



Frequency and  
Trends of  
Above-Cloud  
Aerosols

R. Alfaro-Contreras et al.

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# Investigating the frequency and trends in global above-cloud aerosol characteristics with CALIOP and OMI

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Abstract

Seven and a half years (June 2006–November 2013) of Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) aerosol and cloud layer products are compared with collocated Ozone Monitoring Instrument (OMI) Aerosol Index (AI) data and Aqua Moderate Resolution Imaging Spectroradiometer (MODIS) cloud products, to investigate variability in estimates of bi-annual and monthly above-cloud aerosol (ACA) events globally. The active- (CALIOP) and passive-based (OMI-MODIS) techniques have their advantages and caveats for ACA detection, and thus both are used to get a thorough and robust comparison of daytime cloudy-sky ACA distribution and climatology. For the first time, baseline above-cloud aerosol optical depth (ACAOD) and AI thresholds are derived and examined (AI = 1.0, ACAOD = 0.015) for each sensor. Both OMI-MODIS and CALIOP-based daytime spatial distributions of ACA events show similar patterns during both study periods (December–May) and (June–November). Divergence exists in some regions, however, such as Southeast Asia during June through November, where daytime cloudy-sky ACA frequencies of up to 10 % are found from CALIOP yet are non-existent from the OMI-based method. Conversely, annual cloudy-sky ACA frequencies of 20–30 % are reported over Northern Africa from the OMI-based method, yet are largely undetected by the CALIOP-based method. This is possibly due to a misclassification of thick dust plumes as clouds by the OMI-MODIS based method. An increasing trend of  $\sim 0.5\%$  per year (since 2009) in global monthly cloudy-sky ACA daytime frequency of occurrence is found using the OMI-MODIS based method. Yet, CALIOP-based global daytime ACA frequencies exhibit a near-zero trend. Further analysis suggests that the OMI derived cloudy-sky ACA frequency trend may be affected by OMI row anomalies in later years. A few regions are found to have increasing trends of cloudy-sky ACA frequency, including the Middle-East and India. Regions with slightly negative cloudy-sky ACA frequency trends are found over South America and the Southern Oceans, while remaining regions in the study show a near-zero trend. Global and regional trends are not statistically significant, though, given relatively lacking sam-

### Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ple sizes. A longer data record of ACA events is needed in order to establish a more significant trend of ACA frequency regionally and globally.

## 1 Introduction

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

ACA events are most effectively identified using active-based lidar measurements, which has been demonstrated using the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP; Winker et al., 2010; Kacenelenbogen et al., 2014), the lone such instrument presently in satellite orbit. CALIOP measures backscattered signals at the 532 and 1064 nm wavelengths, including segregated linearly-parallel and orthogonal polarization backscatter states in the former channel. In particular, the active-profiling

### Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



cloud properties (e.g. Yu et al., 2012; Alfaro-Contreras et al., 2014). Further and compared with CALIOP, OMI measurements represent a relatively large surface footprint of 13 km × 24 km at nadir, which limits cloud-clearing efficacies since footprints of this size are prone to sub-pixel cloud contamination (Torres et al., 2007). Collocated Moderate Resolution Imaging Spectroradiometer (MODIS) observations, however, as part of NASA's A-Train satellite constellation, which includes CALIOP (Stephens et al., 2002), can be utilized to distinguish and filter cloudy pixels/scenes within the OMI footprint.

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

## 2 Datasets and methodology

CALIOP Level 2 5 km cloud and aerosol layer products (Winker et al., 2010) and OMI Level 2 Collection 3 UV aerosol products (OMAERUV; Torres et al., 2007) are paired with Aqua MODIS cloud products (MYD06\_L2; King et al., 1997) and Aerosol Robotic Network (AERONET; Holben et al., 1998) Level 2.0 Version 2 cloud-screened data from June 2006 through November 2013.

For identification of ACA, 5 km CALIOP 532 nm cloud and aerosol layer products are used (Winker et al., 2009, 2010) for resolving aerosol extinction above apparent cloud top heights in each respective product file (e.g. Yu et al., 2012; Alfaro-Contreras et al., 2014). The 532 nm above-cloud aerosol optical depth (ACAOD) is then solved by integrating the extinction coefficient over those corresponding bins (Liu et al., 2013;

## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Kacenelebogen et al., 2014). The CALIOP-based trend analysis may be affected by CALIOP signal deterioration over time. Thus, collocated AERONET datasets are used, as the first order approximation, for evaluating instrument-related variations in the long term CALIOP AOD trend. Reported at eight spectral bands ranging from 0.34–1.64  $\mu\text{m}$  (Holben et al., 1998), AERONET AOD datasets are frequently used for validating satellite retrievals (e.g., Zhang et al., 2001; Yu et al., 2003; Kaufman et al., 2005a; Remer et al., 2005; Hahn et al., 2010; Shi et al., 2011; Sayer et al., 2012), as well as model simulated aerosol optical properties (e.g. Zhang et al., 2011, 2014).

The Level 2.0 cloud-screened and quality-assured AERONET AOD data (Eck et al., 1999) from all available coastal and island AERONET sites are used for collocating with CALIOP data. AERONET AOD data are interpolated, based on a method described in Zhang and Reid (2006), to the 0.532  $\mu\text{m}$ , which is the wavelength for CALIOP reported AODs, and are spatio-temporally-located with CALIOP AOD data. Instrument-related trends in the CALIOP AOD are investigated by calculating the global monthly-mean AERONET and CALIOP AODs and comparing the two monthly aerosol loading averages. CALIOP observations found to be within 0.3° latitude/longitude and  $\pm 30$  min of corresponding AERONET observations are considered collocated in space and time. In addition, we have used only pairs that have collocated AERONET AOD (0.532  $\mu\text{m}$ ) data less than 0.2 to exclude major aerosol episodes of continental origin. One additional quality assurance step is applied to exclude pairs with CALIOP AOD of larger than 0.6 for removing potentially noisy CALIOP data. In the case where several CALIOP observations are paired up with a single AERONET retrieval, a one-to-one relationship is established with the closest CALIOP observation.

Besides CALIOP data, OMI AI is used to isolate ACA events in those data. OMI AI and MODIS cloud datasets are spatio-temporally-located, given their position in the NASA “A-Train” constellation (e.g. Stephens et al., 2002), by collocating the two products with respect to overpass times and then identifying all temporally collocated cloudy MODIS pixels located within the boundaries of the OMI footprint. Such methods are described further in Alfaro-Contreras et al. (2014). Cloud fractions from the

## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



MODIS MYD06 product, reported at a 5 km horizontal resolution, are then leveraged for sub-pixel cloud clearing of the OMI AI. The MODIS cloud fraction is computed from the percentage of cloudy 1 km cloud mask product (MOD35) pixels within a given 5 km scene (e.g., Ackerman et al., 1998). With the exception of the cloud-top height restriction from the CALIOP data set, OMI/MODIS and CALIOP data are each filtered and quality-assured (described in detail in Alfaro-Contreras et al., 2014) to calculate respective global ACA distributions. It is known that that the OMI instrument experienced row anomalies beginning in 2008–2009 (<http://www.knmi.nl/omi/research/product/>, accessed on 22 December 2014). Thus the impact of the row anomalies to OMI AI trends is explored later in this paper.

### 3 Above-cloud aerosol baselines and limitations

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

To conceptualize the problem, shown in Fig. 1a are global distributions of mean CALIOP-derived daytime underlying cloud-top height for ACAOD events with optical thickness greater than zero. Figure 1b shows the averaged cloud-top height distributions of the highest cloud located within an atmospheric column regardless of the CALIOP ACAOD. Figure 1c and d depicts the same information as Fig. 1a and b, respectively, during the nighttime analysis. Globally averaged daytime cloud-top heights derived for each method, Fig. 1a and b, are  $\sim 2.0$  and 7.5 km, respectively. Clearly,

applying the CALIOP ACAOD  $> 0$  threshold makes a drastic change in the averaged cloud-top height. Considering that aerosol particles are always present above clouds, thus, results from Fig. 1 may indicate a limitation in CALIOP for detecting very optically thin aerosol particles (e.g. AOD  $< 0.01$ ).

5 Still, given the mean cloud-top height as shown in Fig. 1, we consider the unique AERONET site at Mauna Loa, Hawaii (LAT/LON, 3397 m above mean sea level). This free-tropospheric ground site rests at an altitude roughly within the global mean cloud-top heights discussed previously. Indeed, this physical feature of the site (that is being above the cloud deck below) is one of the key reasons for the importance of the site globally. The yearly mean Level 2.0 AERONET AOD (500 nm) there ranges from 10 0.013–0.023 from 1996–2013, and provides a generalized estimate for potential baseline ACAOD value globally. Kacenelenbogen et al. (2014) report that the CALIOP lidar exhibits limitations in detecting ACA plumes with ACAOD less than 0.02. This lower value may, therefore, represent an effective noise floor, whereby CALIOP algorithm response below it is compromised.

15 Based on Kacenelenbogen et al. (2014) as well as the AOD climatology from the Mauna Loa AERONET site analyses, we arbitrarily set the baseline CALIOP ACAOD value to 0.015. Considering the CALIOP ACAOD baseline is somewhat arbitrarily chosen, we investigate the CALIOP-based ACA frequency distributions by varying the baseline values to 0, 0.01, 0.015 and 0.02 as shown in Fig. 2. Figure 2a–d shows 20 the cloudy-sky global ACA frequency distribution, defined as the number of scenes with AOD greater than our baseline values resolved a top a cloud of optical thickness greater than 0 divided by the number of scenes with column cloud optical depth (COD) greater than 0 per latitude and longitude bin, for the December–May period, for baseline ACAODs of 0 (2a), 0.01 (2b), 0.015 (2c) and 0.02 (2d) respectively, using the CALIOP 25 aerosol layer datasets. Note that different from the cloudy-sky frequency, another way of measuring ACA frequency has been proposed by Devasthale and Thomas (2011) and is referred as the all-sky frequency in this study. The all-sky frequency is defined as number of scenes with ACAOD greater than the baseline resolved over a cloud of

## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



COD > 0 divided by the total number of CALIOP scenes per bin. The difference between the two techniques is discussed in more detail during the trend analysis section.

Shown in Fig. 2, no clear difference is observed in the cloudy-sky ACA frequency by applying various CALIOP ACAOD baselines. Similar conclusion can also be made for the June–November period (Fig. 2e–h). Thus, for the purposes of this paper, the baseline CALIOP ACAOD value of 0.015 (0.532  $\mu\text{m}$ ) is chosen, and the sensitivity of ACA trends to the selection of the baseline CALIOP ACAOD is explored in a later section. Additionally, our selection of CALIOP ACAOD baseline has little effect on the background cloudy-sky ACA frequency, which is for the most part, less than 5% (dark blue) throughout the globe. Thus, we arbitrarily select 5% as the threshold between background and significant cloudy-sky ACA frequencies. For the remainder of the paper, ACA frequencies less than five percent are not considered for global distributions of ACA frequencies.

To derive the corresponding noise floor value for above-cloud OMI AI, a pairwise comparison of collocated above-cloud OMI AI and CALIOP AOD has been performed using one year (2007) of collocated OMI-MODIS and CALIOP data as described in Alfaro-Contreras et al. (2014), though without any limitations on the cloud-top height. Figure 3a depicts the relationship between binned above-cloud OMI AI and CALIOP AOD segregated into six different underlying MODIS-derived CODs (Yu et al., 2012; Torres et al., 2012). The bin averaged CALIOP ACAOD of 0.015, the baseline CALIOP ACAOD value chosen above, corresponds to OMI AI values of 0.7–1.2 for underlying MODIS CODs ranging from 0 to 20. Note that, if CALIOP ACAODs are biased low, the corresponding OMI AI thresholds may also bias low using methods as shown in Fig. 3a.

Still, as suggested from Fig. 3a, baseline values of OMI AI vary from 0.7 to 1.2 depending on the underlying cloud properties. To further explore the issue, detected ACA events are evaluated with the use of different baseline OMI AI values, similar to the CALIOP ACAOD baseline analysis and shown in Fig. 3b–i however using only those bin averages with cloudy-sky ACA frequency greater than five percent. Figure 3b–e

Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



depicts the multi-year (2006–2013) cloudy-sky ACA frequency global average for the December–May period, by applying AI baseline thresholds of 0.7 (3b), 0.8 (3c), 0.9 (3d) and 1.0 (3e) respectively. With the use of the baseline OMI AI value of 0.7, most of the remote southern oceans stand out for significant case numbers. By increasing the AI baseline value to 1.0, in contrast, detected ACA events are significantly reduced. A similar conclusion can also be drawn from the June–November period (Fig. 3f–i). Given that hand-held ship borne sun photometer measurements collected by the Marine Aerosol Network (MAN; Smirnov et al., 2011) show an averaged AOD ( $0.55 \mu\text{m}$ ) of 0.07 or less from 30 to  $60^\circ \text{S}$  (Toth et al., 2013), significant ACA events are not likely over remote southern oceans. Thus, based on Figs. 2 and 3, CALIOP ACAOD of 0.015 and an above-cloud OMI AI of 1.0 are chosen as baselines.

Selection of baseline above-cloud CALIOP AOD and OMI AI is clearly subjective, and done for qualitative analysis in subsequent sections. There are multiple caveats that must be considered before constraining these values more accurately and representatively. First, as mentioned earlier, the CALIOP instrument has issues in detecting optically thin aerosol layers, especially during daytime. Additionally, it is also reported that CALIOP has a decreased sensitivity to stratospheric aerosols layers (Thomason et al., 2007; Winker et al., 2009). Third, besides aerosol loading, OMI AI is also sensitive to parameters such as aerosol vertical distributions, cloud optical depth of underlying cloud and aerosol single scattering albedo (e.g. Yu et al., 2012). Thus, setting a seasonal and regional based baseline for ACA requires a more in depth analysis and should be considered in future studies. Still, this study presents the first ever attempt to solve ACA baselines and the thresholds selected are the best noise floors we can come up with.

#### 4 Comparison of ACA global climatology using two separate techniques

Figure 4a depicts the multi-year gridded mean near-global distribution ( $180^\circ \text{W}$ – $180^\circ \text{E}$ ,  $45^\circ \text{S}$ – $60^\circ \text{N}$ ) of the OMI-derived daytime cloudy-sky ACA frequency for December to

## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



May. This frequency is solved relative to the number of collocated MODIS-OMI cloudy scenes with AI retrieval greater than our noise floor (e.g., 1.0) divided by the number of MODIS cloudy scenes with valid AI retrievals. Figure 4b and c shows corresponding cloudy-sky daytime and nighttime frequencies, respectively, using CALIOP data.

Cloudy-sky CALIOP ACA frequency is defined as the ratio of CALIOP scenes found to have an aerosol plume (with AOD above our previously determined floor noise of 0.015, resolved above the highest cloud layer) to CALIOP observations with COD greater than zero. Figure 4d–f shows the corresponding information to Fig. 4a–c for June to November.

Comparison of daytime cloudy-sky ACA frequency distributions is consistent between the two sensors and seasonal periods are investigated, and depicted in Fig. 4g–j. Some differences are distinct during December–May, as cloudy-sky ACA frequencies as high as 10 % are visible over the Gulf of Mexico from CALIOP, for instance, whereas they are non-existent from OMI-MODIS (Fig. 4a). Cloudy-sky ACA frequencies of 20–30 % are found with OMI-MODIS over high-latitude northern Asia, in contrast with CALIOP that shows no such activity (Fig. 4i). During June–November, both methods resolve ACA events over the west coast of Africa, as well as over the Middle East, of similar magnitude (10–60 %). However, distinct differences can be found between the two datasets. Higher cloudy-sky ACA frequency values of 10–30 % are found over North Africa using OMI-MODIS, in contrast to much lower values of 10–20 % found using CALIOP, for example. An OMI-based ACA study should correspond with a higher noise floor, compared with that of an active sensor, based on OMI's much coarser spatial and vertical resolutions, an inability to resolve non-UV absorbing aerosols, and the fundamental decoupling of column-integrated radiances themselves. Still, if the OMI AI baseline is biased, it may introduce an additional difference between OMI-MODIS- and CALIOP-based ACA frequencies.

Cloudy-sky ACA frequencies as high as 10–30 % are found over North Africa for both periods from OMI-MODIS while CALIOP returns much lower percentages (10–20 %) over the same region. This region is dominated by dust particle transport (Kaufman

## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



et al., 2005b), which is detected by both OMI and CALIOP. Therefore, we suspect that their relative differences as derived in Fig. 4i and j are likely linked to the misidentification of thick dust plumes as clouds by the MODIS cloud-masking scheme over the bright desert surfaces (e.g., Levy et al., 2013). Further differences observed between the two datasets may also be due to the different algorithmic sensitivities exhibited to both the optical depth of the underlying cloud and overlying aerosol plume, as well as the OMI AI and CALIOP AOD noise floors used to define the ACA events. We reexamine this point later in the paper.

Compared with daytime, increases in both the spatial extent and cloudy-sky CALIOP ACA frequencies are observable at night, as seen from Fig. 4b, c, e and f over most regions. Over the most common ACA regions, nighttime cloudy-sky ACA frequencies can be 10–30 % higher than during day, which may partially due to the stronger sensitivity of CALIOP at night allowing for detection of optically thin aerosol plumes. In particular, ACA events are observed with extended frequency over the west coast of North America year round and over the west coast of South America for the June–November period. Cloudy-sky ACA frequencies at night, over both of these regions, are composed of optically-thin aerosol loading cases above our defined noise floor. Nighttime ACA events are also observed over the east coast of Asia year round. One reason for differences in spatial coverage between daytime and nighttime ACA events is plausibly linked to a lower planetary boundary layer that affects the formation of low clouds (e.g. Schrage et al., 2012). Still, the discrepancy between nighttime and daytime ACA events can be partially attributed to the potential detection of relatively optically thin above-cloud aerosol plumes that are more detectable during nighttime compared with day as a result of the higher signal to noise ratio for CALIOP nighttime data (e.g. Kacenelenbogen et al., 2014).

Shown in Fig. 5 are averaged above-cloud OMI AI and CALIOP AOD values for corresponding ACA events from Fig. 4. Figure 5a depicts the mean near-global distribution of OMI AI over MODIS-resolved cloudy skies, defined as OMI-MODIS collocated cloudy pixels (cloud fraction of unity) and OMI AI averaged for each  $1^\circ \times 1^\circ$  grid box,





tion six month frequency exceeds 20 % or more. This indirectly confirms that most ACA outbreaks occur over CALIOP-defined low-level clouds.

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

## 5 Global trend analysis

A global trend analysis of ACA frequency is carried out for five different scenarios. The different scenarios are; OMI daytime cloudy-sky frequency, CALIOP daytime cloudy-sky and all-sky frequencies and CALIOP nighttime cloudy-sky and all-sky frequencies. Figure 8 shows CALIOP daytime cloudy-sky frequency (blue) and all-sky frequency (red), CALIOP nighttime cloudy-sky frequency (purple) and all-sky frequency (orange), and OMI daytime cloudy sky-frequency (green). Each data point represents the global monthly mean ACA frequency of CALIOP and OMI, calculated from 2.5° and 1° gridded ACA frequencies, respectively.

An increase in the OMI cloudy-sky ACA frequency over the study period is apparent in this global dataset, noticeably since 2009. However, this trend is not matched in the CALIOP data. The seasonal variation in ACA frequency is observed from year-to-year for both OMI and CALIOP (dashed lines). However, from the trend lines (showing a percentage change per year), only the OMI daytime cloudy-sky frequency shows a significant increase over this time period (solid lines). The increasing trend in OMI derived daytime global cloudy-sky ACA frequency, not apparent in any of the CALIOP derived trends, is troublesome and may be attributed to any of the different sensitivities

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



of the two techniques, including cloud and aerosol optical properties, aerosol-cloud separation distance, and/or deficiencies in the OMI data products. As will be described below, we further investigate several aspects of the observed OMI AI trend.

Given the unexpected monotonic increase in ACA frequency trend derived using OMI AI data, we examine the trend in OMI daytime cloudy-sky ACA frequency more closely. Figure 8a indicates a near-zero increase in the monthly averages during the first few years of the study, with frequencies increasing at a rate of roughly 0.5 % per year starting in 2009. This time period coincides with the start of OMI data loss due to row anomalies, as mentioned above, leading us to further investigate this as a possible reason for the increase in the observed OMI cloudy-sky ACA frequency. Note that we detected data loss while collocating OMI and CALIOP datasets and found no collocated pixels after 2008; a possible sign that the data loss is likely affecting OMI nadir viewing pixels. This is illustrated in Fig. 9a, which depicts a single swath of OMI AI over the African continent on 1 August 2007 where only OMI pixels with valid AI are shown. The data loss affected a large portion of the OMI AI data near the nadir regions of each OMI AI swath, as shown from a swath in 1 June 2009 (Fig. 9b).

Given that the data loss affects mostly nadir-viewing OMI pixels, OMI AI is evaluated as a function of the OMI sensor's viewing zenith angle (VZA) shown in Fig. 10. All OMI AI pixels for one year (2007) are averaged into one-degree VZA bins. Averaged OMI AI values at the edge of the swath are generally higher by about one AI unit than retrievals taken near the center of the swath. Thus, a meaningful trend cannot be established from the OMI data due to the viewing geometry bias impacting later years of the OMI aerosol products. The remainder of the paper will focus solely on trends derived from CALIOP ACA frequencies, and no further discussion of OMI AI frequencies will be carried out.

Next, AERONET AOD data are used to identify a possible bias in the CALIOP lidar due to potential signal deterioration in the instrument. Figure 11 depicts the clear-sky AOD trends derived using collocated CALIOP-AERONET data over all coastal and island AERONET stations (Zhang and Reid, 2006). Trends similar to those for the collo-

## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



cated AERONET and CALIOP data, as shown in Fig. 11, seem to suggest that potential deterioration issue from CALIOP are rather insignificant to our ACA study.

## 6 Sensitivity study

We next investigate the impact that our noise floor thresholds for overlying CALIOP AOD and/or underlying COD exhibit on derived global CALIOP cloudy-sky ACA frequencies. All CALIOP cloud and aerosol layer datasets are reprocessed such that the following conditions are met: (a) the underlying COD is greater than 0.3 and 2.5, respectively, (b) the AOD of the above-cloud aerosol plume is greater than 0, and (c) both conditions (a) and (b) are true. Passive-based radiance retrievals have been shown to lack sensitivities to optically-thin cloud detection for optical depths less 0.3 (Sassen and Cho, 1992; Ackerman et al., 2008; Holz et al., 2008). Thus, restricting the CALIOP COD to this threshold offers a more direct comparison of CALIOP- and OMI-based ACA frequencies. However, given that this range of optical depth corresponds with relatively high cirrus clouds, for which little contribution to the overall sample is expected, and broken low-level liquid phase clouds that are biased to ambiguously low values from signal aggregation effects in the 5 km product (Leahy et al., 2012; Campbell et al., 2015), this higher threshold provides a more representative basis for evaluation. We re-compute the monthly global mean cloudy-sky frequency for each of the CALIOP-constrained samples defined above during both daytime and nighttime. Respective CALIOP cloudy-sky frequency trends are shown in Fig. 12. Corresponding sample sizes and mean global frequencies are shown in Table 1.

In comparison with the unfiltered data from the daytime (solid red) and nighttime (dotted red) analyses, the various threshold techniques, including the filtering of CALIOP ACAOD according to our floor noise, correspond with significant variance in our results. However all sensitivity tests seem to show the same slightly-negative trend in cloudy-sky ACA frequencies. Although, those ACA events found over optically thicker clouds (COD > 2.5) seem to show more of a null trend over time rather than a slightly-negative

### Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion







cloud aerosol (ACA) events are studied and compared. Active-based profiling is considered an optimal means for identifying ACA occurrence. OMI identification is restricted to ultra-violet (UV)-absorbing ACA events (i.e., smoke), in contrast, through the Aerosol Index (AI) parameter. However, the relatively wide field-of-view of the paired OMI/MODIS datasets, in tandem, provide greater data volume overall, which serves as a relatively well-characterized reference for comparing with CALIOP.

The primary findings of this study are:

1. Baselines values for the passive-based OMI AI as well as active-based CALIOP above-cloud aerosol optical depth (ACAOD) are established in order to distinguish background noise from signal due to significant ACA events such as dust outbreaks and biomass burning. The “baseline” for OMI AI and CALIOP are applied to their respective data sets during processing. However, caution should be exercised when using these baselines, as they are an approximation and will vary depending on ancillary observational parameters for OMI and day vs. nighttime sensitivity for CALIOP.
2. Despite fundamental differences in spatial and vertical samplings, as well as sensitivity to ACA aerosol types, both OMI- and CALIOP-based techniques broadly resolve consistent global/spatial distributions of cloudy-sky ACA frequency. For example, both capture ACA events over the Northwest Coast of Africa and the Arabian Peninsula during the December–May period, and over the North- and South-west Coast, as well as the Southeast Coast of Africa, the Arabian Peninsula and Arabian Sea during the June–November period. Still, discrepancies, as expected, are present. For example, daytime cloudy-sky ACA frequencies of up to 10% are found from CALIOP over Southeast Asia during the June–November period. Such ACA events are none existent using OMI-based method, however we are not certain of the reason for the discrepancy over this region. Over North Africa, cloudy-sky ACA frequencies of around 20% are reported for both periods from the OMI-based method, yet such events are largely undetected by the

Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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

3. CALIOP nighttime data exhibit slightly larger distributions and a 10–20 % greater cloudy-sky ACA frequency annually in comparison to daytime. This may be due to the lowering of the planetary boundary layer at night, influencing frequencies of low-cloud formation, as well as the impact of higher signal-to-noise in nighttime CALIOP datasets for subsequent Level 2 analysis partly controlled for in our study by applying the noise floor. To the latter point, previous study has shown relative stability between day/night CALIOP aerosol products (Campbell et al., 2012). However, the implicit effect on the vertical distribution of aerosol occurrence was not specifically investigated. More detailed study is needed to reconcile this finding.
4. A global trend analysis shows a near-zero negligible slope in the global CALIOP cloudy-sky and all-sky ACA frequencies. However, OMI-MODIS cloudy-sky daytime ACA frequencies show an increase of  $\sim 0.5\% \text{ year}^{-1}$  since 2009 possibly due to a significant loss in the OMI data starting in 2009, mostly for nadir viewing pixels. Investigation of the relationship between OMI Aerosol Index (AI) and satellite viewing zenith angle, suggests a viewing angle dependency of OMI AI. Considering that OMI AI increases near the edge of the viewing swath, it is possible that the overall increase in ACA frequency is due to the significant loss of OMI AI data during later years of the study.
5. A decrease in the cloudy-sky global ACA frequency and data counts ranging from 1–2 % and 1–3 million, respectively, as a result of applying a variety of thresholds to the ACAOD and/or underlying cloud optical depth (COD) during sensitivity analysis. COD thresholds of 0.3 and 2.5 filter high cirrus clouds and non-contiguous low-level water clouds, respectively. Additionally, CALIOP data are reprocessed with no restriction to the ACAOD. Most threshold tests show a reduction in global





## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

**Table 1.** Global cloudy-sky relative frequency and data counts for the sensitivity test carried out in Section 6. 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	103 977 800/	94 461 656/
(Column COD > 0/0.3/2.5)	79 513 688/	73 606 408/
	45 003 964	42 543 896
Data counts/Mean global ACA relative frequency		
COD > 0 and AOD > 0	2 074 636/2.0 %	4 165 264/4.41 %
COD > 0.3 and AOD > 0	1 030 343/1.29 %	3 188 653/4.33 %
COD > 2.5 and AOD > 0	651 730/1.44 %	2 324 228/5.46 %
COD > 0.3 and AOD > 0.015	808 567/1.02 %	1 744 929/2.37 %
COD > 2.5 and AOD > 0.015	498 070/1.11 %	1 280 004/3.0 %
COD > 0 and AOD > 0.015	1 690 221/1.63 %	2 331 364/2.47 %

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

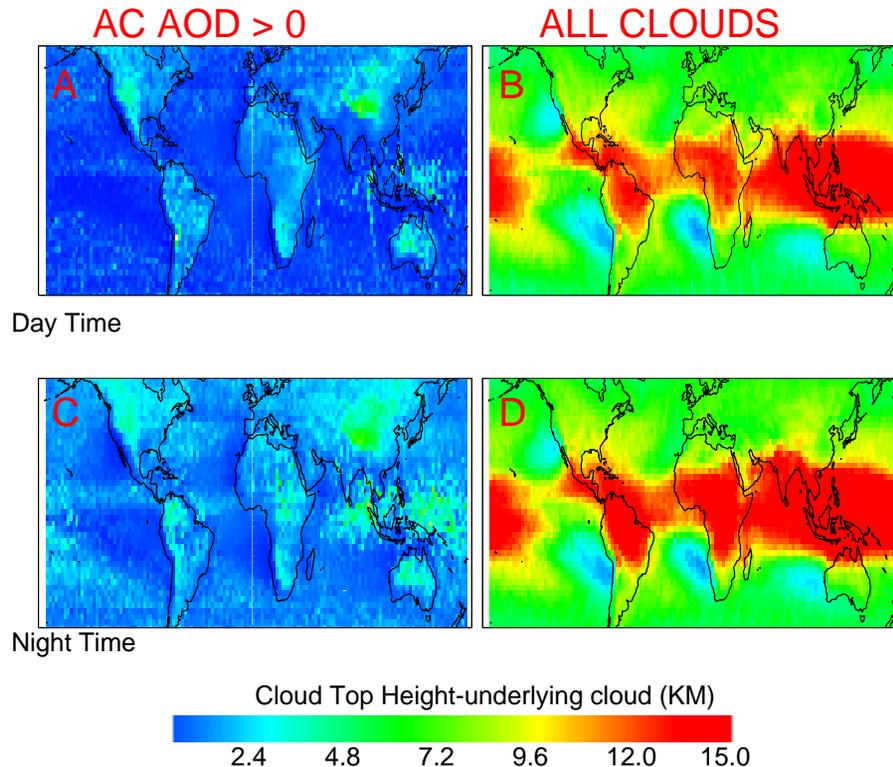
Interactive Discussion



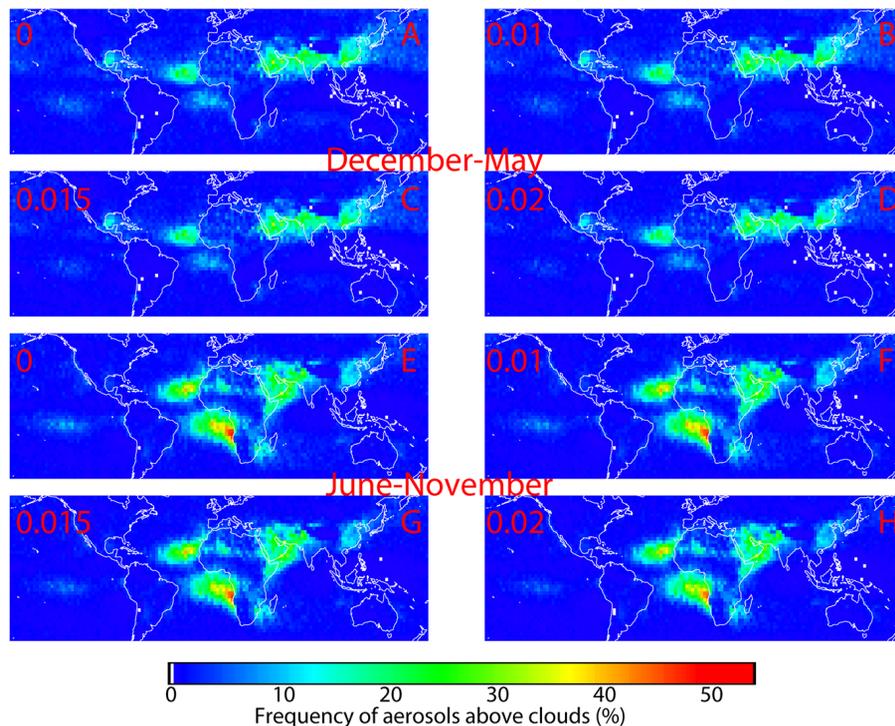


## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.



**Figure 1.** (a) Multi-year (June 2006–November 2013) cloud-top heights (above sea level) of the underlying cloud in the ACA scenarios averaged into  $2.5^\circ \times 2.5^\circ$  bins derived from CALIOP cloud and aerosol layer data sets for the entire year for the daytime observations. Only those clouds with a retrieved aerosol plume (ACAOD > 0) overhead are used in the averaging process for CALIOP daytime observations. (b). Cloud-top heights averaged similar to Fig. 1a however using all CALIOP scenes with column COD > 0 regardless of the AOD. (c and d) show the same information as Figs. 1a and b during nighttime observations.



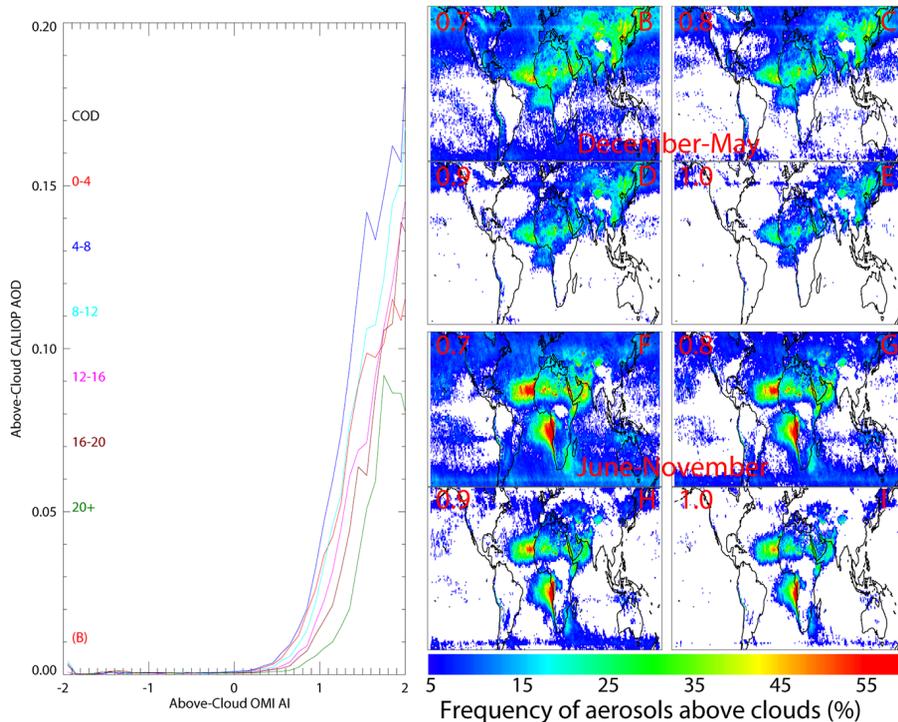
**Figure 2. (a–h)** Multi-year (2006–2013) CALIOP-derived daytime global cloudy-sky ACA frequency applying different CALIOP AODs as the threshold between background and significant aerosol loading. The CALIOP AOD are binned into  $2.5 \times 2.5$  latitude by longitude degree boxes derived using the CALIOP cloud and layer data sets. CALIOP AOD baseline thresholds of 0, 0.010, 0.015 and 0.020 are applied to **(a–d)** respectively for the December–May period. **(e–h)** show the similar results as **(a–d)** but for the June–November period.

Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	

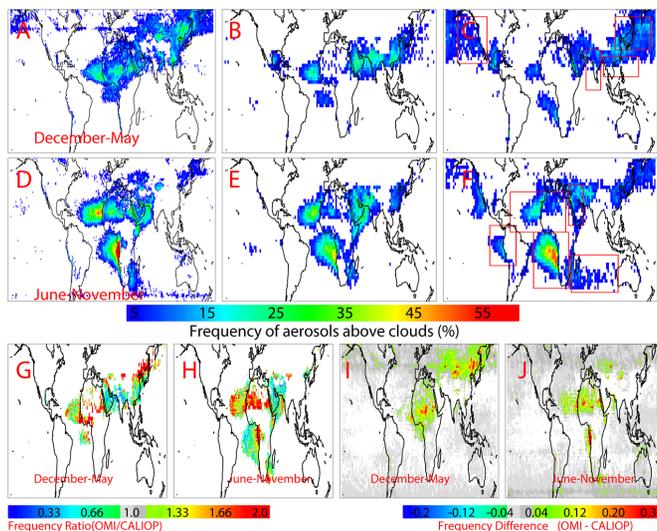




**Figure 3.** (a) Pairwise comparison between collocated OMI and CALIOP observations of above-cloud AI and AOD, respectively, as a function of the underlying MODIS cloud optical depth (COD). CALIOP AOD are averaged into OMI AI bins of 0.1. (b) Multi-year (2006–2013) daytime global cloudy-sky ACA frequency 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 Fig. 2c–f respectively for the December–May period. Figure 2f–i depicts the same information as Fig. 2c–f for the June–November period. ACA frequencies less than 5% are shown in white.

## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.



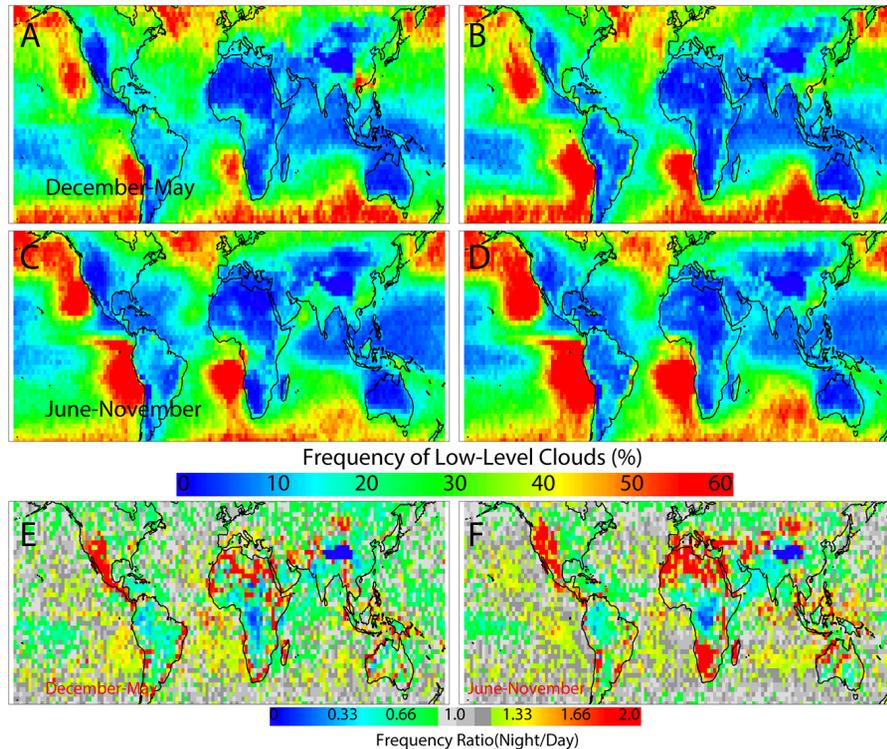
**Figure 4.** (a) Multi-year (December 2006–May 2013) daytime cloudy-sky frequency of occurrence of aerosol above-cloud events during December through May defined from OMI (ratio of opaque MODIS pixels with AI greater than 1.0 to the number of total opaque MODIS pixels). (b) Day-time cloudy-sky frequency of occurrence of above-cloud aerosol events over cloudy skies from CALIOP (ratio of CALIOP pixels with CALIOP AOD<sub>above cloud</sub> > 0.015 to the number of CALIOP pixels with column integrated COD > 0) for the same temporal domain as (a). (c) night-time cloudy-sky frequency of occurrence defined similar to the day time frequency from (b). (d–f) show the same information as (a–c) during June 2006–November 2013. (g and h) 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. (i and j) depict the difference in cloudy-sky frequency used to construct the frequency ratio plots (g and h) 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.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)



## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

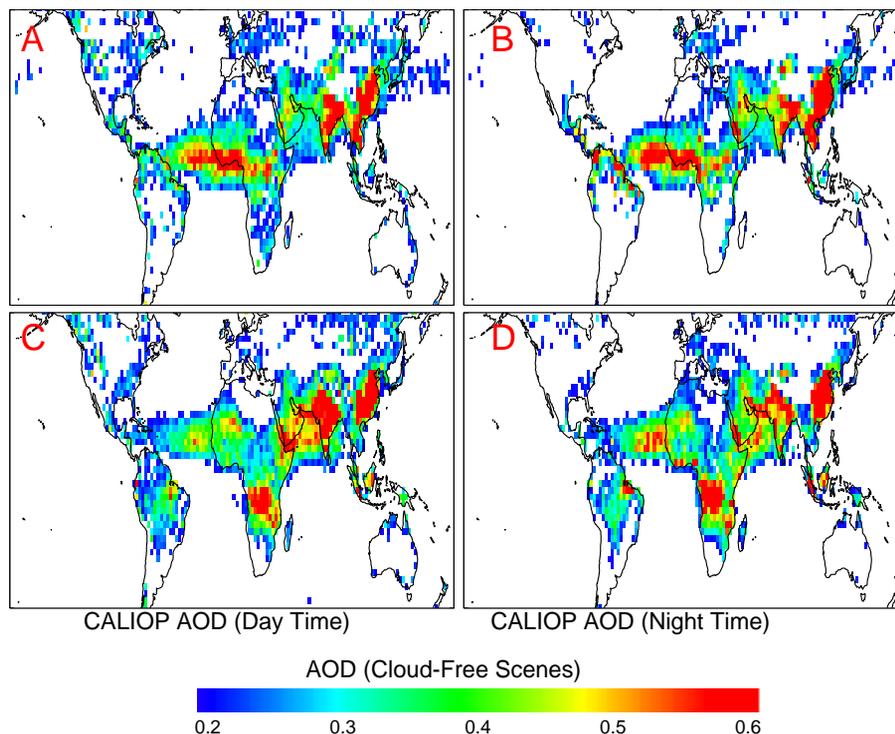


**Figure 6.** Multiyear (June 2006–November 2013) frequency of occurrence of low-level clouds defined by CALIOP as the ratio pixels with COD greater than 0 with cloud-top height < 3 km to the total number of CALIOP scenes within the current  $2.5^\circ \times 2.5^\circ$  bin for **(a)** December to May during day-time observations. **(b)** December to May of night-time observations. **(c)** Daytime frequency of occurrence of low-level cloud decks defined similar to **(a)** during the June–November time frame and **(d)**. Nighttime frequency of occurrence of low-level cloud decks for the same time frame as **(c)**. **(e and f)** depict the night to daytime frequency ratio for the December to May and June to November period, respectively.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

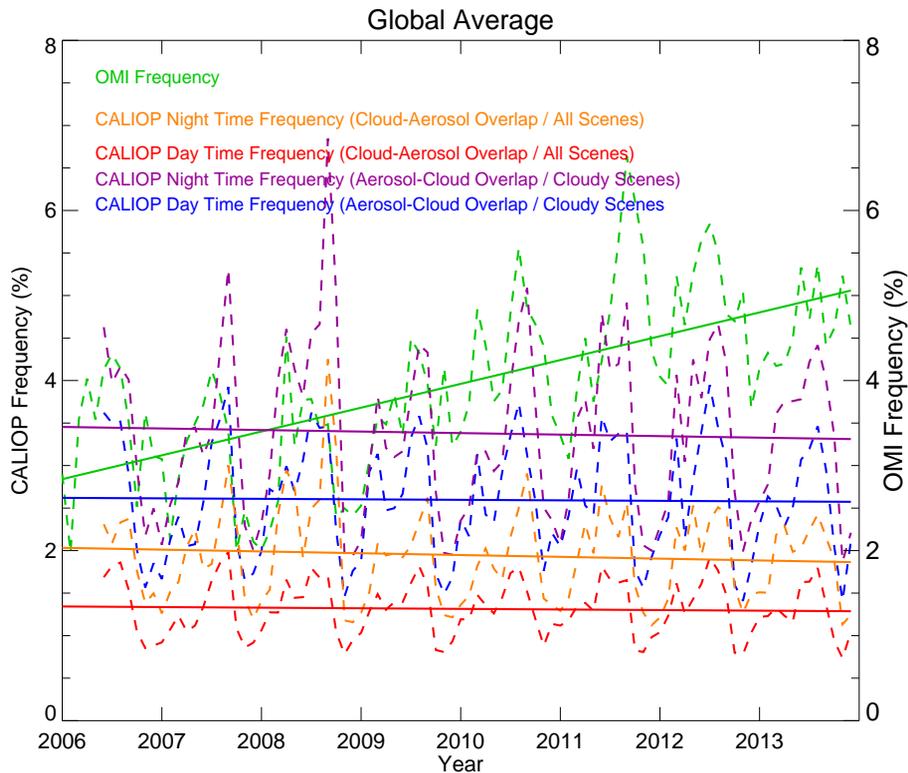
## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.



**Figure 7.** (a) Multi-year (2006–2013)  $2.5^\circ \times 2.5^\circ$  averaged CALIOP day-time aerosol optical depth (AOD) for December through May over completely cloud free scenes derived from CALIOP cloud and aerosol layer products for (a) daytime analysis during the December to May period. (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.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



**Figure 8.** Monthly-averaged global ACA frequencies derived using the OMI-MODIS based method (green) as well as 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 where the solid lines represent the yearly ACA frequency trends: OMI daytime cloudy-sky frequency is shown in green, CALIOP nighttime cloudy-sky frequency is purple, CALIOP nighttime all-sky frequency is orange, CALIOP daytime cloudy-sky frequency is blue and CALIOP daytime all-sky frequency is red.

**Frequency and Trends of Above-Cloud Aerosols**

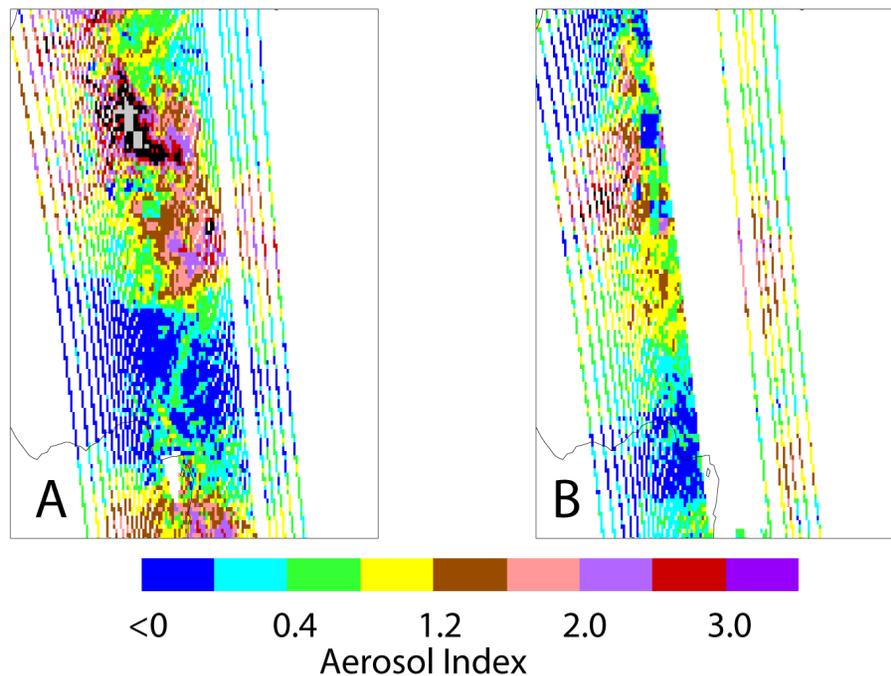
R. Alfaro-Contreras et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

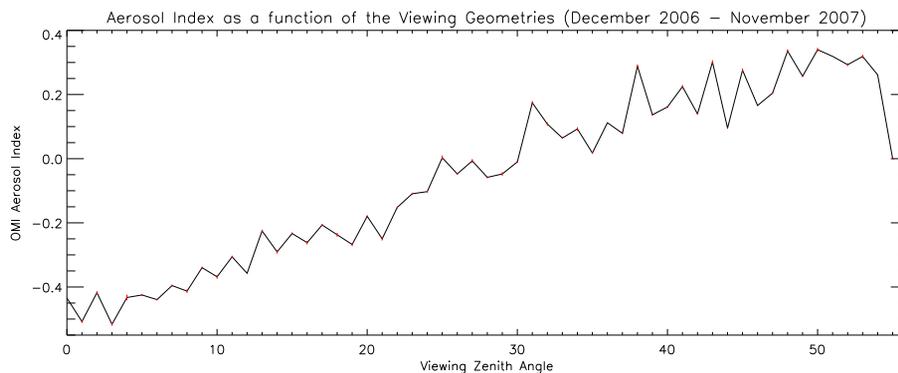


**Figure 9.** (a) A single swath from the OMI instrument over northern Africa on 1 August 2007 before the significant data loss reported in all OMI aerosol products. (b) A single OMI AI swath over the same region as (a) on 1 June 2009 which is affected by the significant data loss.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.

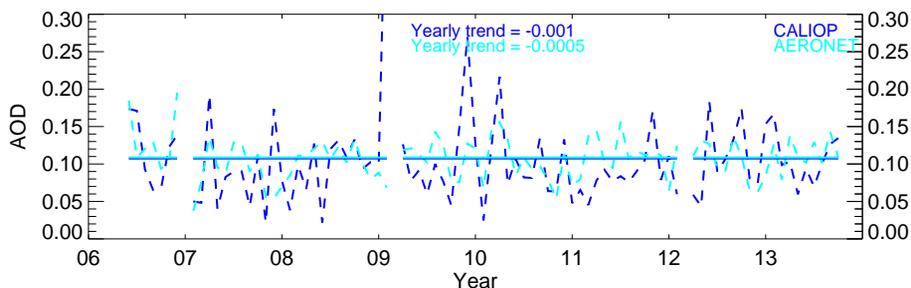


**Figure 10.** The OMI AI as a function of sensor's viewing zenith angle (VZA). All OMI AI data over the course of a year (2007) was binned into  $1^\circ$  VZA increments. The red vertical bars represent the 95 % confidence interval for each  $1^\circ$  bin.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

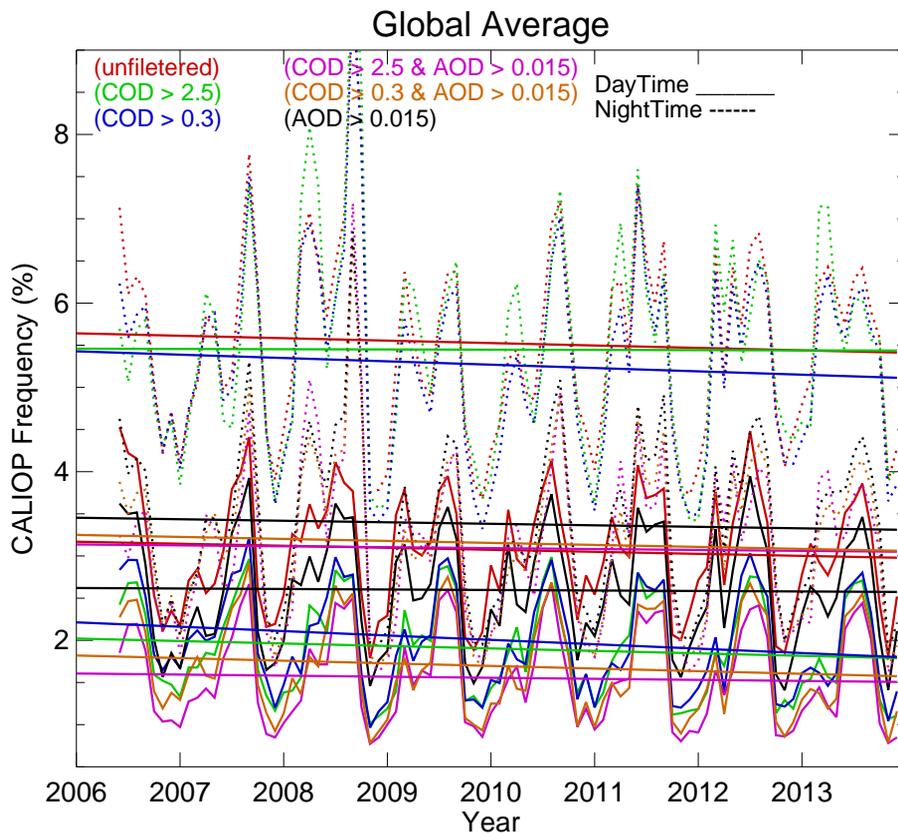
## Frequency and Trends of Above-Cloud Aerosols

R. Alfaro-Contreras et al.



**Figure 11.** Monthly-averaged over ocean clear-sky AODs derived from collocated CALIOP and AERONET data. CALIOP retrievals within  $0.3^\circ$  latitude and longitude and  $\pm 30$  min 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.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



**Figure 12.** Monthly-averaged global CALIOP cloudy-sky frequencies after applying several different threshold techniques to both day and nighttime data. The solid lines show the daytime scenario for each respective case while the dashed lines show the nighttime observations for each case.

## Frequency and Trends of Above-Cloud Aerosols

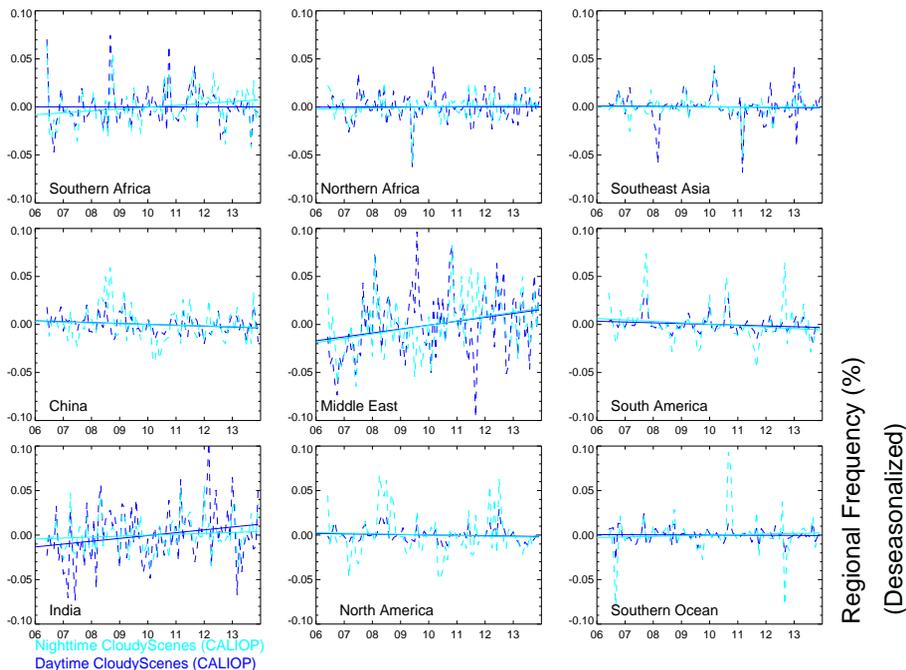
R. Alfaro-Contreras et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



**Frequency and Trends of Above-Cloud Aerosols**

R. Alfaro-Contreras et al.



**Figure 13.** The de-seasonalized monthly- and regionally-averaged cloudy-sky frequency of above-cloud aerosol occurrences for the nine different regions outlined in Fig. 4 and explained in Table 1. The dashed lines shows the monthly frequency over the regions while the solid lines show the trend lines computed for each region with the x axis shows represents the year of the study. CALIOP nighttime is shown in aqua marine while the day-time is shown in dark.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

