

Dear Prof. Stier

Please find attached the response to the reviewer comments. Moreover, I highlighted in red font color the locations of the main changes in the revised text. I did not include very obvious and small changes, typos and so on, in this "red version", but they are done as mentioned in the responses. To the end I placed also supplementary material to indicate what figures would go now to the Supplement in the revised version.

Kind regards,

Antti Arola

We would like to first express our thanks to the **REFEREE #1** for his/her constructive comments. The responses to these are below after the reviewer points that are in bold.

This manuscript uses AERONET AOD products retrieved from the spectral deconvolution algorithm to 1) quantify the AOD enhancement in cloudy-sky conditions, 2) examine the change in Angstrom exponent due to cloudy-sky, and then 3) propose potential explanations that are responsible for the change in Angstrom exponent. While I find the scope of the work is interesting, I have some concerns.

1) The use of "Level 0", the data points that got removed from Level 1 and was not included in Level 2 due to cloud screening: The whole manuscript is based on the assumption (although the authors treated it as a fact) that the fine- mode retrieval that is in Level 1 but not in Level 2 represents aerosol properties in cloudy conditions. The problem is - those retrievals were removed at the first place because we don't know how good those retrievals are and how much they are contaminated by clouds. The authors argue that clouds affect coarse-mode retrievals but not fine-mode, and therefore, the use of fine-mode retrieval is OK. However, I don't see any evidence to show that those retrievals are valid and indeed representative. The paper they cited (Chew et al.) used level 1.5, not level 1, so Chew's conclusions shouldn't be applied directly without caution. In short, as the authors mentioned, it is somewhat surprising that these data have not been fully exploited, but there is a reason for that. It is not scientifically rigorous to use retrievals without checking if those retrievals are meaningful!

We provided a brief note already earlier during the open discussion regarding this reviewer point. Many of these clarifications/justifications are now included also in our revised manuscript to explain the quality assurance included in the AERONET measurement data set that we analyzed and thus to justify their use in our study. We also included some illustrative plots, both in the manuscript and in the Supplement, to indicate how the L1 fine mode AOD is indeed a meaningful measurement also in cloudy conditions.

Minor comment – Page 4: do the authors really mean “the latter for all-sky and the former for clear-sky”? I think it should the other way around.

This is right, it should have been the other way around. This has been now corrected in the revised manuscript.

2) The analysis of seasonal variation and significance:

It would be better to clearly describe the sample size used in each bin, and to include retrieval uncertainty into these analyses. While the data range (figures 5-15) in each month spreads quite widely, it would improve the manuscript greatly by providing more critical discussions about them, rather than simply focusing on means only. Also, the authors throw in something like “For cumulus clouds in the mid-Atlantic US, the AE ... ” or “marine stratocumulus” for Lanai, which needs more care; these statements should be supported by some scientific evidence (a quick way will be to check weather state from ISCCP).

We included the sample size in each bin. Also, the discussion of the monthly plots is now more thorough.

3) Parcel model runs

I thought this part is interesting, but the current descriptions lack logical connections and are very dis-organised. I don't think readers can replicate simulations/results based on the current form, and I would strongly recommend rewriting this part. Here are some specific examples, which hopefully can help the authors understand why the current form could be quite confusing and unclear.

a) Could the authors make it clearer about the initial size and composition distribution used in the simulations? Like, the sentence on Page 14, 'For less hygroscopic aerosol composition ..., e.g., the one we assumed for our Walker Branch simulations', which should be described clearly right in the beginning. What is the growth factor used for Walker Branch? Also, on Page 16, it is mentioned that a very narrow lognormal size distribution, but I don't recall what is used exactly in the control experiments? Perhaps it is mentioned somewhere in the manuscript, but these things should be introduced in a more coherent and organised manner.

The modeling section was strongly modified and re-structured. Therefore these points above, as well as the modeling related reviewer points below, are hopefully adequately addressed by our new revised manuscript.

b) Could the authors make it clearer how the total column AOD and AE are calculated in cloudy and clear-sky conditions? A lot of assumptions are made there and it is unclear where is the justification, and why this will be consistent to observations.

The model calculations are described in more detail in the revised manuscript.

c) Page 17: why using different combinations of wavelengths for Angstrom exponent calculations?

Good point. We agree that it is better to be consistent and changed the wavelengths in these runs. However, as expected, the pattern of AE as a function of size in this plot (and thus the conclusions) did not change.

d) The context of "For instance, for Walker Branch there is ... for sizes above this limit, even close to the cloud top" on Page 17? Also, which part of Figure 18 helps conclude the last sentence of section 3?

These points are clarified in the revised manuscript.

e) Errors on Page 16, "WB@820 refers to the Lanai simulation", legend in Figure 16, and captions for Figure 18.

These are corrected.

We would like to first express our thanks to the **REFEREE #2** for his/her constructive comments. The responses to these are below after the reviewer points that are in bold.

This paper describes an interesting new approach to assess the AOT for fine mode aerosols in cloudy observations from AERONET. Instead of screening for clouds and retrieving fine and coarse mode AOT, the coarse mode AOT in aerosol-cloud mixed observations is attributed to cloud optical thickness, giving the fine mode AOT in cloud conditions. Given this the study tries to assess the magnitude of cloud enhancement in the fine mode.

While the approach is interesting, the paper lacks a sound scientific approach and clear presentation. In my opinion there is a need for a better description of the new approach and the expected impact, results to show the new approach works as expected, or not, and a conclusion. Instead many measurements are presented, explanations are presented, but it is not clear whether the method by itself can be trusted or not. Instead a large number of measurements are shown, which by itself do not prove the underlying new approach.

We provided a brief note already earlier during the open discussion regarding this point “whether the method by itself can be trusted or not”. Many of these clarifications/justifications are now included also in our revised manuscript to explain the quality assurance included in the AERONET measurement data set that we applied and thus to justify their use in our study. We also included some illustrative plots, both in the manuscript and in the Supplement, to indicate how the L1 fine mode AOD is indeed a meaningful measurement also in cloudy conditions.

The paper should be restructured so to follows a clear scientific approach: state the problem, describe how this problem is going to be resolved and then discussion and conclusion. In it’s current form there are too many figures, which do not add to the understanding of the problem, and new discussions are started in the middle of the manuscript. Furthermore, the text is sometimes hard to follow due to strange reasoning or formulations.

We have improved the manuscript as the reviewer suggested. There are only limited number of plots (of key results) in the actual manuscript, while many of the earlier plots are now in the Supplement. The text itself has been also clarified.

Specific comments are below:

line 50: “..(e.g. due to the aqueous process including nitrate or sulfate)”. This is a strange sentence: What processes are meant here? “Nitrate” and “Sulphate” are not ‘processes’.

This is clarified. With these we mean the cloud processing occurring such as formation of sulphate in cloud droplets or redistribution of nitrate aerosol between different sized aerosol particles during cloud droplet formation and evaporation.

lines 59-61: Based on its assumptions, SDA identifies cloud optical depth as the coarse mode AOD component and therefore effectively computes the fine mode AOD also in mixed cloud-aerosol observations.

The assumptions used by the SDA algorithm are not discussed, however, they are vital to assess how this new method works. E.g., from this sentence it is not even clear whether SDA attributes coarse mode AOT to clouds in only mixed phase conditions or all conditions. In the latter case, does the fine mode AOT relate well to the original fine mode AOT (in cloud-free observations)?

In the revised version we have explained, and demonstrated by some example cases, how SDA works in different cases of cloudiness. These examples cover cirrus clouds, when fine mode AOD is not cloud contaminated (case GSFC August 11, 2010; in the Supplement), as well as an example day of rapid cumulus variability (case BLDND from Dragon campaign, July 5, 2011; in the revised manuscript).

line 65: AERONET SDA product has been used to some extent, i.e. for rapid AOD increases in the vicinity of cumulus. Again an unclear sentence. Do you mean to say that SDA has been used 'to study' rapid AOD increases?

This sentence has been clarified.

lines 82-86: The spectral deconvolution algorithm (SDA) product, and its ability to separate coarse and fine mode AOD and provide useful fine mode AOD also in cloudy conditions, is vitally important in our study. O'Neill et al. (2001, 2002) developed SDA algorithm that utilizes spectral total extinction AOD data, ... These sentences are bad English. Please rephrase.

These sentences have been rephrased.

line 98: insert 'mode' after 'coarse'

Done.

line 99: remove the comma

Done.

lines 106-107: we included fine mode AOD and AE at 500 nm, from both Level1 and Level2 SDA measurements, the latter for all-sky conditions and the former for clear-sky conditions. I'm not sure about this one, but I expect this to be the other way around: L1 being all-sky and L2 only clear-sky.

Right, this should have been the other way around. It is now corrected.

lines 122-130 should be part of the Introduction, not results.

This was re-organized, to some extent, and part of this text is now in the methods section.

line 145 and figures 5 to 14. There are far too many figures here that do not add too much information. These figures should be merged into two Figures maximum.

In the revised version there is less figures included in the actual text, and many of the plots from different locations are now placed in the Supplement to make manuscript more easy to read. Also merging is conducted.

Line 299: We performed the analysis on a seasonal basis and found that consistently the highest cloud related 300 AOD enhancements occur in sites in East-Asia.. Please, write concisely, these kind of statements make the manuscript very hard to read: make an analysis on the basis of seasonality and find a region that stands out.

This is clarified.

Assessment of cloud related fine mode AOD enhancements based on AERONET SDA product

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Abstract.

AERONET (AErosol RObotic NETwork), which is a network of ground-based sun photometers, produces a data product called the Aerosol Spectral Deconvolution Algorithm (SDA) that utilizes spectral total aerosol optical depth AOD data to infer the component fine and coarse mode optical depths at 500nm. Based on its assumptions, SDA identifies cloud optical depth as the coarse mode AOD component and therefore effectively computes the fine mode AOD also in mixed cloud-aerosol observations. Therefore, it can be argued that the more representative AOD for fine mode fraction should be based on all direct sun measurements and not only on those cloud-screened for clear-sky conditions, in other words on those from Level 1 (L1) instead of Level 2 (L2) in AERONET. The objective of our study was to assess, including all the available AERONET sites, how the fine mode AOD is enhanced in cloudy conditions, contrasting SDA L1 and L2 in our analysis. Assuming that the cloud-screening correctly separates the cloudy and clear-sky conditions, then the increases in fine mode AOD can be due to various cloud-related processes, mainly by the strong hygroscopic growth of particles in the vicinity of clouds and in-cloud processing leading to growth of accumulation mode particles. We estimated these cloud-related enhancements in fine mode AOD seasonally and found, for instance, that in June-August season the average over all the AERONET sites was 0.011, when total fine mode AOD from L2 data was 0.154, therefore the relative enhancement was 7%. The enhancements were largest, both absolutely and relatively, in East-Asia; for example in June-August season the absolute and relative differences in fine mode AOD, between L1 and L2 measurements, were 0.022 and 10%, respectively. Corresponding values in North-America and Europe were about 0.01 and 6-7%. In some some highly polluted areas the enhancement is greater than these regional averages, e.g. in Beijing region and in JJA season the corresponding absolute values were about

0.1. It is difficult to separate the fine mode AOD enhancements due to in-cloud processing and hygroscopic growth, but we attempted to get some understanding by conducting a similar analysis for SDA-based fine mode Angstrom exponent (AE) patterns. Moreover, we exploited a cloud parcel model, in order to understand in detail the relative role of different processes. We found that in marine conditions, where aerosol concentration is low and cloud scavenging is efficient, the AE changes in opposite direction than in the more polluted conditions, where hygroscopic growth of particles leads to a negative AE change.

30 **1 Introduction**

Aerosol–cloud interactions contribute the largest uncertainty to the total anthropogenic radiative forcing (Myhre, 2013). One of the issues that hinder the measurement-based assessment of aerosol–cloud interactions by remote sensing methods is that typically aerosols and clouds cannot be measured simultaneously by passive remote sensing methods, including ground-based sun-photometers. In these techniques, so-called cloud-screening algorithms are therefore applied to provide aerosol optical depth (AOD) measurements for clear-sky conditions only. Due to this limitation, in aerosol–cloud interaction studies, aerosol and cloud properties have inherently different temporal sampling and therefore additional effects, e.g. impact of meteorology, have necessarily a possible influence in the derived correlations.

Many observational studies have found positive correlations between cloud fraction and AOD (Ignatov et al., 2005; Chand et al., 2012). However, as stressed above, with passive remote sensing the AOD measurements in cloudy conditions are not possible and thus these studies have to rely on cloud-screening technique and therefore the derived cloud aerosol relationships might be linked more to cloud contamination than to real physical processes. On the other hand, active remote sensing of aerosol from lidar measurements does not suffer similarly from this issue of cloud adjacency and these data have been analyzed as well for cloud aerosol interaction effects. Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) lidar data over oceans have been investigated for the relationship between aerosol and clouds (e.g., Várnai and Marshak, 2011; Yang, 2015). These studies have shown a sharp increase in aerosol signal within 4 km towards clouds.

The physical mechanisms contributing to the positive correlation between AOD and cloudiness, in addition to unphysical contamination by undetected clouds, are mainly the following: hygroscopic growth of aerosol particles in the vicinity of clouds, different meteorological conditions with different aerosol properties when clouds are present, and in-cloud processing (e.g. sulfate or nitrate aerosol production). The role of these effects, particularly hygroscopic growth versus meteorological influence, has been debated (e.g., Mauger and Norris, 2007; Engström and Ekman, 2010). However, it is a challenging task to separate the influence of each factor and thus, they have remained poorly known. One of the challenges is related to the time scale of physical processes involved; for-

mation of aerosol mass in gas-to-particle processes occur in minutes in cloud droplets compared to days in cloudless air (sulfate formation as an example), and thus it would be highly important to
60 have same temporal sampling of aerosol and cloud properties.

AERONET (AErosol RObotic NETwork), which is a network of ground-based sun photometers, includes also so-called Aerosol Spectral Deconvolution Algorithm (SDA) that utilizes spectral total extinction AOD data to infer the component fine and coarse mode optical depths at 500nm. Based on its assumptions, SDA identifies cloud optical depth as the coarse mode AOD component and there-
65 fore effectively computes the fine mode AOD also in mixed cloud-aerosol observations. Therefore, these measurements provide interesting insight into the simultaneous aerosol cloud measurements. More specifically, one can obtain and separate aerosol information in clear-sky and cloudy sky conditions, when clouds are thin enough that the direct sun measurements are possible.

AERONET SDA product has been used to some extent, for instance to study rapid AOD increases
70 in the vicinity of cumulus clouds (Eck et al., 2014); but nevertheless it has not been fully exploited yet and thus its unique features offer potential for additional interesting studies. In this paper, we present an analysis of cloud enhanced AOD measurements, based on AERONET SDA product. We included all the good quality data available from all AERONET sites for global analysis. Some of the sites were also selected for a more detailed analysis to demonstrate the usefulness of SDA data.

75 When aerosol climatologies, monthly AOD means or other statistics are formed, then Level 2 data of clear-sky measurements (excluding cloudy cases) are usually only included. This has to be definitely done with passive satellite measurements, in order to avoid cloud contamination in total AOD. However, this can lead to systematic biases due to the sampling; clear-sky conditions are related to particular type of weather patterns, while the excluded cloudy cases may differ systematically also
80 in their aerosol loading. Therefore, we want to stress that by our analysis, we can now obtain a quantitative estimate for the fine mode AOD that is more representative for all-sky conditions and thus also for the enhancement due to these cloudy cases.

2 Data and Methods

2.1 AERONET measurements

85 AERONET (AErosol RObotic NETwork) is a globally distributed network of automatic sun and sky scanning radiometers that measure at several wavelengths, typically centered at 0.34, 0.38, 0.44, 0.50, 0.67, 0.87, 0.94, and 1.02 μm . Each band has a full width of approximately 0.010 μm at half maximum (FWHM), except for 0.34 μm and 0.38 μm channels that have FWHM of 0.002 μm . All of these spectral bands are utilized in the direct Sun measurements, while four of them are used for
90 the sky radiance measurements, 0.44, 0.67, 0.87 and 1.02 μm . Spectral aerosol optical depth (AOD) is obtained from direct sun measurements at high accuracy (0.01 to 0.02 for overhead sun, with the larger errors in the UV (Eck et al., 1999)). The inversion product includes other aerosol optical prop-

erties, such as single scattering albedo (SSA), refractive indices and the column integrated aerosol size distributions above the measurement site provided at the sky radiance wavelengths (Holben
95 et al., 1998; Dubovik et al., 2000).

The ability of spectral deconvolution algorithm (SDA) to separate coarse and fine mode AOD, both in clear-sky and cloudy conditions, plays a key role in our analysis. O'Neill et al. (2001, 2002) developed SDA algorithm that utilizes spectral total extinction AOD data, with the assumption of bimodal aerosol size distributions, to infer the component fine and coarse mode optical depths. An
100 additional fundamental assumption of the algorithm is that the coarse mode Angstrom exponent (AE) and its derivative are assumed to be -0.15 and zero, respectively. The Angstrom exponent and its spectral variation of measured total AOD ($dAE/d\ln WL$) are the measurement inputs to the algorithm. These are determined from spectral AOD measurements at 5 wavelength: 380, 440, 500, 675, and 870 nm.

105 We applied several specific quality checks, in order to select the best quality SDA retrievals. We required that all 5 SDA wavelengths (380, 440, 500, 675, and 870 nm) were available in the Level 2 data and the L1 data were only utilized when L2 data were available within one-week time window, to rule out any instrumental problems. The possible outliers were removed according to the following criterion: $Abs(AOD_{500nm} - AOD_{SDA500nm}) > (0.02 + AOD_{500nm} * 0.005)$. This is the
110 same consistency check between measured AOD at 500 nm and SDA retrieved total AOD at 500 nm that is applied in the quality control checks for AERONET Level 1.5 data for SDA. Additionally, a consistency check of measured AOD compared to SDA retrieved total AOD at 500 nm was applied to both the L1 and L2 SDA data.

It is also noted that there are many internal quality assurance steps already in the regular AERONET
115 processing that ensure the quality and usefulness of both data levels (Level 1 and Level 2). The direct sun measurement data are not included in the AERONET Level 1 data set if the variance of the raw signal is very high within the triplet sequence. The variance threshold applied is based on the root mean square (RMS) differences of the three direct sun triplet measurements relative to the mean of these three values. If the $(RMS/mean) \cdot 100\%$ of the triplet values is greater than 16% then the
120 data will not be used for computation of AOD and the data will not appear in the Level 1 data set. This temporal variance threshold primarily removes data that are affected by clouds with large spatial-temporal variance in cloud optical depth (COD). This effectively removes much of the cumulus cloud contaminated data, although some of the thinner edges with lower COD do remain in the data. In the AERONET Version 2 Level 2 database, if there are only 1 or 2 points remaining in a day after
125 automatic cloud screening (Level 1.5), then none of this data reaches Level 2. In other words, at least 3 AOD observations are needed to pass the Smirnov et al. (2000) cloud screening algorithm in order for the data to reach Level 2 for that day. The Figure S1 in the supplement illustrates this in XiangHe site on August 10, 2010, when only two measurements remained after L1.5 cloud screening and those did not advance to L2 because AERONET requires a minimum of 3 points per day in L2.

130 **2.2 AERONET data selection and analysis**

In our analysis, we included fine mode AOD and AE at 500 nm, from both Level 1 and Level 2 SDA measurements, the former for all-sky conditions and the latter for clear-sky conditions. This data version (Version 2) includes cloud screening of Smirnov et al. (2000), which has been used for all papers using AERONET data since 2000 when this cloud screening algorithm was implemented.

135 New version 3 will be released in 2017, with significantly different cloud screening.

Moreover, we constructed our own specific “Level 0” data set of SDA measurements, including only those cases of Level 1 that were not in Level 2, thus this set includes only cloudy cases, according to the cloud-screening. From these different data sets we calculated the monthly means as follows. First, we calculated hourly means and averaged them to obtain the daily averages. These
140 daily mean values were then used to calculate the monthly averages, by requiring at least 10 days per month. For the seasonal means it was required that all the months had sufficient amount of measurements.

Eck et al. (2014) included several example cases to demonstrate how the AERONET Version 2 Level 1 data include meaningful information for our study as well. In their Figures 16c and 16d, Eck
145 et al. (2014) showed AOD measurements at BLDND site for one day during DRAGON campaign indicating how the large triplet variation data, which is often screened from Level 2, is in fact good fine mode AOD data. As was shown by the MODIS images in Eck et al. (2014), the only cloud type on this site and date was cumulus. Figure 1 shows this same case, but zooming into the afternoon measurements only and includes additionally both fine mode and total Angstrom exponents. During
150 this time period when AOD increased substantially, the Angstrom exponent remained high and relatively constant indicating the dominance of small particles and no significant cloud contamination, the latter point is also supported by very small coarse mode AOD throughout the period.

The strength of SDA algorithm is that fine mode AOD can be obtained also in cloudy conditions, as demonstrated by O’Neill et al. (2002), since it identifies cloud optical depth as the coarse mode
155 AOD component. Moreover, Chew et al. (2011), by comparing AERONET measured spectral AOD with lidar data, showed that SDA is able to effectively separate the fine and coarse so that the latter is only influenced by clouds. Additionally, Kaku et al. (2014) have verified that the SDA technique is also effective in separating the fine and coarse modes from in situ spectral optical measurements. It is likely, however, that the fine mode AOD is underestimated when cirrus ice crystal clouds overlay the
160 aerosol, due to strong forward scattering into the field-of-view of the sun-photometer (A. Smirnov: personal communication, 2016). However cloud screening also occurs when cirrus is not present (high temporal variance in the presence of clouds ; Eck et al. (2014)) and also when very few AOD observations occur on a primarily cloudy day. Nonetheless, since some of the cloud observations occur with cirrus present, the SDA overall provides a lower limit on the enhancement of fine AOD
165 in the presence of clouds. Figure S2 in the Supplement gives an example of one day measurements from GSFC, August 11, 2010, with cirrus clouds present in the afternoon. Non-cloud-screened mea-

surements (L1) between 17-21 UTC illustrate how cloud contamination is in the coarse mode only and fine mode AOD measurements seem to be not cloud contaminated.

2.3 Cloud parcel model simulations

170 The numerical model employed here to study aerosol and cloud droplet microphysics is a so called cloud parcel model which has been described in detail in Kokkola et al. (2003); Romakkaniemi et al. (2006, 2009, 2011). In short, the model solves condensation and evaporation of water between the gas and particle phase. It has a sectional representation of aerosol particle size distribution with a detailed size dependent description of aerosol composition. The model can be employed to study
175 how aerosol size distribution and composition are affecting the wet size of particles and cloud droplet formation in different atmospheric conditions.

For the cloud parcel model simulations the aerosol particle size distribution and composition as well as updraft velocity are needed as an input. In order to obtain the aerosol dry size distribution, we used the monthly mean AERONET-measured size distributions from the Level 2 inversion product. The size distributions from AERONET represent the ambient column averaged volume size
180 distributions. To translate this into dry aerosol size distribution we need to make assumptions on the particle composition and vertical aerosol profiles. The first assumption is that all aerosol is residing in the boundary layer with a height of 1km. In reality this does not hold exactly, however the height of the layer is needed in order to estimate the aerosol number concentration from the columnar aerosol volume distribution. The change in the assumed boundary layer height will translate into
185 change in the aerosol number concentration. The second assumption needed is for aerosol composition. As AERONET provides an estimate of aerosol volume distribution in ambient conditions, the aerosol hygroscopicity information is needed to approximate the dry aerosol size distribution from the wet (ambient) size distribution. Related to this, a value for relative humidity (RH) of air has to
190 be assumed. Here we assume an effective RH in which the modeled wet aerosol size distribution is reproducing the observed AERONET- measured size distribution. Here different assumptions are not independent of each other. For example the change in effective RH could be balanced with a change in the particle dry size to produce similar wet size distribution as aerosol particles grow as a function of RH. This will be discussed in more detail in the results section

195 As an output, the cloud parcel model calculates the ambient size distribution of aerosol particles and cloud droplets at different altitudes. These size distributions are used as an input to Mie calculations, in order to obtain the extinction coefficient and the corresponding Angstrom exponents at each model level together with the integrated columnar estimates of AOD and AE. These calculations are carried out using the Mie calculation tool in LibRadtran (Mayer and Kylling, 2005)
200 version 2.0, assuming purely scattering particles with the real part of refractive index of 1.5. The total column AOD and AE, as Cimel measurement would “see” from these modelled profiles, were estimated both for the cloudy and clear-sky case. In the latter case, the column AOD and AE values

were integrated over the model calculated aerosol profile from the ground level up to a level where RH reached 3 % higher value than effective RH that was used to determine the dry size distribution.

205 This was estimated to correspond to the highest humidity in clear-sky conditions and it was found to approximately reproduce the observed AOD. The remaining layers above this level were integrated assuming that they have this constant extinction, thus estimating the contribution from cloud-free model levels as measured by AERONET.

3 Results

210 3.1 Spatial and temporal patterns of cloud induced AOD and AE

We conducted our analysis first for all the available AERONET sites on a seasonal basis, for the following seasons: March-May (MAM), June-August (JJA), September-November (SON), December-February (DJF). Figure 2 shows these seasonal cases of the difference in AOD between Level 0 and Level 2 data, thus between cases of solely cloudy or clear-sky AOD measurements. We additionally
215 sub-divided our results into the following seven regions, shown also by lines in the plots: North-America, South-America, Europe, North-Africa, South-Africa, Asia, and Australasia. Table 1 shows the seasonal results for each region, e.g. the enhanced fine mode AOD, if sampled only for clear-sky conditions compared to cloudy-sky or all-sky cases. We can see that the AOD enhancements are consistently largest throughout the year in the Asia, reaching values of about 0.1 in many sites. This
220 is a substantial difference that would not have a negligible effect in the radiative effect estimates either, if Level 2 data were used instead. On the other hand, the difference over all the included sites (values given in the Table 2) is rather notable as well, e.g. in JJA fine mode AOD of all-sky data is 0.011 higher than the mean based on Level 2 only (0.154), thus all-sky fine mode AOD being about 7% higher.

225 We also established the AOD differences on a monthly basis separately for each AERONET site. Moreover, we made it similarly for fine mode AE parameter, which was weighted by AOD. We considered this AOD weighting both necessary and useful, in order to produce more robust signal for the seasonality; otherwise the within month AE variability was substantially higher, clearly due to the cases of lowest AOD when the AOD magnitude approaches the uncertainty of the single channel
230 AOD itself.

Figures 3 and 4 show examples of two sites (Walker Branch and Lanai), which will be studied in more detail also by cloud parcel modeling in the next section to understand some of the observed differences, while in the Supplement several other sites are included as well (Arica, Chile; Gwangju GIST, South Korea; Taihu and XiangHe, China). These figures show the monthly AOD-weighted
235 dAE (difference in AE between Level 0 and Level 2 data) and the corresponding annual pattern of $dAOD$ (difference in AOD between Level 0 and Level 2).

Many of the sites exhibit a clear seasonality in $dAOD$ with highest enhancements around JJA season, for instance in Walker Branch, XiangHe, Gwangju GIST. Moreover, often the timing of the highest $dAOD$ is related to slightly negative dAE , perhaps most evidently in XiangHe and Walker
240 Branch. In other words, AE of cloudy cases is slightly smaller, suggesting somewhat larger particles, likely related to swelling in humid conditions. However, these dAE differences are generally relatively small. As shown by Eck et al. (2014), for cumulus clouds in the mid-Atlantic US (Tselioudis et al., 2013) the dAE did not change much despite large changes in AOD on some days. This strongly suggested that particles grew in size from sub-visible Aitken, in addition to larger particle swelling in the high RH environment in and near clouds. In such a polluted environment (e.g. Baltimore-Washington region in Eck et al. (2014)) where it is known that there is SO₂ present it is also highly likely that sulfate particle formation also occurs in the clouds (rapid SO₂ to sulfate conversion in aqueous phase versus relatively slow in non-cloudy environments). Other gas-to-particle conversions are likely in the aqueous phase in cloud droplets (nitrates and organic particles; (e.g.,
245 Hayden et al., 2008; Ervens et al., 2011)). Therefore near zero change in AE could mean both processes are counter-balancing each other.

The dAE differences are generally small and most often negative values. Moreover, these patterns of dAE do not show generally a strong seasonality. However, there were few sites having positive differences systematically, mainly marine sites such as Lanai (Figure 4). The possible reasons for
255 these cases are studied in more detail in the next section.

3.2 Cloud parcel model based investigation of cloud-induced AOD and AE changes.

The fine mode AE differences between cloudy and clear-sky cases shown in the previous section exhibited typically negative values. As discussed above, in these cases the particle growth is likely a more dominant process than cloud activation. The latter process would remove the largest sizes,
260 while the former results in an increase in the effective wet particle size. However, there were about ten sites with clearly positive dAE , all being either Island or coastal sites. As an example of such a case is the weighted fine mode dAE (Figure 4) observed at Lanai; the sixth-largest of the Hawaiian Islands. Although, these positive values are not very large, negative cases seem to be essentially missing particularly during the summer.

Our main interest is to quantitatively understand the processes and their relative importance that could result in prevailing positive fine mode dAE . The sites with positive dAE , are all influenced by marine aerosol that typically has a strong bimodality with relatively small particles in the Aitken/accumulation mode, (e.g., Heintzenberg et al., 2004). With such aerosol size distribution it is possible that the critical size for droplet formation reaches small enough particle sizes to affect
270 the fine mode AOD. Thus the depletion of these particles into cloud droplets could decrease the effective size in the fine mode, being an opposing effect to the growth of aerosols by humidity.

By employing the numerical cloud parcel model, we investigated in more detail the relative role of aerosol hygroscopic growth and cloud activation on the enhancement of AOD and how this can affect the observed AE. This was done for contrasting conditions, namely Lanai and Walker Branch that
275 represent completely different aerosol conditions. For simplicity we assumed the Lanai aerosol to be solely composed of NaCl to represent highly hygroscopic sea salt aerosol. For Walker Branch, the composition was assumed to be 50% insoluble organics and 50% inorganic ammonium sulfate. This composition is assumed to be representative of continental aerosol. For the aerosol size distributions we used AERONET observed monthly mean size distributions from the Level2 inversion product
280 for August 2003.

Apart from the initial size and composition distribution, the initial conditions were assumed to be the same in all simulations: ambient temperature of 288 K and RH of 63 %. After the initialization, model simulated cloud formation for an adiabatically ascending air parcel with a constant updraft velocity w . In Lanai, w was assumed to be 0.2 m s^{-1} which is quite typical for marine stratocumulus
285 clouds. In Walker Branch w was assumed to be 0.5 m s^{-1} , which is typical to broken small cumulus clouds. In the simulations the boundary height was assumed to be 1 km. Assuming that the air parcel ascends adiabatically, the cloud base was reached at 880 m resulting in cloud height of 120 m.

From the simulated wet size distributions, we calculated the optical properties of the aerosol/cloud droplet population for different altitudes. This was done for both Walker Branch (WB) and Lanai
290 aerosol size distributions, with more detailed examination of Lanai case. The numbers within Figure 5 are given for the simulations of two different cases of growth factors (the ratio between the wet and dry diameter) in Lanai, as an estimate of the AERONET-measurement from these profiles in a manner explained in methods section. For example, the first three numbers are for the case of growth factor of 2 as follows: AOD of clear-sky profile, “*AODL2*”; AOD difference between cloudy and clear-sky profiles, “*dAOD1*”; AE difference between cloudy and clear-sky profile, “*dAE1*”.
295 The next three numbers correspond similarly to the case of increased growth factor of 2.2, indicating a positive value for *dAE* and a change of sign if compared to the case with growth factor of 2.

This result of positive *dAE* was achieved with an aerosol size distribution which reproduced observed AERONET distribution at 85 % relative humidity, where the growth factor of particles
300 composed of NaCl is approximately 2.2 (see Figure 1 of Ming and Russell (2001)). To illustrate the effect of hygroscopicity on the aerosol optical properties, we used also another dry size distributions for Lanai: one, where the dry size distribution reproduced the AERONET-observed ambient size distribution at 80 % relative humidity, where the growth factor is approximately 2. Thus, in the latter case, the dry size required to reproduce the AERONET observations was slightly larger than in the
305 case of 85 %. This will directly affect also the wet size of fine mode particles in the cloud, where the growth factor of NaCl particles is between 4-5 depending on the particle size and supersaturation, and now the sign of *dAE* changed.

These simulations demonstrate that dAE can indeed turn to a positive value, but only for highly hygroscopic NaCl aerosols, and when the particle dry sizes are sufficiently small and the supersaturation at the cloud base is high enough to activate these particles. In such a case the largest interstitial aerosol particles are clearly smaller than the estimated size limit for fine mode aerosol classification. For less hygroscopic aerosol composition and higher total aerosol concentration, e.g. the one we assumed for our Walker Branch simulations, did not produce positive dAE cases. In Lanai, the activation of smaller particles than in Walker Branch was further assisted by low number concentration and small dry sizes of aerosol which allowed the maximum supersaturation to reach higher values.

Based on our model simulations, it seems evident that cloud activation can affect and remove particles from the AERONET-measured fine mode AOD, resulting in positive dAE between L0 and L2 measurements. We wanted to additionally assess this threshold of activation size that needs to be reached, so that the effect of the removal of the cloud activated largest size particles in the fine mode, would overcome the hygroscopic growth of the smaller particles in the case of marine aerosol size distribution observed in Lanai. The former process has the overall effect to increase AE in cloudy case, while the latter has an opposing effect. The upper plot of Figure 6 shows the cloud activation (critical) radius as a function critical supersaturation. We repeated our Mie simulations for a range of critical supersaturation from 0 to 0.3%, always removing particles larger than the critical radius corresponding to the critical supersaturation from the aerosol size distribution. The lower panel in Figure 6 shows dAE as a function critical supersaturation. The figure illustrates that only a relatively narrow range of critical supersaturation, corresponding to critical radius of about $0.3 \mu\text{m}$, results in positive AE difference between cloudy and clear-sky column. With higher supersaturations, such small particles are able to activate that the interstitial aerosol is no longer affecting column AOD and thus AE. At low supersaturations, the hygroscopic growth of interstitial particles dominates the column AE, and thus the influence of a cloud scavenging of particles results in negative dAE .

There are cloud related processes that our model simulations do not fully describe, including gas-to-particle conversion and chemical reactions occurring in the aqueous phase. These processes would increase the distance between Aitken and accumulation mode, and thus likely magnify the modelled positive change in AE. In addition, the particle growth in humid regions in surrounding clouds is not included in our 1-D exercise. However, arguably the most important processes of marine cloud environment are included by our cloud parcel modeling study.

Figure 7, in turn, shows the results from our simulations in more detail for several cases. The simulations of both Walker Branch and Lanai are shown for two different model levels, e.g. WB@820 refers to the Walker Branch simulation at the model altitude of 820m, i.e. below the cloud base. To illustrate the effect of cloud activation on the fine mode, results close to the cloud top at 970 m are also shown (“Lanai@970” and “WB@970” for Lanai and Walker Branch, respectively). Finally, for the case of Lanai and close to the cloud top, we included two cases of different aerosol dry sizes, as

discussed above and were shown in the Figure 5. The left-hand-side y-axis shows these cases and
345 the total particle area.

We also estimated AE for a range of single effective sizes by assuming a narrow log-normal size
distribution (with the geometric standard deviation of 1.08) to represent a mono-disperse case for a
size range up to $0.6 \mu\text{m}$, this is shown by a black line corresponding to the right-hand-side y-axis.
This AE estimate was calculated from our modeled extinction efficiencies at 500nm and 340nm.
350 Since the extinction efficiency multiplied by the total particle area gives the total extinction, this
choice of plots in RHS and LHS y-axes gives an opportunity to assess the impact of different particle
sizes in the total AE, with the following interpretation. The relative contribution from any single
particle size to total AE can be estimated from mono-disperse AE weighted by the total particle area
of this given particle size.

355 At 820 m cloud activation has not affected the fine mode size distribution while higher in the
cloud at 970 m particles larger than the activation diameter have been removed from the fine mode.
The activation diameter in the Walker Branch simulation is at around $0.4 \mu\text{m}$ (red solid line) and
it is smaller for our two Lanai cases (blue and green solid lines). As demonstrated in Figure 5, the
growth factor of 2.2 was required for our NaCl simulation to produce positive dAE . And indeed with
360 the help of the Figure 5 this can be understood, if we think about total AE as a convolution between
mono-disperse AE and particle total area over particle sizes. For instance, convolution between black
line and blue line for our Lanai case with the growth factor of 2. The lower the activation limit the
lower is the contribution from sizes with negative AE, and thus larger is the total AE. The green line
case, with the growth factor of 2.2, has reached low enough activation size to produce large enough
365 AE for cloudy case and thus also positive dAE as was shown in the Figure 5.

4 Conclusions

The studies of aerosol–cloud interactions, exploiting remote sensing measurements, are challenging
and therefore many aspects have remained poorly known. Typically the aerosol optical properties
can be measured by passive remote sensing approaches only for clear-sky conditions. Active remote
370 sensing (mainly Lidar) does not suffer equally about cloud adjacency effects, however the coverage
that one can reach currently by active remote sensing is more limited. There exists one remote
sensing product, so called spectral deconvolution (SDA) from AERONET, that can offer a unique
information about the cloud effect on AOD. Therefore, it is somewhat surprising that these data have
not been yet fully exploited for this purpose. We analyzed SDA for different cloud conditions to give
375 quantitative estimates for the cloud enhanced AOD values, using all the available AERONET sites.

We performed the analysis on a seasonal basis and found that regardless of the season the highest
cloud related AOD enhancements occur in East-Asia, reaching levels of AOD of about 0.1. In relative
terms, these values are in range of 10-12% higher if compared to clear-sky (Level 2) fine mode AOD.

This is not insignificant and should be taken into account for, e.g. in the calculations of aerosol
380 radiative effects. On the other hand, the difference over all the included sites is rather notable as
well, e.g. in JJA fine mode AOD of all-sky data is 0.011 higher than the mean based on Level 2 only
(0.154), thus all-sky fine mode AOD being about 7% higher.

We estimated similarly the differences in fine mode AE, between cloudy- and clear-sky cases. In
majority of the cases, negative AE differences were typically prevailing. These cases are likely dom-
385 inated by particle growth in the humid conditions over the cloud activation. There were only about
ten sites of clearly positive dAE , all being strongly affected by marine aerosols. It is noted that the
AE changes were rather small, only few percent. Small new accumulation particles from both growth
of Aitken sized particles and gas-to-particle conversion may counterbalance humidification growth
of some existing accumulation mode particles, thereby resulting in little change in AE. However,
390 a more detailed analysis with a better information on aerosol composition is needed to explore the
strength of competing effects.

Albeit overall dAE was small, in the marine cases the negative dAE cases were essentially
missing, thus suggesting that different processes dominate if compared to the continental cases.
Therefore, we investigated in more detail, with the help of cloud parcel model, the relative role of
395 aerosol hygroscopic growth and cloud activation in different cloudy conditions. Our model sim-
ulations demonstrated that cloud activation can affect and remove particles from the AERONET-
measured fine mode AOD, resulting in positive dAE between L0 and L1 measurements. However,
this requires highly hygroscopic aerosol composition (sea salt) with sufficiently small dry sizes.

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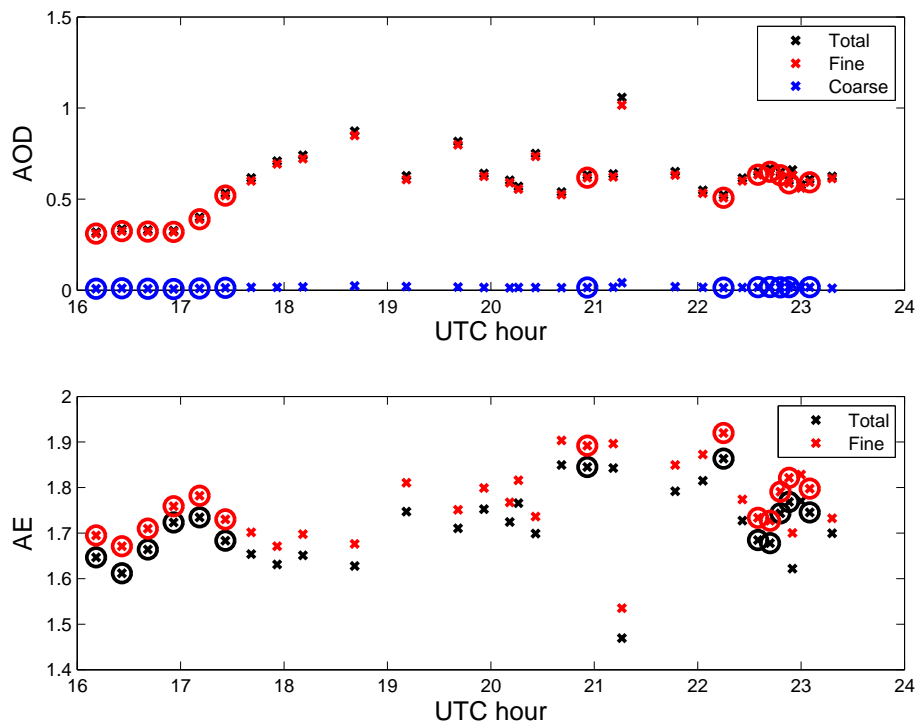


Figure 1. AOD (upper plot) and Angstrom exponent (lower plot) in BLDND site near Essex July 5, 2011. Red, blue and black symbols are for fine mode, coarse mode and total, respectively. Level 1 and Level 2 measurements are indicated by cross and circle, respectively.

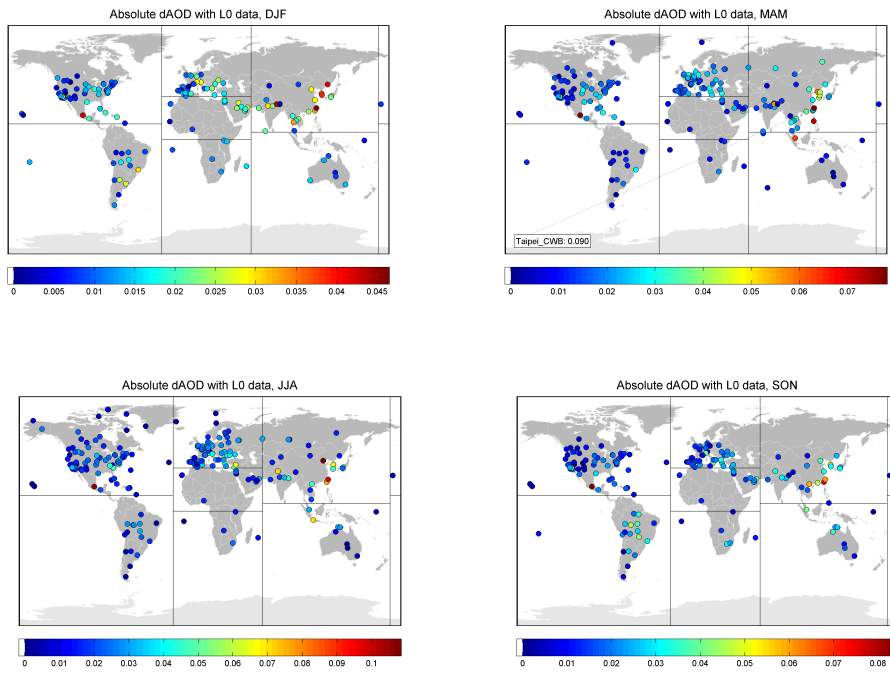


Figure 2. The difference between cloudy-sky and clear-sky (between L0 and L2) fine mode AOD for different seasons: DJF (in the upper left corner), MAM (in the upper right corner), JJA (in the lower left corner), and SON (in the lower right corner).

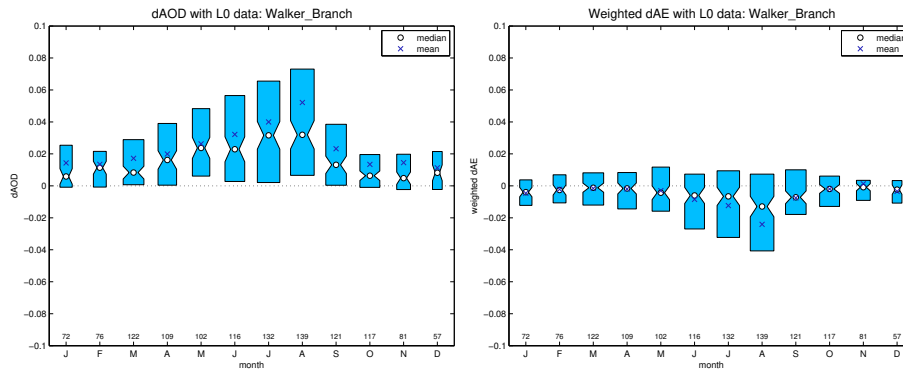


Figure 3. Monthly $dAOD$ and dAE (difference in L0 AE and L2 AE) for Walker Branch, TN, USA. Numbers at the bottom of the figure indicate the amount of measurements per month included in the box-plot.

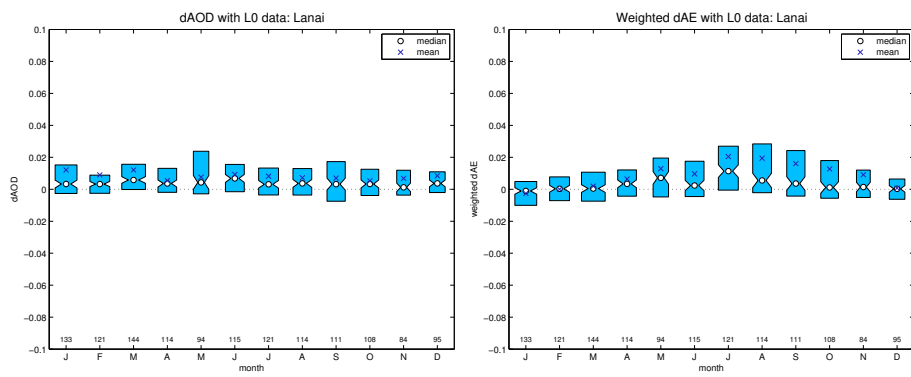


Figure 4. Similar to Figure 3, but for Lanai, Hawaii, USA.

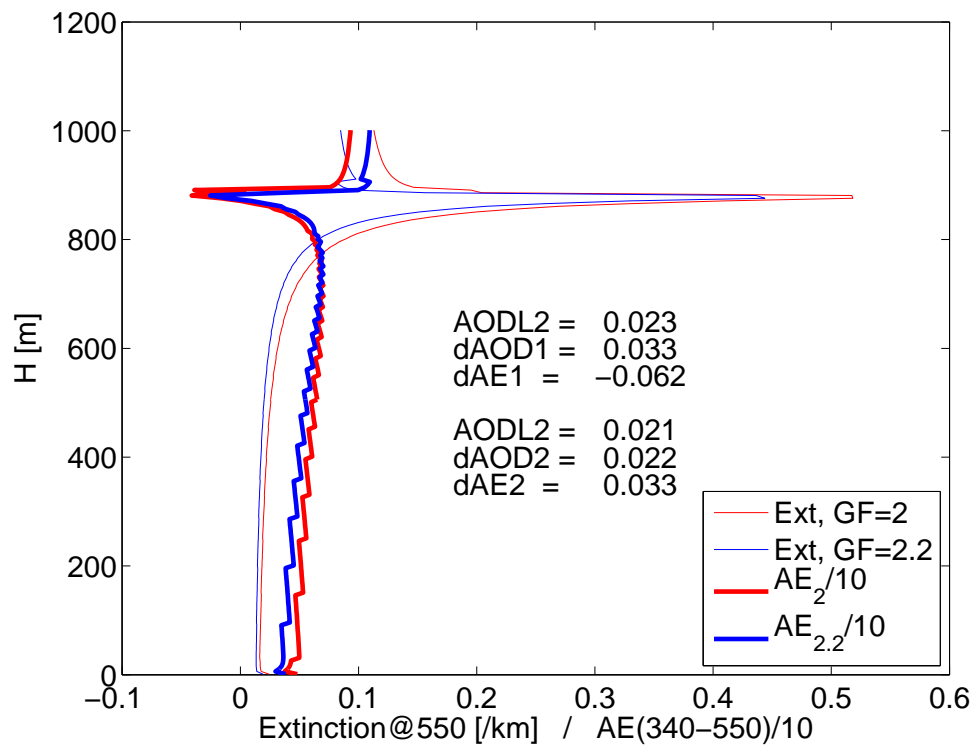


Figure 5. Profile of extinction at 500nm and Angstrom exponent from 340-500nm wavelength pair. The latter is divided by 10, in order to better match the x-scale. The numbers within the figure are given for the simulations of two different cases of growth factors (2 and 2.2). The first three numbers are for the case of growth factor of 2 as follows: AOD of clear-sky profile, “*AODL2*”; AOD difference between cloudy and clear-sky profiles, “*dAOD1*”; AE difference between cloudy and clear-sky profile, “*dAE1*”. The next three numbers correspond similarly to the case of increased growth factor of 2.2,

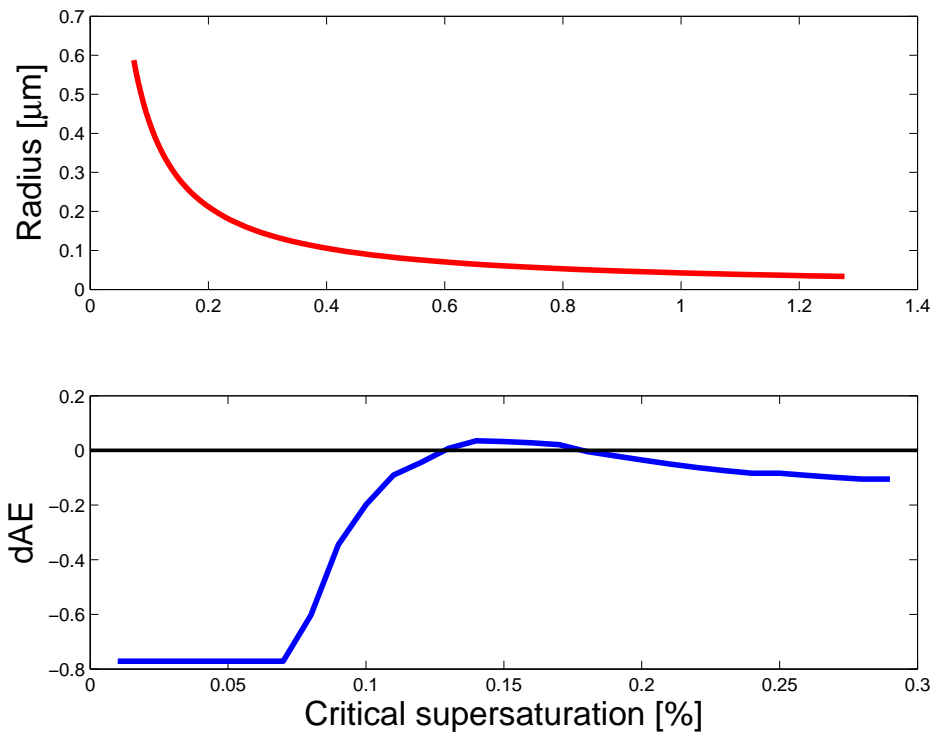


Figure 6. Critical cloud droplet activation radius as a function of critical supersaturation (upper plot) and estimated difference in AE, between cloudy- and clear-sky case (lower plot).

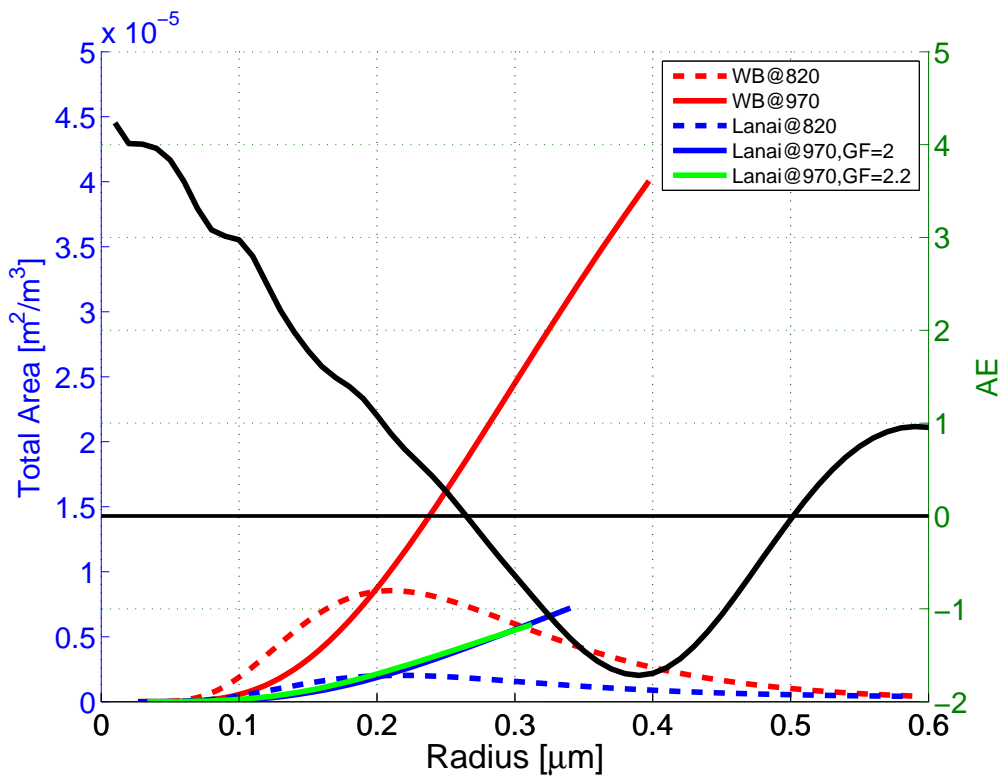


Figure 7. Total aerosol area per volume (LHS y-axis) at different model levels, WB refers to Walker Branch, e.g. WB@820 means Walker Branch case and altitude of 820m. Lanai is shown for two growth factors, 2 and 2.2, as discussed in more details in the text. RHS y-axis shows Angstrom exponent, as a estimated for mono-disperse aerosol of given radius.

Table 1. Seasonal AOD based on sampling for different cloudiness. L1 and L2 refer to Level 1 and Level 2 of AERONET data, respectively. L0 refers to those cases of L1, which did not belong to L2, thus cloudy cases only. Different regions have the following abbreviations: NAm (North America), SAm (South America), Eu (Europe), NAf (North Africa), SAf (South Africa), As (Asia), Aus (Australasia). These regions are indicated by the solid lines in the Figure 2.

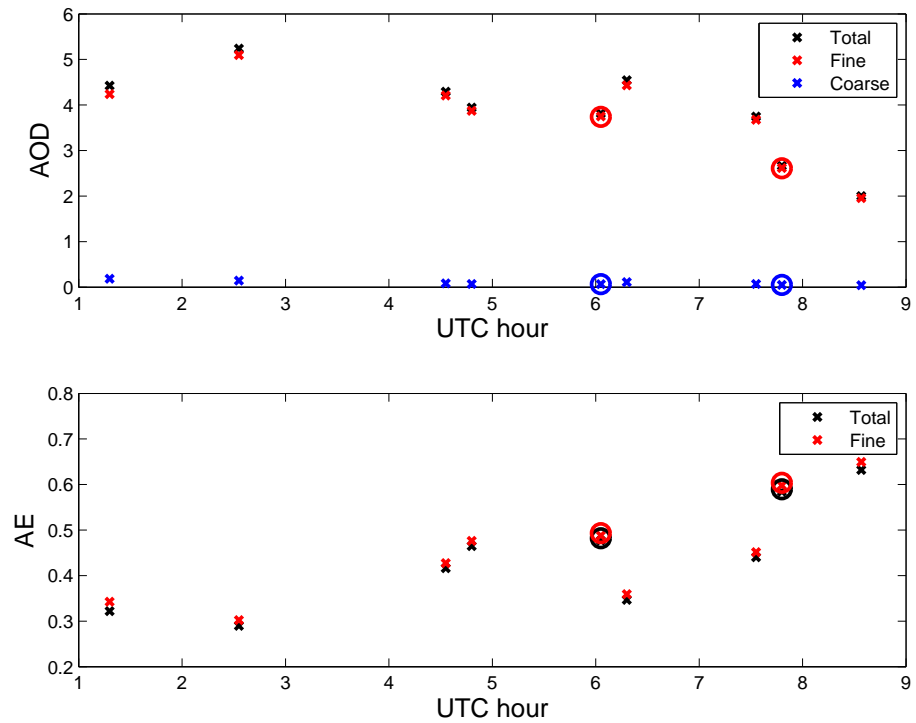
NAm					SAm			
AOD	DJF	MAM	JJA	SON	DJF	MAM	JJA	SON
L1	0.052	0.096	0.147	0.078	0.088	0.065	0.126	0.249
L2	0.047	0.088	0.137	0.073	0.074	0.057	0.119	0.237
L0	0.059	0.105	0.158	0.087	0.095	0.071	0.139	0.262
d12	+0.005	+0.008	+0.010	+0.005	+0.014	+0.008	+0.007	+0.012
d02	+0.012	+0.017	+0.021	+0.014	+0.020	+0.014	+0.020	+0.025
Eu					NAf			
AOD	DJF	MAM	JJA	SON	DJF	MAM	JJA	SON
L1	0.092	0.137	0.152	0.108	0.124	0.112	0.146	0.138
L2	0.089	0.129	0.144	0.103	0.120	0.108	0.142	0.134
L0	0.101	0.149	0.168	0.118	0.136	0.122	0.159	0.153
d12	+0.003	+0.008	+0.009	+0.005	+0.005	+0.004	+0.004	+0.004
d02	+0.013	+0.020	+0.025	+0.015	+0.016	+0.014	+0.017	+0.019
SAf					As			
AOD	DJF	MAM	JJA	SON	DJF	MAM	JJA	SON
L1	0.126	0.084	0.141	0.168	0.289	0.319	0.262	0.268
L2	0.101	0.074	0.135	0.160	0.278	0.304	0.240	0.254
L0	0.139	0.092	0.150	0.174	0.303	0.332	0.273	0.282
d12	+0.026	+0.010	+0.006	+0.008	+0.011	+0.015	+0.022	+0.014
d02	+0.038	+0.018	+0.015	+0.014	+0.025	+0.029	+0.033	+0.028
Aus								
AOD	DJF	MAM	JJA	SON				
L1	0.095	0.093	0.110	0.140				
L2	0.085	0.083	0.099	0.129				
L0	0.102	0.098	0.118	0.148				
d12	+0.010	+0.010	+0.011	+0.011				
d02	+0.016	+0.016	+0.019	+0.019				

Table 2. Otherwise similar to Table 1, but showing overall results for all the sites.

All sites				
AOD	DJF	MAM	JJA	SON
L1	0.119	0.145	0.165	0.143
L2	0.112	0.136	0.154	0.135
L0	0.128	0.155	0.178	0.154
d12	+0.007	+0.009	+0.011	+0.008
d02	+0.016	+0.020	+0.024	+0.018

Supplement of
Assessment of cloud related fine mode AOD
enhancements based on AERONET SDA product

January 4, 2017



2
 Figure S1: AOD (upper plot) and Angstrom exponent (lower plot) in XiangHe August 10, 2010. Red, blue and black symbols are for fine mode, coarse mode and total, respectively. Level 1 and Level 1.5 measurements are indicated by cross and circle, respectively. This day did not include any measurements in Level 2, since AERONET requires a minimum of 3 points per day in Level 2.

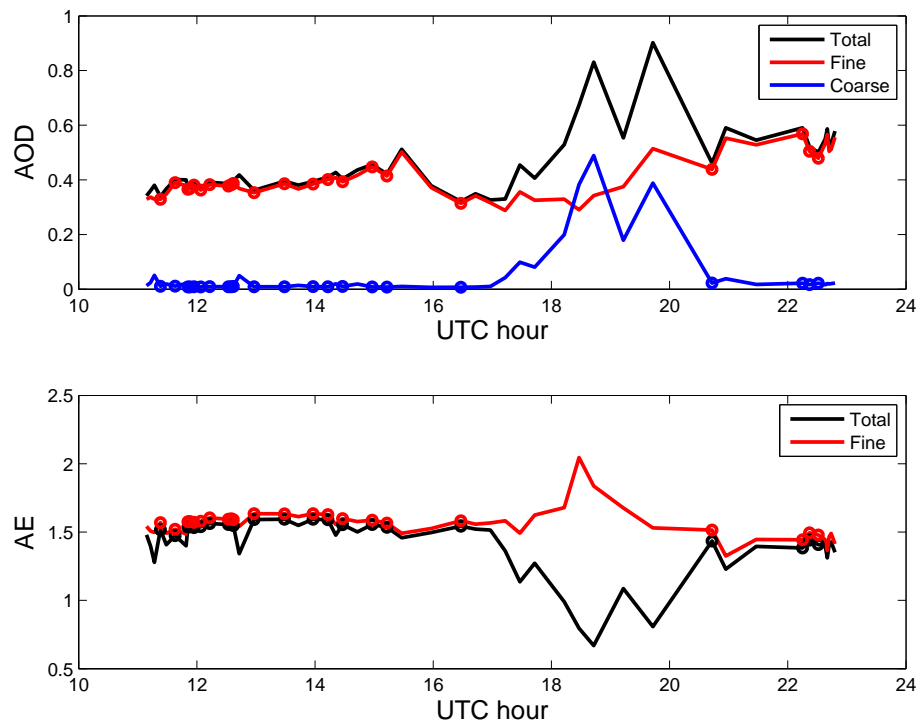


Figure S2: AOD (upper plot) and Angstrom exponent (lower plot) in GSFC August 11, 2010. Red, blue and black symbols/lines are for fine mode, coarse mode and total, respectively. Level 1 and Level 2 measurements are indicated by cross and circle, respectively.

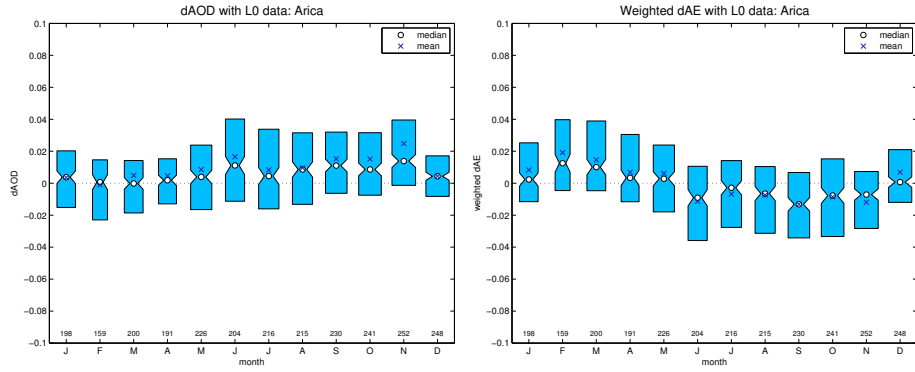


Figure S3: Monthly $dAOD$ and dAE (difference in L0 AE and L2 AE) for Arica, Chile.

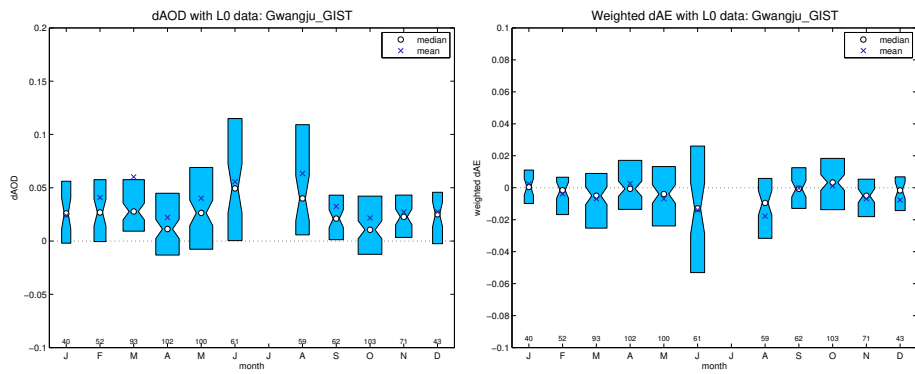


Figure S4: Similar to Figure S3, but for Gwangju GIST, South Korea.

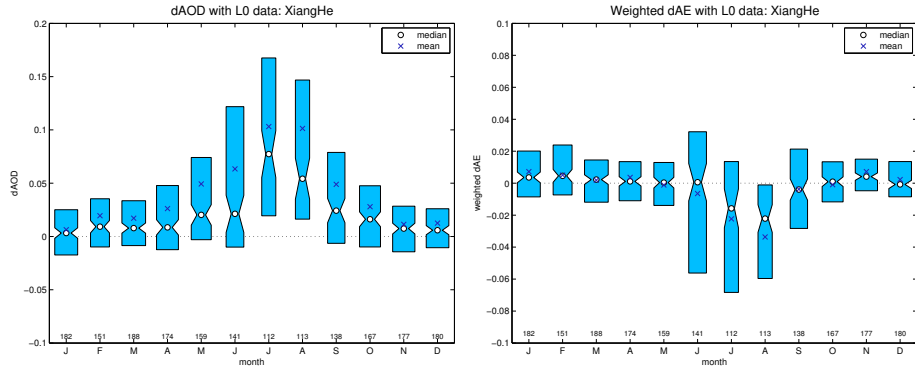


Figure S5: Similar to Figure S3, but for XiangHe, China.

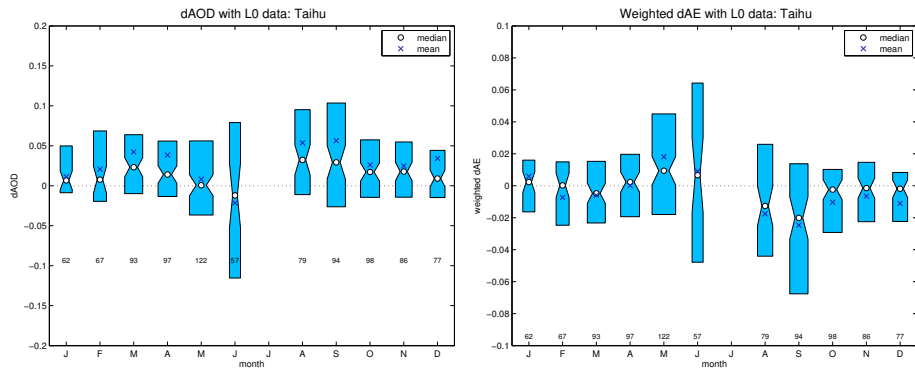


Figure S6: Similar to Figure S3, but for Taihu, China.