

## ***Interactive comment on “Is there a bias in AERONET retrievals of aerosol light absorption at low AOD conditions?” by Elisabeth Andrews et al.***

**Anonymous Referee #3**

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ACPD Review:

Is there a bias in AERONET retrievals of aerosol light absorption at low AOD conditions?

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General comments: The authors present data of situ measurements from aircraft profile flights from which calculations of AOD, single scattering albedo (SSA), and Absorption Aerosol Optical Depth (AAOD) are compared to remote sensing measurements (of AOD) and retrievals of SSA and AAOD from AERONET sun-sky radiometers. These comparisons are made for two sites in the USA and for primarily low AOD levels, mostly less than  $\sim 0.25$  at 440 nm. This is well below the AERONET recommendations for use of absorption parameters from their retrievals ( $>0.4$  at 440 nm is recommended), and

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although the authors discuss this in the text this recommended low AOD threshold is conspicuously absent from both the Abstract and Conclusions sections (and this needs to be remedied). The authors state that in prior publications "... the in-situ derived AOD values tend to be slightly lower than the AOD retrieved from remote sensing measurements." They fail to point out that the in situ measurements rarely if ever measure the total column AOD, which includes both mid- to upper-tropospheric aerosol plus stratospheric AOD. The authors should include some references and discussion on the AOD that is not measured by in situ instruments in the upper troposphere and stratosphere since the aircraft do not fly complete profiles from the surface on upward into the stratosphere. Discussion of the fact that the aircraft profiles presented (with 4.2 km above ground level as the maximum in situ sampling altitude) do not actually measure the total atmospheric column AOD needs to be included in this manuscript. Therefore differences in in situ versus AERONET AOD are indeed expected and the AOD would be expected to be somewhat higher for sunphotometer total column AOD than for in situ in most aircraft sampling strategies. Moreover lidar measurements sometimes show mid to upper altitude aerosol layers that this aircraft sampling strategy (max at 4.2 km agl) would not measure. Additionally, Sunphotometers in general and AERONET instruments in particular measure AOD more directly than any other technique and as such these data are considered by the scientific community to be the gold standard of accurate AOD measurement for the total atmospheric column. AERONET measured AOD represent the ambient aerosol optical properties and do not have to be corrected for RH humidification growth effects, loss of large particle sampling, etc. as is required and/or discussed for in situ data utilized in this paper. Nyeki et al. (2012) found that AERONET measured AOD agrees very well with other well-calibrated sunphotometers. At the Davos, Switzerland site the comparison of the time co-located and matched 500 nm AOD differences between AERONET and GAW-PFR from 2007 through 2010 resulted in a mean AOD difference of  $-0.0024$  and a root-mean square error of  $0.0071$ . These issues should be included in the discussion on AERONET data, and in the section on comparison of AOD from AERONET measurements to in situ

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estimates. Accuracy of AOD is very important in this paper as AAOD is derived from AOD values and the AOD values derived from aircraft profiles (after corrections to make ambient estimations) and also from models (such as within AEROCOM) can be either biased or have significant uncertainties. Furthermore, in order to better understand the comparisons of aircraft profiles to AERONET measurements a scatterplot of computed Extinction Angstrom Exponent (EAE; 440 - 675 nm) estimated from the aircraft data versus AERONET measured EAE needs to be added to Figure 3. This comparison of EAE is pertinent to the discussion in the current text of aircraft inlet sampling issues regarding possible large-sized particle losses.

There is a lack of discussion in the paper of how the uncertainties of the in situ measurements change as aerosol concentrations decrease. All measurement methodologies suffer from issues related to a decrease in signal at low concentrations (relative to potential instrumental noise and offsets), therefore I think that discussion of how the in situ measurement uncertainty changes with aerosol concentration is a very important aspect that needs to be included in the manuscript. Since the paper focuses primarily on low AOD cases, this is a critical issue that is surprisingly neglected in the current manuscript. Additionally it is necessary to summarize in the text a description of the methodology used for computation of profile weighting of the in situ SSA estimates during each aircraft flight. Are these SSA values at each altitude weighted by the extinction coefficient at that altitude, thereby effectively giving higher weighting to the measurements at altitudes that had the highest aerosol concentrations? The AERONET retrievals of SSA are effective optical extinction weighted values for the total atmospheric column, therefore extinction weighting of the in situ data would be the most rigorous way to compare similar quantities. The authors also need to show plots of the in situ aircraft measured/computed SSA altitude profiles to show how SSA varies as a function of altitude for several days of varying AOD magnitude. This is important as it can provide some needed information on how the in situ SSA measurement data look at very low concentrations, especially higher than 3 km above ground level on very low AOD days and also on some moderately high AOD days.

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In the abstract you state: "The tendency of AERONET inversions to overestimate absorption at low AOD values is generally consistent with other published comparisons." However the published comparisons between AERONET retrievals and in situ measured SSA shown in Figure 6 are not for low AOD (the AOD are moderate to high in the Figure) and also the SSA differences are generally within the combined uncertainty estimates of the two different techniques (see numerous additional comments on Figure 6 data below in 'Specific Comments'). Since it has never been established that the in situ measurements of SSA have no bias of their own then it is not possible to say that the AERONET measurements of SSA at moderate to high AOD are biased since there is no absolute benchmark for comparison purposes. Additionally since you state that the science is unclear on absorption enhancement due to coated absorbing particles (Section 2.4.1, line 404-407) you need to give a detailed explanation as to how this unknown factor was incorporated into the uncertainty estimates you made for the in situ measured single scattering albedo (it seems to have been ignored in your estimates). The aircraft sampled aerosols are dried first and therefore true atmospheric ambient state aerosol optical properties are actually not measured directly during the profiles. This is important regarding your claim of relative bias of single scattering albedo from one measurement type versus another since you cannot rigorously state (or indirectly suggest) that the aircraft measurements of single scattering albedo are unbiased given that the ambient state optical properties of the aerosols were not directly measured by your in-situ instruments.

The climatological comparison of in situ and AERONET values in Figure 7 is a very important figure in this paper. This figure suggests that if AERONET data are wisely utilized (as done by Bond et al. (2013), for example) then the SSA differences between the two methodologies can be relatively small. The large differences in the time matched data at Bondville and SGP sites from in situ flights and the AERONET retrievals shown in previous figures (Figs. 4-6) are not nearly as evident in Figure 7, especially for the SGP site. There is a surprising lack of discussion of this apparent discrepancy between the matched aircraft profile/AERONET data and the 'climatologi-

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cal' comparisons and the reasons for it. There is also a surprising lack of emphasis on the SSA comparison results shown in Figure 7 in the Abstract and Conclusions given the importance of this result. Additionally it is very interesting that in Figure 8 the in situ surface measurements of SSA agree quite well with AERONET retrieved SSA for both sites, with excellent agreement at SGP site and within uncertainty bounds for the BND site except for extremely low AOD of less than 0.05. The authors have stated that the RH for the surface measurements are all <40%, although this is somewhat surprising given that the surface RH is typically >40% at this location, or perhaps measurements are never made when RH exceeds 40%? Or maybe the surface data that are shown are for the dried aerosol only? If so, you should apply the same humidification factors to the surface in situ data that you have applied to the aircraft profile data in this analysis to make the comparisons in Figure 8 consistent. A look at climatological data for Enid, Oklahoma and Ponca City, Oklahoma (same region as the SGP site) show daily average surface minimum RH of >40% and average Maximum RH of >75-80% for almost all days of the year. The surface in situ measured SSA to AERONET retrieval comparison result may be particularly interesting since the aerosol concentrations are often highest near the surface and therefore the in situ measurements made at the surface should have less uncertainty than those made at high altitudes where the concentrations may be very low. The authors should also present a comparison of the SSA and Extinction Angstrom Exponent measurements made at the lowest flight altitudes during profiles to those made at the surface by similar in situ instrumentation to show how good the agreement is between these measurements and to prove that the aircraft inlet sampling issues mentioned in the manuscript do not result in significant measurement uncertainties.

Specific Comments:

Abstract: You also state: "We conclude that scaling modeled black carbon concentrations upwards to match AERONET retrievals of AAOD may lead to aerosol absorption overestimates in regions of low AOD." This statement is somewhat simplistic and mis-

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leading since it does not reflect the much better comparisons shown in Figure 7 for 'climatological' analyses. It also ignores the well thought out application of the use of AERONET retrieved SSA values as weighted by higher AOD observations and then applied to highly accurate AOD measurements at all AOD levels from AERONET, similar to the approach of Bond et al. (2013).

Introduction (lines 77-79): You state: "Moreover, by invalidating low AOD cases, the AAOD values that are retained in AERONET Level-2 data may be biased high." Again, it is misleading and simplistic to suggest that careful investigators would take the AAOD values from only Level 2 data and assume that they can be utilized as is. Many researchers have already utilized a much more intelligent approach: first estimate SSA at higher AOD from AERONET, and then apply those values to ALL levels of AOD (see Bond et al., 2013). I suggest that you remove or modify this sentence.

Introduction (lines 142-143): I assume you mean Dubovik et al. (2000). Dubovik et al. (2002) is not in the reference list.

Section 2.1 (lines 185-187): Please elaborate what you mean by improving measurement statistics here. It would seem that the aircraft instruments 10-minute sampling rate at higher altitudes is an attempt to overcome issues associated with low aerosol concentrations and associated limits of instrumental sensitivity. Therefore on very low AOD days it would seem that an even longer time interval than 10 minutes would be justified. Please elaborate on the sampling strategy and state whether it was modified for very low aerosol concentrations (very low AOD days).

Section 2.1 (line 194): "Only complete profiles were used in this analysis." Please state here that complete profiles as made by the aircraft do not equate to complete atmospheric profiles. None of the aerosol from 4.2 km agl through the stratosphere is sampled in the flights. Especially for very some low AOD days and (and also for some moderate-high AOD days in summer with strong convection) it is expected that a significant amount of the AOD actually occurs above 4.2 km agl. These upper aerosol

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layers that are often seen in lidar data may have different optical properties than lower altitude aerosols.

Section 2.1 (line 216-219): Discuss how assuming a constant hygroscopic growth parameter would cause uncertainties when seasonal variation in aerosol type exists. Especially in spring, aerosol type may include biomass burning (crop waste or grass burning) and also dust from the Great Plains region (see Ginoux et al. (2010)), plus pollen from grass and trees.

Section 2.1 (line 267): Please change “column average properties” here to “flight profile average properties” to accurately reflect the fact that the aircraft does not measure the total atmospheric column, as AERONET does.

Section 2.2 (line 290-292): Please add ‘calibrations’ before ‘corrections’ as the consistent high accuracy calibration of AOD and sky radiances are the basis for what makes AERONET data so valuable.

Section 2.2 (line 294): Please add that the AERONET data is Version 2 data, since the Version 3 database will be available in the near future.

Section 2.4.1 (line 356-358): Please discuss whether you accounted for soil dust and biomass burning aerosols in the ‘aerosol chemistry’ mentioned here.

Section 2.4.1 (line 361-364): In your discussion of RH levels during the profile flights please include some mention of the higher RH (RH halos) that typically exist in the vicinity of non-precipitating cumulus clouds that are imbedded within the aerosol layer < 4 km. Higher RH near cumulus clouds and higher AOD in the near Cu cloud environment (likely due to combined humidification, cloud processing of aerosols and rapid gas-to-particle conversions) were observed by Jeong and Li (2010) at the SGP site utilizing both AERONET data and in situ aircraft data. If you only flew aircraft profiles on cloudless time periods or avoided flying near clouds then this needs to be documented in the manuscript, as the sampling could possibly be skewed to specific meteorological

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and/or cloudiness conditions.

Section 2.4.1 (line 401-402): Although biomass burning does not have a consistent influence at these sites it is episodic, therefore did you exclude these biomass burning aerosol episodes from your data analysis? If so how did you identify the biomass burning episodes?

Section 2.4.1 (line 443-445): Please state here that the uncertainty estimate for the in situ SSA of  $\sim 0.04$  is a lower bound since it does not take into account the effect of particle coatings on aerosols since the aerosols are modified (dried) before the measurements are taken, plus some fraction of the coarse mode particles are not sampled.

Section 2.4.1 (line 451-454): You state here that  $\sim 15\%$  of the aerosol in the column is not sampled below the lowest flight altitude (150 m agl) of the aircraft for in situ measurements, and it can also be inferred that possibly another 15% or more is not sampled above the highest flight altitude on very low AOD days or high AOD days with layering from convective vertical aerosol transport. Therefore it is likely that  $\sim 30\%$  of the aerosol in the total atmospheric column is not sampled by your aircraft vertical profiles. This issue needs some discussion in the text and also should be factored into your uncertainties of in situ measured SSA (or clearly state that it has been ignored).

Section 2.4.1 (line 464-467): Please elaborate here on whether the estimates of the percentage of aerosols above the highest flight altitudes as analyzed by Andrews et al. (2004) were comparisons made for all AOD levels and seasons. It would not be surprising for a greater percentage of AOD above flight altitudes to occur in summer when convection is stronger (transporting boundary layer aerosols upwards), or also in all seasons when AOD is very low since there is always some background mid-tropospheric to stratospheric AOD present which constitutes a greater percentage of total AOD when AOD magnitudes are very low.

Figure 3: In Figure 3, please explain how you can have 3 observation points of AERONET measured AOD at 675 nm ranging from  $\sim 0.5$  to  $\sim 1.0$  at INSITU AOD of

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~0.15 when there does not seem to be any corresponding data at 440 nm in the plot above it. This does not seem possible, and should be explained in the text.

Section 3 (line 528-530): Please also add to this paragraph that fact that the in situ under sampling of the total atmospheric column AOD is due to the restricted altitudes of the flight profile measurements (150 meters to 4200 meters).

Section 3.1.2 (line 604-607): Please note that the in situ instrument known cutoff of 5 micron for particle diameter for the aircraft sampling would also contribute to an under sampling of total column AOD, in addition to the incomplete altitudinal atmospheric profile for the total column AOD.

Figure 6 (numerous comments follow regarding some of the referenced data sets plotted in the Figure, especially note the issues regarding aircraft sampling and also the fact that some papers published Version 1 data that were biased due to inaccurate surface albedo assumptions, versus current Version 2 data that became available in 2006): Osborne et al. [2008] compared three cases of aircraft flights (on three different days) over the same site during the same experiment with the same instruments and aircraft but found that the aircraft in situ measured SSA values ranged from 0.04 to 0.07 higher than the AERONET version 2 retrievals. However, for all three of these cases the aircraft measured Angstrom exponents were found to be about 0.40 lower than the AERONET measured values. This discrepancy in AE suggests that the aircraft may have sampled a different fine and coarse mode fraction mixture than the column integrated value measured by AERONET, and the higher SSA in conjunction with lower AE measured by the aircraft is consistent with this possibility. In fact, for the linear fit of SSA versus AE for all aircraft data from DABEX, reported in the work of Johnson et al. [2008], a difference of 0.40 in AE corresponds to a difference in SSA of about 0.06, almost the same value of the bias reported in Osborne et al. [2008].

Johnson et al. [2009] compared in situ measured aerosol optical properties from an aircraft vertical profile flight over the Banizoumbou (Niger) AERONET site on 19 Jan-

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uary 2006. This was a mixed aerosol case with Angstrom exponent (450–700 nm) of approx. 0.8–0.9 and high 550 nm AOD of approx. 0.75, where a shallow dust layer up to 1 km altitude was overlain by a layer of predominantly fine mode smoke. Both aircraft and AERONET measurements of column integrated AOD at 550 nm and of AE were in good agreement for this case, with  $dAOD = 0.08$  and  $dAE = 0.06$ , suggesting that both were sampling the same aerosol mixture. The aircraft measured column mean SSA at 550 nm (from PSAP and nephelometer) was 0.87, in good agreement with the AERONET retrieval of 0.85 (interpolated to 550 nm).

Magi et al. (2005; JAS) Note: Version 1 retrievals were 0.015 lower than V2 retrievals on this day at 1310 UTC at COVE site: From the paper: “Ground-based retrievals of SSA were obtained by the Aerosol Robotic Network (AERONET) sun photometers (e.g., Dubovik et al. 2000) during the CLAMS field campaign from a site known as the Clouds and the Earth’s Radiant Energy System (CERES) Ocean Validation Experiment (COVE; 36.98 N, 75.78 W). The vertical profiles were often spatially located close to COVE. The mean value of SSA at 550 nm from AERONET retrieval data (processed to remove clouds and manually quality assured) is  $0.94 \pm 0.03$ . Therefore, the mean value of SSA retrieved from AERONET agrees with mean value of SSA derived from our in situ airborne measurements ( $0.96 \pm 0.03$ ) to within one standard deviation. On 17 July 2001, measurements were made from the UW aircraft and the COVE site that were both temporally (the aircraft vertical profile was from 1304–1337 UTC and the AERONET retrieval was at 1310 UTC) and spatially (the aircraft was ; 2.5 km from COVE) collocated. The mean value of SSA calculated from the airborne in situ measurements made in polluted layers during this vertical profile was  $0.97 \pm 0.02$ ; the corresponding column-averaged value of SSA for accumulation mode particles retrieved from the AERONET data was  $0.90 \pm 0.03$  (VERSION 1 data). Particle losses in the sampling system for the in situ instruments could have contributed to an underestimate of the absorbing component of the aerosol. Spatial variability may have played a role as well.”

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Mallet et al. (2005): V2 almost same as V1 at 0.932 at 550 nm at 6 UTC: AERONET retrieval (at 0600 UTC ; on June 25, 2001) of the single scattering albedo at Avignon indicated a coherent value (SSA 0.93 at 550 nm) compared to the one obtained from optical measurements for flight 41 (0515–0537 UTC, SSA 0.94 in the PBL).

Haywood et al., (2003): Comparison of aerosol size distributions, radiative properties, and optical depths determined by aircraft observations and Sun photometers during SAFARI 2000 V2 SSA = 0.84, 0.83, 0.81, 0.80 for 440, 675, 870 and 1020 nm V1 SSA = 0.88, 0.87, 0.84, 0.82 for 440, 675, 870 and 1020 nm The corresponding SSA for the mean size distribution used in the calculations derived from the PCASP distributions and from the nephelometer and PSAP on the C-130 is 0.90, 0.87, 0.85, and 0.82 (Table 1).

Figure 7 caption: One detail regarding the caption of Figure 7 that is misleading is the Level 2 AOD shown in the plot. Please note that Level 2 AOD exists for all AOD levels not just for AERONET almucantar retrievals for which AOD is >0.4 at 440 nm. Level 2 almucantar retrievals of size distributions are made for all AOD levels, but refractive indices are only given for AOD>0.4 at 440 nm. Therefore the authors need to clearly describe and accurately label in the figure caption that this data as only associated with AERONET almucantar retrievals for which AOD>0.4 and therefore have error bars on SSA of ~0.03. The actual complete Level 2 AOD data set (for all AOD levels) shows monthly means that are significantly higher in summer with many more days of data sampled and many more partly cloudy to mostly cloudy days sampled also (see Jeong et al., 2010; JGR for a discussion of higher AOD in the near cloud environments at the SGP site).

Section 3.2 (line 692-695): Your statements here assume that the in situ determinations of SSA are un-biased (despite the fact that ambient aerosol properties are not actually measured). This has not been proven in the paper, especially since the in situ data have to be corrected for humidification effects, the total column aerosol is not sampled, and the effects of aerosol coatings are not accounted for (therefore blindly assumed

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have no effect). Please revise or eliminate these sentences.

Section 3.3 (line 754-768): Please note that AOD sampled by AERONET in the Level 2 dataset (not just for the subset that have L2 retrievals) includes many more days of data than the in situ flights, and is therefore a much more statistically robust data sample. Please note that the Level 2 AOD climatology for the SGP site (average of 13-19 years per month) shows significantly higher AOD (440 nm) than shown for L1.5\* in Figure 7, For example for the SGP site the August monthly mean AOD is 0.272 and the September monthly mean is 0.215 at 440 nm. Similarly for the BND site the L2 monthly means of AOD(440 nm) for June, July, August, and September are 0.282, 0.329, 0.343 and 0.283 respectively (computed from 15-17 years of data per month). These monthly means are significantly higher than the AERONET values shown in Figure 7, since the data in Fig 7 are only AOD associated with the Dubovik and King algorithm retrievals.

Section 3.3 (line 781-783): You state: “The AERONET 1.5\* SSA values tend to be quite a bit lower than the other data sets at both sites, which is why the AERONET 1.5\* AAOD values tend to be higher (recall that for AERONET data AAOD is calculated using  $AAOD=(1-SSA)*AOD$ .” No, this is not really accurate, since as shown on Figure 7, at the SGP site the agreement between the AERONET L1.5\* data and in situ measurements of SSA are well within the uncertainty of the measurements for all months (and you have not proved that the in situ is not biased). Please revise this sentence to reflect this fact as presented by the data shown in Figure 7b.

Section 3.4 (line 875-879): Again, you have omitted the fact that for the SGP site the agreement between the in situ estimations of SSA and the AERONET retrievals of SSA are within the uncertainty levels of these data sets over the entire range of AOD shown in Figure 8. The way the paper is written there seems to be a consistent attempt to steer the reader to the conclusion that the AERONET retrievals are biased low despite significant uncertainties in the in situ determinations and despite the fact that the in situ instruments do not measure ambient aerosol properties directly without corrections. Therefore it is not proven that the in situ determinations of SSA are unbiased

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themselves, so the text and title require rewriting to acknowledge this.

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