1967, Tegen et al., 1996, Yu et al., 2006]. This is known as the direct radiative effect (DRE) of 59 dust, which can have a significant influence on the global energy balance [Boucher et al. 2013], 60 as well as regional weather and climate [e.g., Miller and Tegen 1998, Evan et al. 2006, Lau and Kim 2007]. Therefore, it is important to quantify dust DREs as accurate as possible. Moreover, 61 62 mineral dusts can also influence the life cycle and properties of clouds, by altering thermal 63 structure of the atmosphere (known as semi-direct effects) [Ackerman et al., 2000, Hansen et al., 64 1997, Koren et al., 2004], and by acting as cloud condensation nuclei and ice nuclei (known as 65 indirect effects) [Albrecht, 1989, Rosenfeld et al., 1998, Twomey, 1977]. In addition, when African 66 dust aerosols are deposited into Atlantic Ocean and Amazon Basin, they supply essential nutrients 67 for the marine and rainforest ecosystems [Yu et al., 2015], which has important implications for 68 the biogeochemical cycles [Jickells et al., 2005]. In this study, we focus on the quantification of 69 dust direct radiative effect on both SW and LW radiation.

Substantial effort has been made to understand and quantify the DRE of mineral dust since
the 1980s [*Carlson et al.*, 1980, *Cess*, 1985, *Liao et al.*, 1998, *Ramaswamy et al.*, 1985]. Most

72 studies have focused on the SW DRE (DREsw) of mineral dust under clear-sky (cloud free) 73 conditions [Myhre et al., 2003, Tegen et al., 1996, Yu et al., 2006]. Through scattering and 74 absorption, dust aerosols reduce the amount of solar radiation reaching the surface, inducing a 75 negative (cooling) effect at the surface. The DREsw of dust at the top of the atmosphere (TOA) 76 depends also strongly on the albedo of the underlying surface [Keil et al., 2003, Yu et al., 2006]. 77 Over a dark surface, the scattering effect of dust dominates, it leads to a negative DRE at TOA that 78 cools the climate system [Myhre et al., 2003, Tegen et al., 1996]. In contrast, high reflectance of 79 a bright surface enhances the absorption by dust aerosols and could yield a positive dust DREsw 80 (warming effect on the climate system) at TOA when the surface albedo exceeds a critical value 81 [Zhang et al. 2016, Xu et al., 2017]. Different from other aerosol types (e.g., smoke and sulfate 82 aerosols), dust aerosols are large enough to have significant LW direct radiative effect (DRE<sub>LW</sub>) 83 [Sokolik et al., 1999, Sokolik et al., 1998]. Lofted dust aerosols absorb the LW radiation from the 84 warm surface and re-emit the LW radiation usually at lower temperature, thereby reducing the 85 outgoing LW radiation and leading to a positive DRE at TOA that tends to warm the climate 86 system. At the same time, they emit the LW radiation downward that generates a warming effect 87 at the surface. The dust LW effect depends strongly on surface emissivity [Yang et al., 2009] and 88 the vertical profile of atmosphere temperature. The net radiative effect (DRE<sub>net</sub>) of dust is the 89 summation of its DREsw and DRELW. Note that DREsw only acts during daytime, whereas DRELW 90 operates during both day and night.

91 Quantification of the DREsw and DRE<sub>LW</sub> of dust remains challenging and there is a large 92 range of estimates in the literature. Take the Tropical Atlantic for example. Yu et al. [2006] found 93 that the seasonal (JJA) average clear-sky aerosol DREsw at TOA in this region varies from -5.794 W/m<sup>2</sup> to -12.8 W/m<sup>2</sup> based on observations and from -3.7 W/m<sup>2</sup> to -10.4 W/m<sup>2</sup> based on model

95 simulations. An important reason is that dust DRE depends on many factors, including both the 96 microphysical (e.g., dust particle size and shape) and optical (e.g., refractive index) properties, as 97 well as the surface and atmospheric properties (e.g., surface reflectance and temperature, 98 atmospheric absorption). Sokolik et al. [1998] showed that for the sub-micron dust particles, the 99 DREsw is dominant and DRELW is negligible, whereas for super-micron dust particles, DRELW is 100 more important [Sokolik et al., 1996, Sokolik et al., 1999]. Therefore, an accurate measurement of 101 the particle size distribution (PSD) is highly important for estimating the DRE of dust. However, 102 dust PSDs are highly variable and difficult to measure or retrieve, and, as a result, the observations 103 of dust PSD are usually subjected to large uncertainties [see Mahowald et al., 2014 and references 104 therein]. PSD inferred from AERONET observations [Dubovik et al., 2006] relies on observations 105 at shortwave channels, which could bias the dust size low. In fact, more and more observations are 106 emerging to suggest that dust PSD even in regions far from source regions contains substantial 107 fraction of coarse particles. Based on the airborne in-situ measurement of dust PSD in Caribbean 108 Basin from the Puerto Rico Dust Experiment (PRIDE) campaign, Maring et al. [2003] noted that 109 dust particles appear to settle more slowly than expected from the widely used Stokes gravitational 110 settling model. Similarly, recent measurements from the latest Fennec project [*Ryder et al.*, 2013b] 111 and the Saharan Aerosol Long-range Transport and Aerosol-Cloud-interaction Experiment 112 (SALTRACE) [Weinzierl et al., 2017] all suggest that transported dust aerosols in the SAL are 113 significantly coarser than expected based on the Stokes gravitational deposition. Such unexpected 114 existence of coarse particles has important implications for understanding the DRE of dust. In a 115 case of significant fraction of coarse particles, the warming effect on LW radiation (positive) 116 DRELW would partly cancel the DREsw leading to a less negative or even positive DREnet. Most 117 recently, Kok et al. [2017] argue that most of the current global climate models tend to

underestimate the size of dust particles and therefore overestimate the cooling effects of dust. Their estimate of the global mean dust  $DRE_{net}$  is between -0.48 and +0.20 W m<sup>-2</sup>, which includes the possibility that dust causes a net warming of the planet.

121 In addition to dust particle size, particle shape and refractive index also have significant 122 influence on dust DRE. Dust particles are generally nonspherical in shape, which make their 123 single-scattering properties (i.e., extinction efficiency, single-scattering albedo and scattering 124 phase matrix) fundamentally different from those based on spherical models. A few dust particle 125 shape models have been developed [Dubovik et al., 2006, Kandler et al., 2009], which have been 126 increasingly used in aerosol remote sensing and modeling [Levy et al., 2007]. Räisänen et al. [2013] 127 found that replacing the spherical dust models in a GCM with nonspherical model leads to 128 negligible changes in the DRE of dust at TOA. However, a recent GCM-based study by Colarco 129 et al. [2014] suggests that the influence of nonsphericity on dust DRE can be significant at surface 130 and within the atmosphere, depending on the refractive index of dust. Similarly, Kok et al. [2017] 131 argue that a spherical model significantly underestimates the extinction of dust, leading to errors 132 in estimate of dust DRE.

133 Over the past few decades, substantial efforts have been made to measure the spectral 134 refractive index of dust, mostly limited to the SW spectral range [Balkanski et al., 2007, Dubovik 135 et al., 2002, Dubovik et al., 2006, Formenti et al., 2011, Hess et al., 1998, Levoni et al., 1997]. 136 The current widely-used LW refractive index of dust was measured using rather old techniques in 137 the 1970s and 1980s [e.g., Volz 1972, 1973, Fouquart et al. 1987]. Recently, Di Biagio et al. [2014, 138 2017] compiled a comprehensive dust aerosol refractive index database in the LW spectrum 139 ranging from 3 to 15 µm, based on 19 natural samples from 8 dust regions over the globe. This 140 database is the first one as far as we know to document the regional differences in dust LW

refractive index due to the regional characteristics of dust chemical composition. We also call
special attention to a newly developed database of Saharan and Asian dust [Stegmann and Yang,
2017].

144 Satellite observations have long become indispensable for studying the dust aerosols. In 145 particular, the combination of passive (e.g., MODIS and CERES) and active (e.g., CALIPSO) 146 sensors on board of NASA's A-Train satellite constellation provides unprecedented data to study 147 dust aerosols, from long range transport [e.g., Liu et al. 2008, Yu et al. 2015] to dust DRE [e.g., 148 Yu et al. 2006, Zhang et al. 2016]. As A-Train observations become mature, substantial efforts 149 have been made to collocate and fuse the observations from different sensors to make the use of 150 A-Train observations easier for the users. A prominent example is the CERES- CALIPSO-151 CloudSat -MODIS (CCCM) product developed by Kato et al. [2011], which has become a popular 152 dataset for studying the radiative effects of clouds and aerosols and for evaluating GCMs.

153 The present study is inspired and motivated by the latest measurements of the 154 microphysical and optical properties of dust, namely the in-situ dust PSD from the Fennec field 155 campaign [Ryder et al. 2013a, 2013b] and the dust LW refractive index from Di Biagio [2014, 156 2017], as well as the recent studies (e.g., Kok et al. [2017]) suggesting that cooling effects of dust 157 is overestimated in most climate models due to the underestimation of dust size. The study is 158 carried out in three steps, each with a distinct objective. First, we attempt to derive a set of 159 observation-based instantaneous dust DREsw and DRELw for the tropical North Atlantic based on 160 the A-Train satellite observations reported in the CCCM product, without imposing any 161 assumptions on dust size, shape or refractive index. Here, the instantaneous dust DRE represents 162 dust DRE derived under the conditions (e.g., solar position, atmospheric condition) at the measured 163 or computed time to distinguish from the diurnally averaged DRE in Section 4. Then, we perform

164 multiple sets of radiative transfer computations of the *instantaneous* dust DRE in the North 165 Atlantic region based on the same dust extinction profiles from CCCM in combination with 166 different dust physical and optical properties. The objective is to understand the sensitivity of dust 167 DREsw and DRELW to the PSD, nonsphericity, and refractive index of dust and to obtain a set of 168 dust properties that yield the best agreement with satellite flux observations (e.g., CERES). In the 169 third step, we use the derived dust properties and extend the radiative transfer computations to 170 diurnal mean and to DRE at surface. The rest of this paper is organized as follows: Section 2 171 describes the data and model used. Section 3 presents the sensitivity of dust DRE to dust size, 172 shape and refractive index. Section 4 discusses diurnally averaged net DRE of dust aerosols and 173 uncertainty analysis. Section 5 concludes the article.

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- 175 **2. Data and Models**

#### 176 2.1 The CERES- CALIPSO-CloudSat -MODIS (CCCM) product

177 To estimate instantaneous dust DRE, we use aerosol and radiation remote sensing products 178 from the A-Train satellite sensors, namely, the integrated CERES, CALIPSO, CloudSat, MODIS 179 merged product (CCCM) developed by [Kato et al., 2011]. In the CCCM product, high-resolution 180 CALIOP, CloudSat and MODIS retrievals are collocated with 20-km CERES footprints. For each 181 CERES footprint, the CCCM product provides the TOA flux observations (both SW and LW) 182 from CERES, aerosol (MOD04 "Dark Target" product [Remer et al., 2005]) and cloud (MOD06 183 [Platnick et al., 2003]) properties retrieved from MODIS, aerosol optical thickness for each aerosol 184 layer from CALIOP [Winker et al., 2010] and cloud vertical profile from the combination of 185 CALIOP and CloudSat [Kato et al., 2010]. Up to 16 aerosol layers identified by CALIOP are kept 186 within a CERES footprint. Figure 1 shows the JJA mean aerosol optical depth (AOD) from the

187 CALIOP observations reported in the CCCM product. Clearly, the transported dust aerosols lead188 to enhanced AOD in the tropical North Atlantic region.

189 In addition to the "raw" retrievals, the CCCM product also provides post-processed flux 190 computations for each CERES pixel based on derived aerosol and/or cloud extinction profiles, 191 which is done in the following steps. First, the CALIOP aerosol retrievals within each CERES 192 pixel are averaged to obtain the aerosol extinction profile at the 0.5 µm reference wavelength. 193 Then, the aerosol type and associated spectral optical properties, e.g., extinction coefficient, single-194 scattering albedo, and asymmetry factor are specified mostly based on the aerosol type simulations 195 from the Model of Atmospheric Transport and Chemistry (MATCH [Collins et al., 2001], with 196 the exception of dust aerosols. If CALIOP observes dust aerosols (dust and polluted dust), the 197 aerosol type is set to dust. This is based on the consideration that the depolarization observation 198 capability of CALIOP is ideal for dust detection because the nonsphericity of dust can cause 199 significant depolarization in contrast to most other types of aerosols. Finally, the aerosol extinction 200 profiles and the aerosol spectral optical properties are used to compute the broadband fluxes at 201 both TOA and surface and for both SW and LW under 2 conditions: 1) with aerosol, 2) without 202 aerosol, so that the aerosol DRE can be derived from the difference of the two conditions. 203 Temperature and humidity profiles used in flux computations are from the Goddard Earth 204 Observing System (GEOS-5) Data Assimilation System reanalysis [*Rienecker et al.*, 2008].

205

206 2.2 Dust Physical and Optical models

To investigate the sensitivity of dust DRE to microphysical and optical properties of particles, we use several sets of widely used or newly obtained dust size distribution, dust shape distribution and dust refractive index.

210 Two dust particle size distributions (PSD) shown in *Figure 2*, are considered in this study. 211 One PSD is inferred based on AERONET ground-based retrievals at Cape Verde site (16°N, 22°W) 212 from [Dubovik et al., 2002] (referred to as "AERONET" PSD). The other dust PSD is obtained 213 from the recent airborne measurements of transported Saharan dust from the Fennec 2011 field 214 campaign over both the Sahara (Mauritania and Mali) and the eastern Atlantic Ocean, between the 215 African coast and Fuerteventura. Ryder et al. [2013a] separate the PSD measurements from this 216 campaign into three broad categories: fresh, aged, SAL (acronym for "Saharan Air Layer"). The 217 fresh category over the Sahara represents dust uplifted no more than 12 hours prior to measurement; 218 the aged category over the Sahara represents dust aerosols mobilized 12 to 70 hours prior to 219 measurement; the SAL category represents dust aerosols transported over the adjacent east 220 Atlantic, mostly from flights over Fuerteventura, Canary Islands (28°N, 13°W). All these 221 categories come from the mean of vertical profile observations (excluding the marine boundary 222 layer for SAL categories). The Fennec airborne PSD dataset is particularly novel, in that larger 223 particle sizes were measured than has been done previously in dust layers, with the exception of 224 Weinzierl et al., 2011, and that errors due to sizing uncertainties have been specifically quantified 225 (see Ryder et al., 2013b and Ryder et al., 2015 for full details). Because this paper focuses on the 226 Tropical Atlantic Ocean region, we use dust size distribution in the SAL category (referred to as 227 the "Fennec-SAL PSD"). Evidently from Figure 2, the Fennec-SAL PSD, which peaks around 228 5~6  $\mu$ m and has a significant fraction of particles with  $r > 10\mu$ m, is much coarser than the 229 AERONET PSD, which peaks around  $1 \sim 2 \mu m$  and has almost no particles  $r > 10 \mu m$ .

230 The dust refractive indices are taken from three sources:

(1) The Optical Properties for Aerosols and Clouds database (OPAC) [*Hess et al.*, 1998],
which has been widely used in climate models and satellite remote sensing algorithms.

233 (2) A merger of remote sensing based estimates of dust refractive indices in the shortwave 234 from 0.5 µm to 2.5 µm [Colarco et al., 2014], drawn from Kim et al. [2011] in the visible, and 235 Colarco et al. [2002] in the UV and (referred to as "Colarco-SW"). Kim [2011] collected the 236 AERONET (V 2) retrievals from 14 sites over North Africa and the Arabian Peninsula. Then the 237 dust refractive index is derived from the dust dominant cases for these sites selected based on the 238 combination of large aerosol optical depth (AOD  $\ge$  0.4 at 440 nm) and small Ångström exponent 239  $(Å_{ext} \le 0.2)$  to select the dust cases. Colarco et al. [2002] derived the dust refractive index in the UV 240 by matching the simulated dust radiative signature in the UV with the satellite observations from 241 the Total Ozone Mapping Spectrometer.

(3) The refractive indices in the LW from 3µm ~15µm from Di Biagio et al. [2017]
(referred to as "Di-Biagio-LW"). This database is based on the laboratory measurements of 19
natural soill sample from 8 regions: northern Africa, the Sahel, eastern Africa and the Middle East,
eastern Asia, North and South America, southern Africa, and Australia. The refractive index from
the Mauritania site is selected for this study because it is geographically close to the Fennec field
campaign.

*Figure 3* compares the real and imaginary parts of the refractive index for each of these data sets. In the SW, the imaginary part of the OPAC refractive index is much greater than that of Colarco-SW, which implies that dust aerosols based on the OPAC refractive index is more absorptive. In the LW, the Di-Biagio-LW refractive index is smaller than the OPAC values in terms of both the real and imaginary parts.

Dust aerosols are generally nonspherical in shape. Spheroids have proven to be a reasonable first-order approximation of the shape of nonspherical dust [*Dubovik et al.*, 2006, *Mishchenko et al.*, 1997]. The shape of a spheroid particle is determined by the so-called aspect

256 ratio, i.e., ratio of the polar to equatorial lengths of the spheroid. In our study, two spheroidal shape 257 distributions are used for computing the optical properties of non-spherical dust: (1) a size-258 independent aspect ratio distribution from Dubovik et al. [2006] (see Figure 4a) and (2) a size-259 dependent aspect ratio distribution extracted from Table 2 in Koepke et al. [2015], which is 260 discretized from measurement data of Kandler et al. [2009] (Figure 4b). The Dubovik et al. [2006] 261 shape distribution employs both oblate (aspect ratio < 1) and prolate (aspect ratio > 1) spheroids, 262 while the Kandler et al. [2009] shape distribution considers only prolate spheroids. For comparison 263 purpose, we also include spherical dust in our sensitivity studies. We use the Lorenz-Mie theory 264 code of Wiscombe [1980] to compute the optical properties of spherical dust particles. The optical 265 properties of spheroidal dust particles are derived from the database of Meng et al. [2010]. Note 266 that we assume volume equivalent radius for the AERONET-PSD to be consistent with Dubovik 267 et al. [2006] and the maximum dimension for Fennec-SAL PSD to be consistent with Ryder et al. 268 [2013b].

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#### 270 **2.3 Radiative transfer modeling**

The Rapid Radiative Transfer Model (RRTM) [*Mlawer et al.*, 1997] is used to compute both SW and LW radiative fluxes for both clear and dusty atmospheres. RRTM retains reasonable accuracy in comparison with line-by-line results for single column calculations. It divides the solar spectrum into 14 continuous bands ranging from 0.2  $\mu$ m to 12.2  $\mu$ m and the thermal infrared (3.08  $\mu$ m -1000  $\mu$ m) into 16 bands. We explicitly specify the spectral AOD,  $\omega$  and g of dust aerosols for every band in the radiative transfer simulations.

#### **3. Case Selection and Observation-based Estimate of Instantaneous Dust DRE**

278 **3.1 Selection of cloud-free and dust-dominant cases in the CCCM product** 

279 In this study, we focus on the Saharan dust outflow region in North Atlantic marked by the box in Figure 1 (10° N ~ 30° N, 45° W ~ 20° W). This selection is based on several considerations. 280 281 Firstly, during the summer months (JJA) this region is dominated by transported dust aerosols 282 from Sahara. Secondly, because the ocean surface is dark, dust aerosols have a strong negative 283 DREsw in this region. Thirdly, the abovementioned AERONET Cape Verde and Fennec-SAL PSD 284 measurements are made in the vicinity of this region. Finally, the dust DREs in this region have 285 been extensively studied in the literature, making it easier for us to compare our results with 286 previous work.

287 We first select cloud-free and dust-dominant CERES pixels in the region from four summer 288 seasons (2007~2010) of the available CCCM product. Within each CERES pixel, the CCCM 289 product report two cloud masks, one from CALIOP and the other from MODIS. The former is 290 more sensitive to optically thin clouds but has a very narrow spatial sampling rate available only 291 along the CALIOP ground track. The latter provides the cloud mask for the entire CERES pixel 292 but may miss thin clouds. Because of the relative large footprint size (~20 km), the cloud-free 293 condition actually poses a very strong constraint on the CERES product. Out of the total 36165 of 294 CERES pixels in this region from 4 seasons of data, we found 1663 (only 5%) of cloud-free pixels 295 according to the CALIOP cloud mask. The sampling is further reduced to 464 (only 1.3%) if the MODIS cloud mask is used to ensure the entire CERES footprint is cloud-free. This result is not 296 297 surprising because the MODIS cloud mask is more "clear-sky conservative", i.e., it tends to label 298 a pixel as cloudy if there is any ambiguity in its cloud mask test [Ackerman et al., 1998]. A 299 comparison of collocated CALIOP and MODIS cloud mask along the CALIOP track by [Holz et 300 al., 2008] reveals that MODIS masks more pixels as clear-sky than CALIOP does in the tropical 301 Atlantic dust outflow region (see their Fig. 3a), which is consistent with our result.

302 After selecting the cloud-free cases, we use the aerosol type information in the CCCM 303 product to further select dust-dominant cases (i.e., more than 90% of the aerosols within a given 304 CERES pixel are attributed to dust, in terms of area coverage). As aforementioned, the CCCM 305 product relies on CALIOP observations for detecting dust aerosols. After imposing the dust-306 dominant condition, we are left with a total of 607 and 245 cloud-free and dust-dominant CERES 307 pixels, if CALIPSO and MODIS cloud mask are used, respectively. Furthermore, we found that 308 within these selected pixels 153 out of 607 cases and 87 out of 245 cases have both CALIOP and 309 MODIS aerosol optical depth (AOD) retrievals in the CCCM product, and the rest (454 out of 607 310 cases and 158 out of 245 cases) have AOD retrievals only from the CALIOP, but no AOD retrieval 311 from MODIS. The reason for this is unclear and beyond the scope of this study, but perhaps due 312 to the more rigorous quality control used in the passive aerosol retrieval from MODIS [Remer et 313 al., 2005].

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#### 315 **3.2 Observation-based estimate of instantaneous dust DRE**

316 Many previous studies have shown that the aerosol DRE<sub>SW</sub> over the dark ocean surface is 317 approximately linear with the AOD. The increasing rate of the magnitude of DRE<sub>SW</sub> with AOD is 318 called the DREsw efficiency which is an important and useful quantity in many applications such 319 as aerosol model evaluation [Zhou et al., 2005]. We note that DREsw depends on solar zenith angle 320 (SZA). However, Because the selected region is relatively small, the SZA at the A-Train overpass 321 time in the domain only varies slightly among our selected cases, from 20° to 28°. Considering 322 the limited sample size and the small SZA variation, we therefore estimate DRE efficiency based 323 on the combination of all selected cases without breaking them into smaller SZA intervals. 324 Because of the nearly linear relationship between DREsw and AOD, the CERES TOA flux

325 observation and the collocated AOD retrievals from either CALIOP or MODIS can be combined 326 to derive an observation-based estimate of the instantaneous dust DRE. Figure 5 shows linear 327 regressions of CERES measured upward SW flux at TOA with satellite retrieved AOD for the 328 selected cloud-free and dust dominant cases. Black dots and lines are for selected cases using the 329 CALIOP cloud mask. For the 153 cases with both CALIOP and MODIS AOD retrievals, the 330 combination of CERES and MODIS (*Figure 5a*) leads to a DREsw efficiency of dust  $-49.7 \pm 7.1$ W/m<sup>2</sup>/AOD (AOD is at 0.5  $\mu$ m) with a linear regression  $R^2$  value of 0.69. The uncertainty, i.e., 331 332  $\pm$ 7.1 W/m<sup>2</sup>/AOD, associated with the regression line coefficients is estimated based on the 1-  $\sigma$ (one standard deviation) errors following Hsu et al. [2000]. The combination of CERES flux and 333 334 CALIOP AOD (*Figure 5b*) leads to a DRE<sub>sw</sub> efficiency of  $-36.5 \pm 4.8 \text{ W/m}^2/\text{AOD}$  based on 1-  $\sigma$ 335 error with a  $R^2$  value of 0.5. To investigate the impact of different cloud mask, we also show the 336 regression results derived from the cases selected based on MODIS cloud mask in Figure 5 (red 337 dots and lines). We notice that the results are very similar to those based on the CALIOP cloud 338 mask. Therefore, we conclude that the selection of cloud mask has negligible impact on our 339 estimation of DRE and the main uncertainty is associated with the AOD retrieval. Considering that 340 the MODIS and CALIOP aerosol retrievals are based on completely different methods, some 341 difference between the two are not surprising. The tighter correlation between MODIS AOD and 342 TOA upward SW flux is expected because MODIS retrieval is based on the reflected spectral solar 343 radiation, whereas the CALIOP AOD retrievals are based on the inversion of backward scattering 344 lidar signals. The potential reasons for the differences between CALIOP and MODIS AOD 345 retrievals are beyond the scope of this study. Interested readers are referred to a couple of recent 346 comparison studies by Kim et al. [2013] and Ma et al. [2013].

In summary, the *instantaneous* dust DREsw efficiency in the selected region during summer season is  $-49.7 \pm 7.1 \text{ W/m}^2/\text{AOD}$  based on CERES-MODIS observations and  $-36.5 \pm 4.8$ W/m<sup>2</sup>/AOD based on CERES-CALIOP observations. With the DREsw efficiency the DREsw can be easily derived from the AOD observations. The *instantaneous* DREsw estimated from the CERES-MODIS and CERES-CALIOP data is  $-14.2 \pm 2.0 \text{ W/m}^2$  and  $-10.4 \pm 1.4 \text{ W/m}^2$ , respectively (see Table 1).

353 In addition to the SW flux measurement, the CCCM product also provides the CERES 354 measurement of LW flux at TOA. Figure 6 shows the histograms of the broadband outgoing 355 longwave radiation (OLR) measured by CERES for the selected cases. Note that besides dust AOD, 356 OLR also strongly depends on other factors such as surface temperature, atmospheric profiles and 357 dust altitude. As a result, there is a high variability in those abovementioned factors among the 358 selected 607 cases. Therefore, it is not possible to derive the DRELW efficiency and DRELW in the 359 same way as we did for the SW. Here we use a different method. To estimate the DRE<sub>LW</sub>, we first 360 computed the dust-free OLR based on ancillary data of surface temperature and atmospheric 361 profiles reported in the CCCM which is from the GEOS Model of NASA's Global Modeling and 362 Assimilation Office (GMAO) [Kato et al. 2011]. Then, the DRELW can be estimated from the 363 difference between CERES observed OLR (i.e., blue solid line in Figure 6) and the computed dust-364 free OLR (i.e., black dashed line in Figure 6). We refer to this method as "semi-observation based" 365 as it is based on the combination of observed dust-laden OLR and computed dust-free OLR. To 366 test if our computed dust-free OLR has any potential bias due to, for example, errors in the 367 ancillary data (i.e., atmospheric gas and temperature), we selected 75 cloud free cases in the same 368 region and season with no dust detected by CALIPSO. Note that because of the small dust loading 369 in these cases the computed OLR at TOA mainly depends on the accuracy of ancillary data of

surface temperature and atmospheric profiles. Therefore, the comparison between the computed OLR and CERES measurements of those cases can inform us if there is any potential bias in our computation of dust-free OLR. It turns out that the difference between RRTM and CERES OLR has a mean value around 0.7 W/m<sup>2</sup> with standard deviation around 3.8 W/m<sup>2</sup> (not shown). Therefore, in the following analysis all our dust-free OLRs are reduced by 0.7 W/m<sup>2</sup> to account for this positive bias, which leads to a semi-observation-based *instantaneous* DRE<sub>LW</sub> of dust at 2.7  $\pm$  0.32 W/m<sup>2</sup> with the 95% confidence level.

## **4. Sensitivity of Dust DRE to Microphysical and Optical Properties of**

378 **Particles** 

379 The cloud-free and dust-laden cases from the CCCM product facilitate an ideal testbed for 380 investigating the sensitivity of dust DREs to the microphysical (i.e., PSD and shape) and optical 381 (i.e., refractive index) properties of dust. We use the aerosol extinction profiles at the 0.5  $\mu$ m from 382 the CCCM product (which is based on CALIOP/CALIPSO observations) and different 383 combinations of the dust properties to drive multiple sets of radiative transfer simulations of dust 384 DREs. Through comparisons of the radiative transfer simulations with CERES observation, we 385 study how the physical and optical properties influence both the DRE<sub>SW</sub> and DRE<sub>LW</sub> of dust. It 386 should be mentioned here that the CCCM product also use the same methodology to generate the 387 aforementioned post-processed flux profile. In the analysis, we will also compare our dust DRE 388 simulations with the results provided in the CCCM products.

### 389 **4.1 Sensitivity to dust size and refractive index**

390 In the first sensitivity study, we study the influences of dust size and refractive index on 391 the dust scattering properties and consequently dust DREs. Based on different combinations of the 392 PSDs (AERONET vs. Fennec-SAL) and SW refractive index (OPAC vs. Colarco-SW), we 393 simulate four sets dust spectral scattering properties (Figure 7), and correspondingly four sets of 394 dust DRE<sub>sw</sub> efficiency (Figure 8). In the simulations, dust particles are assumed to be spheroidal 395 and the aspect ratio distribution from Dubovik et al. [2006] (see Figure 4a) is used. The OPAC-396 LW refractive index is used. The impacts of dust shape distribution and LW refractive index on 397 dust DRE will be discussed later.

398 Figure 7 shows the scattering properties for the four different combinations of dust PSD 399 and refractive index. The extinction efficiency  $(Q_e)$  based on the Fennec-SAL PSD is significantly 400 larger than that based on the AERONET PSD (Figure 7a). The spectral shape is also different. The 401  $Q_e$  based on the Fennec-SAL PSD is rather flat in the SW region due to its large size whereas the  $Q_e$  based on the AERONET PSD decreases with wavelength. The  $Q_e$  shows no sensitivity to 402 403 refractive index in Figure 7a. It is because the Colarco-SW and OPAC-SW are different only in 404 the imaginary part (see Figure 3) which has minimal influence on  $Q_e$ . In contrast, the single 405 scattering albedo (SSA) in Figure 7b shows more sensitivity to refractive index. As expected, the 406 Fennec-SAL PSD and OPAC-SW combination (i.e., larger size and more absorptive refractive 407 index) has the smallest SW SSA while the AERONET PSD and Colarco-SW i.e., smaller size and 408 less absorptive refractive index) has the largest SW SSA. The other two combinations yield similar 409 SW SSA that are in between the abovementioned two extremes. The asymmetry factor (g) in 410 Figure 7c shows a primary sensitivity to size and a secondary sensitivity to refractive index.

Figure 7d shows spectral variation of dust AOD normalized with respect to AOD at  $0.5\mu m$ . The peak wavelength of solar radiation ( $0.5\mu m$ ) and peak wavelength of terrestrial thermal radiation ( $10\mu m$ ) are highlighted with dashed lines. The  $0.5\mu m$  AOD is used as the reference for normalization because as aforementioned, we use the  $0.5\mu m$  aerosol extinction profile in the 415 CCCM derived from CALIOP to drive our radiative transfer simulations. After spectral 416 normalization, one can see that given the same 0.5 µm AOD the 10 µm AOD based on the Fennec-417 PSD is much larger than that based on the AERONET PSD by around 80%. This is an important 418 feature that has important implications for the DRELW of dust. The SW reflection of dust depend 419 not only on AOD, but also SSA and g. Figure 7e shows spectral variation of AOD\*SSA\*(1-g), 420 where AOD indicates dust load, is multiplied by SSA to take the scattered fraction, and then 421 multiplied by (1-g) to take the backscattered portion. It is a quantity more relevant for 422 understanding dust SW reflection. Evidently, this index suggests that the combination of smaller 423 size (AERONET PSD) and less absorptive refractive index (Colarco-SW) leads to most reflective 424 dust among the four sets of simulations, whereas the larger size (Fennec PSD) and more absorptive 425 refractive index (OPAC) combination generates least reflective dust. The other two combinations 426 are in between and somewhat similar.

427 Figure 8 shows the four sets of simulated TOA upward SW fluxes as a function of the input 428 AOD at 0.5 µm. For comparison purpose, the DREsw efficiency regression results based on 429 observations in *Figure 5*, as well as the results reported in the CCCM products, are also plotted. 430 Focusing on our computations first, we note that as expected the most reflective dust based on the 431 combination of AERONET PSD and Colarco-SW refractive index leads to the largest DREsw 432 efficiency (-70.5 W/m<sup>2</sup>/AOD), while the least reflective dust based on the combination of Fennec-433 SAL PSD and OPAC ref yields the smallest DRE<sub>sw</sub> efficiency (-30.6 W/m<sup>2</sup>/AOD). Clearly, these 434 results are outside of the range based on observations (i.e.,  $-36.5 \pm 4.8 \sim -49.7 \pm 7.1 \text{ W/m}^2/\text{AOD}$ ), 435 suggesting they are too extreme. The other two combinations, i.e. AERONET PSD+OPAC-SW 436 and Fennec-SAL PSD + Colarco-SW, generate similar DREsw efficiency at -47.6 and -53.3 437 W/m<sup>2</sup>/AOD, respectively, both comparable to the CERES-MODIS based value. Interestingly, the

DREsw efficiency based on the flux computations reported in the CCCM product is -81 W/m<sup>2</sup>/AOD, even larger than that based on AERONET PSD + Colarco refractive index, suggesting that the dust model used in the CCCM flux computations is too reflective in the SW. The *instantaneous* DREsw and DREsw efficiency at surface for the two combinations that agree with the CERES observation, i.e., AERONET PSD+OPAC-SW and Fennec-SAL PSD + Colarco-SW, are given in the *Table 2*.

444 One additional point to note in Figure 8 is that, the TOA flux vs. AOD relations based on 445 the radiative transfer computations are much less scattered than those based on observations. The 446  $R^2$  value for the computation-based regressions all exceed 0.95, much higher than the observation-447 based results in Figure 5. This is because, in reality the TOA flux is influenced not only by AOD, 448 but also many other factors, such as surface reflectance variation, boundary layer aerosols that 449 might not be undetected by satellite, uncertainty in satellite retrieval algorithm. Most of these 450 factors are not accounted for in the radiative transfer computations, leading to a near perfect 451 correlation between TOA flux and input AOD. This should not be interpreted as a lack of 452 variability, rather than a smaller uncertainty.

On one hand, the results in Figure 8 are encouraging, as they suggest that a relatively simple 453 454 combination of dust size and refractive index can enable us to simulate the dust DREsw that are 455 comparable with observations. On the other hand, the fact that two different dust models lead to 456 similar DREsw efficiency simulation, both comparable with observation, points to a long-lasting 457 problem in aerosol remote sensing. That is, different combinations of aerosol microphysical and 458 optical properties can lead to similar radiative signatures. The combination of smaller dust size 459 with more absorptive refractive index is as good as the combination of larger size with less 460 absorptive refractive index, as long as DREsw is concerned.

But are the two combinations also equal in terms of closing the LW radiation? This is an important question, because ideally an appropriate dust model should close both SW and LW radiation. To address this question, we extend our radiative transfer simulations to the LW. It is important to point out that the LW and SW dust radiative properties are not independent but related through the physical properties of dust. For example, the AOD at a given wavelength  $\lambda$  in LW is related to the visible AOD through

$$AOD(\lambda) = AOD(0.5\mu m) \frac{Q_e(\lambda)}{Q_e(0.5\mu m)},$$
(1)

467 where  $Q_e$  is the extinction efficiency that is determined by dust size, shape and refractive index. 468 The dust size and shape are obviously independent of wavelength and therefore connect the SW 469 and LW. Even the refractive index in the SW and LW regions should be physically self-consistent 470 because refractive index is determined by the chemical composition of dust. Unfortunately, 471 because the refractive index measurements are often made either for SW only or LW only, there 472 is a lack of measurement of dust refractive index measurement from visible all the way to thermal 473 infrared.

474 In our computations, we first use the LW dust refractive index from OPAC to compute the 475 dust LW scattering properties and the corresponding OLR. Based on the same OPAC-LW 476 refractive index, the Fennec-SAL PSD yields an instantaneous DRE<sub>LW</sub> of +3.0 W/m<sup>2</sup> at TOA and 477  $+7.7 \text{ W/m}^2$  at surface (see *Table 3*). The results based on the AERONET PSD are significantly 478 smaller,  $+1.8 \text{ W/m}^2$  at TOA and  $+4.7 \text{ W/m}^2$  at surface. This difference between the two PSDs can 479 be easily understood with Figure 7b. Given the same visible AOD, the coarser Fennec PSD has a 480 larger infrared AOD than the AERONET PSD, and therefore stronger warming effects in the LW. 481 The more important question is which one, Fennec or AERONET PSD, leads to OLR 482 simulations that agree better with the CERES observation? The differences between the computed

483 OLRs and the CERES measurements of OLR for the selected dust cases are shown in Table 4, 484 together with the significance test results, i.e., 't-score' and 'p-value' from the Student's t-test. 485 Interestingly, the OLRs based on the combination of AERONET PSD + OPAC-LW refractive 486 index are systematically warmer (larger) than CERES measurements by an average of 0.9 W/m<sup>2</sup>. 487 The high t-score of 2.36 and low p-value of 0.02 indicate this warm bias to be statistically 488 significant. In contrast, the OLRs based on the combination of Fennec PSD + OPAC-LW refractive 489 index have a bias only at  $-0.5 \text{ W/m}^2$  and a p-value (0.55) significantly larger than the commonly 490 used 0.05 threshold, which means that OLR of this dust model is statistically indistinguishable 491 from the CERES measurements. Then, to investigate the sensitivity of the computation to LW dust 492 refractive index, we performed the computations again based on the Di Biagio et al. LW refractive 493 index. As shown in Table 4, the OLR based on Fennec PSD is still better than that based on the 494 AERONET PSD, even though both sets deteriorate slightly in comparison with the results based 495 on the OPAC LW refractive index. Overall, the size difference is the primary reason for the fact 496 that the OLR based on Fennec PSD is systematically smaller than that based on the AERONET 497 PSD. As shown in Figure 7, due to size difference, the  $Q_e$  based on the Fennec-SAL PSD (coarser) 498 decreases at a slower rate than that based on the AERONET PSD (finer). As a result, according to 499 Eq. (1) given the same SW AOD, the Fennec-SAL has a larger LW AOD and therefore less OLR 500 than the AERONET PSD. In comparison with our results, the OLRs reported in the CCCM product 501 (not shown here) are on average 3.1 W/m<sup>2</sup> larger than CERES measurements. This warm OLR 502 bias of CCCM product in the LW is consistent with its "too reflective" bias in the SW in Figure 8. 503 The LW result in Table 4 is interesting and important. First of all, it suggests that the LW 504 spectral region provides useful information content on dust properties that is complementary to 505 SW. As we see from Figure 8, the Fennec-SAL PSD + Colarco-SW refractive index and

AERONET PSD + OPAC-SW SW refractive combinations yield very similar SW radiation simulations. However, only Fennec PSD can lead to reasonable LW radiation simulation. Secondly, although the main point here is more about the usefulness of the information content in LW, the fact that the coarser Fennec PSD leads to better OLR simulation than AERONET PSD and CCCM product (based on MATCH) aligns with the recent studies (e.g., Kok et al. [2017]) arguing that dust size tends to be underestimated in the aerosol simulation models.

512 Finally, as expected, the combination of Fennec PSD + OPAC-LW also yields the best 513 simulation of the dust DRE<sub>LW</sub>, at  $3.0 \text{ W/m}^2$ , in comparison with the result derived from the CERES 514 OLR observations and RRTM dust-free OLR computation with ancillary data provided by CCCM 515 product (i.e.,  $+3.4\pm0.32 \text{ W/m}^2$  based on CERES-CALIPSO combination).

516

#### 517 **4.2 Sensitivity to dust shape**

518 In this section, we investigate the sensitivity of dust DRE to the shape (or shape distribution) 519 of dust. For all the computations in the last section, we have used the spheroidal dust model with 520 the aspect ratio distribution from Dubovik et al. [2006] (See Figure 4a). Now, we replace this 521 model with another spheroidal dust model by Kandler et al. [2009] shown in Figure 4b. For 522 comparison purpose, we also carry out another set of computation assuming spherical dust. For 523 dust size and refractive index, we use the Fennec-SAL and Colarco-SW/OPAC-LW refractive 524 index since dust DREs based on this combination has shown the best agreement with the 525 observations.

In Figure 9, we compare the scattering properties of dust based on three different shape models. Overall, the two spheroidal models are very similar and both significantly different from the spherical model. More specifically, in the SW the  $Q_e$  based on spheroidal models is

significantly larger than that based on spherical dust model. In the LW it is the opposite. The  $\omega$ in Figure 9b suggest that the spherical dust is more absorptive than spheroidal dust in the SW region, when other things are equal. Figure 9d and e show the normalized the AOD with respect to AOD(0.5  $\mu$ m) and the spectral variation of the scattering index AOD\*SSA\*(1-g). From Figure 9d we can see that given the same SW AOD, the spherical model has the larger LW AOD than the two spheroidal models. The comparison in Figure 9e reveals that the spherical dust model is less reflective than the spheroidal model in the SW.

536 Figure 10 shows the radiative transfer simulations for the selected cases based on the three 537 dust shape models. The DREsw efficiency based on the Kandler et al. [2009] is -48.3 W/m<sup>2</sup>/AOD, 538 which almost identical to the  $-47.6 \text{ W/m}^2/\text{AOD}$  based on the Dubovik et al. [2006] model. In 539 contrast, the DREsw efficiency based on the spherical dust model is much smaller -39.8 540  $W/m^2/AOD$ , which can be expected from the results in Figure 9e (i.e., spherical dust is less 541 reflective). Because the DRE<sub>SW</sub> efficiencies based on all three shape models are within the 542 observation-based values, we cannot tell if the spherical dust model is better or worse than the 543 spheroidal models.

544 As mentioned above, the two spheroidal dust models yield very similar OLR simulations 545 and are both statistically indistinguishable from the CERES observations. In contrast, the OLR 546 simulations based on the spheroidal dust models has a statistically significant  $-0.8 \text{ W/m}^2$  cold bias, 547 with a p-value of 0.03 (See Table 4). Overall, the r esults in Figure 10 and Table 4 indicate that 548 the two spheroidal models provide a slightly better, especially in LW, agreement with the 549 observations. Note that different shape models may have different angular and/or spectral signature 550 in terms of radiance, which is more important for satellite remote sensing. But this is beyond the scope of this study and will be investigated in future work. 551

#### 552 **5. Diurnally Mean Dust DRE in North Atlantic**

553 The DRE computations in the last section (i.e., Table 1~ Table 3) are instantaneous values 554 corresponding to the overpassing time of Aqua around 1:30PM local time. The strong solar 555 insolation makes the instantaneous DRE<sub>sw</sub> much larger than DRE<sub>LW</sub> in terms of magnitude, 556 leading to a strong negative DRE<sub>net</sub> (cooling) of dust. However, the DREsw operates only during 557 daytime, while the DRE<sub>LW</sub> operates both day and night. In addition, because of the availability of 558 satellite observations only at TOA, we have focused only on the DRE at TOA in the analyses 559 above. To appreciate the relative magnitude of DRELW with respect to DRESW we extend our DRE 560 simulations and analysis from *instantaneous* to *diurnal mean*, and also from TOA to surface. Over 561 tropical ocean, the OLR is most sensitive to sea surface temperature (SST). Our sensitivity study 562 based on the 3-hour MERRA (Modern-Era Retrospective analysis for Research and Applications) 563 data suggests that the diurnal SST variation in the tropical North Atlantic region is so small that 564 the diurnal mean OLR is close to the *instantaneous* value. Similarly, we also found that the diurnal 565 variation of atmospheric profile (e.g., water vapor) has negligible impact on the diurnal DREsw 566 computation. Therefore, we only compute the diurnal variation of DREsw due to the change of 567 solar zenith angle and ignore the small diurnal variation of DRE<sub>LW</sub> as well as the impacts of 568 atmospheric profile change on DRE<sub>sw</sub>.

Table 5 summarizes the key results of the diurnal mean DREsw and DREsw efficiency at TOA, as well as at surface. In the SW, the two most reasonable combinations of PSD and refractive index, Fennec-SAL PSD + Colarco-SW and AERONET-PSD + OPAC-SW leads to similar TOA DREsw efficiency around  $-29 \text{ W/m}^2/\text{AOD}$ , which is at the center of the  $-16 \sim -41 \text{ W/m}^2/\text{AOD}$ range reported in Yu et al. [2006]. At the surface, the DREsw efficiency based on these two combinations are around  $-83 \text{ W/m}^2/\text{AOD}$ , which is significantly stronger than the  $-27 \sim -68$ W/m<sup>2</sup>/AOD range reported in Yu et al. [2006]. It should be noted that we have limited this study to dust-dominant cases, whereas the values in Yu et al. [2006] are based on simple domain averageand include other types of aerosol.

578 By combining the information in Table 3 and Table 5, we can easily derive the net DRE<sub>net</sub> 579 of dust in the North Atlantic during summer. The TOA DREnet based on the combination of 580 Fennec-SAL PSD + Colarco-SW + OPAC-LW refractive indices gives a regional mean DRE<sub>net</sub> of -6.9 W/m<sup>2</sup> and -18.3 W/m<sup>2</sup> at TOA and surface, respectively. In comparison, the corresponding 581 582 values based on the combination of AERONET PSD + OPAC-SW + OPAC-LW refractive indices 583 are  $-8.5 \text{ W/m}^2$  and  $-22.5 \text{ W/m}^2$ , respectively. It is interesting and important to point out that the 584 DRELW is significant, about 17% ~ 36% (depending on the choice of PSD and refractive index) in 585 terms of magnitude with respect to the DREsw, and therefore not negligible in the DREnet 586 regardless whether for TOA or surface.

#### 587 **6. Summary and Discussions**

588 In this study, we use A-Train satellite observations reported in the CCCM product and 589 recent in situ measurements of dust properties to investigate the DREs of the dust aerosols in the 590 North Atlantic African dust outflow region during summer months. First, we select about 600 591 cloud-free and dust-dominant CERES pixels from 5 seasons of CCCM product. Based on these 592 cases, we first derive a set of observation-based instantaneous (corresponding to Aqua overpassing 593 time) DRE<sub>sw</sub> efficiency and DRE<sub>sw</sub> using the combination of CERES-measured TOA flux and 594 MODIS or CALIPSO retrieved dust AOD. The DREsw efficiency and DREsw based on CERES-595 MODIS observation are  $-49.7 \pm 7.1$  W/m<sup>2</sup>/AOD and  $-14.2 \pm 2$  W/m<sup>2</sup>, respectively. The values 596 based on the CERES-CALIOP combination are  $-36.5\pm4.8$  W/m<sup>2</sup>/AOD and  $-10.4\pm1.4$  W/m<sup>2</sup>, 597 respectively. Using the combination of CERES-measured OLR (i.e., with dust) and computed 598 dust-free OLR based on ancillary data, we also derive a set of semi-observation-based TOA 599 DRE<sub>LW</sub> between  $2.38 \sim 3.72 \text{ W/m}^2$ .

600 In the follow-up sensitivity study, we use the RRTM radiative transfer model to compute 601 the DRE of dust using the observed 0.5µm dust extinction profiles from CALIPSO under various 602 different assumptions of dust PSD, refractive index and shape distributions. We find that two dust 603 models, one based on Fennec-SAL PSD and Colarco-SW refractive index and the other on 604 AERONET PSD and OPAC-SW refractive index, provide the best fit to the observation-based 605 DREsw efficiency and DREsw. However, only the one based on the Fennec-SAL PSD, which is 606 much coarser than the AERONET-PSD, can also provide reasonable fit to the observation-based 607 DRELW. We also find that the DREs based on the two spheroidal dust models are quite similar to 608 each other, but more different from those based on spherical dust, suggesting that the detailed 609 shape distribution is less important in the calculation of dust DRE. Based on the dust model that 610 provides the best fit to the observation-based DRE, we estimate the diurnal mean dust DREsw 611 efficiency in the North Atlantic region during summer months (JJA) from 2007 to 2010 to be 612 around -28 and -82 W/m<sup>2</sup>/AOD at TOA and surface, respectively. The corresponding DRE<sub>SW</sub> is 613  $-9.9 \text{ W/m}^2$  and  $-26 \text{ W/m}^2$  at TOA and surface, respectively. The diurnal mean DRE<sub>LW</sub> is about 3  $W/m^2$  at TOA and 7.7  $W/m^2$  at surface. As dust aerosol properties varies temporally and spatially, 614 615 DREs of dust aerosols also have high spatio-temporal variation. Therefore, it is worthy to extend 616 the analysis to other regions and years in future studies.

617 Our estimation of the instantaneous TOA DREsw efficiency is in reasonable agreement 618 with the values reported in a recent study by Mishra et al. [2017]. Their observations are from a 619 satellite instrument similar to CERES, called Megha-Tropiques-ScaRaB (MT- ScaRaB). Flying in 620 a low-inclination orbit, this instrument is able to observe the TOA radiation in the tropical region

621 at various local times. Using 4 years MT- ScaRaB radiation and MODIS AOD observation, Mishra 622 et al. [2017] estimate that the instantaneous TOA DRE<sub>sw</sub> corresponding to a solar zenith angle of 623 ~40° in the North Atlantic region is about  $-40 \pm 3 \text{ W/m}^2/\text{AOD}$ , which is in between our range of  $-49.7\pm7.1$  W/m<sup>2</sup>/AOD and  $-36.5\pm4.8$  W/m<sup>2</sup>/AOD. Our estimation of the diurnal mean TOA 624 625 DRE<sub>SW</sub> efficiency ( $-28 \text{ W/m}^2/\text{AOD}$ ) is in between the  $-18 \text{ W/m}^2/\text{AOD}$  reported in Mishra et al. 626 [2017] and  $-35 \text{ W/m}^2/\text{AOD}$  reported in Li et al. [2004]. The difference may result from different 627 selection of cases and domain. Note that our analysis is limited to cloud-free and dust-dominant cases that are selected based on MODIS and CALIOP observations. 628

629 Due to the lack of study on dust DRE<sub>LW</sub> in this region, it is difficult to find a comparable 630 result the literature to validate our estimate of DRELW. Nevertheless, our result that the positive 631 DRE<sub>Lw</sub> cancels about 30% of the negative DRE<sub>sw</sub> in the computation of the diurnal mean net dust 632 DRE is in agreement with many previous studies attesting the importance of dust DRE<sub>LW</sub> (e.g., 633 Zhang et al. 2003, Haywood et al. 2005). Note that over land, e.g., the Sahara Desert, the brighter 634 surface reflectance will reduce the cooling effect of DREsw or even leads to warming (positive) 635 DREsw. At the same time, the hot surface temperature during daytime may result in DRE<sub>LW</sub> 636 significantly larger than that over ocean. Therefore, the DRELW is expected to be even more 637 significant in comparison with DREsw, over land than over ocean, which is an interesting topic for 638 future studies.

Another interesting result from this study is that given the same visible AOD dust particle size and dust absorption in the SW can compromise each other in determining dust DREsw. As a result, it is difficult to specify both variables using the SW radiation alone. In such case, the LW radiation could provide complementary and important information on dust properties, especially dust particle size. Most of the current aerosol property retrieval algorithms use only SW radiation observations. There are also a few algorithms to retrieve dust properties using only LW radiation
observation [e.g., Pierangelo et al., 2004, DeSouza-Machado et al. 2006, Peyridieu et al., 2010].
It is worth exploring in future studies the possibility and benefit of retrieving dust properties
utilizing both the SW and LW observations.

648 Finally, as discussed in Section 3.1, because the selected region is quite cloudy, and the 649 footprint of CERES is relatively large, the sampling rate of cloud-free and dust dominant cases is 650 very low. An important question arises from the low sampling is whether our results are representative. More specifically, one may wonder if our cloud-free cases are also representative 651 652 of the clear-sky part of those cloudy CERES pixels. To address this question, we investigated if 653 the dust properties (e.g., AOD and dust temperature) and meteorological conditions (e.g., surface 654 temperature and precipitable water) have any correlation with the cloud fraction. If the statistics 655 of the dust properties and meteorological conditions from our clear-sky cases are similar to those 656 from the cloudy cases, then our results are arguably representative of not only the clear-sky dust-657 dominant CERES pixels, but also the clear-sky part of cloudy dust dominant CERES pixels. To 658 this end, we first check the AOD. This time we selected all the dust-dominant cases based on 659 CALIOP observations regardless of the cloud fraction. Then, we divided all the cases into 5 groups 660 according to the cloud fraction within the CERES pixel, i.e., 0~20%; 20~40% 40~60% 60~80% 661 and >80%. As shown in Figure 11a, the dust AOD from the cloudy groups is statistically larger 662 than that from our cloud-free cases, which also means larger DRE<sub>sw</sub> if the DRE efficiency remains 663 the same. We don't know whether other dust properties, such as size, shape and refractive index, 664 are correlated with cloud fraction. Investigating this is extremely challenging, if not impossible, 665 using satellite observations. We have to leave this for future studies using other types of 666 measurements (e.g., in situ). In addition to dust AOD, we also checked the surface temperature,

667 the dust layer temperature (weighted by the dust extinction coefficient from CALIOP) and the total 668 amount of water vapor in the column. These quantities are potentially important for the DRE<sub>LW</sub>. 669 As shown in the Figure 11, in terms of the surface temperature (Figure 11 b) and dust layer 670 temperature (Figure 11 c), the cloudy dust-dominant cases are almost identical to our cloud-free 671 dust-dominant cases. However, not surprisingly, we found that the cloud-free cases are drier than 672 the cloudy cases (Figure 11 c). Note that, given the same dust properties, an increasing of water 673 vapor increases the atmospheric opacity in the LW, which tends to reduce the dust DRE<sub>LW</sub>. In 674 summary, if the dust particles properties (i.e., dust size, shape and refractive index) remain the 675 same, then the DREsw of dust in the clear-sky part of cloudy CERES pixels would be slightly 676 larger than that based on our results because of the larger AOD. In the LW, the larger AOD of the 677 clear-sky part of cloudy CERES pixels would lead to a larger DRE<sub>LW</sub>, but on the other hand, the 678 increased humidity under cloudy conditions counteract the effect of larger AOD. The net result is 679 dependent on the relative importance of these two competing factors.

# 682 Figures and Tables:

683

684

Table 1 Observation-based instantaneous (at A-Train overpassing time) DRE and DREsw

686 Efficiency at the top of atmosphere (TOA). The values in the parenthesis for DRE<sub>LW</sub> are based

687 on the assumption of 0.7  $W/m^2$  bias in our clear-sky OLR computation. See text for detail.

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	TOA DRE <sub>SW</sub> Efficiency $[W \cdot m^{-2} \cdot AOD^{-1}]$	$\frac{\text{TOA DRE}_{SW}}{[W \cdot m^{-2}]}$	$\frac{\text{TOA DRE}_{LW}}{[W \cdot m^{-2}]}$
CERES-MODIS AOD	-49.7±7.1	-14.2±2.0	3.1±0.60 (2.4±0.60)
CERES-CALIPSO AOD	-36.5±4.8	-10.4±1.4	3.4±0.32 (2.7±0.32)

Table 2 Instantaneous DREsw and DREsw Efficiency at TOA and Surface based on different dust models (e.g., PSD, refractive index, and shape). 

PSD	Refractive Index	Shape	TOA DRE <sub>SW</sub> efficiency (W/m²/AOD <sub>0.5μm</sub> )	TOA DRE <sub>sw</sub> (W/m <sup>2</sup> )	Surface DRE <sub>SW</sub> efficiency (W/m²/AOD <sub>0.5µm</sub> )	Surface DRE <sub>SW</sub> (W/m²)
Fennec-SAL	Colarco-SW	Dubovik	-47.6	-13.5	-179.4	-51.5
AERONET	OPAC-SW	Dubovik	-53.3	-15.5	-190.1	-55.0
Fennec-SAL	Colarco-SW	Sphere	-39.8	-11.4	-200.4	-58.2

725	Table 3 Instantaneous DRELw based on different dust models. Note that the diurnal mean values
726	are almost identical to the instantaneous results due to small diurnal variation in the LW.

PSD	Refractive Index	Shape	TOA DRE <sub>LW</sub> efficiency (W/m <sup>2</sup> /AOD <sub>0.5μm</sub> )	TOA DRE <sub>LW</sub> (W/m <sup>2</sup> )	Surface DRE <sub>LW</sub> efficiency (W/m²/AOD <sub>0.5µm</sub> )	Surface DRE <sub>LW</sub> (W/m <sup>2</sup> )
Fennec-SAL	OPAC-LW	Dubovik	10.5	3.0	26.9	7.7
AERONET	OPAC-LW	Dubovik	6.3	1.8	16.4	4.7
Fennec-SAL	Di-Biagio-LW	Dubovik	8.4	2.4	18.9	5.4
Fennec-SAL	OPAC-LW	Sphere	12.6	3.6	32.9	9.4

731	Table 4 The difference in OLR between our computations and the CERES measurements for the
732	selected dust cases. The values in the table are based on the assumption of 0.7 W/m <sup>2</sup> bias in our

732 clear-sky OLR computation.

PSD	<b>Refractive Index</b>	Shape	Mean Difference	Standard Deviation	T-score	P-value
Fennec-SAL	OPAC-LW	Dubovik	-0.2	3.8	-0.62	0.55
Fennec-SAL	Di-Biagio-LW	Dubovik	0.3	3.7	0.83	0.41
Fennec-SAL	OPAC-LW	Kandler	-0.4	3.9	-0.9	0.54
Fennec-SAL	OPAC-LW	Sphere	-0.8	4.0	-2.1	0.033
AERONET	OPAC-LW	Dubovik	0.9	3.7	2.36	0.02
AERONET	Di-Biagio-LW	Dubovik	1.5	3.7	3.94	8.5e-5

738 Table 5 Diurnally mean  $\mathsf{DRE}_{\mathsf{SW}}$  and  $\mathsf{DRE}_{\mathsf{SW}}$  Efficiency at TOA and Surface

PSD	Refractive index	Shape	TOA DRE <sub>sw</sub> Efficiency (W/m²/AOD)	TOA DRE <sub>SW</sub> (W/m <sup>2</sup> )	Surface DRE <sub>SW</sub> Efficiency (W/m²/AOD)	Surface DRE <sub>SW</sub> (W/m <sup>2</sup> )
Fennec-SAL	Colarco-SW	Dubovik	-28	-9.9	-82.1	-26.0
AERONET	OPAC-SW	Dubovik	-29.4	-10.3	-85.7	-27.2
Fennec-SAL	Colarco-SW	Spherical	-22.8	-8.2	-89.6	-28.5



766 Figure 1 CALIPSO derived seasonal mean (JJA) dust aerosol optical depth (AOD) at 0.5 µm averaged over five summers (2007~2010) in cloud free sky condition from the integrated CALIPSO, CloudSat, CERES, MODIS merged product (CCCM).




779 Figure 2 Size distributions of mineral dust used in this study. Fennec-SAL curve is from a new in-situ measurement of Saharan dust taken during the Fennec 2011 aircraft campaign [Ryder et al. 2013]. The solid curve represents desert dust size distribution retrieved from AERONET observations at Cape Verde site reported in Dubovik et al. [2002].









Figure 4 Two spheroidal dust shape distributions models a) shows aspect ratio distributions from Dubovik et al. [2006]. The ln $\epsilon$ -interval is 0.09. b) shows aspect ratio distributions as function of particle radius interval discretized from measurement of Kandler et al. (2009). The first point of each line covers the measurement data from  $\epsilon$ =1.0 to 1.3, the last point of each line covers  $\epsilon >$ 2.9 and the other points cover  $\epsilon$ -intervals of 0.2 Koepke et al. [2015].

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Figure 5 Linear regressions of CERES measured upward SW flux at TOA with satellite retrieved
AOD for the selected cloud-free and dust dominant cases (Black points represent the cases selected
using CALIPSO cloud mask along the ground track, red points represent the cases selected using
MODIS cloud mask over the entire CERES footprint). a) shows the regression results based on
MODIS AOD for cases (153 black points and 87 red points) with MODIS AOD retrievals. b) is
for all cases (607 black points and 245 red points) with CALIPSO AOD retrievals.



849 Figure 6 PDF of observed OLR from CERES (i.e., with dust) and computed dust-free OLR based

on the atmospheric profiles and surface temperature reported in CCCM. 







based on different combination of PSD and refractive index. PSD type and refractive index typeare indicated in legends.



874Dust AOD  $@ 0.5 \mu m$ 875Figure 8 The four sets of simulated TOA upward SW fluxes as a function of the input AOD at 0.5876 $\mu m$ . For comparison purpose, the DREsw efficiency regression results based on observations in877Figure 5, as well as the results reported in the CCCM products, are also plotted.



Figure 9 Extinction efficiency (Qe), b) single scattering albedo (SSA), c) asymmetry factor (g) d) normalized AOD with respect to AOD @ 0.5 µm, and e) AOD\*SSA\*(1-g) of dust aerosols based on different combination of PSD and refractive index. PSD type and refractive index type are indicated in legends.



916 Figure 10 shows the radiative transfer simulations for the selected cases based on the three dust917 shape models.



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 924 Figure 11 Histograms of a) dust AOD, b) surface temperature c) dust temperature and d) total
 925 column water vapor of dust dominant CERES pixels with different cloud fractions.

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