

1 **Investigating the Frequency and Inter-Annual Variability in Global Above-Cloud Aerosol**  
2 **Characteristics with CALIOP and OMI**

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## ABSTRACT

23  
24 Seven and a half years (June 2006-November 2013) of Cloud-Aerosol Lidar with Orthogonal  
25 Polarization (CALIOP) aerosol and cloud layer products are compared with collocated Ozone  
26 Monitoring Instrument (OMI) Aerosol Index (AI) data and Aqua Moderate Resolution Imaging  
27 Spectroradiometer (MODIS) cloud products, to investigate variability in estimates of bi-annual  
28 and monthly above-cloud aerosol (ACA) events globally. The active- (CALIOP) and passive-  
29 based (OMI-MODIS) techniques have their advantages and caveats for ACA detection, and thus  
30 both are used to derive a thorough and robust comparison of daytime cloudy-sky ACA  
31 distribution and climatology. For the first time, baseline above cloud aerosol optical depth  
32 (ACAOD) and AI thresholds are derived and examined ( $AI = 1.0$ ,  $ACAOD = 0.015$ ) for each  
33 sensor. Both OMI-MODIS and CALIOP-based daytime spatial distributions of ACA events  
34 show similar patterns during both study periods (December – May) and (June – November).  
35 Divergence exists in some regions, however, such as Southeast Asia during June through  
36 November, where daytime cloudy-sky ACA frequencies of up to 10% are found from CALIOP  
37 yet are non-existent from the OMI-based method. Conversely, annual cloudy-sky ACA  
38 frequencies of 20-30% are reported over Northern Africa from the OMI-based method, yet are  
39 largely undetected by the CALIOP-based method. Using a collocated OMI-MODIS-CALIOP  
40 dataset, our study suggests that the cloudy-sky ACA frequency differences between the OMI-  
41 MODIS- and CALIOP-based methods are mostly due to differences in cloud detection capability  
42 between MODIS and CALIOP as well as QA flags used. An increasing inter-annual-variability  
43 of ~0.3-0.4% per year (since 2009) in global monthly cloudy-sky ACA daytime frequency of  
44 occurrence is found using the OMI-MODIS based method. Yet, CALIOP-based global daytime  
45 ACA frequencies exhibit a near-zero inter-annual-variability. Further analysis suggests that the  
46 OMI derived inter-annual-variability of cloudy-sky ACA frequency may be affected by OMI row

47 anomalies in later years. A few regions are found to have increasing slopes in inter-annual  
48 variability of cloudy-sky ACA frequency, including the Middle-East and India. Regions with  
49 slightly negative slopes of the inter-annual variability of cloudy-sky ACA frequencies are found  
50 over South America and China, while remaining regions in the study show nearly zero change in  
51 ACA frequencies over time. The inter-annual variability of ACA frequency are not statistically  
52 significant on both global and regional scales, though, given relatively lacking sample sizes. A  
53 longer data record of ACA events is needed in order to establish significant trends of ACA  
54 frequency regionally and globally.

55

## 56 1. Introduction

57 The above-cloud aerosol (ACA) phenomenon, wherein significant active-based  
58 backscatter and passive-based scattered solar radiances are induced by particles above what are  
59 predominately lower tropospheric clouds, has gained an increased amount of attention from the  
60 scientific community (e.g. Haywood et al., 2004; Wilcox et al., 2009; Coddington et al., 2010;  
61 Devasthale and Thomas, 2011; Wilcox, 2012; Kacenelenbogen et al, 2014). In particular,  
62 whereas passive-based atmospheric retrievals are compromised by a binding inability to  
63 decouple aerosol, cloud and atmospheric radiances in the ACA scenario, corresponding cloud  
64 property retrievals are uniquely biased (Wilcox et al., 2009; Meyer et al., 2013; Alfaro-Contreras  
65 et al., 2014; Li et al., 2014). ACA further perturbs regional radiation budgets by absorbing and  
66 reflecting radiation from the cloud layers underneath the unidentified aerosol particle layer (e.g.,  
67 Haywood et al., 2004), which again must be accounted for when estimating global cloud and  
68 aerosol forcing budgets and regional semi-direct impact on static stability and cloud feedback.  
69 Global oceans are covered with clouds nearly 70% of the time (e.g. Rossow and Schiffer, 1999),  
70 with almost non-existent corresponding ground-based verification data of ACA phenomena.  
71 This exacerbates the impact of ACA effects globally, limiting characterization of any  
72 quantitative impact and frequency of occurrence almost exclusively to satellite-based  
73 measurements.

74 ACA events are most effectively identified using active-based lidar measurements, which  
75 has been demonstrated using the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP;  
76 Winker et al., 2010; Kacenelenbogen et al, 2014 ), one of the few such instruments presently in  
77 satellite orbit. CALIOP measures backscattered signals at the 532 and 1064 nm wavelengths,  
78 including segregated linearly-parallel and orthogonal polarization backscatter states in the former

79 channel. In particular, the active-profiling element is essential for decoupling aerosol and cloud  
80 scattering contributions in ACA events (Devasthale and Thomas, 2011). Utilizing four years of  
81 CALIOP Level 2 data (Winker et al., 2009), Devasthale and Thomas (2011) evaluated seasonal  
82 and latitudinal patterns of ACA for liquid water cloud events. Alfaro-Contreras et al. (2014)  
83 describe seasonal frequencies in ACA over the southern Atlantic Ocean off the West African  
84 coastline, as well as over the Gulf of Tonkin in Southeast Asia where high ACA loading  
85 episodes were found during the summer and fall months and early spring months, respectively.

86 Whereas limited process studies have helped raise awareness of the ACA problem  
87 overall, year to year variability in global ACA frequency distribution has not yet been developed  
88 with CALIOP. Despite a nearly eight-year (2006-present) CALIOP data archive available, one  
89 must be considerate of the fact that satellite lidar profiling is constrained presently to a single  
90 laser-illuminated curtain and roughly sixteen daily orbits of the planet. Questions thus arise about  
91 the representativeness of CALIOP datasets for some climatological analyses, like ACA, given its  
92 temporal persistence and spatial extent (Devasthale and Thomas, 2011; Yu et al., 2012).  
93 Additionally, for CALIPSO-based ACA studies to be meaningful, the potential impacts of signal  
94 deterioration to CALIOP derived aerosol optical depth (AOD) values need to be known. Despite  
95 the practical limitations of applying passive sensors for studying phenomena like ACA, then, the  
96 relatively wide field-of-view on passive imagers renders far greater data volume, which makes  
97 them more ideal options for a long-term study.

98 Ozone Monitoring Instrument (OMI) measurements have also been used for studying  
99 ACA events (e.g., Wilcox et al., 2009; Yu et al., 2012; Alfaro-Contreras et al., 2014). In  
100 particular, the OMI Aerosol Index (AI), computed using the difference between observed and  
101 calculated ultraviolet (UV) radiances (Torres et al., 2007), has been used to locate UV-absorbing

102 aerosols suspended over bright cloud decks (e.g. Yu et al., 2012, Torres et al., 2012). This  
103 technique, originally used on the Total Ozone Mapping Spectrometer (TOMS), can only be used  
104 to detect UV-absorbing aerosols, such as biomass burning smoke and desert dust aerosols and is  
105 sensitive to underneath cloud properties (e.g. Yu et al., 2012; Alfaro-Contreras et al., 2014).  
106 Further and compared with CALIOP, OMI measurements represent a relatively large surface  
107 footprint of 13x24 km at nadir, which limits cloud-clearing efficacies since footprints of this size  
108 are prone to sub-pixel cloud contamination (Torres et al., 2007). Collocated Moderate Resolution  
109 Imaging Spectroradiometer (MODIS) observations, however, as part of NASA's A-Train  
110 satellite constellation, which includes CALIOP (Stephens et al., 2002), can be utilized to  
111 distinguish and filter cloudy pixels/scenes within the OMI footprint.

112 Comparison of active vs. passive based sensors for evaluating the spatio-temporal  
113 coverage of ACA events, and for studying inter-annual variability of ACA occurrence on  
114 regional and global scales, represents a conservative means for conceptualizing the breadth of the  
115 problem. The goal of this work is, therefore, to compare and contrast distributions in global and  
116 regional ACA frequencies and their year-to-year variability using both CALIOP- and OMI-  
117 based approaches. Caveats to each approach are specifically identified, and thus qualified within  
118 the discussion so as to keep comparison as consistent and robust as possible. We highlight  
119 regions particularly susceptible to ACA occurrence, establishing a baseline for future ACA-  
120 induced biases in satellite cloud property retrievals overall.

121

## 122 **2. Datasets and Methodology**

123 CALIOP Level 2 5-km cloud and aerosol layer products (Winker et al., 2010) and OMI  
124 Level 2 Collection 3 UV aerosol products (OMAERUV; Torres et al., 2007) are paired with  
125 **Collection 5.1** Aqua MODIS cloud products (MYD06\_L2; King et al., 1997) from June 2006

126 through November 2013. **Aerosol Robotic Network (AERONET; Holben et al., 1998) Level 2.0**  
127 **Version 2 cloud-screened data are also used to assist the analysis.**

128 For identification of ACA, 5-km CALIOP 532 nm cloud and aerosol layer products are  
129 used (Winker et al., 2009, 2010) for resolving aerosol extinction above apparent cloud top  
130 heights in each respective product file (e.g. Yu et al., 2012; Alfaro-Contreras et al., 2014). The  
131 532 nm above cloud aerosol optical depth (ACAOD) is then solved by integrating the extinction  
132 coefficient over those corresponding bins (Liu et al., 2013; Kacenelebogen et al., 2014). The  
133 CALIOP-based inter-annual variability analysis may be affected by CALIOP signal deterioration  
134 over time. Thus, collocated AERONET datasets are used, as first order approximation, for  
135 evaluating instrument-related variation in the year-to-year variability of CALIOP AOD.  
136 Reported at eight spectral bands ranging from 0.34  $\mu\text{m}$  – 1.64  $\mu\text{m}$  (Holben et al., 1998),  
137 AERONET AOD datasets are frequently used for validating satellite retrievals (e.g., Zhang et al.,  
138 2001; Yu et al., 2003; Kaufman et al. 2005; Remer et al., 2005; Hahn et al., 2010; Shi et al. 2011;  
139 Sayer et al. 2012), as well as model simulated aerosol optical properties (e.g. Zhang et al., 2011;  
140 2014).

141 The Level 2.0 cloud-screened and quality-assured AERONET AOD data (Eck et al.,  
142 1999) from all available coastal and island AERONET sites are used for collocating CALIOP  
143 data. AERONET AOD data are interpolated, based on a method described in Zhang and Reid  
144 (2006), to the 0.532  $\mu\text{m}$  CALIOP wavelength and are spatio-temporally-located with  
145 CALIOP AOD data. Year-to-year changes in AOD retrieved from the CALIOP instrument are  
146 investigated by calculating the global monthly-mean AERONET and CALIOP AODs and  
147 comparing the two monthly aerosol loading averages. CALIOP observations found to be within  
148 0.3 degrees latitude/longitude and  $\pm 30$  minutes of corresponding AERONET observations are

149 considered collocated in space and time (see Omar et al., 2013 for a summary of the limitations  
150 pairing CALIOP and AERONET observations). In addition, we have used only pairs that have  
151 collocated AERONET AOD ( $0.532 \mu\text{m}$ ) data less than 0.2 to exclude major aerosol episodes of  
152 continental origin. One additional quality assurance step is applied to exclude pairs with  
153 CALIOP AOD of larger than 0.6 for removing potentially noisy CALIOP data. In the case where  
154 several CALIOP observations are paired up with a single AERONET retrieval, a one-to-one  
155 relationship is established with the closest CALIOP observation.

156 OMI AI are used to isolate ACA events in those data. OMI AI and MODIS cloud  
157 datasets are spatio-temporally-collocated, given their position in the NASA “A-Train”  
158 constellation (e.g. Stephens et al., 2002), by collocating the two products with respect to  
159 overpass times and then identifying all temporally-collocated cloudy MODIS pixels located  
160 within the boundaries of the OMI footprint. Such methods are described further in Alfaro-  
161 Contreras et al. (2014). Cloud fractions from the Collection 5.1 MODIS MYD06 product, which  
162 are used to determine the opaqueness of the MODIS scenes, are reported at a 5 km horizontal  
163 resolution, are then leveraged for sub-pixel cloud clearing of the OMI AI. The MODIS cloud  
164 fraction is computed from the percentage of cloudy 1-km cloud mask product (MOD35) pixels  
165 within a given 5-km scene (e.g., Ackerman et al., 1998). It should be noted that this MODIS  
166 cloud fraction is reported regardless of the success of cloud optical property retrievals (e.g.,  
167 cloud optical depth and Liquid Water Path). The, OMI and MODIS data are each filtered and  
168 quality-assured (described in detail in Alfaro-Contreras et al., 2014) to calculate respective  
169 global ACA distributions. The OMI and MODIS data are spatially and temporally collocated,  
170 and the collocated OMI AIs are assigned to 100% cloudy MODIS scenes (as determined by  
171 MODIS, with a COD > 0). This collocation process and methods are further described in Alfaro-



172 Contreras et al. (2014). However, cloud inhomogeneity is not considered, and we leave the topic  
173 for another study.

174 If multi-layer clouds exist, MODIS can only resolve the highest cloud layer most of the  
175 time. Thus, we focus on the highest level clouds in any given atmospheric column using  
176 CALIOP cloud layer products for a more accurate representation between the two techniques.  
177 The CALIOP data are filtered based on the study by Yu et al. (2012), where aerosol layers found  
178 with ‘medium’ or ‘high’ confidence are used. Note that, initially, cloudy scenes are defined as  
179 CALIOP COD > 0 for the CALIOP-based method and no QA steps are applied to the CALIOP  
180 cloud layer products to ensure the detection of all possible ACA events. The effect of QA flags  
181 from the CALIOP cloud layer products to the detected CALIOP ACA frequency is further  
182 explored in Section 4.2 (as well as shown in Devasthale and Thomas, 2011). It is known that  
183 that the OMI instrument has experienced row anomalies since 2008-2009  
184 (<http://www.knmi.nl/omi/research/product/>, accessed on 22 Dec. 2014). Thus, the impact of the  
185 row anomalies on the inter-annual variability of ACA occurrence derived from OMI AI is  
186 explored later in this paper.

187

### 188 **3. Above-Cloud Aerosol Baselines and Limitations**

189 There are always aerosol particles above clouds (a fact that quickly becomes lost when  
190 discussing the basic physics of ACA relative to satellite observation). Therefore, there exists  
191 some baseline thresholds by which active backscatter and/or passive radiances become  
192 significant relative to a given physical process or retrieval (i.e., radiative forcing, heating rates,  
193 transmission estimates, cloud microphysical retrievals, etc.). Accordingly, each of the  
194 instruments subject to the ACA phenomenon in this study exhibit fundamental sensitivities to  
195 ACA detection, which impact our ability to characterize the problem fully. Therefore, the

196 baseline thresholds for significant ACA events need to be identified for both OMI- and CALIOP-  
197 based ACA studies.

198 To conceptualize the problem, we look at the globally averaged cloud-top height for  
199 clouds located under aerosol plumes, which is found to be roughly 2.0 km and compares well  
200 with previous studies (Devasthale and Thomas, 2011). Thus, we consider the unique AERONET  
201 site at Mauna Loa, Hawaii (LAT/LON, 3397 m above mean sea level). This free-tropospheric  
202 ground site rests at an altitude roughly within the global mean cloud top heights. Indeed, this  
203 physical feature of the site (that is, **most commonly** being above a cloud deck below) is one of  
204 the key reasons for the importance of the site globally. The yearly mean Level 2.0 AERONET  
205 AOD (500 nm) there ranges from 0.013-0.023 (500 nm) from 1996-2013, and provides a  
206 generalized estimate for potential baseline ACAOD value globally. Kacenelenbogen et al.  
207 (2014) report that the CALIOP lidar exhibits limitations in detecting ACA plumes with ACAOD  
208 less than 0.02. This lower value may, therefore, represent an effective noise floor, whereby  
209 CALIOP algorithm response below it is compromised.

210 Based on Kacenelenbogen et al. (2014), combined with the AOD climatology from the  
211 Mauna Loa AERONET site analyses, we arbitrarily set the baseline CALIOP ACAOD value to  
212 0.015. Still, the CALIOP ACAOD baseline of 0.015 is arbitrary. We thus investigate CALIOP-  
213 based ACA frequency distributions by varying the baseline values to 0, 0.01, 0.015 and 0.02 as  
214 shown in Fig. 1. Figures 1a-d show the cloudy-sky global ACA frequency distribution from  
215 CALIOP, defined in Table 1, for the Dec.- May period, for baseline ACAODs of 0 (1a), 0.01  
216 (1b), 0.015 (1c) and 0.02 (1d) respectively, using the CALIOP aerosol layer datasets. Note that  
217 different from the cloudy-sky frequency, another way of measuring ACA frequency has been  
218 proposed by Devasthale and Thomas (2011) and is referred as the all-sky frequency from

219 CALIOP in this study, also defined in Table 1. The difference between the two techniques is  
220 discussed in more detail during the section analyzing the year to year variation in ACA  
221 frequency occurrence.

222 Shown in Fig. 1, no clear difference is observed in the cloudy-sky ACA frequency by  
223 applying various CALIOP ACAOD baselines. A similar conclusion can also be made for the  
224 June- Nov period (Figs. 1e-1h). Thus, for the purposes of this paper, the baseline CALIOP  
225 ACAOD value of 0.015 (0.532  $\mu\text{m}$ ) is chosen, and the sensitivity of ACA inter-annual variability  
226 to the selection of the baseline CALIOP ACAOD is explored in a later section. Additionally, our  
227 selection of CALIOP ACAOD baseline has little effect on the background cloudy-sky ACA  
228 frequency, which is for the most part less than 5 % (dark blue) globally. Thus, we arbitrarily  
229 select 5% as the threshold between background and significant cloudy-sky ACA frequencies. For  
230 the remainder of the paper, ACA frequencies less than five percent are not considered for global  
231 distributions of ACA frequencies (except for sensitivity and case studies).

232 To derive the corresponding noise floor value for above-cloud OMI AI, a pairwise  
233 comparison of collocated above-cloud OMI AI and CALIOP AOD has been performed using  
234 one year (2007) of collocated OMI-MODIS and CALIOP data, as described in Alfaro-Contreras  
235 et al. (2014), though without any limitations on the cloud-top height. Figure 2a depicts the  
236 relationship between binned above-cloud OMI AI and CALIOP AOD segregated into six  
237 different underlying MODIS-derived CODs (Yu et al., 2012, Torres et al., 2012). The bin-  
238 averaged CALIOP ACAOD of 0.015, which reflects the baseline CALIOP ACAOD value  
239 chosen above, corresponds with OMI AI values of 0.7 - 1.2 for underlying MODIS CODs  
240 ranging from 0 to 20. Note that, if CALIOP ACAODs are biased low, the corresponding OMI  
241 AI thresholds may bias high using methods as shown in Fig. 2a.

242 Still, as suggested from Fig. 2a, baseline values of OMI AI vary from 0.7 to 1.2  
243 depending on the underlying cloud properties. To further explore the issue, detected ACA events  
244 are evaluated using different baseline OMI AI values, similar to the CALIOP ACAOD baseline  
245 analysis and shown in Figs. 2b-2i, though using only those bin averages with cloudy-sky ACA  
246 frequency greater than five percent. Figures 2b-2e depict the multi-year (2006-2013) cloudy-sky  
247 ACA frequency global average for the Dec.-May period, by applying AI baseline thresholds of  
248 0.7 (2b), 0.8 (2c), 0.9 (2d) and 1.0 (2e) respectively. With the use of the baseline OMI AI value  
249 of 0.7, most of the remote southern oceans stand out for significant case numbers. By increasing  
250 the AI baseline value to 1.0, in contrast, detected ACA events are significantly reduced. A  
251 similar conclusion can also be drawn from the June-Nov. period (Figs. 2f-i). Given that hand-  
252 held ship borne sun photometer measurements collected by the Marine Aerosol Network (MAN;  
253 Smirnov et al., 2011) show an averaged AOD ( $0.55 \mu\text{m}$ ) of 0.07 or less from  $30^\circ$  to  $60^\circ$  S (Toth  
254 et al., 2013), significant ACA events are not likely over remote southern oceans. Thus, based on  
255 Figs. 1 and 2, CALIOP ACAOD of 0.015 and an above-cloud OMI AI of 1.0 are chosen as  
256 baselines. As we have now defined our baseline thresholds for ACA from both OMI and  
257 CALIOP, this enables us to create definitions of the various ACA frequencies used throughout  
258 this study, which are shown with further detail in Table. 1.

259 Selection of baseline CALIOP ACAOD and OMI AI is clearly subjective, and done for  
260 qualitative analysis in subsequent sections. There are multiple caveats that must be considered  
261 before constraining these values more accurately and representatively. First, as mentioned  
262 earlier, the CALIOP instrument has issues in detecting distinct optically-thin aerosol layers,  
263 especially during daytime. Additionally, it has been reported that CALIOP has a decreased  
264 sensitivity to stratospheric aerosols layers (Thomason et al., 2007; Winker et al., 2009). Third,

265 besides aerosol loading, OMI AI is also sensitive to parameters such as aerosol vertical  
266 distribution, optical depth of underlying cloud and aerosol single scattering albedo (e.g. Yu et al.,  
267 2012). Thus, setting a seasonal and regional based baseline for ACA requires a more in depth  
268 analysis and should be considered in future studies. Still, this study presents the first ever  
269 attempt to solve ACA baselines, and the thresholds selected are the best noise floors we can  
270 derive with the given inputs.

271

#### 272 **4. Comparison of ACA Global Climatology using Two Separate Techniques**

##### 273 **4.1 ACA Global Climatology from all available MODIS, OMI and CALIOP data**

274 Figure 3a depicts the multi-year gridded mean near-global distribution (180°W - 180°E,  
275 45°S - 60°N) of the OMI-derived daytime cloudy-sky ACA frequency (defined in Table 1.) for  
276 December to May. Figures 3b and 3c show corresponding cloudy-sky daytime and nighttime  
277 frequencies, respectively, using CALIOP data (defined in Table.1). Figures 3d-3f show the  
278 corresponding information to Figs. 3a-3c for June to November.

279 Comparison of daytime cloudy-sky ACA frequency distribution is consistent between the  
280 two sensors and seasonal periods investigated, and depicted in Figs. 3g-3j. Some differences are  
281 distinct during December-May, as cloudy-sky ACA frequencies as high as 10 % are visible over  
282 the Gulf of Mexico from CALIOP, for instance, whereas they are non-existent from OMI-  
283 MODIS (Fig. 3a). Cloudy-sky ACA frequencies of 20-30 % are found with OMI-MODIS over  
284 high-latitude northern Asia, in contrast with CALIOP that shows no such activity (Fig. 3i).  
285 During June-November, both methods resolve ACA events over the west coast of Africa, as well  
286 as over the Middle East, of similar magnitude (10-60%). However, distinct differences can be  
287 found between the two datasets. Higher cloudy-sky ACA frequency values of 10-30% are found  
288 over North Africa using OMI-MODIS, in contrast with much lower values of 10-20% found

289 using CALIOP, for example. An OMI-based ACA study should correspond with a higher noise  
290 floor compared with that of an active sensor, based on OMI's much coarser spatial and vertical  
291 resolutions, an inability to resolve non-UV absorbing aerosols, and the fundamental decoupling  
292 of column-integrated radiances themselves. Still, if the OMI AI baseline is biased, it may  
293 introduce an additional difference between OMI-MODIS- and CALIOP-based ACA frequencies.

294 Cloudy-sky ACA frequencies as high as 10-30 % are found over North Africa for both  
295 periods from OMI-MODIS while CALIOP returns much lower percentages (10-20%) over the  
296 same region. This region is dominated by dust particle transport (Kaufman et al. 2005), which is  
297 detected by both OMI and CALIOP. Therefore, we suspect that their relative differences as  
298 derived in Figs. 3i and 3j are likely linked to the misidentification of thick dust plumes as clouds  
299 by the MODIS cloud-masking scheme over bright desert surfaces (e.g., Levy et al., 2013). Such  
300 a misclassification is also illustrated in a case study (July 8, 2007, over the Saharan region) as  
301 shown in Fig. 4. Over a section of CALIOP detected cloud free aerosol polluted regions (15-  
302 30°N, 2-6°W), where OMI AI values are above 2, cloud fraction of a unit (fully cloud cover) is  
303 reported by the MODIS MYD06 product, indicating a potential misclassification of aerosol  
304 plumes as clouds in the MODIS MYD06 product. Further differences observed between the two  
305 datasets may also be due to different algorithmic sensitivities exhibited relative to both the  
306 optical depth of the underlying cloud and overlying aerosol plume, the OMI AI and CALIOP  
307 AOD noise floors used to define the ACA events, the particular QA settings applied to any of our  
308 datasets, differences in cloud-detection techniques between CALIOP and MODIS or the inability  
309 of OMI to detect all aerosol types. We have further explored this issue in section 4.2.

310 Compared with daytime, increases in both the spatial extent and cloudy-sky CALIOP  
311 ACA frequencies are observable at night, as seen from Figs. 3b, c, e and f, over most regions.

312 Over the most common ACA regions, nighttime cloudy-sky ACA frequencies can be 10-30 %  
313 higher than during day, which may partially due to the stronger sensitivity of CALIOP at night  
314 allowing for detection of optically thin aerosol plumes. In particular, ACA events are observed  
315 with extended frequency over the west coast of North America year round and over the west  
316 coast of South America for the June-Nov. period. Cloudy-sky ACA frequencies at night, over  
317 both of these regions, are composed of optically-thin aerosol loading cases above our defined  
318 noise floor. Nighttime ACA events are also observed over the east coast of Asia year round.  
319 One reason for differences in spatial coverage between daytime and nighttime ACA events is  
320 plausibly linked to a lower planetary boundary layer that affects the formation of low clouds  
321 (e.g., Schrage et al., 2012). Still, the discrepancy between nighttime and daytime ACA events  
322 can be partially attributed to the potential detection of relatively **diffuse ACA** plumes that are  
323 more detectable during nighttime compared with day as a result of the higher signal to noise ratio  
324 for CALIOP nighttime data (e.g. Kacenelenbogen et al., 2014).

325 Shown in Fig. 5 are averaged above-cloud OMI AI and CALIOP AOD values for  
326 corresponding ACA events from Fig. 3. Figure 5a depicts the mean near-global distribution of  
327 OMI AI over MODIS-resolved cloudy skies, defined as OMI-MODIS collocated cloudy pixels  
328 (cloud fraction of unity) and OMI AI averaged for each  $1^\circ \times 1^\circ$  grid box, during December to  
329 May. Only bins with averaged AI greater than 1.0 are plotted in accordance with our defined  
330 noise floor. **Also for December-May**, Fig. 5b depicts multi-year mean gridded daytime  
331 CALIOP ACAOD averaged for each  $2.5^\circ \times 2.5^\circ$  grid box for CALIOP-defined cloudy pixels  
332 (COD > 0), using only bin averaged ACAOD greater than 0.015. Figure 5c **features** the same  
333 information as Fig. 5b, now for nighttime CALIOP retrievals.

334 During the Dec.-May period, elevated OMI AI values are observed over the Saharan  
335 desert region of northern Africa, as well as in Southeast Asia off the coast of northern Vietnam.  
336 In comparison with OMI AI, CALIOP AOD shows a much broader distribution of AODs greater  
337 than the baseline ( $ACAOD > 0.015$ ) for the entire globe. Bin averaged AIs greater than the  
338 baseline ( $AI > 1.0$ ) are sparse during the winter and spring months. Additionally, optically thin  
339 aerosol plumes are observed over the Northern Pacific Ocean during the CALIOP nighttime  
340 analysis (Fig. 5c), when compared to the daytime (Fig. 5b).

341 Figures 5d-5f depict the same information as Figs. 5a-5c, now for the Jun.-Nov. period.  
342 This period exhibits a relatively large overall distribution of ACA events. In addition to the  
343 Saharan dust outbreaks, elevated AI and AOD values over the southern Africa smoke region are  
344 also found from both OMI and CALIOP datasets, respectively. This period exhibits large  
345 aerosol loading and ACA frequency over Southern Africa and the southeast Atlantic Ocean.  
346 High values of ACAOD are also found over the Indian Ocean and Arabian Sea, due likely to the  
347 transport of dust aerosols from the east Saharan and Arabian Gulf regions (Satheesh et al., 2006).  
348 Comparing Figs. 3 and 5 over regions such as the west coast of South and North America, it is  
349 clear that cloudy-sky ACA frequencies are mostly attributable to relatively low aerosol loading  
350 events. Figure 5 shows a drastically reduced distribution of averaged OMI AIs above the AI  
351 baseline (1.0) in comparison to averaged CALIOP ACAODs above the AOD baseline (0.015).

352 Again, differences are visible here between day and nighttime CALIOP AOD  
353 distributions. Off the Southwest coast of Africa, the development of marine stratus-type clouds,  
354 as suggested from Fig. 6, may lead to higher ACAOD values at night. Over India and the  
355 Middle East, we suspect that higher daytime ACAOD values may exist. Still, lower CALIOP



356 signal-to-noise during daytime may be a limiting factor that contributes significantly to the  
357 difference.

358 It is likely that most ACA events occur over low-level liquid-phase cloud decks.  
359 Therefore, spatial distributions of CALIOP-derived low-level clouds are investigated. Figure 6a  
360 (6b) depicts the daytime (nighttime) multi-year mean distribution of low-level clouds (defined as  
361 the ratio of CALIOP scenes with a COD > 0 and cloud-top height < 3km over total number of  
362 CALIOP scenes) during December – May, for years 2006 to 2013. CALIOP cloud layer data are  
363 gridded into 2.5° x 2.5° bins. Figures 6c and 6d depict the same information as Figs. 6a and 6b,  
364 now from June – Nov., for years 2006 to 2013. Figures 6e and 6f depict the ratio between  
365 daytime and nighttime low-level cloud frequencies per bin for the Dec.-May period and June-  
366 Nov. periods, respectively. The ratio is as high as 2.0 over the Northern and Southern Africa  
367 regions during June-Nov., as well as over the Western US annually. Such a high ratio between  
368 day and nighttime data leads to a nighttime frequency of 10-20 % low-level cloud coverage  
369 increase over most regions compared with daytime observations, plausibly due to diurnal  
370 boundary layer effects.

371 A significant percentage of CALIOP-derived low-level clouds are plausibly  
372 stratocumulus clouds, which are frequently observed over the west coasts of major continents  
373 (e.g. Wood et al., 2012). Qualitative comparison of Figs. 5 and 6 indicates reasonable  
374 consistency between high frequencies of CALIOP-defined low-level cloud formation and ACA  
375 loading. With the exception of the Saharan region, again due to the possible misclassification of  
376 thick aerosol plumes as clouds by MODIS discussed earlier, most ACA loading cases are found  
377 where the CALIOP-defined low-level cloud formation six month frequency exceeds 20 %. This  
378 indirectly confirms that most ACA outbreaks occur over CALIOP-defined low-level clouds.

379 It is also useful to evaluate ACA frequency relative to mean clear-sky AOD. Figures 7a-7d  
380 depict the multi-year mean clear-sky CALIOP AOD for the same temporal and spatial domains  
381 as Figs 5b, c, e and f, respectively. As opposed to the cloud-sky ACA aerosol loading (Fig. 5),  
382 AOD loading over clear-skies shows more activity inland, as the formation of low-level clouds is  
383 more common over coastal regions (ICCP, 2007). An inter-comparison among Figs. 5, 6 and 7  
384 suggests that ACA events do not necessarily follow clear sky AOD patterns but rather those  
385 above-cloud aerosol-polluted regions with a high frequency of low-cloud presence.

386

#### 387 **4.2 ACA Global Climatology from the collocated MODIS, OMI and CALIOP dataset**

388 As illustrated in Fig. 3 for the Dec.-May daytime period, ACA events over North Africa  
389 as derived from the OMI-MODIS-based method are not found from the CALIOP-based method.  
390 Also, ACA events over India, as reported from the CALIOP-based method, are not visible from  
391 the OMI-MODIS-based method. Similarly, for the June-November period, ACA events over  
392 North Africa reported from the CALIOP-based method are not as frequent as those seen from the  
393 OMI-MODIS-based method. Yet the ACA events detected from the CALIOP-based method  
394 over Southern China are not visible from the OMI-MODIS-based method.

395 To identify differences between the OMI-MODIS- and CALIOP-based cloudy-sky ACA  
396 global climatology, a collocated dataset has been constructed that includes spatially and  
397 temporally-collocated MODIS, OMI and CALIOP data for the period of June 2006 – November  
398 2008. Note that no collocated data are available after Nov. 2008 due to the row anomaly of the  
399 OMI instrument. All three sensors are on board the A-train constellation, making temporal  
400 collocation less of an issue, and we require the observational times of the three datasets to be  
401 within +/- 30 minutes to be considered. To spatially collocate the three datasets, only MODIS

402 (OMI) observations within 0.04 degrees (0.2 degrees) of the center of a CALIOP data point  
403 (from the 5-km CALIOP aerosol and cloud layer products) are used. Using the collocated OMI-  
404 MODIS-CALIOP data set, differences in cloudy-sky OMI-MODIS- and CALIOP-based ACA  
405 frequencies are studied as functions of CALIOP cloud and aerosol QA flags (Devastahale and  
406 Thomas, 2011), the differences between MODIS and CALIOP reported cloud coverages, and  
407 aerosol properties (UV-absorbing versus non UV-absorbing aerosols).

408         Similar to Figs. 3a and b, Figs. 8a and d show the cloudy-sky ACA frequency as detected  
409 by the CALIOP- and OMI-MODIS-based methods respectively, but with use the OMI-MODIS-  
410 CALIOP collocated dataset, for the Dec.-May period. Figures 8b (8e) and 8c (8f) show the all-  
411 sky ACA frequency and cloudy sky frequency for the CALIOP- (OMI-MODIS-) based methods.  
412 As mentioned in Table 1, all-sky ACA frequency is defined as the number of ACA events  
413 divided by all data points. Thus, Figs 8b (8e) and 8c (8f) can also be considered as ACA event  
414 data counts and cloudy-sky data counts for the CALIOP- (OMI-MODIS-) based method.  
415 Readers should be aware that the spatial distribution of MODIS cloud fraction, as shown in Figs.  
416 8f and l, differ from the spatial distribution of cloud fraction obtained from the standard MODIS  
417 cloud products (e.g. King et al., 2013) for a few reasons. First, Figs. 8f and 8l are constructed  
418 using the MODIS cloud fraction values from a collocated OMI, CALIOP and MODIS dataset,  
419 and thus, only near-nadir MODIS cloud mask data are used. Also, cloud fraction values (at a 5  
420 km resolution) from the MODIS MYD06 product are used. To be consistent with the OMI-  
421 MODIS analysis, only the 5-km granules that are 100% cloudy (or 25 1-km MODIS pixels  
422 within a 5-km granule are all cloudy) are counted as cloudy granules. Thus, broken and/or non-  
423 contiguous clouds may be excluded in the cloud fraction calculation.

424           The first thing to notice from these data is that cloudy sky frequency from the CALIOP-  
425 based method is higher than that of the OMI-MODIS-based method. The differences in cloudy  
426 sky frequencies are not unexpected, as the CALIOP-based method can detect optically-thin  
427 clouds (such as thin cirrus clouds) for which the OMI-MODIS-based method is limited (e.g.,  
428 Toth et al., 2013). Also, the all-sky ACA frequencies from CALIOP- and OMI-MODIS-based  
429 methods show similar magnitudes for both the Dec.-May and June-Nov. periods. Thus, the  
430 higher cloud-sky ACA events over North Africa, as reported from the OMI-MODIS-based  
431 methods, are likely due to the differences in cloud detection capability among the different  
432 sensors.

433           For the Dec.-May period, higher all-sky ACA frequency is reported from the CALIOP-  
434 based method over India. A similar situation is also found for the June-Nov. period over  
435 Southeast Asia. While one would suspect that the greater number of ACA events over India and  
436 Southeast Asia regions could be due to the fact that the OMI-MODIS based method is only  
437 sensitive to non UV-absorbing aerosols, we also evaluated the issue with respect to CALIOP QA  
438 flags. Figure 9 shows global plots of **cloudy sky frequency and all-sky ACA** frequencies from  
439 the original QA metrics used to generate Fig. 3, as well as global plots altering CALIOP aerosol  
440 and cloud QA flags to ‘lenient’, ‘intermediate’ and ‘strict’. Here, the CAD scores and Feature  
441 Classification Flags are used to define the quality of each retrieval. In order for a feature to be  
442 considered lenient, intermediate or stringent quality, its CAD score absolute value must be  
443 greater than either 0, 20 and 70, respectively. In addition, the feature flag must also return at least  
444 ‘low’, ‘medium’ and ‘confident’ result for the ‘lenient’, ‘intermediate’ and ‘stringent’ QA levels,  
445 respectively as defined in Liu et al. (2009).

446 Figures 9a-9c show the distributions of **cloudy sky frequency** with the use of lenient,  
447 intermediate and strict CALIOP cloud QAs, respectively. These data reflect how cloud QA  
448 exhibits only a minor effect on the spatial distribution of **cloudy sky frequencies**. Figures 9d-9f  
449 show the spatial distribution of **all-sky ACA frequency** with the ‘lenient’ aerosol QA setting but  
450 with the cloud QA levels of ‘lenient’, ‘intermediate’ and ‘strict’, respectively. Clearly seen from  
451 Figs. 9d and 9e, with the changing of cloud QA setting from ‘lenient’ to ‘intermediate’, are that  
452 the CALIOP-based **all-sky ACA frequencies** are much reduced over North Africa, the Middle-  
453 East, India and Southern China. This indicates that a portion of the observed differences  
454 between the OMI-MODIS- and CALIOP-based methods may due to cloud QA. Similarly, when  
455 we hold the cloud QA setting constant at ‘lenient’ while varying the aerosol QA setting from  
456 ‘lenient’ to ‘intermediate’ and ‘strict’ (Figs. 9g-9i), no significant changes in **all-sky ACA**  
457 **frequencies** are found. We repeat the process for the June-November period, as shown in Figs.  
458 **9j-9r**, and similar conclusions are found.

459 The CALIPSO Level 2 cloud and aerosol layer products include cloud retrievals  
460 conducted using horizontal averages at the three extended settings (e.g. 5, 20 or 80 km averages).  
461 While 5 km averaging detects the most “reliable” cloud and aerosol signals, the 80-km averaging  
462 locates features with “weaker” signals (Vaughan et al., 2009). Since the CALIPSO Level 2  
463 cloud and aerosol layer products are used in this study, the results presented here shall include  
464 horizontal averages from the three setting as mentioned. In addition, using CALIOP’s ability to  
465 distinguish different aerosol types, we find that absorbing aerosols (dust, smoke and polluted  
466 dust) constitute about 80 % of ACA particles over southeast Asia during June – Nov., and more  
467 than 90 % over India during the Dec. – May period. **Thus, OMI’s inability to detect all aerosol**  
468 **types may play a minor role in** the observed ACA frequency differences over these regions. On

469 the contrary, the differences in cloud detection capability, the QA settings and their arbitrary  
470 thresholds used are instead likelier be the primary causes for the discrepancies between OMI-  
471 MODIS- and CALIOP-based methods. Still, aerosol type discrimination from CALIOP  
472 measurements has its own limitations, and we leave this topic for a future paper to explore.

473

## 474 **5. Inter-Annual Variability of Global ACA Frequency**

475 An analysis of the year-to-year variation in global cloud-sky ACA frequency is carried  
476 out for five different scenarios. The different scenarios are: OMI daytime cloudy-sky frequency,  
477 CALIOP daytime cloudy-sky and all-sky frequencies and CALIOP nighttime cloudy-sky and all-  
478 sky frequencies. As suggested from Section 4.2, only CALIOP data with both cloud and aerosol  
479 QA settings as either ‘medium’ or ‘highest’ confidence levels are used hereafter. Figure 10  
480 shows CALIOP daytime cloudy-sky frequency (red) and all-sky frequency (blue), CALIOP  
481 nighttime cloudy-sky frequency (orange) and all-sky frequency (purple), and OMI daytime  
482 cloudy sky-frequency (green). Each data point represents the global monthly-mean ACA  
483 frequency of CALIOP and OMI calculated from 2.5° and 1° gridded ACA frequencies,  
484 respectively.

485 An increase in the OMI cloudy-sky ACA frequency over the study period is apparent in  
486 this global dataset, most noticeably since 2009. However, this inter-annual variability is not  
487 matched in the CALIOP data. The seasonal variation in ACA frequency is observed from year-  
488 to-year for both OMI and CALIOP (dashed lines). However, from the year-to-year variation  
489 lines (showing a percentage change per year), only the OMI daytime cloudy-sky frequency  
490 shows a significant increase over this time period (solid lines). The increasing inter-annual  
491 variability in OMI derived daytime global cloudy-sky ACA frequency, which is not apparent in

492 any of the CALIOP derived global cloudy-sky ACA frequencies, is troublesome and may be  
493 attributed to any of the different sensitivities of the two techniques, including cloud and aerosol  
494 optical properties, aerosol-cloud separation distance, and/or deficiencies in the OMI data  
495 products. As will be described below, we further investigate several aspects of the observed  
496 increasing in inter-annual variability in the OMI derived daytime cloudy-sky global ACA  
497 frequency.

498         Given the unexpected monotonic increase in global ACA frequency derived using OMI  
499 AI data over the course of our study, we examine the inter-annual variability in the OMI daytime  
500 cloudy-sky ACA frequency more closely. Figure 10 indicates a near-zero increase in the seasonal  
501 averages during the first few years of the study, with frequencies increasing at a rate of roughly  
502 0.3-0.4% per year starting in 2009. This time period coincides with the start of OMI data loss  
503 due to row anomalies, as mentioned above, leading us to further investigate this as a possible  
504 reason for the increase in the observed OMI cloudy-sky ACA frequency. Note that we detected  
505 data loss while collocating OMI and CALIOP datasets and found no collocated pixels after 2008;  
506 a possible sign that the data loss is likely affecting OMI nadir viewing pixels. This is illustrated  
507 in Fig. 11a, which depicts a single swath of OMI AI over the African continent on 1 August 2007  
508 where only OMI pixels with valid AI are shown. The data loss affected a large portion of the  
509 OMI AI data near the nadir regions of each OMI AI swath, as shown from a swath in 1 June  
510 2009 (Fig. 11b).

511         Given that the data loss affects mostly nadir-viewing OMI pixels, OMI AI are evaluated  
512 as a function of the OMI sensor's viewing zenith angle (VZA) shown in Fig. 12. All OMI AI  
513 pixels for one year (2007) are averaged into one-degree VZA bins. Averaged OMI AI values at  
514 the edge of the swath are generally higher by about one AI unit than retrievals taken near the

515 center of the swath. Thus, our analysis, which examines inter-annual variability in the OMI-  
516 derived ACA frequency, is compromised due to viewing geometry bias impacting later years of  
517 the OMI aerosol products. The remainder of the paper will focus solely on year to year variation  
518 derived from CALIOP ACA frequencies, and no further discussion of OMI AI frequencies will  
519 be carried out.

520 Next, AERONET AOD data are used to identify possible bias in the CALIOP lidar due to  
521 potential signal deterioration in the instrument. Figure 13 depicts the year-to-year variation in the  
522 clear-sky AOD derived using collocated CALIOP-AERONET data over all coastal and island  
523 AERONET stations (Zhang and Reid, 2006). The inter-annual variability in global AOD similar  
524 to those for the collocated AERONET and CALIOP data as shown in Fig. 13 seems to suggest  
525 that potential deterioration issue from CALIOP are rather insignificant to our ACA study.

526

## 527 **6. Sensitivity Study**

528 We next investigate the impact that our noise floor thresholds for overlying CALIOP  
529 AOD and/or underlying COD exhibit on derived global CALIOP cloudy-sky ACA frequency.  
530 All CALIOP cloud and aerosol layer datasets are reprocessed such that the following conditions  
531 are met: (a) the underlying COD is greater than 0.3 and 2.5, respectively, (b) the AOD of the  
532 above-cloud aerosol plume is greater than 0, and (c) both conditions (a) and (b) are true.  
533 Passive-based radiance retrievals have been shown to lack sensitivities to optically-thin cloud  
534 detection for optical depths less 0.3 (Sassen and Cho, 1992; Ackerman et al., 2008; Holz et al.,  
535 2008). Thus, restricting the CALIOP COD to this threshold offers a more direct comparison of  
536 CALIOP- and OMI-based ACA frequencies. However, given that this range of optical depth  
537 corresponds with relatively high cirrus clouds, for which little contribution to the overall sample  
538 is expected, and broken low-level liquid phase clouds that are biased to ambiguously low values



539 from signal aggregation effects in the 5-km product (Leahy et al., 2012; Campbell et al., 2015),  
540 this higher threshold provides a more representative basis for evaluation. We re-compute the  
541 monthly global mean cloudy-sky frequency for each of the CALIOP-constrained samples  
542 defined above during both daytime and nighttime. The inter-annual variability in global cloudy-  
543 sky ACA frequency derived from CALIOP are shown in Fig. 14. Corresponding sample sizes  
544 and mean global frequencies are shown in Table 2.

545 In comparison with the unfiltered data from the daytime (solid red) and nighttime (dotted  
546 red) analyses, the various threshold techniques, including the filtering of CALIOP ACAOD  
547 according to our floor noise, correspond with significant variance in our results. However, all  
548 sensitivity tests seem to show the same slightly-negative trend in cloudy-sky ACA frequency.  
549 Although, those ACA events found over optically thicker clouds ( $COD > 2.5$ ) seem to show  
550 more of a null inter-annual variability over time rather than a slightly-negative inter-annual  
551 variability in the CALIOP global ACA frequency. The COD threshold tests raises the daytime  
552 mean global cloudy-sky frequency from 1.8% to 2.0 and 2.5% for the 0.3 and 2.5 COD  
553 thresholds, respectively. This corresponds with a reduction in the sample size of approximately  
554 0.4 ( $COD < 0.3$ ) and 0.6 ( $COD < 2.5$ ) million scenes when compared with the unfiltered  
555 methods. During the nighttime analysis, the global mean cloudy-sky frequency is changed from  
556 4.5% to 6.1 and 8.1%, respectively, while data counts change to 2.8 and 2.1 million globally for  
557 the corresponding nighttime COD threshold tests. Setting a noise floor threshold on the AOD  
558 reduces mean global cloudy-sky ACA frequencies by 0.33 and 1.9% for day and nighttime  
559 analyses, respectively, corresponding with a reduction of global data counts of 0.3 and 1.6  
560 million scenes. After screening out millions of samples during this sensitivity analysis, the same  
561 near-zero or decreasing trend is found for CALIOP ACA frequencies, which is indication that

562 neither cloud or aerosol thresholds, or lack there-of, have a major impact on the inter-annual  
563 variability of global CALIOP cloudy-sky ACA frequency.

564

## 565 **7. Regional Year-to-Year Variation Analysis**

566 A regional analysis of cloudy-sky ACA frequency is also conducted, consistent with  
567 methods described above for global analysis. Regional analyses were chosen over high ACA  
568 frequency regions, as indicated from Fig. 3. The nine regions of interest, shown in Table 3 and  
569 indicated by the red boxes in Fig. 3, are: Northern Saharan Africa, Southern Africa, Southeast  
570 Asia, China, the Middle East, South America, India, North America, and the Southern Oceans.  
571 Figure 15 shows the regional cloudy-sky de-seasonalized ACA frequency for CALIOP daytime  
572 (blue) and nighttime (teal) analyses, along with linear regression lines (described earlier for the  
573 global analysis). Positive inter-annual variability in the cloudy-sky ACA frequency is found  
574 over the Middle East and India for both daytime and nighttime. In contrast, decreasing inter-  
575 annual variability in the cloudy-sky ACA frequency are found over China and South America for  
576 both daytime and nighttime. All other regions correspond with a negligible change in cloudy-  
577 sky ACA frequency during the study period.

578 A regional analysis of variation of cloud coverage over time is also conducted in order to  
579 investigate whether the observed increases in ACA frequency over time are a result of cloud  
580 coverage or aerosol loading. Positive AOD trends are observed from both regions (Zhang and  
581 Reid, 2010; Hsu et al., 2012). Cloud cover frequency exhibits an insignificant trend over India  
582 indicating that the ACA frequency increase may be due to aerosol loading increase over the  
583 region. The observed increase in cloudy-sky ACA frequency over the Middle-East, however,  
584 may be also due to the aerosols, as a slight decrease cloud coverage frequency is observed over  
585 time over this region. Inter-annual variability and its significance are also calculated for each of

586 the regional and global analyses shown in Table 3 using methods described by Weatherhead et  
587 al. (1998). As is apparent from Table 3, none of the trends are statistically significant (i.e., trend  
588 significance  $> 2$ ) with a confidence interval of 95%. Applying methods described in  
589 Weatherhead et al., (1998), we determine that an ACA data record spanning 37 and 36 years is  
590 needed to detect a 1 % yearly change with 95 % confidence, in cloudy-sky ACA frequency for  
591 day and nighttime, respectively.

592 Inter-annual variability for both ACAOD and cloud-free AOD are also calculated  
593 globally and for all regions shown in Table 3. Globally, the inter-annual variabilities of clear-sky  
594 AOD and ACAAOD are slightly positive, while the ACA frequency is negative during both day  
595 and night. Regions corresponding with a negative trend of all three parameters (ACA frequency,  
596 ACAOD, and clear-sky AOD) include: Southeast Asia (nighttime) and South America  
597 (nighttime). The Middle East (day and night) and India (day) regions exhibit positive trends for  
598 all three parameters. The remaining regions exhibit a combination of positive, negative or near-  
599 zero trends in all three parameters.

600

## 601 **8. Conclusions**

602 Using Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) layer products and  
603 collocated Ozone Monitoring Instrument (OMI) Aerosol products and Aqua Moderate  
604 Resolution Imaging Spectroradiometer (MODIS) cloud products data from June 2006 –  
605 December 2013, spatial distributions, including global and regional variabilities, of above cloud  
606 aerosol (ACA) events are studied and compared. Active-based profiling is considered an optimal  
607 means for identifying ACA occurrence. OMI identification is restricted to ultra-violet (UV)-  
608 absorbing ACA events (i.e., smoke), in contrast, through the Aerosol Index (AI) parameter.  
609 However, the relatively wide field-of-view of the paired OMI/MODIS datasets, in tandem,

610 provide greater data volume overall, which serves as a relatively well-characterized reference for  
611 comparing with CALIOP.

612 The primary findings of this study are:

- 613  
614 1. Baseline values for passive-based OMI AI and active-based CALIOP above-cloud  
615 aerosol optical depth (ACAOD) are established in order to distinguish background  
616 noise from signal due to significant ACA events such as dust outbreaks and  
617 biomass burning. The “noise floor” for OMI AI and CALIOP are applied to their  
618 respective data sets during processing. However, caution should be exercised  
619 when using these baselines, as they are an approximation and will vary depending  
620 on ancillary observational parameters for OMI and day versus nighttime  
621 sensitivity for CALIOP.
- 622 2. Despite fundamental differences in spatial and vertical samplings, as well as  
623 sensitivity to ACA aerosol types, both OMI- and CALIOP-based techniques  
624 broadly resolve consistent global distributions of cloudy-sky ACA frequency. For  
625 example, both capture ACA events over the Northwest Coast of Africa and the  
626 Arabian Peninsula during the December-May period, and over the North- and  
627 South-west Coast, as well as the Southeast Coast of Africa, the Arabian Peninsula  
628 and Arabian Sea during the June - November period. Still, discrepancies, as  
629 expected, are present. For example, daytime cloudy-sky ACA frequencies of up  
630 to 10% are found from CALIOP over Southeast Asia during the June-Nov. period  
631 while such ACA events are none existent using OMI-based method. Over North  
632 Africa, cloudy-sky ACA frequencies of around 20-30% are reported for both  
633 periods from the OMI-based method, yet such events are largely undetected by

634 the CALIOP-based method. We suspect that heavy dust plumes may be  
635 misidentified as clouds by the passive-based method, thus causing an unexpected  
636 rise in the passive-based derived cloudy-sky ACA frequency over that region.

637 3. The differences between the OMI- and CALIOP-based daytime cloudy-sky ACA  
638 frequencies are explored using a collocated OMI-MODIS-CALIOP dataset for the  
639 period of June 2006 – November 2008. Our analysis shows that the difference in  
640 cloud detectability between the MODIS and CALIOP instruments, as well as the  
641 QA flags applied, are the major reasons for the differences. Although the OMI-  
642 MODIS-based method is only sensitive to UV-absorbing aerosols and the  
643 CALIOP-based method is capable of detecting ACA events of all aerosol types,  
644 we did not find this to be one of the major reasons for the difference in ACA  
645 frequencies.

646 4. CALIOP nighttime data exhibit slightly larger distributions and a 10-20 % greater  
647 cloudy-sky ACA frequency annually in comparison with daytime. This may be  
648 due the subsidence of the planetary boundary layer at night, influencing  
649 frequencies of low-cloud formation, as well as the impact of higher signal-to-  
650 noise in CALIOP datasets for subsequent Level 2 analysis partly controlled for in  
651 our study by applying the noise floor. To the latter point, previous study has  
652 shown relative stability between day/night CALIOP aerosol products (Campbell  
653 et al., 2012). However, the implicit effect on the vertical distribution of aerosol  
654 occurrence was not specifically investigated. More detailed study is needed to  
655 reconcile this finding.

656 5. **We find** a near-zero negligible slope in the global CALIOP cloudy-sky and all-  
657 sky ACA frequencies. However, OMI-MODIS cloudy-sky daytime ACA  
658 frequencies show an increase of  $\sim 0.3\text{-}0.4\%$  / year since 2009, possibly due to a  
659 significant loss in the OMI data starting in 2009 mostly for nadir viewing pixels.  
660 Investigation of the relationship between OMI Aerosol Index (AI) and satellite  
661 viewing zenith angle, suggests a viewing angle dependency of OMI AI.  
662 Considering that OMI AI increases near the edge of the viewing swath, it is  
663 possible that the overall increase in ACA frequency is due to the significant loss  
664 of OMI AI data during later years of the study.

665 6. Changes in the cloudy-sky global ACA frequency and data counts ranging from  
666 2-4 % and 1-3 million, respectively, are found as a result of applying a variety of  
667 thresholds to the ACAOD and/or underlying cloud optical depth (COD) during  
668 sensitivity analysis. COD thresholds of 0.3 and 2.5 filter high cirrus clouds and  
669 non-contiguous low-level water clouds, respectively. CALIOP data are further  
670 reprocessed with no restriction to the ACAOD. Most threshold tests show a  
671 reduction in global ACA frequencies however those ACA events located over  
672 optically thick clouds ( $\text{COD} > 2.5$ ) show a near zero slope in the ACA frequency  
673 variability. However, a significant change over time to CALIOP global day or  
674 nighttime ACA frequency is not apparent.

675 7. Globally, clear-sky AOD and ACAOD temporal variations are slightly positive  
676 while cloudy-sky ACA frequency exhibits a slightly negative inter-annual  
677 variability in both the day and night times. Some select regions examined  
678 globally, selected for their relatively high ACA frequency overall, exhibit a

679 consistent inter-annual variability in all three parameters. For example,  
680 statistically significant increases in clear sky AOD are found over India and  
681 Middle-East from various passive based analysis (e.g. Zhang and Reid, 2010).  
682 Increasing in cloudy-sky ACA frequencies are also found for the two regions for  
683 the study period of 2006-2013. Other regions exhibit agreement between some,  
684 but not all, parameters. However, neither the regional or global trends of any of  
685 the three parameters are statistically significant. An ACA data record spanning at  
686 least 30 years is needed in order to report a 10% per decade change in ACA  
687 frequency with 95% confidence.

688 This study confirms that significant (i.e., resolvable with the techniques applied) ACA  
689 events occur with a frequency of 1-8% globally and as high as 30-50 %, regionally, over some of  
690 the most ACA-abundant regions. The two complementary techniques applied to locate ACA  
691 events and derive global and regional distributions and both exhibit strengths and weaknesses.  
692 This study shows that, when used simultaneously, combined passive/active analysis can help  
693 present a more comprehensive analysis of ACA than a single-sensor analysis alone. However,  
694 the analysis strongly reinforces the use of active-based lidar profiling for distinguishing aerosol  
695 presence that perturbs passive-based column-integrated radiative parameters. The vertical  
696 distribution and optical properties of aerosol and cloud layers are fundamental to accurate  
697 column radiative closure. The effects cloud-aerosol overlap can exhibit on cloud and aerosol  
698 property retrieval techniques demands some coordinated active/passive observation for ensuring  
699 clarity and limiting bias in top-of-atmosphere retrievals.

700 Due to the extensive spatial coverage and consistency of retrieved datasets from space-  
701 borne instruments, trend analyses, and the need for consistent multi-sensor profiling, should

702 become primary motivating factors behind mission design and life expectancy in orbit. Our  
703 analysis shows that in a few decades, proper analysis of ACA trends are possible through  
704 continuation of a CALIOP/OMI-like paradigm. Ultimately, this work, paired with Alfaro-  
705 Contreras (2014) and others, have broadly conceptualized the ACA problem globally. Past  
706 studies have shown that ACA events represent a fundamental climate phenomenon on a global  
707 scale (Peters et al. 2011), thus ACA requires specific long term monitoring. Trend analysis,  
708 then, will help ultimately distinguish this attribute, and thus whether or not ACA is simply noise  
709 or a radiatively-significant process that is sensitive to changes in land-use globally and a  
710 fluctuating frequency and distribution of elevated aerosol particles over time. Future satellite  
711 mission designs should emphasize extending the life of these instruments for application to  
712 environmental parameter inter-annual variability studies.

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## **Figure Captions**

**Figure 1.** (A-H) Multi-year (2006-2013) CALIOP-derived daytime global cloudy-sky ACA frequency applying different CALIOP AODs as the threshold between background and significant aerosol loading. The CALIOP AOD are binned into  $2.5^\circ \times 2.5^\circ$  bins derived using the CALIOP cloud and layer data sets. CALIOP AOD baseline thresholds of 0, 0.010, 0.015 and 0.020 are applied to Figs. 1A, 1B, 1C and 1D respectively for the Dec.-May period. Figures 1E-1H show the similar results as Fig. 1A-1D but for the June-Nov. period.

**Figure 2.** (A) Pairwise comparison between collocated OMI and CALIOP observations of above-cloud AI and AOD, respectively, as a function of the underlying MODIS cloud optical depth (COD). CALIOP AOD are averaged into OMI AI bins of 0.1. (B-I) Multi-year (2006-2013) daytime global cloudy-sky ACA frequency applying several different OMI AIs as the threshold between background and significant aerosol loading. The OMI AIs are binned into  $1^\circ \times 1^\circ$  bins derived from the MODIS-OMI collocated data set. OMI AI baseline thresholds of 0.7, 0.8, 0.9 and 1.0 are applied to Figs. 2B, 2C, 2D and 2E respectively for the Dec.-May period. Figures 2F-2I depict the same information as Figs. 2B-2E for the June-Nov. period. ACA frequencies less than 5 % are shown in white.

**Figure 3.** (A) Seven year (December 2006- May 2013) daytime cloudy-sky frequency of occurrence of aerosol above-cloud events during December through May defined from OMI (ratio of totally cloudy MODIS pixels with AI greater than 1.0 to the number of totally cloudy MODIS pixels). (B) Day-time cloudy-sky frequency of occurrence of ACA events over cloudy skies from CALIOP (ratio of CALIOP pixels with  $\text{CALIOP AOD}_{\text{above cloud}} > 0.015$  to the number of CALIOP pixels with column integrated  $\text{COD} > 0$ ) for the

same temporal domain as Fig. 3A. (C) Night-time cloudy-sky frequency of occurrence defined similar to the daytime frequency from Fig. 3B. (D-F) Shows the same information as Figs. 3A-3C during June 2006-November 2013. Figures 3G-3H depict the ACA frequency ratio defined as the OMI-MODIS daytime cloudy-sky frequency divided by the CALIOP derived daytime cloudy-sky frequency for the December to May and June to November period, respectively. Figures 3I-3J depict the difference in cloudy-sky frequency used to construct the frequency ratio plots (3G and 3H) for the same temporal ranges. The red boxes show the areas selected for regional studies. Only OMI and CALIOP bins with frequency of 5% or higher are shown in this analysis.

**Figure 4.** (a) Aqua MODIS true color image on July 8, 2007 over the Saharan Desert in Northern Africa (b) The same domain as Figure 4a but for Cloud Fraction from the MODIS MYD06 data. (c) Similar to Figure 3a but for OMI Aerosol Index (AI). Fig. 4c is created by averaging all OMI observations into  $0.25^\circ \times 0.25^\circ$  grids over the region shown. (d) The vertical profile for CALIOP backscatter, where the instrument overpass is superimposed on Figure 4a. Also, the CALIOP track shown in Fig. 4a is constructed by highlighting those MODIS observations that are collocated with CALIOP observations.

**Figure 5.** (A) Multi-year (2006-2013) daytime AI averaged into  $1.0^\circ \times 1.0^\circ$  bins constructed from collocated MODIS and OMI AI over strictly MODIS cloudy scenes during December through May. The averaged OMI AI is neglected below 1.0 in accordance with the AI ground floor determined in Fig. 2. (B) Multi-year (2006-2013) daytime ACAOD averaged into  $2.5^\circ \times 2.5^\circ$  bins derived from CALIOP cloud and aerosol layer products. Averaged CALIOP ACAOD below 0.015 are considered below the noise floor for the study and thus are not shown. (C) Shows the CALIOP ACAOD similar to Fig. 5B except

for night-time observations. (D-F) Shows the same information as Figs. 5A-5C during the summer and fall months (June-November).

**Figure 6.** Multi-year (June 2006 - November 2013) frequency of occurrence of low-level clouds defined by CALIOP as the ratio of pixels with COD greater than 0 with cloud-top height < 3km to the total number of CALIOP scenes within the current  $2.5^\circ \times 2.5^\circ$  bin for (A) December to May during daytime observations, (B) December to May of night-time observations, (C) Daytime frequency of occurrence of low-level cloud decks defined similar to Fig 6A. during the June-November time frame and (D). Nighttime frequency of occurrence of low-level cloud decks for the same time frame as Fig. 6C. Figures 6E and 6F depict the night to daytime frequency ratio for the December to May and June to November periods, respectively.

**Figure 7.** Multi-year (2006-2013)  $2.5^\circ \times 2.5^\circ$  averaged CALIOP day-time AOD for (A) December through May over completely cloud free scenes derived from CALIOP cloud and aerosol layer daytime analysis, (B) Nighttime analysis during the December to May period, (C) Daytime analysis for the June to November period and (D) nighttime analysis for the June to November period. Only scenes which contained an averaged AOD > 0.2 with a column COD = 0 were used in the analysis.

**Figure 8.** (A) Two-and-a-half-year (June 2006 – November 2008) daytime CALIOP cloudy-sky ACA frequency during the December through May period, using the collocated OMI-MODIS-CALIOP dataset (defined in Table 1). (B) The same as Fig. 8A, however for the all-sky CALIOP ACA frequency. (C) CALIOP cloudy sky frequency, which is defined as the number of collocated CALIOP observations with COD > 0 over the total number of collocated CALIOP observations. (D-F) Similar to Figs. 8a-8c but using the OMI-

MODIS-based method (defined in Table 1). It should be noted that the cloudy sky frequency from the OMI-MODIS technique is defined as the number of observations with cloud fraction equals to one divided by the total number of observations (defined in Table 1). (G-J) Depict the same information as 8A-F except for the June – November (2006-2008) period.

**Figure 9.** (A) Two-and-a-half-year (June 2006 – November 2008) daytime CALIOP cloudy-sky frequency during the December through May period, using the collocated OMI-MODIS-CALIOP dataset with the application of the most lenient cloud QA. (B-C) Depict the same information as Fig. 9A, however now using intermediate and strict cloud QA settings, respectively. (D) Depicts the all-sky frequency using the same data set as Fig. 9A, now using lenient cloud and aerosol QAs. (E-F) Depict the same information as Fig. 9D varying the cloud QA to intermediate and strict. (G-I) Similar to Figs. D-F but holding the lenient cloud QA while varying the aerosol QA from lenient to intermediate and strict, respectively. (J-R) Depict the same information as Figs. 9A-I for the June to November period (2006 – 2008).

**Figure 10.** Monthly-averaged global ACA frequencies derived using the OMI-MODIS based method (green) as well as the CALIOP-based method as described in the text. The corresponding baseline thresholds are applied to both CALIOP and OMI data. Dashed lines represent monthly variations in ACA frequencies and the solid lines represent the yearly ACA frequency trends: OMI daytime cloudy-sky frequency is shown in green, CALIOP nighttime cloudy-sky frequency is orange, CALIOP nighttime all-sky frequency is purple, CALIOP daytime cloudy-sky frequency is red and CALIOP daytime all-sky frequency is blue.



**Figure 11.** (A) A single swath from the OMI instrument over northern Africa on August 1, 2007 before the significant data loss reported in all OMI aerosol products. (B) A single OMI AI swath over the same region as Fig. 11A on June 1, 2009 which is affected by the significant data loss.

**Figure 12.** The OMI AI as a function of the sensor's viewing zenith angle (VZA). All OMI AI data over the course of a year (2007) were binned into  $1^\circ$  VZA increments. The red vertical bars represent the 95% confidence interval for each  $1^\circ$  bin.

**Figure 13.** Monthly-averaged over ocean clear-sky AODs derived from collocated CALIOP and AERONET data. CALIOP retrievals within  $0.3^\circ$  latitude and longitude and  $\pm 30$  minutes of the corresponding AERONET station and observation are considered collocated. AERONET and CALIOP AODs above 0.2 and 0.6, respectively, are not included in order to avoid high aerosol loading cases and exclude noisy CALIOP data.

**Figure 14.** Monthly-averaged global CALIOP cloudy-sky frequencies after applying several different threshold techniques to both day and night time data. The solid lines show the daytime scenario for each respective case while the dotted lines show the nighttime observations for each case.

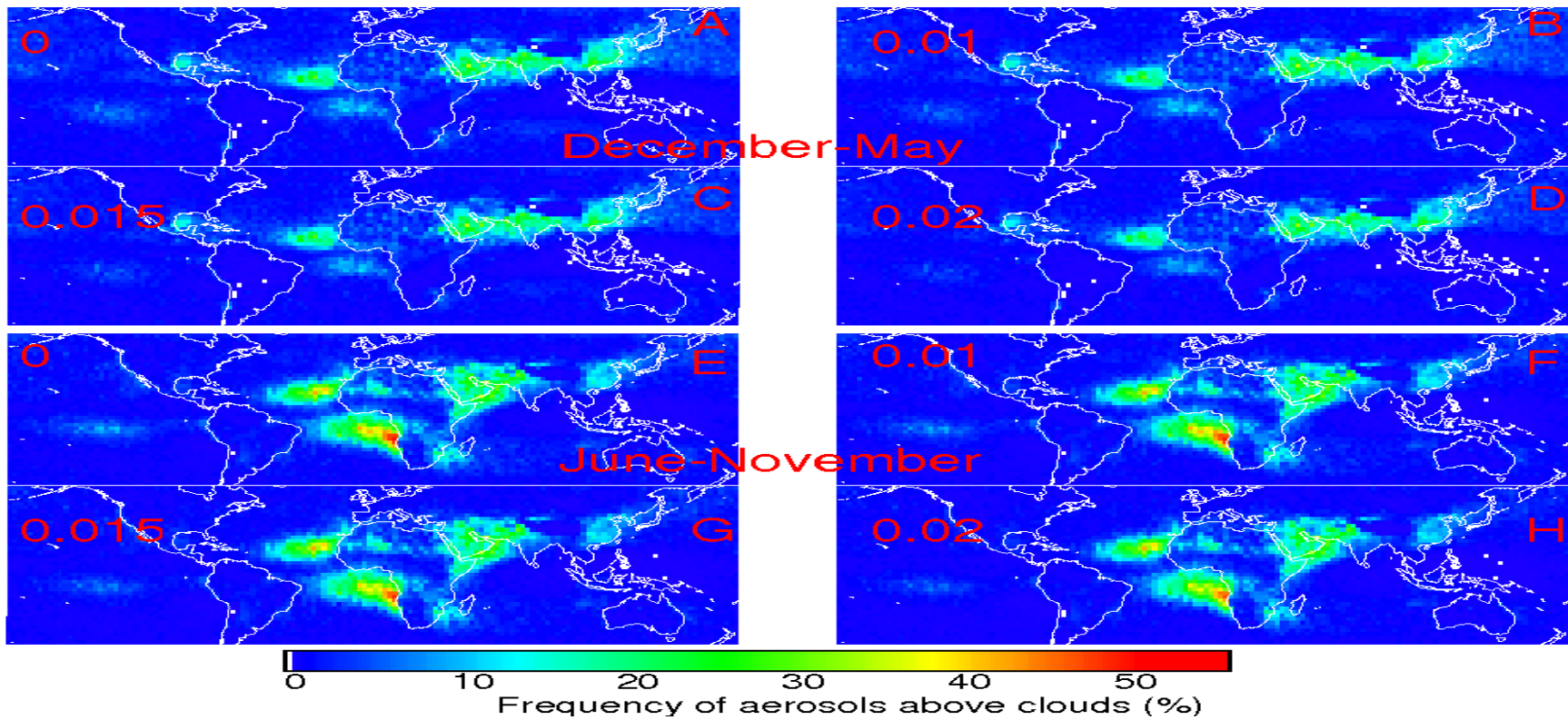
**Figure 15.** The de-seasonalized monthly- and regionally-averaged cloudy-sky frequency of ACA occurrences for the nine different regions outlined in Fig. 3 and explained in Table 3. The dashed lines show the monthly frequency over the regions and the solid lines show the trend lines computed for each region with the x-axis representing the year of the study. The CALIOP nighttime analysis is shown in aqua marine and the day-time analysis is shown in dark blue.

## **Table Captions**

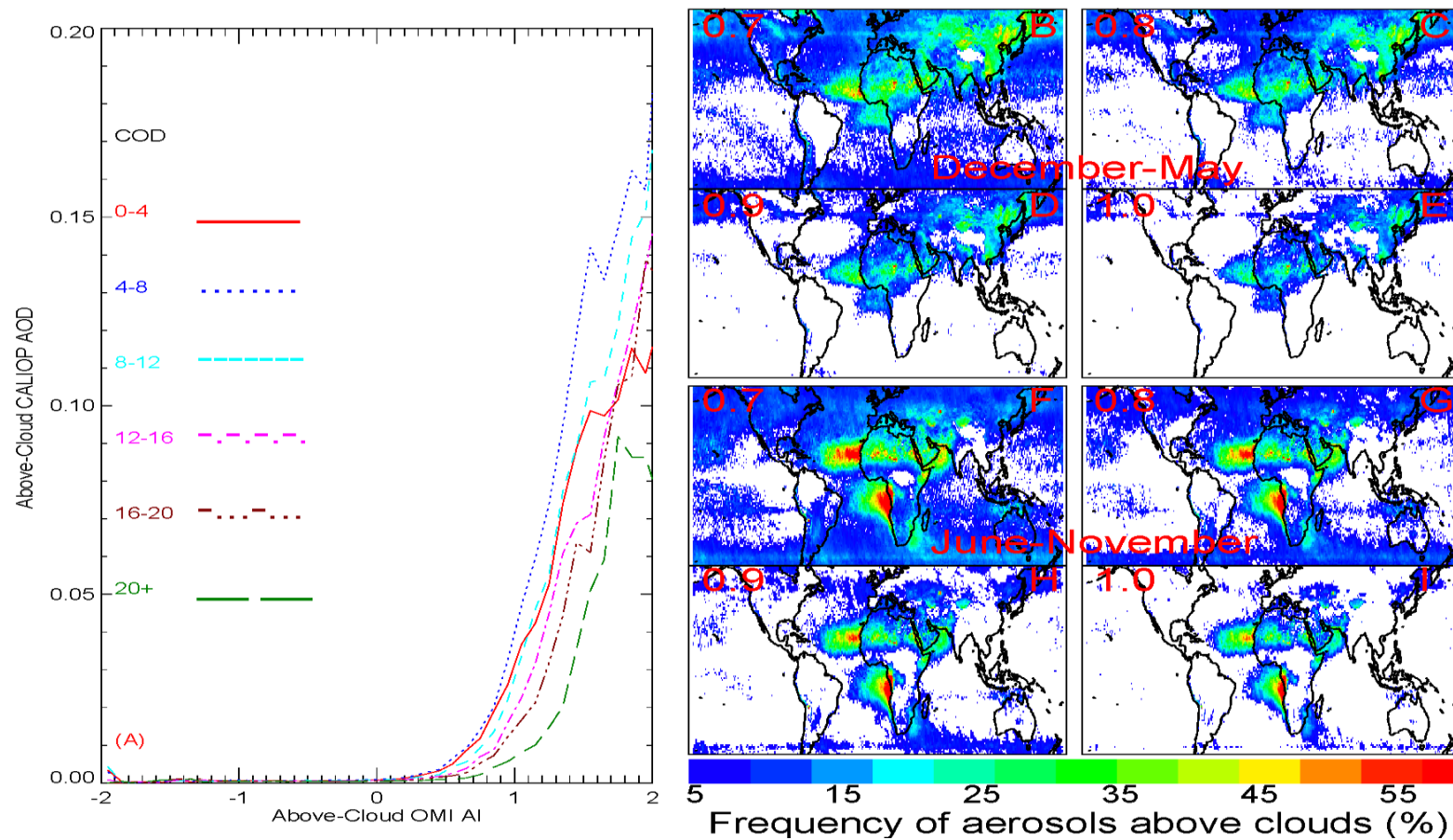
**Table 1.** Various definitions of frequency of above-cloud aerosols (ACA) used throughout the study.

**Table 2.** Global cloudy-sky relative frequency and data counts for the sensitivity test carried out in Sect. 5. Aerosol and cloud layers retrieved with ‘intermediate’ or ‘strict’ QA metrics are considered in this analysis. A total of five different threshold tests are applied to both day and nighttime CALIOP cloud and aerosol layer products.

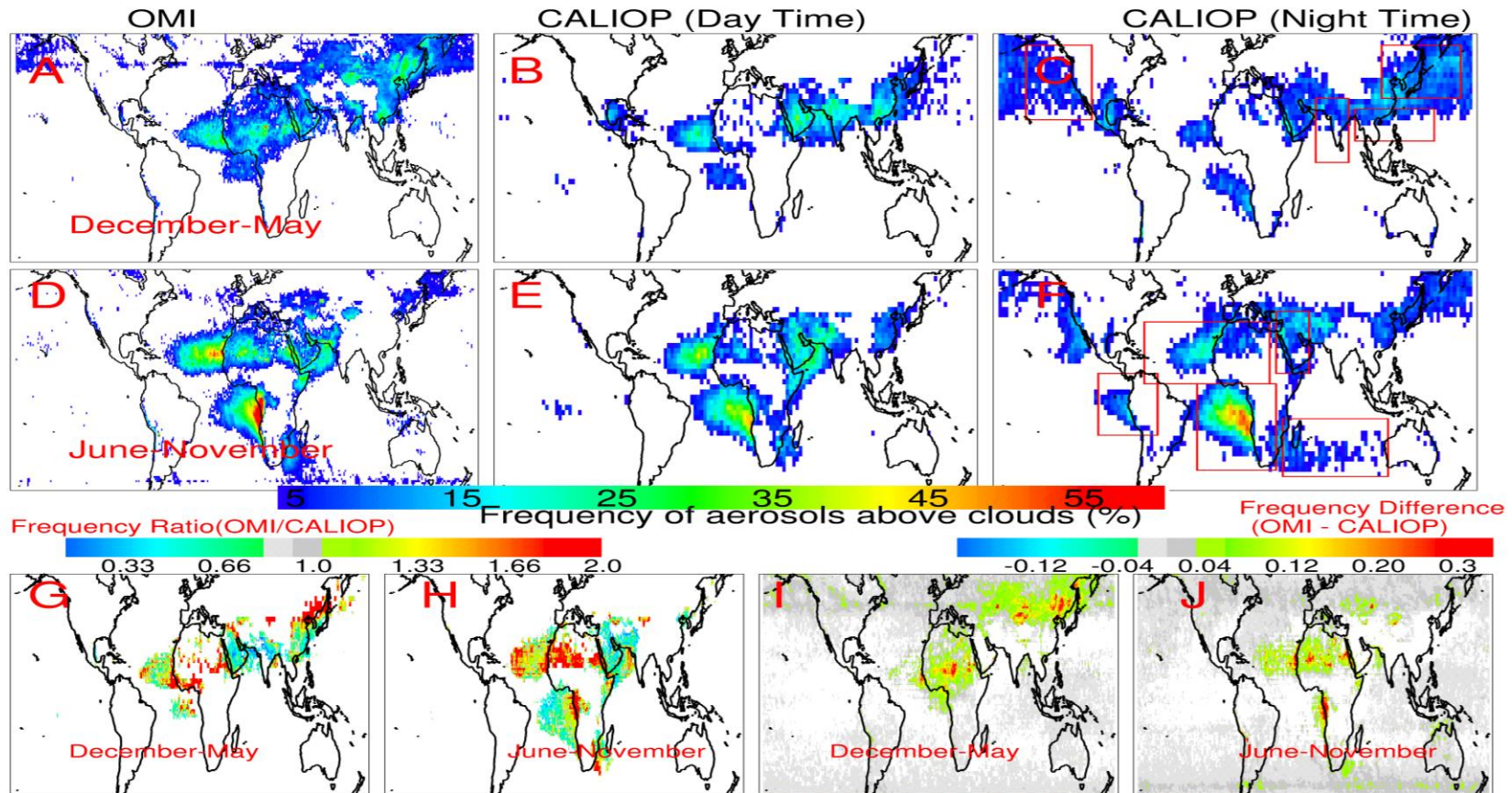
**Table 3** Seven and a half year above-cloud aerosol cloudy-sky frequency, ACAOD and clear-sky AOD inter-annual variability analysis for the selected target regions. Aerosol and cloud layers retrieved with ‘intermediate’ or ‘strict’ QA metrics are considered in this analysis. Yearly variation for the entire globe is also included. For each region, inter-annual variability (frequency change per year) for the three parameters, the ACA cloudy-sky frequency, ACAOD and clear-sky AOD values are reported. Note that the inter-annual variability for clear sky AODs is estimated using 100% cloud free data from the CALIOP cloud and aerosol layer products.



**Figure 1.** (A-H) Multi-year (2006-2013) CALIOP-derived daytime global cloudy-sky ACA frequency applying different CALIOP AODs as the threshold between background and significant aerosol loading. The CALIOP AOD are binned into  $2.5^\circ \times 2.5^\circ$  bins derived using the CALIOP cloud and layer data sets. CALIOP AOD baseline thresholds of 0, 0.010, 0.015 and 0.020 are applied to Figs. 1A, 1B, 1C and 1D respectively for the Dec.-May period. Figures 1E-1H show the similar results as Fig. 1A-1D but for the June-Nov. period.



**Figure 2.** (A) Pairwise comparison between collocated OMI and CALIOP observations of above-cloud AI and AOD, respectively, as a function of the underlying MODIS cloud optical depth (COD). CALIOP AOD are averaged into OMI AI bins of 0.1. (B-I) Multi-year (2006-2013) daytime global cloudy-sky ACA frequency applying several different OMI AIs as the threshold between background and significant aerosol loading. The OMI AIs are binned into  $1^\circ \times 1^\circ$  bins derived from the MODIS-OMI collocated data set. OMI AI baseline thresholds of 0.7, 0.8, 0.9 and 1.0 are applied to Figs. 2B, 2C, 2D and 2E respectively for the Dec.-May period. Figures 2F-2I depict the same information as Figs. 2B-2E for the June-Nov. period. ACA frequencies less than 5 % are shown in white.



**Figure 3.** (A) Seven year (December 2006- May 2013) daytime cloudy-sky frequency of occurrence of aerosol above-cloud events during December through May defined from OMI (ratio of totally cloudy MODIS pixels with AI greater than 1.0 to the number of totally cloudy MODIS pixels). (B) Day-time cloudy-sky frequency of occurrence of ACA events over cloudy skies from CALIOP (ratio of CALIOP pixels with CALIOP  $AOD_{above\ cloud} > 0.015$  to the number of CALIOP pixels with column integrated COD > 0) for the same temporal domain as Fig. 3A. (C) Night-time cloudy-sky frequency of occurrence defined similar to the daytime frequency from Fig. 3B. (D-F) Shows the same information as Figs. 3A-3C during June 2006-November 2013. Figures 3G-3H depict the ACA frequency ratio defined as the OMI-MODIS daytime cloudy-sky frequency divided by the CALIOP derived daytime cloudy-sky frequency for the December to May and June to November period, respectively. Figures 3I-3J depict the difference in cloudy-sky frequency used to construct the frequency ratio plots (3G and 3H) for the same temporal ranges. The red boxes show the areas selected for regional studies. Only OMI and CALIOP bins with frequency of 5% or higher are shown in this analysis.

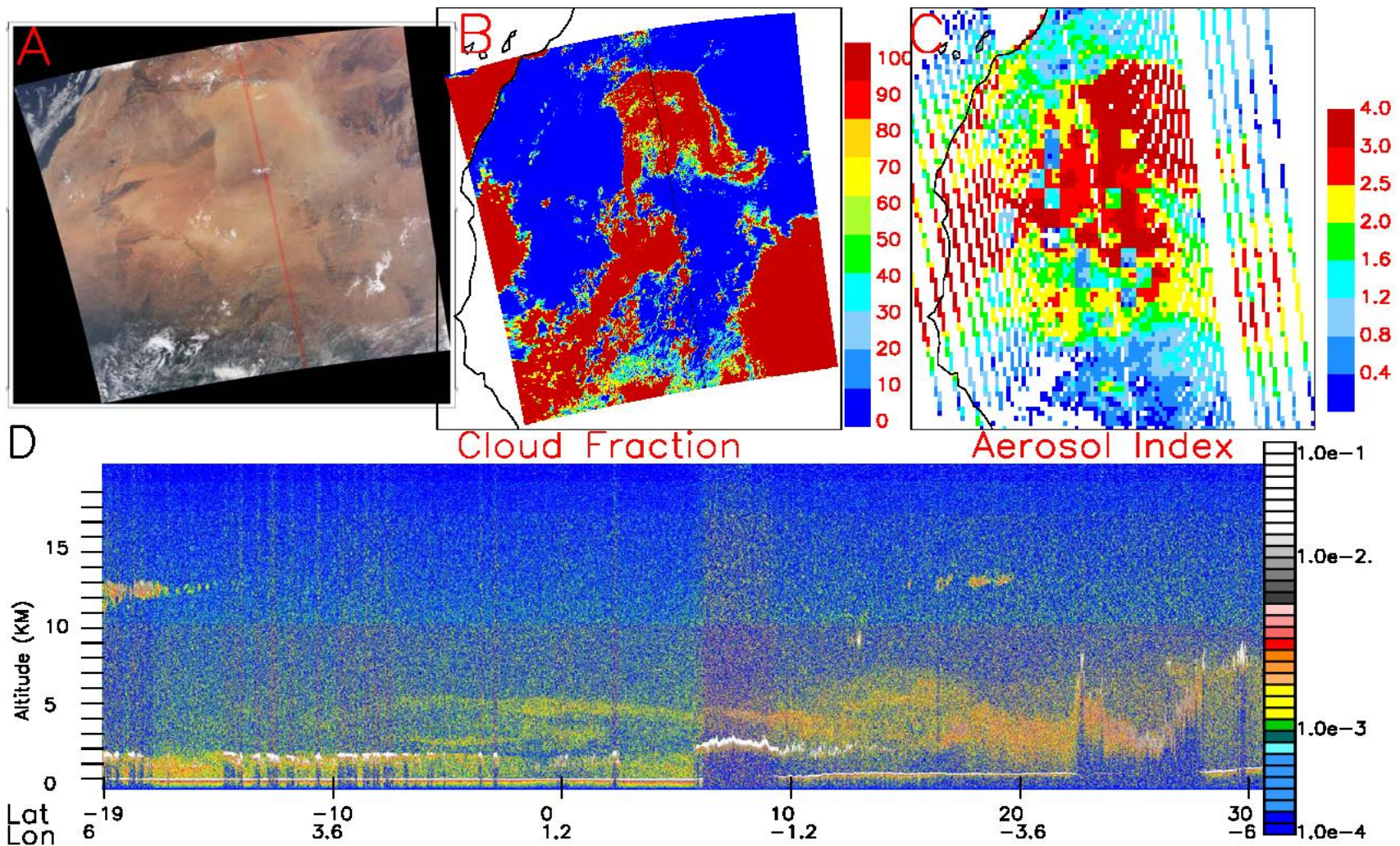
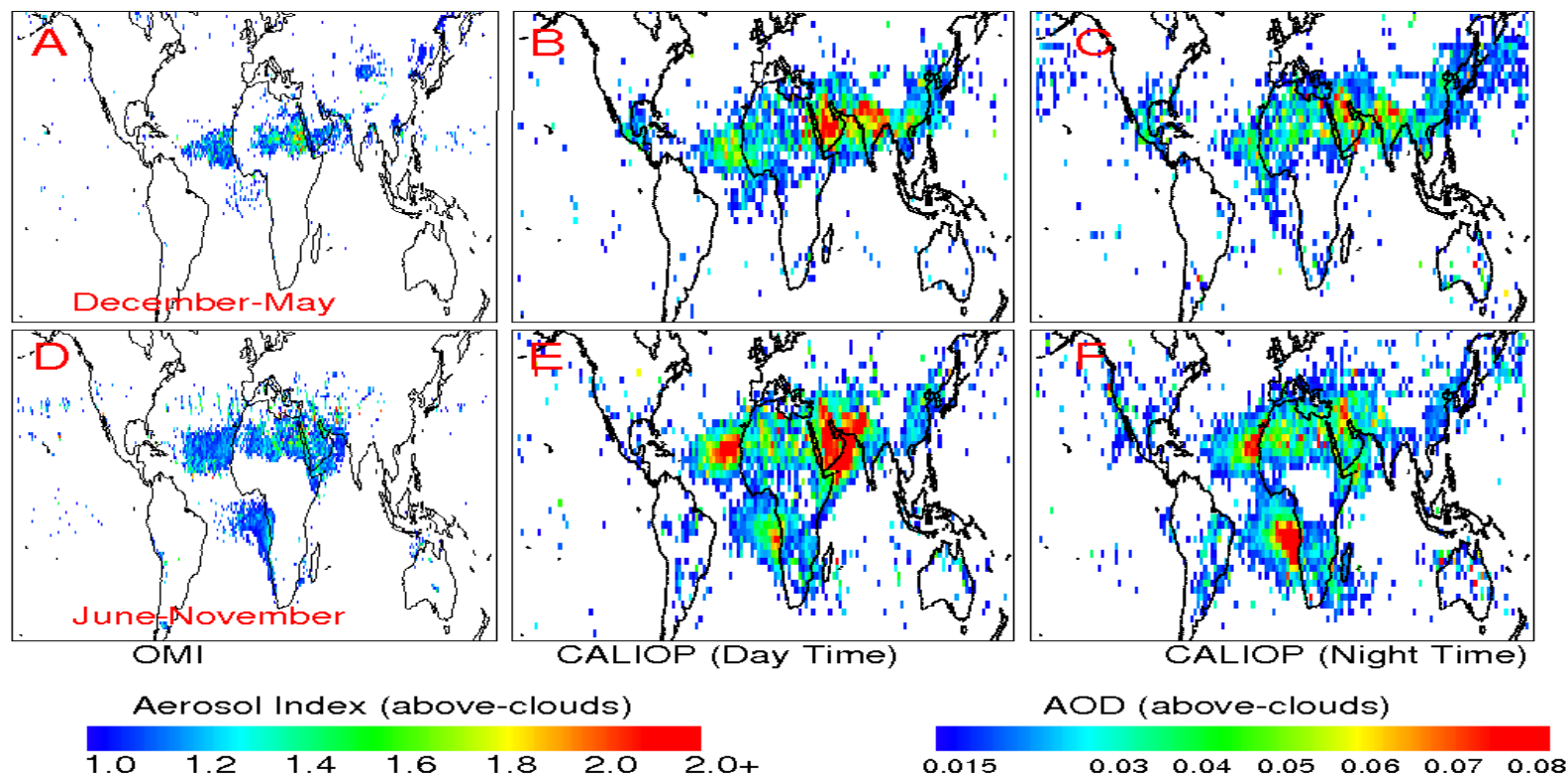
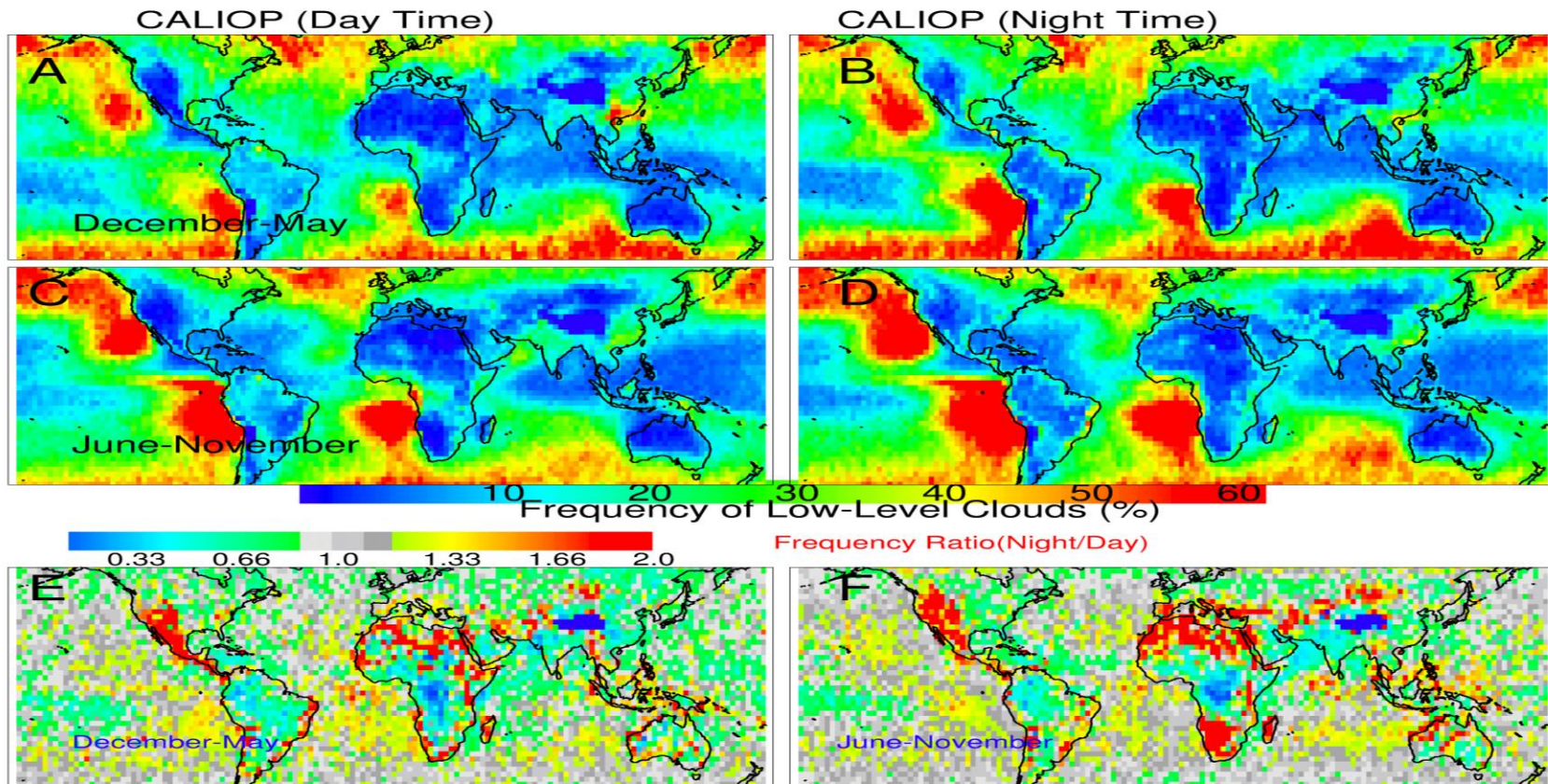


Figure 4. (a) Aqua MODIS true color image on 8 July 2007 over the Saharan Desert in Northern Africa (b) The same domain as Figure 4a but for Cloud Fraction from the MODIS MYD06 data. (c) Similar to Figure 3a but for OMI Aerosol Index (AI). Fig. 4c is created by averaging all OMI observations into  $0.25^\circ \times 0.25^\circ$  grids over the region shown. (d) The vertical profile for CALIOP backscatter, where the instrument overpass is superimposed on Figure 4a. Also, the CALIOP track shown in Fig. 4a is constructed by highlighting those MODIS observations that are collocated with CALIOP observations.

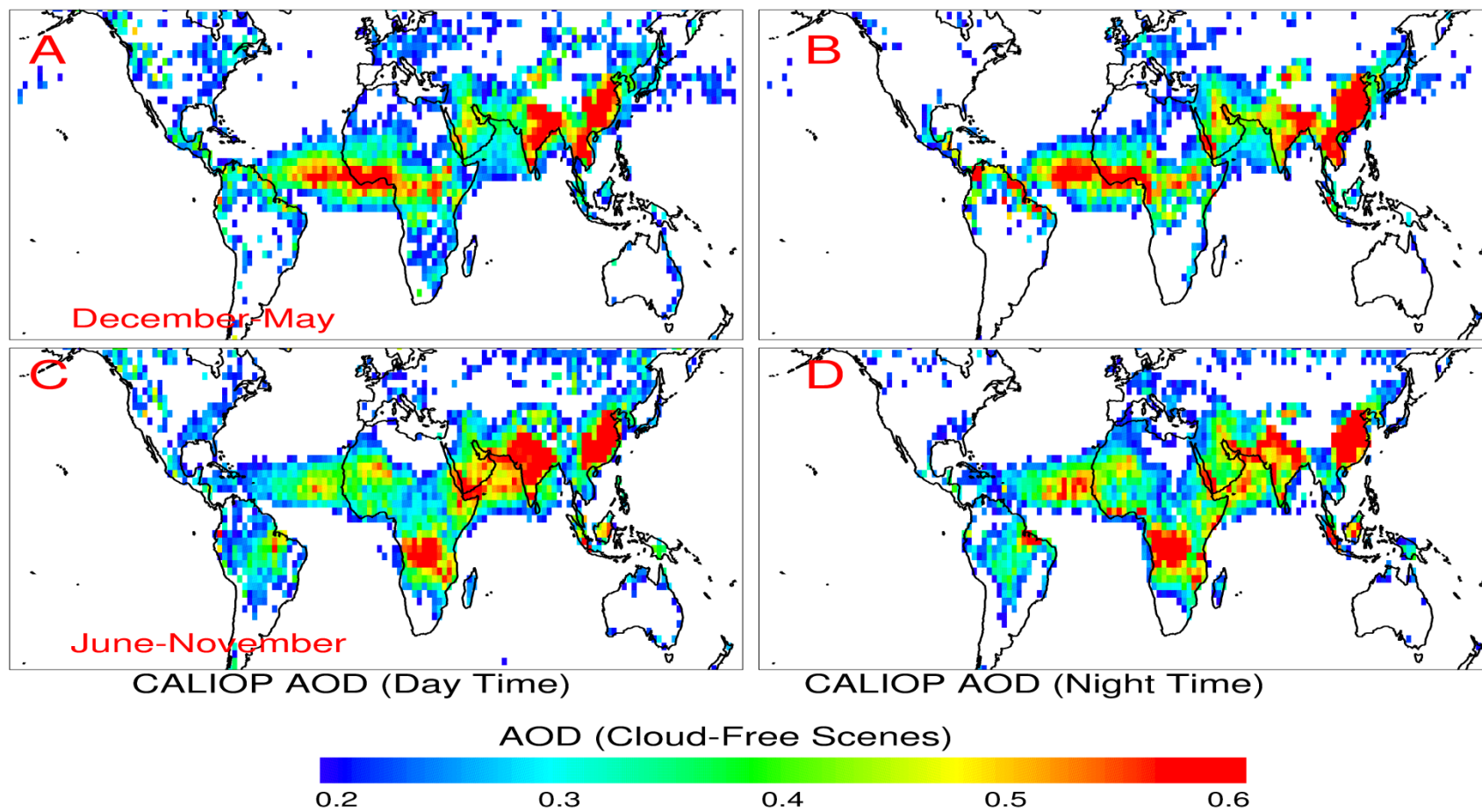


**Figure 5.** (A) Multi-year (2006-2013) daytime AI averaged into  $1.0^\circ \times 1.0^\circ$  bins constructed from collocated MODIS and OMI AI over strictly MODIS cloudy scenes during December through May. The averaged OMI AI is neglected below 1.0 in accordance with the AI ground floor determined in Fig. 2. (B) Multi-year (2006-2013) daytime ACAOD averaged into  $2.5^\circ \times 2.5^\circ$  bins derived from CALIOP cloud and aerosol layer products. Averaged CALIOP ACAOD below 0.015 are considered below the noise floor for the study and thus are not shown. (C) Shows the CALIOP ACAOD similar to Fig. 5B except for night-time observations. (D-F) Shows the same information as Figs. 5A-5C during the summer and fall months (June-November).

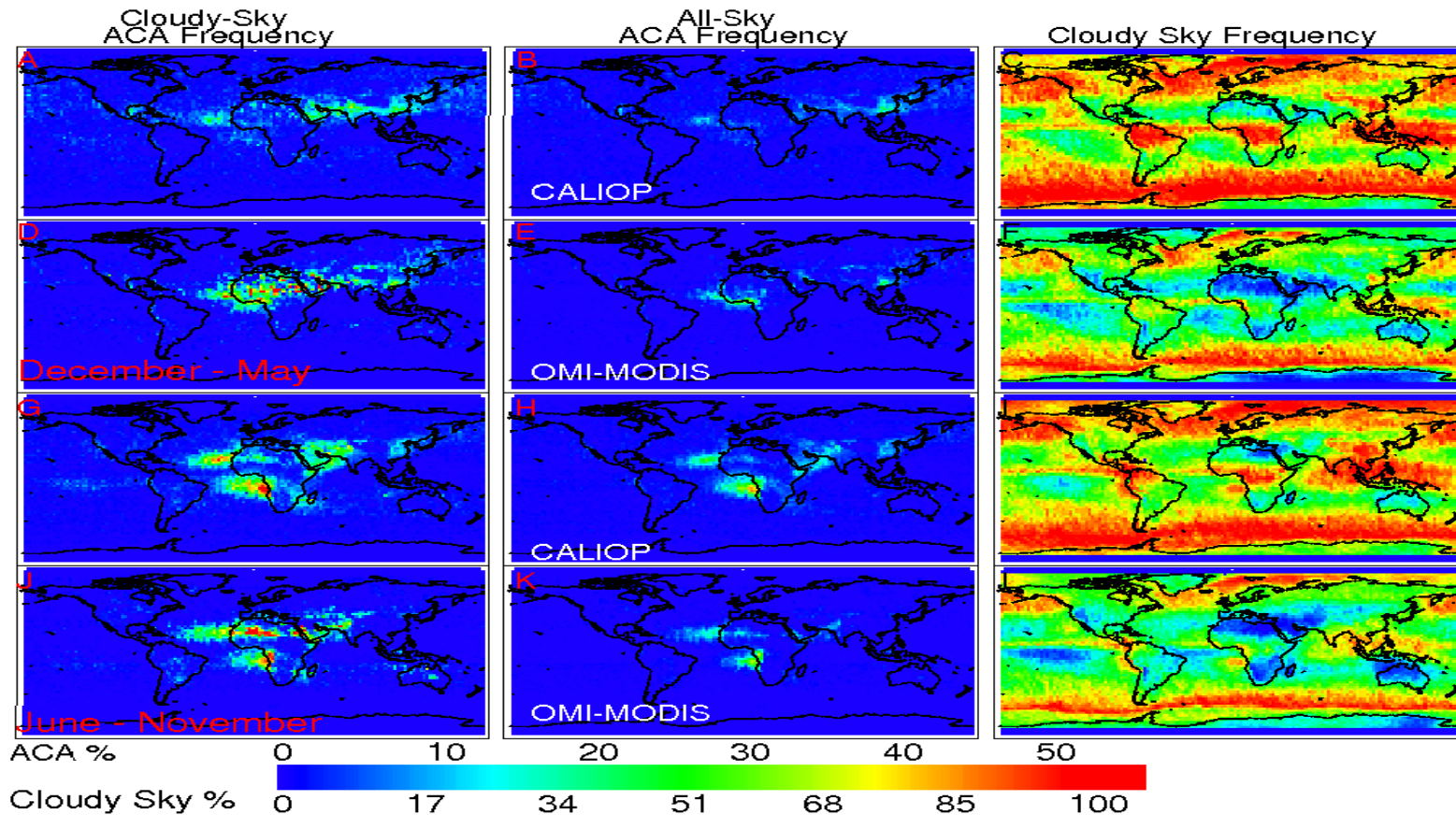


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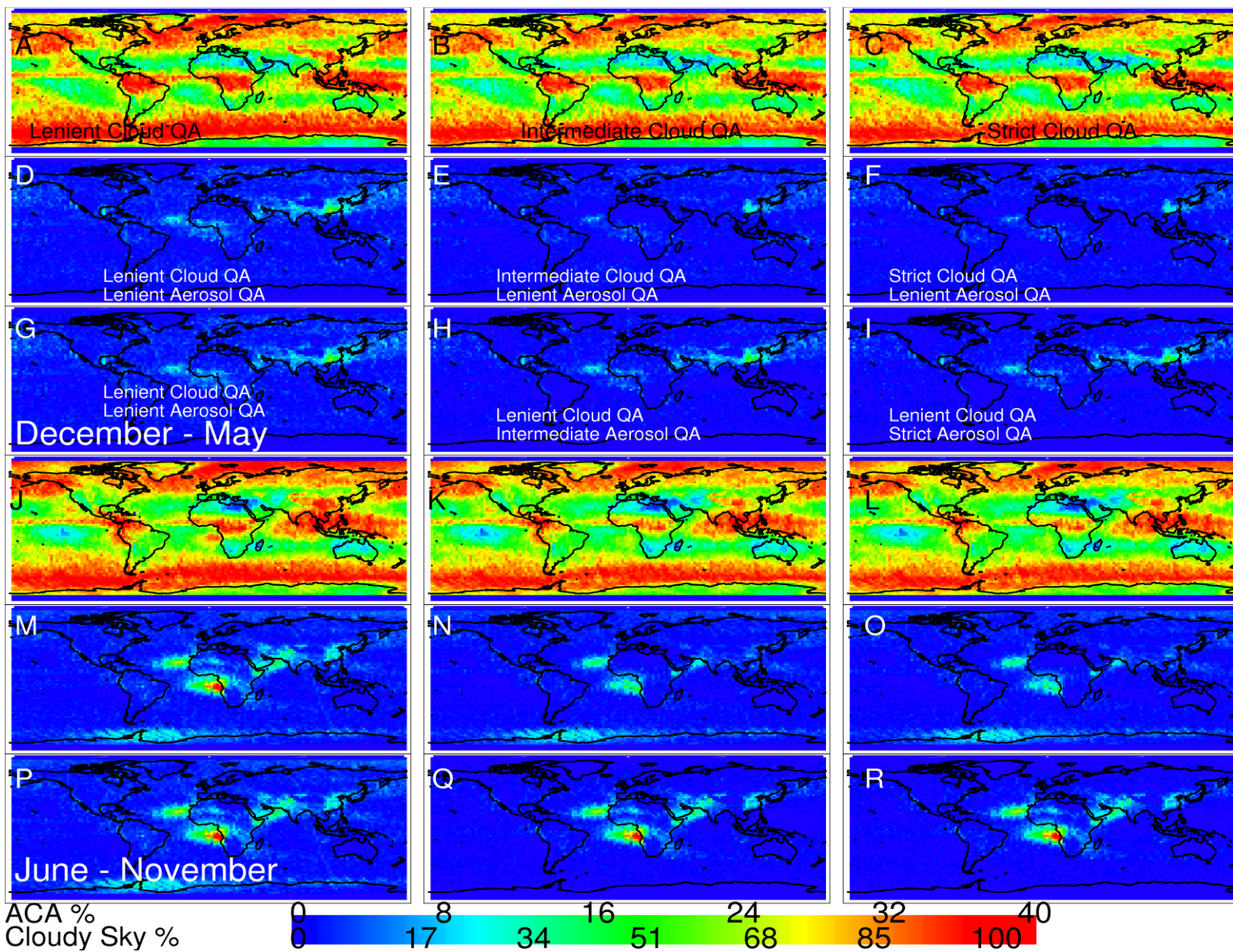




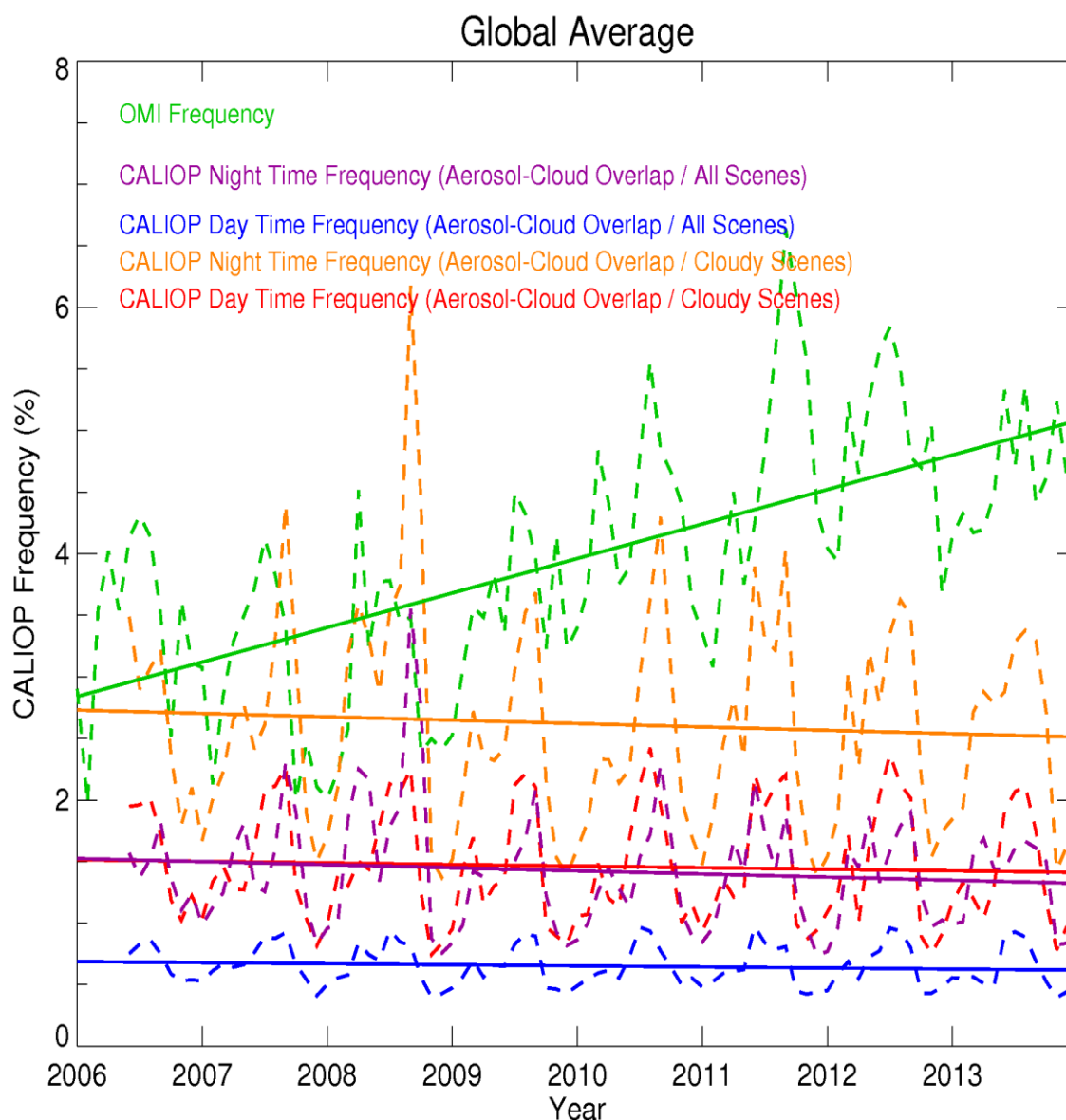
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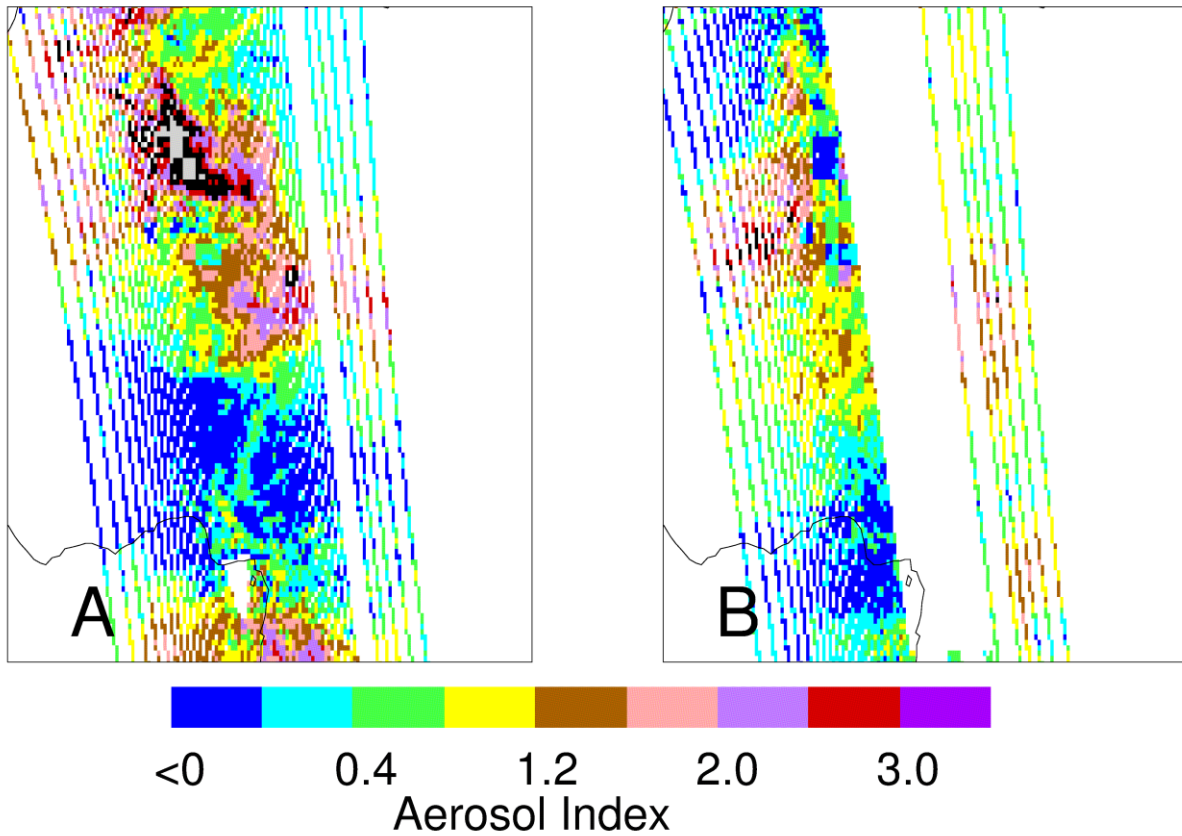
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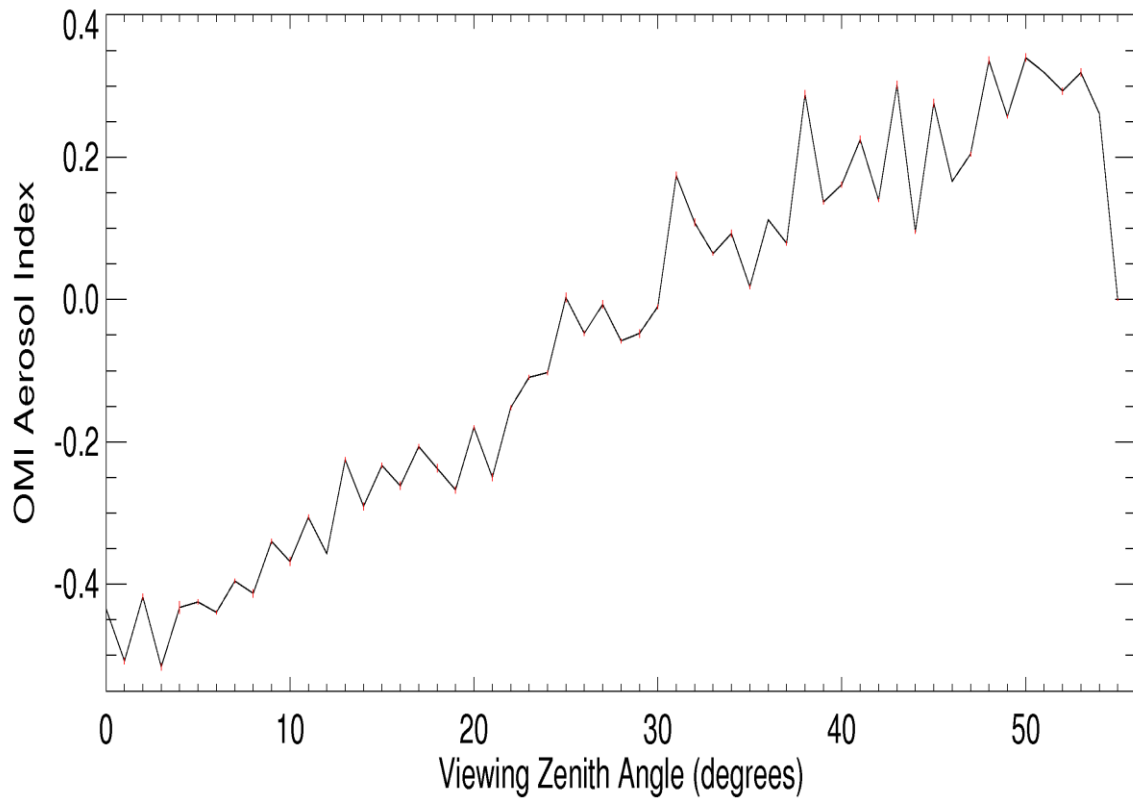
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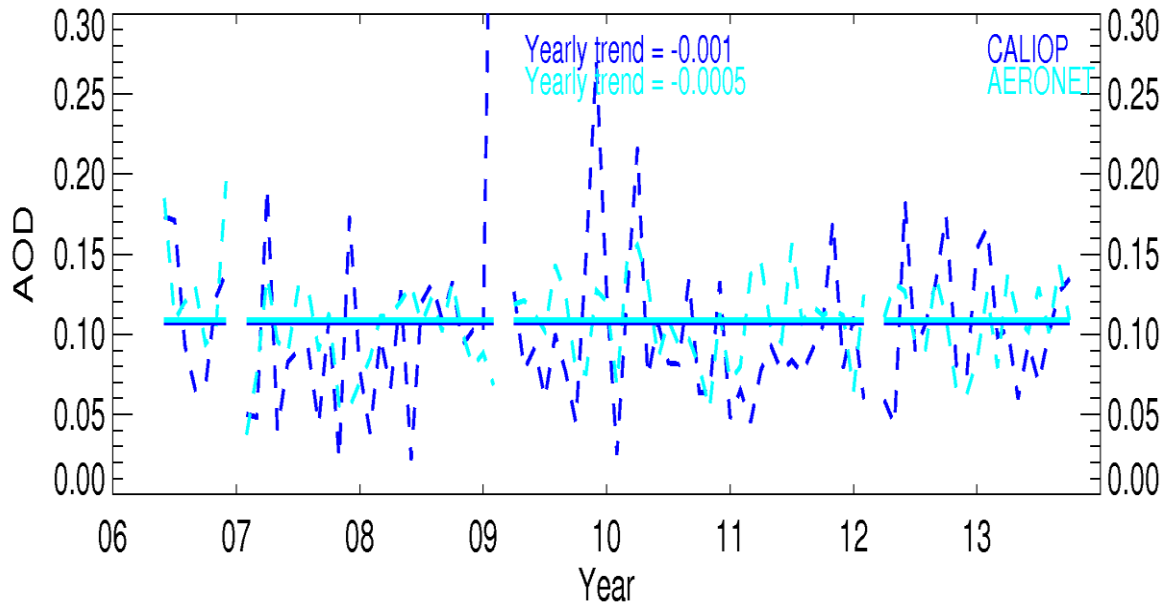
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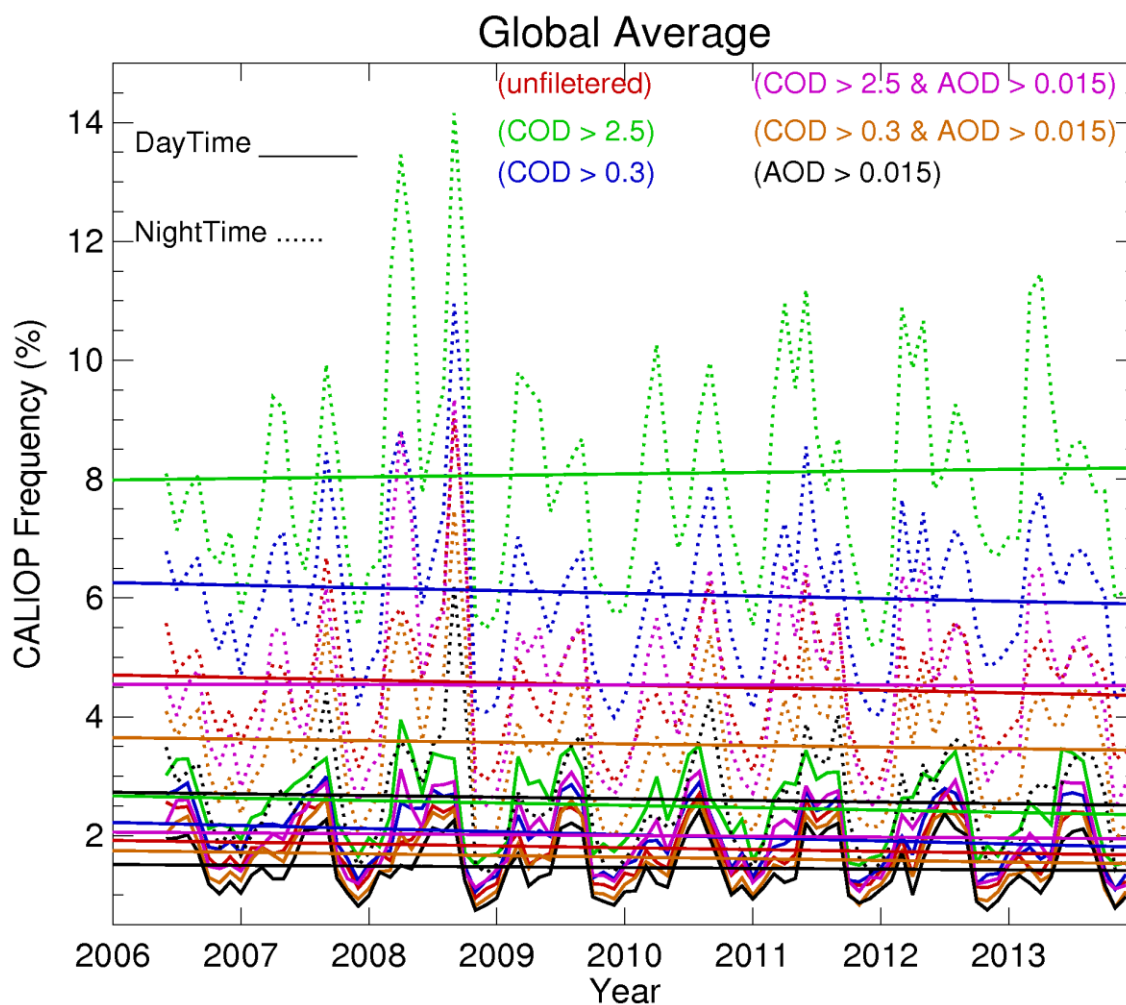


**Figure 12.** The OMI AI as a function of the sensor's viewing zenith angle (VZA). All OMI AI data over the course of a year (2007) were binned into 1° VZA increments. The red vertical bars represent the 95% confidence interval for each 1° bin.

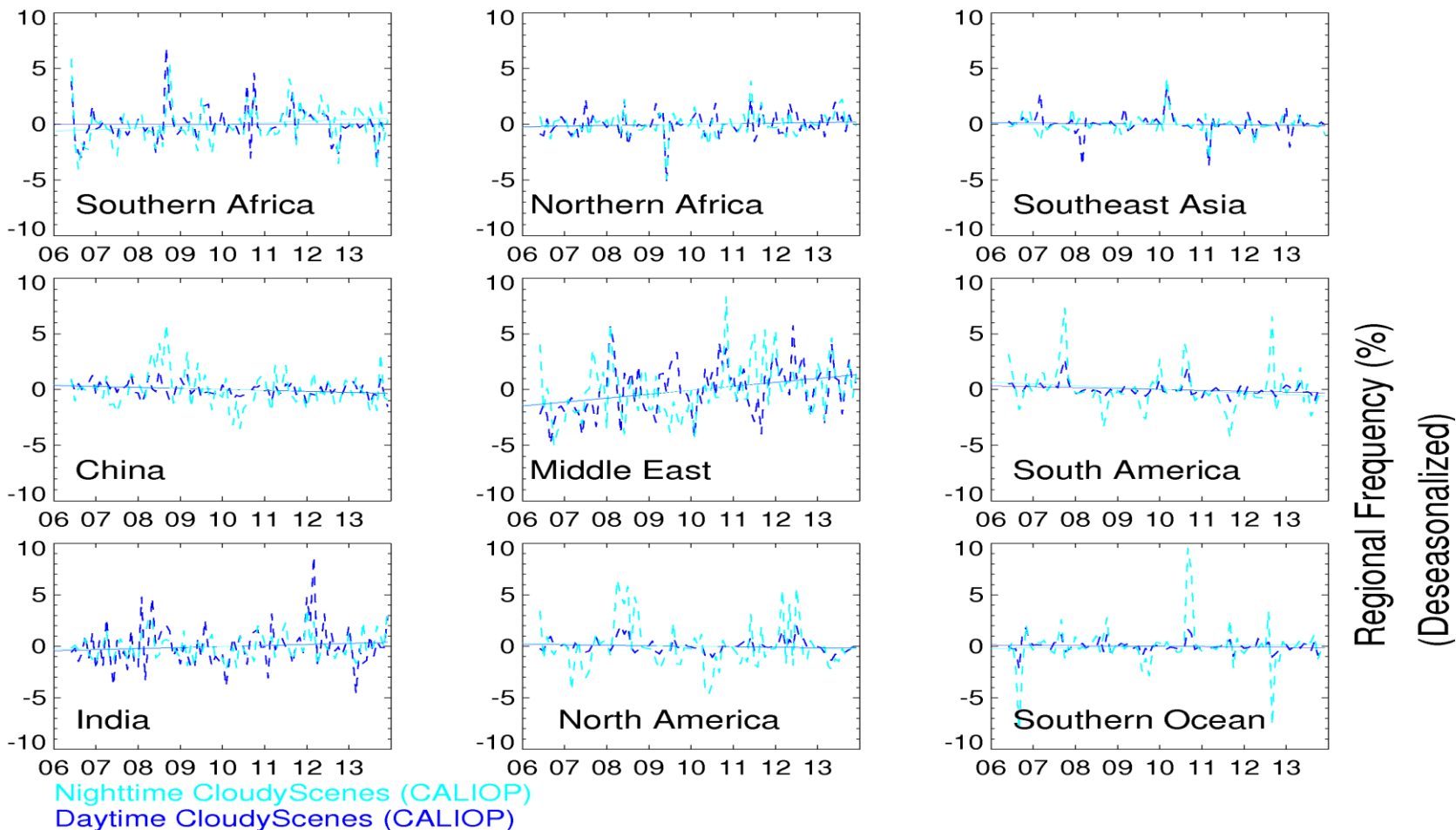


**Figure 13.** Monthly-averaged over ocean clear-sky AODs derived from collocated CALIOP and AERONET data. CALIOP retrievals within  $0.3^\circ$  latitude and longitude and  $\pm 30$  minutes of the corresponding AERONET station and observation are considered collocated. AERONET and CALIOP AODs above 0.2 and 0.6, respectively, are not included in order to avoid high aerosol loading cases and exclude noisy CALIOP data.





**Figure 14.** Monthly-averaged global CALIOP cloudy-sky frequencies after applying several different threshold techniques to both day and night time data. The solid lines show the daytime scenario for each respective case while the dotted lines show the nighttime observations for each case.



**Figure 15.** The de-seasonalized monthly- and regionally-averaged cloudy-sky frequency of ACA occurrences for the nine different regions outlined in Fig. 3 and explained in Table 3. The dashed lines show the monthly frequency over the regions and the solid lines show the trend lines computed for each region with the x-axis representing the year of the study. The CALIOP nighttime analysis is shown in aqua marine and the day-time analysis is shown in dark blue.

<b>Table 1.</b> Various definitions of frequency of above-cloud aerosols (ACA) used throughout the study.		
Name	Data Set	Definition
Cloudy-Sky ACA Frequency (Passive)	OMI-MODIS	(# of MODIS observation with assigned AI > AI baseline and cloud fraction equal to one) / (# of MODIS observations with cloud fraction equal to one and valid AI retrieval) per latitude and longitude grid over given time period
All-Sky ACA Frequency (Passive)	OMI-MODIS	(# of MODIS observation with assigned AI > AI baseline and cloud fraction equal to one) / (# of MODIS observations with valid AI retrieval) per latitude and longitude grid over given time period
Cloud-Sky ACA Frequency (Active)	CALIOP	(# of CALIOP observations with AOD > AOD baseline located above a cloud with COD > 0) / (# of CALIOP observations with COD > 0) per latitude and longitude grid box over given time period
All-Sky ACA Frequency (Active)	CALIOP	(# of CALIOP observations with AOD > AOD baseline located above a cloud with COD > 0) / (total # of CALIOP observations) per latitude and longitude grid box over given time period
Cloudy-Sky Frequency	MODIS	(# of observations with Cloud Fraction of unity (CF =1) / total # of MODIS observations)

**Table 2.** Global cloudy-sky relative frequency and data counts for the sensitivity test carried out in Sect. 5. Aerosol and cloud layers retrieved with ‘intermediate’ or ‘strict’ QA metrics are considered in this analysis. A total of five different threshold tests are applied to both day and nighttime CALIOP cloud and aerosol layer products.

	Day	Night
Total Cloudy Scenes (Column COD > 0 /0.3/2.5)	100,028,240/ 54,801,072/ 28,559,920	91,828,232/ 52,634,300/ 25,897,344
<b>Data Counts / Mean Global ACA Relative Frequency</b>		
COD > 0 & AOD > 0	1,193,048/ 1.79 %	3,368,351 / 4.5 %
COD > 0.3 & AOD > 0	789,652/ 2.0 %	2,795,442 / 6.1 %
COD > 2.5 & AOD > 0	556,097/ 2.5 %	2,091,310/ 8.09 %
COD > 0.3 & AOD > 0.015	597,917/ 1.63 %	1,516,547/ 3.54 %
COD > 2.5 & AOD > 0.015	420,778/ 2.0 %	1,167,569/ 4.52 %
COD > 0 & AOD > 0.015	904,892 / 1.46 %	1,765,620 / 2.6 %

**Table 3** Seven and a half year above-cloud aerosol cloudy-sky frequency, ACAOD and clear-sky AOD inter-annual variability analysis for the selected target regions. Aerosol and cloud layers retrieved with ‘intermediate’ or ‘strict’ QA metrics are considered in this analysis. Yearly variation for the entire globe is also included. For each region, inter-annual variability (frequency change per year) for the three parameters, the ACA cloudy-sky frequency, ACAOD and clear-sky AOD values are reported. Note that the inter-annual variability for clear sky AODs is estimated using 100% cloud free data from the CALIOP cloud and aerosol layer products.

Region	Latitude (°)	Longitude (°)	Slope /per year (CALIOP day-time) (%)	Trend Significance CALIOP day-time ( $\frac{\dot{\omega}}{\sigma_{\dot{\omega}}}$ )	Slope /per year (CALIOP night-time) (%)	Trend Significance CALIOP night-time ( $\frac{\dot{\omega}}{\sigma_{\dot{\omega}}}$ )
ACA cloudy-sky frequency (%) / Above-cloud aerosol AOD / clear-sky AOD						
Southern Africa	37°S - 5°N	30°W - 30°E	0.007/ -0.001/ -0.0004	0.009/ 0.18/ 0.04	0.148/ 0.0005/ 0.0009	0.159/ 0.067/ 0.08
Northern Africa	5°N - 35°N	70°W - 25°E	0.05/ -0.0006/ -0.001	0.116/ 0.035/ 0.07	0.07/ -0.0001/ -0.002	0.133/ 0.005/ 0.09
Southeast Asia	10°N - 25°N	90°E - 150°E	-0.04/ 0.004/ -0.002	0.080/ 0.17/ 0.1	-0.010/ -0.0012/ -0.0004	0.026/ 0.07/ 0.02
China	30°N - 55°N	110°E - 160°E	-0.084/ 0.0006/ 0.0009	0.238/ 0.090/ 0.05	-0.10/ -0.0006/ 0.0002	0.088/ 0.10/ 0.008
Middle East	10°N - 40°N	30°E - 55°E	0.36/ 0.004/ 0.006	0.239/ 0.15/ 0.16	0.339/ 0.004/ 0.005	0.238/ 0.09/ 0.13
South America	20°S - 10°N	105°W - 60°W	-0.078/ 0.0018/ -0.0016	0.189/ 0.18/ 0.12	-0.157/ -0.0002/ -0.0019	0.109/ 0.03/ 0.09
India	0° - 30°N	60°E - 85°E	0.10/ 0.001/ 0.0084	0.106/ 0.08/ 0.20	0.08/ -0.0035/ 0.010	0.110/ 0.064/ 0.19
North America	20°N - 60°N	160°W - 110°W	-0.05/ 0.0005/ 0.00002	0.082/ 0.06/ 0.003	-0.074/ -0.0005/ -0.0003	0.045/ 0.10/ 0.04
Southern Oceans	40°S - 12°S	35°E - 115°E	-0.04/ 0.0004/ 0.0012	0.120/ 0.083/ 0.29	0.05/ 0.0004/ 0.0008	0.037/ 0.078/ 0.21
Global			-0.004/ 0.0004/ 0.0006	0.049/ 0.16/ 0.13	-0.02/ 0.0004/ 0.0007	0.05/ 0.15/ 0.18

