An overview of aerosol properties in clear and cloudy sky based on the CALIPSO observations

Yulan Hong¹ and Larry Di Girolamo¹

¹University of Illinois at Urbana Champaign

November 21, 2022

Abstract

A full understanding of the climatological properties of aerosols is an important step towards characterizing their effects on the regional and global climate. Utilizing the observations from Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO), we study cloud-free and cloudy aerosol properties with attentions on aerosol and cloud layer vertically relative positions. On a global scale, the cloud-free aerosols account for 55.9% of all detected aerosols with a mean optical depth () and uncertainty of 0.135 {plus minus} 0.047. The cloudy aerosols, accounting for 44.1%, have a larger and uncertainty of 0.143 {plus minus} 0.074 compared to the cloud-free aerosols. The above-cloud aerosols (4.2%), primarily composed of elevated smoke, dust/volcanic ash and polluted dust, have a much smaller (0.056 0.038). The below-cloud aerosols (21.3%) have ~ 0.165 0.087, showing close probability density distributions, aerosol types, backscatter-depolarization ratio diagram, and lidar ratio-color ratio diagram with the cloud-free aerosols. In addition, 27.4% of the detected aerosol profiles are found to have cloud layers vertically connected to aerosol layers. The lidar backscatter profiles of these aerosols have larger median values than the cloud-free, above-cloud and below-cloud aerosols. The seasonal variability of the cloud-free and the cloudy aerosols significantly varies with regions. This study implies that quantifying the impact of clouds, particularly cirrus due to the wide coverage of cirrus-aerosol overlap, on aerosol direct radiative effect is crucial to assess aerosols' roles in the Earth-climate system.

1	
2	
3 4	An overview of aerosol properties in clear and cloudy sky based on the CALIPSO observations
5	Yulan Hong ^a , Larry Di Girolamo ^a
6 7	a. Department of Atmospheric Sciences, University of Illinois at Urbana-Champaign, Urbana, Illinois, USA
8	Correspondence: Yulan Hong (yulanh@illinois.edu)
9	
10	
11	
12	Key Points:
13 14 15 16 17 18	 The study reveals that the below-cloud aerosols may share similar properties with those without clouds overlying. The wide coverage of the below-cloud aerosols indicates the significance of cirrus's impact on aerosol direct radiative effect. Regional and seasonal variations of the cloud-free and cloudy aerosols strongly depend on local climate system and long-track transport.
19	
20	
21	
22	
23	
24	
25	
26	
27	
28	
29	
30	
31	
32	

33 Abstract

- A full understanding of the climatological properties of aerosols is an important step towards
- characterizing their effects on the regional and global climate. Utilizing the observations from
- 36 Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO), we study cloud-
- 37 free and cloudy aerosol properties with attentions on aerosol and cloud layer vertically relative
- positions. On a global scale, the cloud-free aerosols account for 55.9% of all detected aerosols
- with a mean optical depth ($\bar{\tau}_a$) and uncertainty of 0.135 ± 0.047. The cloudy aerosols, accounting for 44.1%, have a larger $\bar{\tau}_a$ and uncertainty of 0.143 ± 0.074 compared to the cloud-free
- 40 aerosols. The above-cloud aerosols (4.2%), primarily composed of elevated smoke, dust/volcanic
- 42 ash and polluted dust, have a much smaller $\bar{\tau}_a$ (0.056±0.038). The below-cloud aerosols (21.3%)
- have $\bar{\tau}_a \sim 0.165 \pm 0.087$, showing close τ_a probability density distributions, aerosol types,
- backscatter-depolarization ratio diagram, and lidar ratio-color ratio diagram with the cloud-free
- 45 aerosols. In addition, 27.4% of the detected aerosol profiles are found to have cloud layers
- vertically connected to aerosol layers. The lidar backscatter profiles of these aerosols have larger
- 47 median values than the cloud-free, above-cloud and below-cloud aerosols. The seasonal
- variability of the cloud-free and the cloudy aerosols significantly varies with regions. This study
- implies that quantifying the impact of clouds, particularly cirrus due to the wide coverage of
- 50 cirrus-aerosol overlap, on aerosol direct radiative effect is crucial to assess aerosols' roles in the
- 51 Earth-climate system.
- 52

53 1. Introduction

54 Aerosols impact on Earth weather and climate system by their direct and indirect effects, 55 which has been intensely studied in the past few years (Andreae & Rosenfeld, 2008; Chand et al., 2009; Ten Hoeve & Augustine, 2016; Lohmann & Feichter, 2005; Matus et al., 2015; 56 57 Rosenfeld et al., 2014; Tan et al., 2017). According to the most recent Intergovernmental Panel on Climate Change (IPCC) report, the radiative forcing due to aerosols from observation-based 58 evidence was assessed to be -0.4 ± 0.4 W m⁻² (IPCC, 2021), which is likely able to offset part of 59 the warming induced by the emission of greenhouse gases (Rosenfeld et al., 2019), but with the 60 uncertainty in the same order as the estimated value. 61

Whether aerosols directly warm or cool locally not only depends on aerosol physical and 62 63 chemical properties but only relies on the properties of the vertically overlapping clouds. For instance, biomass burning aerosols can switch from a net cooling to a net warming effect at the 64 top of atmosphere (TOA) with an increase of underlying cloud fraction (Chand et al., 2009; Keil 65 & Haywood, 2003). Similarly, increase of underlying cloud fraction and optical depth was found 66 to enhance the warming effect of dust aerosols (Xu et al., 2017). A cirrus layer overlying an 67 aerosol layer is able to cause modest change of the radiative effect of absorbing or nonabsorbing 68 69 aerosols at TOA (Liao & Seinfeld, 1998). Considering the wide coverage of cirrus clouds globally (Hong & Liu, 2015), cirrus impact on aerosol direct radiative effect could be 70

71 extraordinary, which by now has not been assessed over the globe.

Incomplete knowledge of cloud-aerosol interactions also challenges an accurate estimate
of aerosol direct radiative effect. Cloud active aerosols indirectly interact with clouds by
modifying cloud microstructure such as reducing liquid cloud effective radius (Ross et al., 2018;
Tan et al., 2017), suppressing precipitation (Andreae & Rosenfeld, 2008; Rosenfeld et al., 2014),

increasing cloud cover in precipitating clouds or the opposite in non-precipitating clouds

77 (Christensen & Stephens, 2011; Lebsock et al., 2008). These effects in turn modify aerosol direct

radiative effect with the extent of modification depending on cloud-aerosol relative positioning

in the atmosphere. Nevertheless, the fundamental understanding of aerosol-cloud-precipitation

80 interactions and their representation in large-scale models is insufficient. For instance, IPCC

81 AR6 reports that the total aerosol forcing including aerosol-radiation and aerosol-cloud

interaction vary widely between different climate models from -1.50 to -0.68 W m⁻² (IPCC, 2021).

Additionally, clouds modify aerosol properties. For instance, aerosols exposed near cloud edge were found with the lidar backscatter and extinction coefficient enhanced resulted from humidification effects on aerosol properties (Rauber et al., 2013; Tackett & Di Girolamo, 2009). While aerosol loading above low-level cloud top is posited to have no significant difference with adjacent clear sky over the Southeast Atlantic according to the ORACLES (ObseRvations of Aerosols above Clouds and their intEractionS) filed campaign data (Shinozuka et al., 2020).

However, Chung et al., (2016) stated that aerosol optical depth (AOD or τ_a) above low-level

91 cloud is generally smaller than clear sky in the nearby over the globe as showed by the

92 CALIPSO observations. The disagreement between these two studies indicates how clouds

modify aerosol properties near cloud top locally or globally is not clear. This incomplete

knowledge also posts challenges in reducing the uncertainties in aerosol radiative effects.

Characterizing the amount and distribution of aerosols and their co-occurrence with 95 clouds from observations is an important step to improve the estimate of aerosol direct and 96 indirect effects on the Earth climate system. Global or regional aerosol climatologies which show 97 aerosol loadings, seasonal variations, land-sea contrast derived from passive sensors have been 98 displayed in a series of studies (Kim et al., 2007; Mao et al., 2014; Mehta et al., 2016; 99 Mishchenko et al., 2007). These climatologies represent the cloud-free aerosols, which has been 100 applied for a wide range of purposes, such as assessment of their effects on climate (Loeb & 101 Manalo-Smith, 2005) or model evaluation (Palacios-Penã et al., 2019). A climatology only 102 considering the clear-sky aerosols could be biased if aerosols occurring in cloudy sky 103

significantly differ from those in clear sky.

Active sensors probing into atmosphere with cloud and aerosol vertical structures resolved are able to detect aerosols in cloudy sky. For instance, by utilizing the CALIPSO lidar, aerosol and cloud layers are decoupled, and a 3-D aerosol climatology have been derived in both clear and cloudy skies (Winker et al., 2013). Also, attentions have been paid to the global distributions of the above-cloud aerosols with the help of the CALIPSO observations (Alfaro-Contreras et al., 2016; Devasthale & Thomas, 2011). Insufficient studies target to the aerosols appearing below clouds detected from space with the concern of the lidar signal attenuation and

additional uncertainty induced from the cloud layers (Chung et al., 2016; Tackett & Di

113 Girolamo, 2009). The CALIPSO aerosol algorithm has considered appropriate signal correction

through first retrieving the overlying layers and then using the retrieved attenuation for the

- attenuation correction below (Winker et al., 2009).
- In this study, using the CALIPSO observations, our main objective is to characterize the
 properties of aerosols in clear and cloudy skies with their uncertainties considered. The
- 118 distributions of cloudy aerosols are studied by considering the relative positions of cloud and
- aerosol layers. Overall, we focus on the following scientific questions:
- 120 1. How the spatial distributions of the aerosols occurring above, below or vertically adjunct to
- 121 clouds differ from the cloud-free aerosols;

- 122 2. What are the optical properties and lidar parameters for the aerosols above, below or adjunct
- to cloud layers and the cloud-free aerosols;
- 3. What are the differences in the regional and seasonal variabilities of the cloudy and the cloud-free aerosols.
- 126

127 2. Data and Methodology

- 128 **2.1 CALIPSO**
- 129

CALIPSO launched in April 2006 carries three primary instruments: the Imaging Infrared 130 Radiometer (IIR), the Wide Field Camera (WFC) and the near-nadir view Cloud-Aerosol Lidar 131 with Orthogonal Polarization (CALIOP). The CALIOP operates at three channels measuring the 132 backscatter data at 1064 nm and the two orthogonal polarization components at 532 nm that 133 provide near global measurements of aerosol and cloud profiles (Winker et al., 2003). The 134 CALIPSO level 2 dataflow starts from a search of atmospheric features (cloud or aerosol layers) 135 utilizing the 532 nm attenuated backscatter profiles (Vaughan et al., 2009). The scene 136 classification algorithm (SCA) identifies the atmospheric features as clouds (liquid or ice) or 137 aerosols (aerosol types) (Liu et al., 2009). Profiles of extinction coefficients and particle 138 backscatter are retrieved within the layers by the hybrid extinction retrieval algorithm (HERA) 139 (Winker et al., 2009; Young & Vaughan, 2009). The retrieved profiles are compiling into the 140 141 profile products, while the layer products report the layer top and base, integrated optical properties, etc. Regions with low loadings of aerosols that undergo undetected by CALIOP are 142 reported as 'clear air', which are assigned to zero extinction coefficient (Winker et al., 2009). 143 The CALIPSO level 2 aerosol and cloud merged layers product, version 4-20 144 (CAL LID L2 05kmMLay-Standard-V4-20) is the main dataset used in this study. This merged 145 layer product reports column properties of aerosol and cloud in 5-km resolution, including layer 146 top and base altitude, integrated attenuated backscatter, volume depolarization ratio, color ratio, 147 optical depth and the corresponding uncertainties calculated based on error propagation (Winker 148 et al., 2009; Young et al., 2008, 2013). A CAD (cloud and aerosol discrimination) score is also 149 reported for each layer, which adopts lidar backscatter, color ratio, and altitude to produce a 150 possibility for discriminating cloud and aerosol (Liu et al., 2009, 2019). The CAD score also 151 represents a confidence level for cloud and aerosol classification with larger absolute values for a 152 higher confidence. The level 2 aerosol profile product (CAL_LID_L2_05kmAPro-Standard-V4-153 20) is also used to obtain the lidar backscatter profiles at 532 nm for our climatology study. Data 154 from 2007.01-2020.06 is used for this work. Table 1 summarizes the data products and variables 155

- used in this study.
- 157 Table 1. Data products and the corresponding variables used in this study.

Product	Variables
CAL_LID_L2_05kmMLay-	Column_Optical_Depth_Tropospheric_Aerosols_532
Standard-V4-20	Column_Opitcal_Depth_Troposperic_Aerosols_Uncertainty_532
	CAD_Score
	Layer_Top_Altitude
	Layer_Base_Altitude
	ExtinctionQC_532
	Integrated_Attenuated_Backscatter_532
	Integrated_Volume_Depolarization_Ratio
	Final_532_Lidar_Ratio

	Integrated_Attenuated_Total_Color_Ratio
	Day_Night_Flag
	IGBP_Surface_Type
	Minimum_Laser_Energy_532
	Latitude
	Longitude
CAL_LID_L2_05kmAPro-	Total_Backscatter_Coefficient_532
Standard-V4-20	

159 2.2 Data Quality Control

CALIOP is experiencing low energy laser shots above the South Atlantic Anomaly 160 (SAA) region since 2016. To exclude those low energy shots, profiles containing 532 nm laser 161 energy less than 80 mJ are not considered as recommended by the CALIPSO team (https://www-162 calipso.larc.nasa.gov/resources/calipso_users_guide/advisory/advisory_2018-06-163 12/CALIPSO Laser Energy Technical Advisory.pdf). Similar to Tackett et al. (2018) and 164 Winker et al. (2013), our study also applies several filters to the CALIPSO data to ensure data 165 quality. The data passing the filters must meet the following criterions. First, the CAD scores 166 must be in the range of -100 to -20, representing intermediate to high confidence levels -- CAD 167 filter; second, extinction_QC_flag_532 values are equal to either 0 (unconstrained retrieval; 168 initial lidar ratio unchanged during solution process), or 1 (constrained retrieval), or 16 (opaque 169 layers and unchanged lidar ratio), or 18 (opaque layers and changed lidar ratio) – Ext OC filter; 170 third, aerosol layer above 4 km should not be adjunct to clouds which may be misclassified from 171 cirrus fringes – Ci filter. Note that when a column has one aerosol layer being filtered, the whole 172 column will be excluded in our analysis. As summarized in Table 2, aerosols in cloudy sky are 173 influenced by the filters more than in clear sky. The Ext QC filter modifies the most on AOD 174 175 compared to other filters.

	All filters	No CAD filter	No Ci filter	No Ext_QC filter	
Cloud-free	0.135	0.138	0.135	0.157	
Cloudy	0.143	0.159	0.141	0.209	
All aerosol	0.139	0.148	0.138	0.180	

176 Table 2. Summary of global mean aerosol optical depth with different filters applied.

177

178 **2.3 Classification of Aerosols**

Figure 1 shows how we group the detected aerosols (i.e. those aerosols with $\tau_a > 0$) for a 179 better characterization of aerosols in cloudy sky. The quality-controlled aerosol data is divided 180 into two main groups based on cloud presence: cloud-free and cloudy aerosols. In each cloudy 181 sky profile, by checking the gap of the closest aerosol and cloud layer, we deduce whether there 182 are any aerosol and cloud layers vertically connected. If the gap is less than 0.5 km, the profile 183 184 will be classified as *connected-cloud* aerosol (Figure 1b), otherwise, it will be grouped as *isolated-cloud* aerosol (Figure 1c). According to the vertically relative locations of aerosol and 185 cloud layers, the *connected-cloud* aerosols have four subcategories. When there is only one 186 aerosol and one cloud layer in the column and they are vertically connected, this profile is 187 classified as the one-layer aerosol-cloud connected group. The aerosol layer can be above or/and 188 below cloud layer. An example is shown in Figure 1b1. The second group of the connected-189 190 cloud aerosol is aerosol-cloud connected layer below cloud layer (Figure 1b2), where the overlying cloud can be multiple layers. Aerosol-cloud connected layer above cloud layer is the 191

third group of the connected-cloud aerosol (Figure 1b3), where the underlying cloud can be 192 multiple layers. All the connected-cloud aerosol profiles not beyond to the first three groups will 193 be included in the four subcategories (*other connected-cloud* aerosol), including the profiles with 194 more than one aerosol-cloud connected layers, or multi-layer aerosols or/and multi-layer clouds 195 with at least one aerosol-cloud connected layer. Figure 1d4 gives an example of the other 196 connected-cloud aerosol profile. The isolated-cloud aerosol is grouped in a similar way. The 197 below-cloud (Figure 1c1) aerosol can have multiple layers for the overlying clouds or the 198 underlying aerosols. Similarly, the above-cloud aerosol (Figure 1c2) can also contain multi-layer 199 underlying clouds or multi-layer overlying aerosols. Any profiles of the isolated-cloud aerosols 200 that don't beyond to the first two subgroups will be grouped as other isolated-cloud aerosols. 201 Figure 1c3 gives an example. 202





204

Figure 1. Aerosol scenarios in clear sky (a) and cloudy sky (b-c). (b) Aerosols in the column 205 206 where there are at least one aerosol and cloud layer vertically connected (connected-cloud aerosol): (b1) only one aerosol and one cloud layer in the column but vertically connected (one-207 layer aerosol-cloud connected), (b2) the aerosol-cloud connected layer below cloud layer, (b3) 208 209 the aerosol-cloud connected layer above cloud layer, and (b4) an example for multi-layer aerosol or/and multi-layer cloud that doesn't beyond to (b1-b3) (other connected-cloud aerosol). (c) 210 Aerosols in the column where there are no aerosol and cloud layers vertically connected: (c1) 211 212 below-cloud aerosol, (c2) above-cloud aerosol, and (c3) an example for multi-layer aerosol or/and multi-layer cloud that doesn't beyond to (c1-c2) (other isolated-cloud aerosol). 213 214

Figure 2 shows a CALIPSO curtain applied our aerosol classification method as described in Section 2.3. The shape in red indicates aerosol, while in blue indicates cloud. One can clearly see the below-cloud aerosol in the regions marked by the red dots at 20 km, the above-cloud aerosol indicated by the pink dots, and the group of the one-layer aerosol-cloud connected aerosols in green.





Figure 2. (a) Cloud-aerosol groups for a case on Jul. 24th, 2019 when CALIPSO overpassed above the Atlantic Ocean as shown in (b). The filled circles marked at 20 km flag the aerosol

classification as in Figure 1: black for the cloud-free, green for the one-layer aerosol-cloud

connected, blue for the aerosol-cloud connected layer below cloud layer, cyan for the aerosol-

cloud connected layer above cloud layer, purple for the other connected-cloud aerosol; red for

- the below-cloud aerosol, pink for the above-cloud aerosols, and orange for the other isolated-cloud aerosol.
- 227 228

229 3. Results and Discussion

230 **3.1 Global Distributions of The Cloud-free and The Cloudy Aerosols**

Figure 3 shows the global distributions of aerosol fraction (AF or f_a) in clear (Figure 3a) 231 and cloudy (Figure 3b) skies. Aerosol fraction (Equation 1) is defined as the samples of one 232 aerosol group such as the cloud-free or the cloudy aerosols, divided by all samples with $\tau_a > 0$ 233 that pass the quality filters as discussed in Section 2.2. As expected, the global distributions of 234 the cloud-free and the cloudy aerosols detected by CALIOP follow the climatology of clear and 235 cloudy skies. Specifically, in the subtropical regions, the cloud-free aerosols are dominated with 236 f_a higher than 60%. For the cloudy aerosols, they widely spread in the tropical regions where 237 associate with ubiquitous cirrus clouds (Hong & Liu, 2015; Sassen et al., 2008) and in the 238 subtropics such as west of South Africa where stratocumulus deck is prevailing (Sassen & Wang, 239

- 240 2008). As a result, negative f_a difference (cloudy cloud-free) is observed over most of the
- subtropical regions and positive f_a difference occurs over the tropics (Figures 3e and 3g).



Figure 3. Global distributions of aerosol fraction and optical depth in cloud-free and cloudy skies shown in (a)-(d). Zonal averages of aerosol fraction (e) and optical depth (f): black for the cloudfree aerosol and red for the cloudy aerosols. (g) and (h) are for the aerosol fraction and optical depth differences between the cloudy and the cloud-free aerosols.

247

248 Mean AOD in each 6° long by 5° lat grid is calculated as the summation of optical depth divided by the total sample with $\tau_a > 0$ for each aerosol group. As shown in Figures. 3c and 3d, 249 250 both the cloud-free and the cloudy aerosols display relatively large values (e.g. $\tau_a > 0.4$) over Africa, Center Atlantic Ocean, Arabian and Indian Peninsula, and East Asia. We also observe 251 some large τ_a values (~ 0.2) over the trade wind (easterly, such as Pacific ocean) and midlatitude 252 storm track (westerly) regions than the nearby ocean, corresponding to the sea-salt aerosols 253 254 (Grythe et al., 2014). This pattern is consistent with the aerosol climatology derived from 255 MODIS (Remer et al., 2008). The cloudy τ_a is generally higher over the globe, especially over Arabian Sea and its surrounding regions, whereas over the Southeast Asia and Center Africa, 256 257 cloud-free τ_a is larger (Figure 3h).

Over the globe, aerosols detected by CALIOP have about 55.9% samples occurring in clear sky, and another 44.1% occurring in cloudy sky. Cloudy aerosols occupy a smaller proportion because aerosols with low concentrations or under opaque clouds undergo undetectedby CALIOP.

The global mean aerosol optical depth ($\bar{\tau}_a$), calculated as Equation (2), and the global mean τ_a uncertainty for clear sky are about 0.135±0.047, while in cloudy sky, they are about 0.143±0.074. For all aerosols, $\bar{\tau}_a$ is about 0.139±0.059. An averaged AOD difference of 0.08 between the cloudy and cloud-free aerosols is significant based on t-test analysis. The uncertainty of the cloudy-sky aerosol is much larger than the cloud-free aerosol, indicating bigger challenge to retrieve aerosols in cloudy sky (Winker et al., 2009).

- 268
- 269 270

 $f_{ai} = n_{ai}/N_{ai} \quad (1)$ $\bar{\tau}_a = \sum_i \tau_{ai} n_{ai}/\sum_i n_{ai} \quad (2)$

271

where i represents the ith grid, n_{ai} and N_{ai} are the samples for one aerosol group and total aerosol samples with quality control in that grid.

Figure 4 displays the profiles of aerosol samples and lidar backscatter (β) for cloud-free

275 (black) and cloudy (red) skies globally. As displayed, aerosols are mostly detected in the

boundary layer (< 2 km) with a peak occurrence at ~ 1 km, and evident aerosols are also

observed between 2 - 4 km. The average lidar backscatter profiles (solid lines in Figure 4b) show

that the cloudy aerosols have stronger backscatter along all altitudes than the cloud-free aerosols.

279 Yet the median lidar backscatter profiles show slightly stronger backscatter for the cloudy

aerosols only under the boundary layer. Above the boundary layer, the median lidar backscattersare quite consistent between the cloudy and the cloud-free aerosols. Larger lidar backscatter in

cloudy sky agrees with larger cloudy τ_a .



283

Figure 4. Number of samples (a) and lidar backscatter profiles (b): red for the cloudy aerosol,
black for the cloud-free aerosol, solid line for mean values and dashed line for median values.

The vertical resolution is same as Level-2 aerosol profile data, i.e. 30 m below 8.2 km and 60 m above 8.2 km.

288

289 3.2 Global Distributions of The Connected-cloud and The Isolated-cloud Aerosols

To investigate the detailed characteristics of aerosols in cloudy sky, Figures 5 and 6 display the global distributions of f_a and τ_a for the connected-cloud and the isolated-cloud aerosols.

As showed, the connected-cloud aerosols are mainly occupied by the one-layer aerosol-293 294 cloud connected and the aerosol-cloud connected layer below cloud layer groups, which account for about 8.8% and 3.4% among all aerosols detected by CALIOP, respectively (Figures 5a1 and 295 5a2). For the one-layer aerosol-cloud connected aerosols, relatively large f_a primarily occurs 296 over the west of continents where marine stratocumulus are prevailing, including Center and 297 North Atlantic where are polluted by the westward transferred Sahara dust (Devasthale & 298 Thomas, 2011; Yang et al., 2012; Zhang et al., 2016), Southeast Atlantic where biomass burning 299 smoke is advected from South Africa during the local winter/spring seasons (Devasthale & 300 Thomas, 2011; Zhang et al., 2016), as well as West of North and South America, and West of 301 Australia. Also, the one-layer aerosol-cloud connected aerosols are frequent over East Asia 302 where low-level stratocumulus are prevailing during spring and winter (Hong & Di Girolamo, 303 2020; Klein & Hartmann, 1993), providing high change for clouds embedding to aerosol layers 304 in the lower troposphere. For the aerosol-cloud connected layer below cloud layer group, they 305 primarily distribute over the tropics and midlatitude storm tracks where cirrus clouds are 306 prevailing (Hong & Liu, 2015; Sassen et al., 2008). The aerosol-cloud connected layer above 307 cloud layer and other connected-cloud aerosols account for about 1.6% and 2.9%, respectively, 308

among all detected aerosols.





Figure 5. Aerosol fraction (a1-a4) and optical depth (b1-b4) for the four subgroups of the connected-cloud aerosol: first row for the one-layer aerosol-cloud connected, second row for the aerosol-cloud connected layer below cloud layer, the third row for the aerosol-cloud connected layer above cloud layer, and the bottom row for the other connected-cloud aerosol.

For optical depth, the one-layer aerosol-cloud connected (Figure 5b1) and the aerosolcloud connected layer below cloud layer aerosols (Figure 5b2) show similar spatial patterns with the cloud-free aerosols but their global means are smaller. The aerosol group of the aerosol-cloud connected layer above cloud layer has the smallest mean τ_a (Figure 5b3), while the other connected-cloud aerosols have the largest τ_a , particularly over ocean and over the 'dust belt' cross Sahara eastward to Arabia and Southwest of Asia (Figure 5b4). Also, the aerosol-cloud connected layer below cloud layer (Figure 5b2) and the other connected-cloud aerosols (Figure 5b4) have larger τ_a uncertainties than the other two connected-cloud aerosol groups, i.e. the onelayer aerosol-cloud connected (Figure 5b1) and the aerosol-cloud connected layer above cloud layer (Figure 5b3). The overlying clouds induce additional uncertainty to the lower aerosol layers as discussed in Section 1 (Winker et al., 2009).

Figure 6 shows the distributions of the isolated-cloud aerosols. The below-cloud aerosols 327 (Figure 6a1) account for about 21.2% among all aerosols, frequently distributing in the tropical 328 regions where occur ubiquitous cirrus clouds (Hong & Liu, 2015; Sassen et al., 2008). The 329 fraction of the above-cloud aerosols (Figure 6a2) are about 4%, and are primarily distributed 330 over ocean, i.e. West of the Africa and America continents, as well as East of Asia continent, 331 agreeing with the results from previous research that focused on the above-cloud aerosols 332 (Devasthale & Thomas, 2011; Yang et al., 2012; Zhang et al., 2016). For the other isolated-cloud 333 aerosols (Figure 6a3), their global average fraction is about 2%. The $\bar{\tau}_a \sim 0.165$ of the below-334 cloud aerosols is larger than that of the cloud-free aerosols ($\bar{\tau}_a \sim 0.135$). The above-cloud 335 aerosols have smaller $\bar{\tau}_a$ (0.056) than the cloud-free aerosols primarily due to a shallower 336 integrated column than the cloud-free aerosols. Also, aerosol optical depth uncertainties of the 337 below-cloud and other isolated-cloud groups tend to be larger than the cloud-free or the above-338 339 cloud aerosols. The retrieved cloud attenuation used for the underlying layer signal correction 340 responds for the additional uncertainty to aerosol layers (Chung et al., 2016; Winker et al., 2009).

341



342

Figure 6. Same as Figure 5 but for the isolated-cloud aerosol: the upper panels for belowcloud aerosol, the middle panels for above-cloud aerosol, and the lower panels for the other isolated-cloud aerosol.

It has been noted that the cloudy aerosols are primarily occupied by the below-cloud 347 aerosols ($f_a \sim 21\%$), which account for almost half of the detected cloudy aerosols ($f_a \sim 44\%$). 348 Considering that aerosols under opaque clouds are missed by CALIOP, the below-cloud aerosols 349 are more spread than what we observe from Figure 6a1. Liao and Seinfeld, (1998) stated that a 350 cirrus cloud layer causes modest changes of aerosol radiative effect at TOA, which is about 1 W 351 m⁻² change compared to no cirrus presence in their radiative transfer simulations. The wide 352 coverage of the below-cloud aerosols indicates that the impact of cirrus to aerosol direct radiative 353 354 effect could be significant over the globe.

It is also noted that the τ_a uncertainties are larger for those aerosols with overlying cloud layers in the column, i.e. the aerosol-cloud connected layer below cloud layer and below-cloud aerosols, compared to the situations without overlying cloud presence, i.e. the one-layer aerosolcloud connected and the cloud-free aerosols. In Section 3.3, we further check more details of τ_a distributions and other aerosol properties to investigate whether there is any similarity between aerosols with and without overlying cloud layers.

362 **3.3 Optical Property and Lidar Variables**

363 3.3.1 Optical Depth

The probability density functions (PDF) of τ_a are shown in Figure 7 for the cloud-free, 364 the connected-cloud and the isolated-cloud aerosols. The cloud-free τ_a PDF is nearly a 365 lognormal distribution with a peak at $\tau_a \sim 0.083$ (Table 3). For the connected-cloud aerosols, 366 their PDFs are generally fatter than the cloud-free aerosols and have higher frequencies at small 367 τ_a regions. The one-layer aerosol-cloud connected and the aerosol-cloud connected layer below 368 cloud layer aerosols show close PDFs (Figure 7a, pink and red), close values in their means, 369 370 modes and medians (Table 3, b1 and b2) with a comparison with other aerosol groups. The PDF of the aerosol-cloud connected layer above cloud layer aerosols (Figure 7a, green) shows much 371 higher frequency when $\tau_a < \sim 0.04$ with the smallest median, mode and median values among 372 373 all the connected-cloud aerosol groups.

Similarly, for the isolated-cloud aerosols, the below-cloud aerosols show a very close PDF with the cloud-free aerosols but with slightly larger mode (~ 0.089), median and mean values (Table 3, c1). In contrast, the above-cloud aerosols have much smaller mode values than the cloud-clear aerosols as shallower aerosol columns contribute to a smaller τ_a .



378

Figure 7. The probability distribution functions of AOD for global aerosols: (a) the
connected-cloud aerosols and (b) the isolated-cloud aerosols with a contrast to the cloud-free
aerosols shown in black curve in both (a) and (b).

382

Table 3 The AOD medians, modes and means for the cloud-free, the connected-cloud and the
 isolated-cloud aerosols. b1-b4 and c1-c4 represent the same aerosol categories as Figures 1b1-b4
 and Figures c1-c4, respectively.

	Cloud-	Connected-cloud			Isolated-cloud			
	free	b1	b2	b3	b4	c1	c2	c3
Median	0.076	0.048	0.043	0.027	0.093	0.083	0.024	0.050
Mode	0.083	0.075	0.052	0.038	0.110	0.089	0.017	0.048
Mean	0.135	0.130	0.127	0.079	0.201	0.165	0.056	0.132

386

387 **3.3.2 Depolarization Ratio and Lidar Backscatter**

388 The integrated attenuated backscatter and depolarization ratio (δ) are two important variables for constraining aerosol type selections (Omar et al., 2009). Figure 8 shows the two-389 dimensional (2-D) histogram of β versus δ for the detected aerosol layers. As displayed, most of 390 391 the one-layer aerosol-cloud connected (Figure 8a), the aerosol-cloud connected layer below cloud layer (Figure 8c), the cloud-free (Figure 8b), and the below-cloud aerosols (Figure 8d) 392 have $\delta < 0.02$. In contrast, the aerosol-cloud connected layer above cloud layer (Figure 8e) and 393 the above-cloud aerosols (Figure 8f) show tremendous samples with $\delta > 0.02$. These two groups 394 395 of aerosols distribute in the regions where are frequently affected by dust and biomass burning (Figures 5a3 and 6a2), being consistent with that relatively large δ indicates desert dust and 396 biomass burning aerosols (Burton et al., 2012; Omar et al., 2009). Moreover, the connected-397 cloud aerosols tend to have lidar backscatter (β) less than 0.001 Sr⁻¹km⁻¹, agreeing with higher 398 frequency at small τ_a region in Figure 7a. For the cloud-free and the below-cloud aerosols, they 399 have most samples with $\beta > 0.001 \text{ Sr}^{-1} km^{-1}$, indicating marine and polluted continental 400 aerosols (Omar et al., 2009). The above-cloud aerosols rarely have $\beta > 0.001 \text{ Sr}^{-1} km^{-1}$. Again, 401 the cloud-free and the below-cloud aerosols share similar $\beta - \delta$ diagram with the samples 402 concentrated in the regions of $\beta > 0.001 \text{ Sr}^{-1} km^{-1}$ and $\delta < 0.02$. It is also similar for the one-403 layer aerosol-cloud connected and the aerosol-cloud connected layer below cloud layer aerosols 404 who have a similar pattern of $\beta - \delta$ diagram with the samples concentrated in two regions: 1. 405 $\beta \sim 0.0003 - 0.001 \text{ Sr}^{-1} km^{-1}$ and $\delta < 0.02$ and 2. $\beta \sim 0.003 \text{ Sr}^{-1} km^{-1}$ and $\delta < 0.02$. 406



Figure 8. Two-dimensional histogram of lidar backscatter (in log₁₀ scale) vs. depolarization ratio
for the cloud-free, the connected-cloud and the isolated-cloud aerosols. The total frequency of
each diagram is equal to 100.

411

412 **3.3.3 Lidar Ratio and Color Ratio**

Lidar ratio (Sr), i.e. the extinction coefficient to lidar backscatter ratio, is essential to obtain optical depth. The CALIPSO aerosol type classification assigns one Sr to one aerosol type (Omar et al., 2009). In the V4 data, lidar ratio has been updated to better represent aerosol types

(Kim et al., 2018). Six main aerosol types are manifested in Figure 9, i.e.: clean marine (Sr =416 23), dusty marine (S = 37), dust/volcanic ash (Sr = 44), clean continental (Sr = 53), polluted 417 dust (Sr = 55) and polluted continental/smoke/elevated smoke (Sr = 70) aerosol. The dots and 418 error bars in Figure 9 represent means and standard deviations of color ratio (γ) for each aerosol 419 type. The number near each aerosol type is obtained by integrating the frequency over y axis, i.e. 420 color ratio, representing the proportion of each aerosol type within that aerosol group. For 421 instance, Figure 9b shows that the cloud-free aerosols contain about 45.9% samples being 422 classified as clean marine aerosols. 423 The proportions of aerosol types in each aerosol group are closely related to aerosol 424 distributions over the globe. Clean marine aerosols are the primary aerosol type for the cloud-425 free (Figure 9b, 45.9%), the one-layer aerosol-cloud connected (Figure 9a, 36.5%), the aerosol-426 cloud connected layer below cloud layer (Figure 9c, 32.5%) and the below-cloud (Figure 9d, 427 49.8%) aerosols. In particular, the below-cloud aerosols show a similar $Sr - \gamma$ pattern with the 428 cloud-free aerosols. The cloud-free aerosol group is slightly larger in the proportions of 429 430 dust/volcanic ash and polluted dust aerosols, while the below-cloud aerosol group is about 4% and 1.3% higher in clean marine and dusty marine aerosols. For the above-cloud (Figure 9f) and 431 the aerosol-cloud connected layer above cloud layer aerosols (Figure 9e), they share similar 432 433 aerosol types as well. Dust/volcanic ash (> 30%), polluted dust (~ 24%) and elevated smoke (~ 434 25%) are the three dominated aerosol types, but the clean marine or dusty marine aerosols are rare (<5%) for these two aerosol groups. The mean and standard deviation of color ratio 435 436 between the cloud-free and the below-cloud aerosols are similar with the mean color ratio of

437 about 0.5 for clean marine, dusty marine and dust/volcanic ash aerosols, about 0.4 for polluted

dust and polluted continental/smoke/elevated smoke, and about 0.2 for clean continentalaerosols.



Figure 9. 2-D histograms of lidar ratio vs. color ratio. The gray dots and error bars represent the
mean and standard deviation of color ratio, while the number represents the total fraction of each
aerosol type.

444

445 Overall, the τ_a distributions, the $Sr - \gamma$ and $\beta - \delta$ diagrams have shown similarity 446 between the one-layer aerosol-cloud connected and the aerosol-cloud connected layer below 447 cloud layer aerosols as well as between the cloud-free and the below-cloud aerosols. Some 448 differences in these properties between aerosols with and without overlying clouds could result 449 from their contrast in spatial distributions as discussed in Section 3.2, from the accuracy of 450 cloud-aerosol discrimination, from additional uncertainties induced by the overlying cloud layers451 as well as from a modification of aerosol properties by cloud layers.

452 **3.4 Regional and Seasonal Analysis**

460

As displayed in Section 3.2, aerosols unevenly distribute over the globe. To characterize the regional aerosol variability, we show aerosol properties in cloudy and clear skies for nine selected locations as indicated in Figure 10 where are significantly affected by anthropogenic and natural activities. Figures 11 and 12 display the timeseries of monthly \bar{f}_a and $\bar{\tau}_a$ for the

457 cloud-free (red), the cloudy (orange, a combination of the connected-cloud and the isolated-

458 cloud), the connected-cloud (green), the isolated-cloud (grey) aerosols. Figures 13 and 14 show

the profiles of aerosol samples and median lidar backscatters.



Figure 10. Selected areas for studying the seasonal variations of aerosol fraction and optical
depth derived from CALIOP: EAS short for East Asian region, MIN for Malaysian-Indonesia
region, IND for Indian region, EUR for European region, CAO for Center Atlantic Ocean, SAO
for Southeast Atlantic Ocean, EUS for East US, WUS for west of US, AMZ for Amazonian
region.



466

Figure 11. The timeseries of aerosol fraction for the selected regions as displayed in Figure 10:
red- the cloud-free aerosol, orange – the cloudy aerosol, green – the connected-cloud aerosol,
grey- the isolated-cloud aerosol.

For the whole globe, the cloudy and the cloud-free $\bar{\tau}_a$ values show seasonal variations ranging roughly from 0.1 to 0.15, being larger during boreal summer and smaller during boreal winter (Figure 12a). The seasonal variations agree with results from passive sensor

472 white (Figure 12a). The seasonal variations agree with results from passive sensor
 473 measurements such as MODIS and MISR (Mehta et al., 2016; Mishchenko et al., 2007; Remer et

al., 2008). In contrast, the cloud-free and cloudy aerosol fractions are quite flat (Figure 11a).

In different regions, the variations of $\bar{\tau}_a$ and \bar{f}_a differ significantly between the cloud-free and the cloudy aerosols which rely on the variations of cloud systems, local emissions, and longtrack transport. For example, in the Malaysia-Indonesia regions (Figures 11-14i), the fraction of aerosols in cloudy sky, primarily contributed by the below-cloud aerosols (Figure 6a), is twice more than that of the cloud-free aerosols due to the ubiquitous cirrus in this region (Figure 11i). The cloudy $\bar{\tau}_a$ is slightly larger than the cloud-free $\bar{\tau}_a$, while the connected-cloud $\bar{\tau}_a$ is the

- 481 largest in this region (Figure 12i). The climate impact in this region is observable the cloud-
- 482 free aerosols account for a higher fraction during the winter-spring seasons of 2009-2010, 2015-
- 483 2016 and 2019-2020 when El Niño phase dominated; during the La Niña years such as 2010,
- 484 2017 and 2018, cloudy aerosols have larger fractions. The detected aerosols primarily occur
- below 4 km (Figure 13i). The median backscatter profiles (Figure 14i) also display that the
- 486 connected-cloud aerosols tend to have the largest backscatter, extending up to 4 km. The
 487 isolated-cloud and the cloud-free aerosols have their median backscatter profiles nearly
- 487 isolated-cloud and the cloud-free acrossis have then median backscatter promes hearly
 488 overlapped below 4 km, indicating the similarity between aerosols with and without clouds
- 489 overlying.



490 491 Fig

Figure 12. Same as Figure 11 but for the timeseries of aerosol optical depth.

In the Southeast Atlantic region, aerosols frequently transport above stratocumulus
during the biomass burning season (local winter) (Zhang et al., 2016) when the cloud-free
aerosols account for almost 60% among all detected aerosols (Figure 11e). The cloudy aerosols
up to 80% during local summer are primarily contributed by the isolated-cloud (above-cloud)

- 496 aerosols. The cloud-free aerosols show that \bar{f}_a peaks in May-Aug. $\bar{\tau}_a$ in this region is larger in
- 497 local winter and smaller during summer (Figure 12e). The connected-cloud and the isolated-
- 498 cloud τ_a are smaller than the cloud-free aerosols in this region because aerosols are frequently
- 499 transferred above stratocumulus so that cloudy τ_a is being integrated in a shallower column than
- 500 in clear sky. This is also revealed in Figure 13e that shows a peak located at ~ 3 km, i.e. above
- 501 boundary layer, and another peak in boundary layer (~ 1 km). It is obvious that there are two
- 502 bumps in the median backscatter profiles (Figure 14e) one below 1 km and another one
- between 2-6 km, contributing by the boundary-layer and the elevated aerosols, respectively.



504



Similarly, over the Center Atlantic Ocean region (Figures 11-14d), aerosols originated 506 from Africa dust are often elevated and transported westward (Yang et al., 2012). As displayed 507 in Figures 13d and 14d, aerosols are concentrated in the boundary layer below 2 km but with 508 another pump between 2-6 km. In this region, the cloud-free aerosols account for more than 50% 509 through the year with no obvious seasonal cycles. However, the connected-cloud aerosol fraction 510 displays seasonal periodicity that relates to the cumulus which appear from Mach – August each 511 year (Figure 11d) (King et al., 2013). The timeseries of both the cloud-free and 512 cloudy $\bar{\tau}_a$ overlaps well, being larger in boreal summer season (Figure 12d) that is associated 513 with the outbreak of Saharan air layer (Xian et al., 2020). 514



Figure 14. Same as Figure 11 but for profiles of the median values of lidar backscatters at 532nm.

518 In the East Asian and Indian regions, aerosol characteristics are impacted by the Asian monsoon. The cloud-free aerosols occupy a higher fraction than the cloudy aerosols during the 519 winter monsoon season in both regions, and vice versa during the summer monsoon season 520 521 (Figures 11g and 11h). In the Indian region, $\bar{\tau}_a$ peaks in summer as the prevailing Southwest surface wind transports dust particles from the desert from Asia and Africa to India and its 522 surrounding regions (Sivaprasad & Babu, 2014). The cloud-free τ_a is generally larger than the 523 connected-cloud $\bar{\tau}_a$, while the isolated-cloud $\bar{\tau}_a$ is the smallest (Figure 12g). In East Asia, the 524 525 connected-cloud aerosols are more prevalent during winter-spring season associating with the occurrence of the prevailing low stratiform clouds (Hong & Di Girolamo, 2020; Klein & 526 Hartmann, 1993), while the isolated-cloud aerosols (primary below-cloud aerosol,) are being 527 528 more frequent in summer (Figure 11h). Similar to Indian region, the cloud-free $\bar{\tau}_a$ is larger than the cloudy $\bar{\tau}_a$ as well, with the connected-cloud $\bar{\tau}_a$ being the smallest. In both regions, aerosols 529 are detected up to 6 km (Figures 13g and 13h), whereas in India, aerosol backscatters are large 530 up to 6 km and in East Asia, they decrease quicky above 2 km (Figures 14g and 14h). 531

In the other four regions- Europe (Figures 11f-14f), West of US (Figures 11b-14b), East US (Figures 11c-14c) and Amazon (Figures 11j-14j), the seasonal variations and the behaviors of the cloud-free and the cloudy aerosols also heavily rely on local weather and climate. For instance, in the Amazonian regions, the $\bar{\tau}_a$ and \bar{f}_a peak in the dry season, and cloudy aerosols are dominated by the below-cloud aerosols (> 40%, Figure 6) due to ubiquitous cirrus; In the West of US, aerosols are detected below 2 km and rare aerosols are elevated above boundary layer, while in Europe, East US and Amazon, aerosols can be detected up to 4 km.

539 Overall, the characteristics of the cloud-free and the cloudy aerosols as well as their 540 seasonal variations significantly vary with regions. In some regions such as in East Asia, the 541 cloud-free $\bar{\tau}_a$ tends to be the largest, while in some other regions such as in Malaysia-Indonesia 542 region, the cloud-free $\bar{\tau}_a$ is the smallest. The difference in the cloudy and the cloud-free $\bar{\tau}_a$ in a 543 region relies on their differences in aerosol types, geometrical thickness of the aerosol layers as 544 well as the retrieved τ_a uncertainties. One should be noted that the geometrical thickness of

- aerosol layers is overall larger in clear sky than in cloudy sky, which should result in larger
- cloud-free $\bar{\tau}_a$. This is observed when the aerosol concentration is heavy such as in the Southeast
- 547 Atlantic Ocean and Indian region. However, aerosol-cloud interactions and the larger $\bar{\tau}_a$
- uncertainties in cloudy sky could also contribute to a larger cloudy $\bar{\tau}_a$. This can be revealed in
- Figure 12 when the aerosol concentration is low ($\bar{\tau}_a < 0.2$), the cloudy $\bar{\tau}_a$ tends to be larger than
- 550 the cloud-free $\bar{\tau}_a$. To what degree is contributed by aerosol-cloud interaction or retrieval
- 551 uncertainty is not inferred in this study.

Regardless of regional τ_a differences, there are some common features. The median lidar backscatter profiles of the cloudy and the cloud-free aerosols are almost overlapped in all regions. However, the connected-cloud median backscatter profiles are the largest among all aerosol groups for all regions. The larger lidar backscatter of the connected-cloud aerosols could result from an enhancement of aerosol backscatter near cloud edge (Tackett & Di Girolamo, 2009). Misclassification of clouds, especially cloud detrainments near cloud boundary, to

aerosols likely induce larger lidar backscatter to the connected-cloud aerosols as well.

559 5. Summary

560 This study examines the climatological properties of aerosols detected by CALIOP 561 occurring in cloud-free and cloudy skies. Considering that clouds above or below aerosol layers 562 influence aerosol direct radiative effects in a different way, cloudy aerosols are further classified 563 into the connected-cloud and the isolated-cloud aerosols based on the vertically relative locations 564 of aerosol and cloud layers. The differences between the cloud-free and the cloudy aerosols are 565 emphasized by examining their spatial distributions, optical property, seasonal and regional 566 variations.

567 Overall, for all the detected aerosols by CALIOP that pass our quality filters, the cloudfree aerosol samples account for about 55.9%, and the cloudy aerosol samples occupy about 568 44.1% over the globe. The cloudy aerosol samples are underestimated as CALIOP misses very 569 570 thin aerosols and aerosols under opaque clouds. The global average optical depth ($\bar{\tau}_a$) and its uncertainty of the cloud-free and the cloudy aerosols are 0.135 ± 0.047 and 0.143 ± 0.074 , 571 respectively. The connected-cloud aerosols, occurring frequently in the marine stratocumulus 572 and East Asian regions, have a $\bar{\tau}_a \sim 0.138 \pm 0.063$. The below-cloud aerosols, frequently occur 573 in the regions covered by ubiquitous cirrus, have a $\bar{\tau}_a \sim 0.165 \pm 0.087$, whereas the above-cloud 574 aerosols tend to have smaller $\bar{\tau}_a \sim 0.056 \pm 0.039$. 575

The cloud-free, the below-cloud, the one-layer aerosol-cloud connected and the aerosol-576 cloud connected layer below cloud layer aerosols primarily consist of marine aerosols, while the 577 aerosol-cloud connected layer above cloud layer and the above-cloud aerosols contain more 578 elevated smoke, polluted dust and volcano ash aerosols. We find similar τ_a PDFs, $\beta - \sigma$ and 579 $Sr - \gamma$ diagrams between the one-layer aerosol-cloud connected and the aerosol-cloud connected 580 581 layer below cloud layer aerosols, as well as between the below-cloud and the cloud-free aerosols. These similarities may indicate that the aerosol climatology from passive sensors may be well 582 representative globally. However, considering the additional uncertainties caused by the 583 584 overlying cloud layers to the underlying aerosols observed from space-borne lidar, it is necessary 585 to utilize ground-based lidar observations to confirm the property similarity of aerosols with and without cloud layers above. 586

Also, this study reveals a wide coverage of the below-cloud aerosols that are frequently
distributed over the tropical regions, which account for about ~ 21.2% among all detected

aerosols. Due to the fact that a cirrus layer is able to modify aerosol direct radiative effect at the TOA and heating rates in the atmosphere, it is important to quantify to what degree cirrus clouds change the underlying aerosol direct effect in order to obtain an accurate estimate of Earth radiation budget. The ground-based observations such as from the Atmospheric Radiation Measurement providing important measurements of clouds, aerosols and radiation will serve for simulating and constraining aerosol direct radiative effect at the TOA in both clear and cloudy skies regionally as demonstrated in two recent studies (Balmes & Fu, 2021; Wu et al., 2021).

Our results also suggest that regional and seasonal variations of the cloud-free and the 596 cloudy aerosol fraction and optical depth are strong, which are affected by local emissions and 597 large-scale climate systems. In East Asia and Southeast Atlantic Ocean, the cloud-free $\bar{\tau}_a$ is the 598 599 largest, and the connected-cloud $\bar{\tau}_a$ is the smallest, vice versa in East US, West US and Malaysia-Indonesia regions. Though aerosol variations are huge in different regions. The 600 connected-cloud lidar backscatter profiles (median values) are the largest among different 601 602 aerosol groups in all selected regions. As aerosol and cloud layers are vertically adjunct, aerosol properties can be affected by clouds- hygroscopic aerosols might grow in a more humid 603 environment near clouds (Rauber et al., 2013). Detrainment/entrainment cloud process could also 604 alter aerosol properties Also, small cloud segments near cloud boundary also post challenges for 605 the aerosol-cloud discrimination, leading to some cloudy pixels being identified as aerosols. To 606 what degree the cloud contamination contributes to the connected-cloud profile is unknow. 607 Finally, this study has provided a comprehensive analysis of the cloud-free and the 608

cloudy aerosols, which will aid the understanding and better quantifying the Earth radiationbudget.

611

612 Acknowledgements

This research has been funded by the NASA CAMP²Ex (Cloud, Aerosol and Monsoon
 Processes-Philippines Experiment) field campaign under the research grant 80NSSC18K0144 and
 80NSSC21K1449. We acknowledge the CALIPSO science team for providing the aerosol
 products, which is available at https://search.earthdata.nasa.gov/search.

617

618 **References**

- Alfaro-Contreras, R., Zhang, J., Campbell, J. R., & Reid, J. S. (2016). Investigating the
 frequency and interannual variability in global above-cloud aerosol characteristics with
 CALIOP and OMI. *Atmospheric Chemistry and Physics*, 16(1), 47–69.
- 622 https://doi.org/10.5194/acp-16-47-2016
- Andreae, M. O., & Rosenfeld, D. (2008). Aerosol-cloud-precipitation interactions. Part 1. The
 nature and sources of cloud-active aerosols. *Earth-Science Reviews*, 89(1–2), 13–41.
 https://doi.org/10.1016/j.earscirev.2008.03.001

Balmes, K. A., & Fu, Q. (2021). All-Sky Aerosol Direct Radiative Effects at the ARM SGP Site. *Journal of Geophysical Research: Atmospheres*, *126*(17).
https://doi.org/10.1029/2021JD034933

Burton, S. P., Ferrare, R. A., Hostetler, C. A., Hair, J. W., Rogers, R. R., Obland, M. D., et al. (2012). Aerosol classification using airborne High Spectral Resolution Lidar measurements-

- methodology and examples. *Atmospheric Measurement Techniques*, 5(1), 73–98.
 https://doi.org/10.5194/amt-5-73-2012
- Chand, D., Wood, R., Anderson, T. L., Satheesh, S. K., & Charlson, R. J. (2009). Satellite derived direct radiative effect of aerosols dependent on cloud cover. *Nature Geoscience*,
 2(3), 181–184. https://doi.org/10.1038/ngeo437
- 636 Christensen, M. W., & Stephens, G. L. (2011). Microphysical and macrophysical responses of
 637 marine stratocumulus polluted by underlying ships: Evidence of cloud deepening. *Journal*638 *of Geophysical Research Atmospheres*, *116*(3), 1–10.
 639 https://doi.org/10.1029/2010JD014638
- Chung, C. E., Lewinschal, A., & Wilcox, E. (2016). Relationship between low-cloud presence
 and the amount of overlying aerosols. *Atmospheric Chemistry and Physics*, *16*(9), 5781–
 5792. https://doi.org/10.5194/acp-16-5781-2016
- 643 Devasthale, A., & Thomas, M. A. (2011). A global survey of aerosol-liquid water cloud overlap
 644 based on four years of CALIPSO-CALIOP data. *Atmospheric Chemistry and Physics*,
 645 *11*(3), 1143–1154. https://doi.org/10.5194/acp-11-1143-2011
- Grythe, H., Ström, J., Krejci, R., Quinn, P., & Stohl, A. (2014). A review of sea-spray aerosol source functions using a large global set of sea salt aerosol concentration measurements. *Atmospheric Chemistry and Physics*, 14(3), 1277–1297. https://doi.org/10.5194/acp-14-1277-2014
- Ten Hoeve, J. E., & Augustine, J. A. (2016). Aerosol effects on cloud cover as evidenced by
 ground-based and space-based observations at five rural sites in the United States. *Geophysical Research Letters*, 43(2), 793–801. https://doi.org/10.1002/2015GL066873
- Hong, Y., & Di Girolamo, L. (2020). Cloud Phase Characteristics Over Southeast Asia from ATrain Satellite Observations. *Atmospheric Chemistry and Physics*, (20), 8267–8291.
 https://doi.org/https://doi.org/10.5194/acp-20-8267-2020
- Hong, Y., & Liu, G. (2015). The characteristics of ice cloud properties derived from CloudSat
 and CALIPSO measurements. *Journal of Climate*, 28, 3880–3901.
 https://doi.org/10.1175/JCLI-D-14-00666.1
- IPCC. (2021). Climate Change 2021: The Physical Science Basis. Contribution of Working
 Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change.
 Cambridge University Press.
- Keil, A., & Haywood, J. M. (2003). Solar radiative forcing by biomass burning aerosol particles
 during SAFARI 2000: A case study based on measured aerosol and cloud properties. *Journal of Geophysical Research: Atmospheres*, *108*(13), 1–10.
 https://doi.org/10.1029/2002jd002315
- Kim, M.-H., Omar, A. H., Tackett, J. L., Vaughan, M. A., Winker, D. M., Trepte, C. R., et al.
 (2018). The CALIPSO version 4 automated aerosol classification and lidar ratio selection
 algorithm. *Atmospehric Measurment Techniques*, *11*, 6107–6135.
 https://doi.org/10.5194/amt-11-6107-2018
- Kim, S. W., Yoon, S. C., Kim, J., & Kim, S. Y. (2007). Seasonal and monthly variations of

- 671 columnar aerosol optical properties over east Asia determined from multi-year MODIS,
- 672 LIDAR, and AERONET Sun/sky radiometer measurements. *Atmospheric Environment*,
- 673 *41*(8), 1634–1651. https://doi.org/10.1016/j.atmosenv.2006.10.044
- King, M. D., Platnick, S., Menzel, W. P., Ackerman, S. A., & Hubanks, P. A. (2013). Spatial and
 temporal distribution of clouds observed by MODIS onboard the Terra and Aqua satellites. *IEEE Transactions on Geoscience and Remote Sensing*, *51*, 3826–3852.
- 677 https://doi.org/10.1109/TGRS.2012.2227333
- Klein, S. A., & Hartmann, D. L. (1993). The seasonal cycle of low stratiform clouds. *Journal of Climate*. https://doi.org/10.1175/1520-0442(1993)006<1587:TSCOLS>2.0.CO;2
- Lebsock, M. D., Stephens, G. L., & Kummerow, C. (2008). Multisensor satellite observations of
 aerosol effects on warm clouds. *Journal of Geophysical Research Atmospheres*, *113*(15), 1–
 12. https://doi.org/10.1029/2008JD009876
- Liao, H., & Seinfeld, J. H. (1998). Effect of clouds on direct aerosol radiative forcing of climate.
 Journal of Geophysical Research Atmospheres, 103(D4), 3781–3788.
 https://doi.org/10.1029/97JD03455
- Liu, Z., Vaughan, M., Winker, D., Kittaka, C., Getzewich, B., Kuehn, R., et al. (2009). The
 CALIPSO lidar cloud and aerosol discrimination: Version 2 algorithm and initial
 assessment of performance. *Journal of Atmospheric and Oceanic Technology*, 26(7), 1198–
 1213. https://doi.org/10.1175/2009JTECHA1229.1
- Liu, Z., Kar, J., Zeng, S., Tackett, J., Vaughan, M., Avery, M., et al. (2019). Discriminating
 between clouds and aerosols in the CALIOP version 4 . 1 data products, 2006, 703–734.
- Loeb, N. G., & Manalo-Smith, N. (2005). Top-of-atmosphere direct radiative effect of aerosols
 over global oceans from merged CERES and MODIS observations. *Journal of Climate*,
 18(17), 3506–3526. https://doi.org/10.1175/JCLI3504.1
- Lohmann, U., & Feichter, J. (2005). Global indirect aerosol effects: a review. *Atmos. Chem. Phys*, 5, 715–737. https://doi.org/10.5194/acp-5-715-2005
- Mao, K. B., Ma, Y., Xia, L., Chen, W. Y., Shen, X. Y., He, T. J., & Xu, T. R. (2014). Global
 aerosol change in the last decade: An analysis based on MODIS data. *Atmospheric Environment*, 94, 680–686. https://doi.org/10.1016/j.atmosenv.2014.04.053
- Matus, A. V., L'Ecuyer, T. S., Kay, J. E., Hannay, C., & Lamarque, J.-F. (2015). The role of
 clouds in modulating global aerosol direct radiative effects in spaceborne active
 observations and the community earth system model. *Journal of Climate*, 28(8), 2986–
 3003. https://doi.org/10.1175/JCLI-D-14-00426.1
- Mehta, M., Singh, R., Singh, A., Singh, N., & Anshumali. (2016). Recent global aerosol optical
 depth variations and trends A comparative study using MODIS and MISR level 3 datasets.
 Remote Sensing of Environment, 181, 137–150. https://doi.org/10.1016/j.rse.2016.04.004
- Mishchenko, M. I., Geogdzhayev, I. V., Cairns, B., Carlson, B. E., Chowdhary, J., Lacis, A. A.,
 et al. (2007). Past, present, and future of global aerosol climatologies derived from satellite
 observations: A perspective. *Journal of Quantitative Spectroscopy and Radiative Transfer*,
 106(1–3), 325–347. https://doi.org/10.1016/j.jqsrt.2007.01.007

- Omar, A. H., Winker, D. M., Kittaka, C., Vaughan, M. A., Liu, Z., Hu, Y., et al. (2009). The
 CALIPSO automated aerosol classification and lidar ratio selection algorithm. *Journal of Atmospheric and Oceanic Technology*, 26(10), 1994–2014.
 https://doi.org/10.1175/2009JTECHA1231.1
- Palacios-Penã, L., Jiménez-Guerrero, P., Baró, R., Balzarini, A., Bianconi, R., Curci, G., et al.
 (2019). Aerosol optical properties over Europe: An evaluation of the AQMEII Phase 3
 simulations against satellite observations. *Atmospheric Chemistry and Physics*, 19(5), 2965–
- 718
 2990. https://doi.org/10.5194/acp-19-2965-2019
- Rauber, R. M., Zhao, G., Girolamo, L. Di, & Colón-Robles, M. (2013). Aerosol size distribution,
 particle concentration, and optical property variability near caribbean trade cumulus clouds:
 Isolating effects of vertical transport and cloud processing from humidification using
 aircraft measurements. *Journal of the Atmospheric Sciences*, 70(10), 3063–308.
 https://doi.org/10.1175/JAS-D-12-0105.1
- Remer, L. A., Kleidman, R. G., Levy, R. C., Kaufman, Y. J., Tanré, D., Mattoo, S., et al. (2008).
 Global aerosol climatology from the MODIS satellite sensors. *Journal of Geophysical Research Atmospheres*, *113*(14), 1–18. https://doi.org/10.1029/2007JD009661
- Rosenfeld, D., Andreae, M. O., Asmi, A., Chin, M., Leeuw, G., Donovan, D. P., et al. (2014).
 Global observations of aerosol-cloud-precipitation- climate interactions. *Reviews of Geophysics*, 52, 750–808. https://doi.org/10.1002/2013RG000441
- Rosenfeld, D., Zhu, Y., Wang, M., Zheng, Y., Goren, T., & Yu, S. (2019). Aerosol-driven
 droplet concentrations dominate coverage and water of oceanic low-level clouds. *Science*,
 363(6427), eaav0566. https://doi.org/10.1126/science.aav0566
- Ross, A. D., Holz, R. E., Quinn, G., Reid, J. S., Xian, P., Joseph Turk, F., & Posselt, D. J.
 (2018). Exploring the first aerosol indirect effect over Southeast Asia using a 10-year
 collocated MODIS, CALIOP, and model dataset. *Atmospheric Chemistry and Physics*, *18*,
 12747–12764. https://doi.org/10.5194/acp-18-12747-2018
- 737 Sassen, K., & Wang, Z. (2008). Classifying clouds around the globe with the CloudSat radar: 1738 year of results. *Geophysical Research Letters*, *35*, L04805.
 739 https://doi.org/10.1029/2007GL032591
- Sassen, K., Wang, Z., & Liu, D. (2008). Global distribution of cirrus clouds from
 CloudSat/Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO)
 measurements. *Journal of Geophysical Research*, *113*, D00A12.
- 743 https://doi.org/10.1029/2008JD009972
- Shinozuka, Y., Kacenelenbogen, M., Burton, S., Howell, S., Zuidema, P., Ferrare, R., et al.
 (2020). Daytime aerosol optical depth above low-level clouds is similar to that in adjacent
 clear skies at the same heights: airborne observation above the southeast Atlantic. *Atmospheric Chemistry and Physics*, (January), 1–26. https://doi.org/10.5194/acp-20191007
- Sivaprasad, P., & Babu, C. A. (2014). Seasonal variation and classification of aerosols over an
 inland station in India. *Meteorological Applications*, 21(2), 241–248.
 https://doi.org/10.1002/met.1319

- Tackett, J. L., & Di Girolamo, L. (2009). Enhanced aerosol backscatter adjacent to tropical trade
 wind clouds revealed by satellite-based lidar. *Geophysical Research Letters*, *36*(14), 1–5.
 https://doi.org/10.1029/2009GL039264
- Tackett, Jason L., Winker, D. M., Getzewich, B. J., Vaughan, M. A., Young, S. A., & Kar, J.
 (2018). CALIPSO lidar level 3 aerosol profile product: Version 3 algorithm design. *Atmospheric Measurement Techniques*, 11(7), 4129–4152. https://doi.org/10.5194/amt-114129-2018
- Tan, S., Han, Z., Wang, B., & Shi, G. (2017). Variability in the correlation between satellitederived liquid cloud droplet effective radius and aerosol index over the northern Pacific
 Ocean. *Tellus B: Chemical and Physical Meteorology*, *69*(1), 1391656.
 https://doi.org/10.1080/16000889.2017.1391656
- Vaughan, M. A., Powell, K. A., Kuehn, R. E., Young, S. A., Winker, D. M., Hostetler, C. A., et
 al. (2009). Fully automated detection of cloud and aerosol layers in the CALIPSO lidar
 measurements. *Journal of Atmospheric and Oceanic Technology*, *26*(10), 2034–2050.
 https://doi.org/10.1175/2009JTECHA1228.1
- Winker, D. M., Tackett, J. L., Getzewich, B. J., Liu, Z., Vaughan, M. A., & Rogers, R. R.
 (2013). The global 3-D distribution of tropospheric aerosols as characterized by CALIOP. *Atmospheric Chemistry and Physics*, *13*(6), 3345–3361. https://doi.org/10.5194/acp-133345-2013
- Winker, David M., Pelon, J. R., & McCormick, M. P. (2003). The CALIPSO mission:
 Spaceborne lidar for observation of aerosols and clouds. *Lidar Remote Sensing for Industry and Environment Monitoring III*, 4893, 1–11. https://doi.org/10.1117/12.466539
- Winker, David M., Vaughan, M. A., Omar, A., Hu, Y., Powell, K. A., Liu, Z., et al. (2009).
 Overview of the CALIPSO mission and CALIOP data processing algorithms. *Journal of Atmospheric and Oceanic Technology*, 26(11), 2310–2323.
 https://doi.org/10.1175/2009JTECHA1281.1
- Wu, X., Balmes, K. A., & Fu, Q. (2021). Aerosol Direct Radiative Effects at the ARM SGP and
 TWP Sites: Clear Skies. *Journal of Geophysical Research: Atmospheres*, *126*(5), 1–24.
 https://doi.org/10.1029/2020JD033663
- Xian, P., Klotzbach, P., Dunion, J., Janiga, M., Reid, J., Colarco, P., & Kipling, Z. (2020).
 Revisiting the relationship between Atlantic dust and tropical cyclone activity using aerosol optical depth reanalyses: 2003–2018. *Atmospheric Chemistry and Physics*, 1–43. https://doi.org/10.5194/acp-2020-287
- Xu, H., Guo, J., Wang, Y., Zhao, C., Zhang, Z., Min, M., et al. (2017). Warming effect of dust
 aerosols modulated by overlapping clouds below. *Atmospheric Environment*, *166*, 393–402.
 https://doi.org/10.1016/j.atmosenv.2017.07.036
- Yang, W., Marshak, A., Várnai, T., Kalashnikova, O. V., & Kostinski, A. B. (2012). CALIPSO
 observations of transatlantic dust: Vertical stratification and effect of clouds. *Atmospheric Chemistry and Physics*, *12*(23), 11339–11354. https://doi.org/10.5194/acp-12-11339-2012
- Young, S. A., & Vaughan, M. A. (2009). The retrieval of profiles of particulate extinction from

- cloud-aerosol lidar infrared pathfinder satellite observations (CALIPSO) data: Algorithm 792 793 description. Journal of Atmospheric and Oceanic Technology, 26(6), 1105–1119. https://doi.org/10.1175/2008JTECHA1221.1 794
- Young, S. A., Winker, D. M., Hu, Y., & Kuehn, R. E. (2008). CALIOP algorithm theoretical 795 basis document, Part 4: Extinction retrieval algorithms. *Theoretical Basis Documents*. 796 Retrieved from 797 798 http://scholar.google.com/scholar?hl=en&btnG=Search&q=intitle:CALIOP+Algorithm+Th eoretical+Basis+Document+Part+3+:+Scene+Classification+Algorithms#0 799 Young, S. A., Vaughan, M. A., Kuehn, R. E., & Winker, D. M. (2013). The retrieval of profiles 800 of particulate extinction from Cloud-Aerosol Lidar and Infrared Pathfinder Satellite 801 Observations (CALIPSO) data: Uncertainty and error sensitivity analyses. Journal of 802 Atmospheric and Oceanic Technology, 30(3), 395–428. https://doi.org/10.1175/JTECH-D-803 12-00046.1 804 Zhang, Z., Meyer, K., Yu, H., Platnick, S., Colarco, P., Liu, Z., & Oreopoulos, L. (2016). 805
- 806 Shortwave direct radiative effects of above-cloud aerosols over global oceans derived from 8 years of CALIOP and MODIS observations. Atmospheric Chemistry and Physics, 16(5),
- 807
- 2877-2900. https://doi.org/10.5194/acp-16-2877-2016 808