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Atmospheric Chemistry and Physics Discussions

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 #1

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This work compares AERONET column retrievals of aerosol single-scatter albedo to column single-scatter albedos obtained from in situ flight profiles. This is an important topic, because modeling groups regularly use the AERONET absorption products to infer or constrain atmospheric black carbon concentrations. This work focuses especially on assessing the capability of the Level 1.5 AERONET retrievals that are obtained at low aerosol optical depth (AOD(440) < 0.4) common to many locations throughout the world, which has received little or no attention thus far. The paper is clear and well written, scientifically solid, and thoroughly covers nearly all issues associated with this topic. I have a few topical comments and a laundry list of small details that will make the paper stronger, in my opinion.

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Discussion paper

Column Single-scatter Albedos

Once concern is the hygroscopic growth corrections for the nephelometer and PSAP measurements. Since particles are dried prior to entering the aircraft instruments, the authors employ an empirical correction to the scattering measurements and no correction to the absorption measurements. The scattering correction is an exponential function of RH (Eq. 1), and the exponents that they choose for the corrections are based upon aerosol chemistry measurements at nearby IMPROVE sites. This is all very standard in the business, and the pitfalls of empirical scattering corrections at high RH are well documented. The authors argue that they are mostly operating at low RH, so that their column AODs are not very sensitive to the hygroscopic growth corrections.

This is all fine, except that Figure 3 shows a significant amount of scatter in the author's AOD comparisons that undoubtedly works its way into the SSA comparisons. This is unavoidable when applying climatological hygroscopic growth corrections to in situ scattering profiles, but why not use AERONET AODs to compute column SSAs? Unlike AERONET's absorption and size distribution *retrievals*, AERONET AOD is a robust *measurement* that is widely accepted by the community. Thus, the authors could compute a "hybrid" SSA, whereby

$$SSA_{hybrid} = \frac{AOD_{aeronet} - AAOD_{psap}}{AOD_{aeronet}}.$$

This will isolate the differences in SSA caused by absorption, which is of primary interest to the community. This approach will also remove the uncertainty associated with the empirical hygroscopic growth factors applied to the scattering measurements, and it will remove the scattering uncertainty associated with undersampling the coarse mode. Since AAOD is very well-correlated at SGP (per Fig 4), I suspect that this approach will tighten up the SSA scatter at that site in Figure 5.

Now, some readers may find issues with this approach as well, so I suggest includ-

ing this approach as an additional alternative to the material already presented in the paper. For some of us, this approach will remove doubts about hygroscopic growth corrections and inlet size cutoffs for the scattering measurements.

Merging of in situ and AERONET data

Figure 3-6: The authors use multiple AERONET AODs and AAODs for each flight profile (i.e., the number of AERONET AODs at BND is 662, but there are only 72 flight profiles). This is not appropriate, in my opinion. The flight profiles occur over a 2-hour period, so I argue that they are comparing quasi 2-hour averages to AERONET's quasi-instantaneous retrievals. The atmosphere can easily change during this timeframe, especially since the authors allow an additional 1 hour before the flight and 3 hours after the end of the flight for including additional AERONET retrievals. Apparently the spread of AOD for a given flight profile can be quite large over this 6 hour period, too, as seen in the upper left panel of Figure 3 (for instance the AERONET AOD(440) ranged from 0.45 to 0.55 when in situ AOD(440) = 0.3). Additionally, the authors have argued that aerosols sampled within a 3-4 hour period have high autocorrelations, per their Figure 2. Since the premise of their comparison is that they are essentially sampling homogeneous air masses on average, why not use averaged AERONET values for comparison to the flight profiles? (After all, there is a heck of a lot of averaging going on for each in situ flight leg.) This will reduce the number of points in figures 3,4,5, and 6, but I believe that this will provide a more accurate presentation of the data. Having said all that, I am OK with multiple AODs per in situ measurement (red points in Fig 3), as this provides an indication of the variability in aerosol optical properties that occur in the atmosphere during the flight profiles. A comment from the authors pointing this out would be helpful, though (if I've got this correct). Multiple AERONET AAODs and SSAs per flight in Figures 4 and 5 serve no purpose, though.

The authors present autocorrelations in Figure 2, which is a robust way to identify acceptable collocation lag times. They conclude that a 3-hour lag is acceptable for their analysis. They mention that the auto-correlations of scattering is greater than 0.8 for

up to a 4-5 hr time lag (lines 316-317), but they do not specify that the auto-correlation of absorption is only 0.75 at BND and 0.55 at SGP. This should be explicitly stated here, because it is important for putting Figure 4 into context. If the auto-correlations in figure 2 are robust values computed with single instruments, then Figure 2 represents the maximum correlation that we can realistically expect to achieve in a comparison of two different instruments. Thus, $R^2 = 0.77$ at SGP in Figure 4 (or, $R = 0.87$) is actually an excellent result for AERONET, as the correlation is substantially greater than the auto-correlation in Figure 2. It is also an excellent result for the authors, as it demonstrates that they have put great care into producing a clean analysis. This is something worth mentioning in the article. Surprisingly, the BND site has higher 3-hour autocorrelations for absorption than SGP (r = 0.75 for BND and r = 0.55 for SGP, per figure 2), but the results for BND in Fig 4 are not as clean as SGP. Nonetheless, the correlation coefficients for BND in Figure 4 (R = 0.53-0.58) are not that far from the auto-correlation of 0.75 for absorption at BND in Fig 2.

This brings up another confusing point: The authors define R^2 as the correlation coefficient on lines 518, but R^2 is usually reserved for the coefficient of determination, and R is the correlation coefficient. The authors also use “r" for the auto-correlation in Figure 2, which adds to the confusion. Thus, I am unsure if the R^2 values in Figures 3-5 indicate correlation coefficients or coefficients of determinations. Additionally, how does one determine either parameter (correlation coefficient or coefficient of determination) when the number of points on the y-axis is different than the number of points on the x-axis (e.g., R2 = 0.84 for 56 AERONET retrievals and only 24 flights at BND for the upper left panel of Figure 3, etc.)? The authors need to explain how they were able to do this (mathematically) in both the text and in the figure captions.

Mixing State

It is easy to demonstrate (with publicly available Mie codes) that an internally-mixed absorbing aerosol particle has a much higher absorption coefficient than an identical externally-mixed counterpart. The amount of absorption enhancement varies with the

particle size and coating thickness, but factors of two or more absorption enhancements are reasonable. Recent measurements utilizing the SP2 instrument indicate that roughly half of biomass burning and background aerosols are internally mixed (e.g., Schwarz, GRL, 2008).

It is not clear to me that a filter-based instrument can differentiate between internally and externally mixed particles because the EM field on the particle changes when it touches the substrate. Therefore, any enhanced absorption associated with internal mixing of atmospheric carbon particles might not be detectable with a filter measurement. The altering of aerosol absorption is further aggravated by the drying process, which can modify aerosols by removing coatings that are composed of semi-volatile compounds, inorganics, and certainly water.

The authors state on line 406 that "measured absorption Ångström exponents are quite low (close to 1) suggesting little influence of coatings." This is a weak argument, however, because the absorption Ångström exponent is measured *after* the drying process, which could have easily removed semi-volatile coatings. Additionally, coated carbonaceous particles of about 0.1 μm diameter and a wide range of coating thicknesses can easily have absorption Ångström exponents close to 1, per Gyawali (ACP, 2009, Figure 8).

Finally, the authors conclude on line 434 that ". . . it is not possible to estimate the actual uncertainty in the in-situ light absorption measurements reported here due to coating effects." Since accurately estimating this uncertainty is not possible, the authors seem to be choosing an uncertainty of zero; I believe that many readers will not be satisfied with this approach.

Hence, I suggest that the authors include a more conservative uncertainty of 50% for the PSAP absorption coefficient (rather than 25%) to account for all possible PSAP errors (you've already included the 50% value in much of your discussion on pages 10-11). This number still comes out of thin air (just like zero), but you can point to the

50% enhancement factor recommended by Bond and Bergstrom (AST, 2006). Thus, I recommend including a comment on line 445 that if the PSAP absorption uncertainty is 50%, the SSA uncertainty at SSA=0.9 is approximately 0.06.

Optical and Aerodynamic sizes

The authors state that their inlet samples particles with aerodynamic diameters less than 7 μm, and that the particle diameter for 50% sampling efficiency is 5 μm (lines 196-198). They go on to address this issue on lines 601-617 by using the AERONET size distributions to estimate the fraction of aerosol extinction that occurs at AERONET diameters of less than 5 μm. By doing this, the authors are assuming that aerodynamic diameter is equal to volume equivalent diameter, which is not generally the case (unless the aerosols are spherical and the aerosol density equals 1). Since $D_{vol} \propto D_{aer}/\sqrt{\rho}$, the authors should use a much smaller cutoff diameter than 5 μm in their equation on line 606. For instance, if one assumes $\rho = 2$, then D_{vol} = 3.53 for spheical aerosols; if $\rho = 2.7$, then D_{vol} = 3.04. These smaller cutoff values will decrease their estimated extinction fractions of the particles entering their inlets. However, the authors correctly argue on lines 612-617 that under-sampling large particles probably lowers the in situ SSA, and that correcting this artifact will probably make the discrepancy between AERONET and in situ worse. Nonetheless, some recognition of the optical/aerodynamic size difference will indicate to readers that you considered this issue.

Tables and Figures

I like Table 1, but the authors need to include the number of AERONET retrievals and the number of in situ flights that are used to compute these statistics. Standard deviations of the means (SDOM) would be nice, too, so that readers can quickly see that the difference between the averages are statistically significant.

I am having difficulty understanding Table 2. You list 56 retrievals with Level 2 AOD + almucantar at BND, but only 6 retrievals with Level 2 + almucantar + $AOD_{440} > 0.2$.

The 2nd number should be greater than the first number, right? (The first number only includes AOD > 0.4, but the 2nd number includes all retrievals with AOD > 0.2). Unless... do you mean “size distribution retrieval" instead of “almucantar retrieval?" The almucantar products include size, AAOD, SSA, and complex refractive index; size distributions are the only Level 2 almucantar products available at AOD(440) < 0.4.

The legend in Figure 1 is confusing. The legend contains two solid black lines (Direct RF BCFF and Global), but there is only one solid black line in the figure. The legend also contains two variations of dashed black lines (Land and Sea), but there are no black dashed lines in the figure. The figure contains two red dashed lines that are not shown in the legend.

The authors need to explain that “r" represents auto-correlation in Figure 2, as it is not obvious from the caption. Also, presumably the correlograms correspond to scattering and absorption coefficients measured with the airborne nephelometer and PSAP; that should also be stated in the caption.

Line-by-line Details

The authors lament on line 27 that the terminology for absorbing aerosols are imprecise, but Andreae and Galencsér (2006) provide precise definitions for soot, soot carbon, brown carbon, light absorbing carbon, elemental carbon, apparent elemental carbon, black carbon, and equivalent black carbon. It would be nice if the community embraced this paper as the “go-to" article for carbonaceous aerosol definitions.

Line 77-78: the authors state “...by invalidating low AOD cases, the AAOD values that are retained in the AERONET Level-2 data may be biased high." This is not quite correct. It would be more accurate to state that “...the averaged AAODs" or “...the climatological AAODs" are biased high.

Lines 82, 93: Authors mention several Aerocom models, but do not tell us which ones or which Aerocom experiments they are pulling the data from.

Line 185: How much horizontal distance is covered in these 5- and 10-minute flight legs? This is important for readers to understand the auto-correlation within the profiles.

Lines 192-194 states that there are 253 complete profiles at BND and 132 complete profiles at SGP. However, only 24 and 14 flights were used in the analysis of Figures 3-6. I think that it is important to mention this here, because this description sounds like you have a much larger dataset available than you really do. This might be a good opportunity to mention that the stringent AERONET cloud screening procedure drastically reduces the number of possible comparisons, and this must also be considered by modelers when they utilize AERONET retrievals.

Lines 322-325: The authors state:
“Because the profiles are “stair-step" descents from 4600 m asl down to 450 m asl (e.g., see Figure 4 in Sheridan et al., 2012), matching with AERONET retrievals at the end of the profile means that the matches are more closely aligned with when the airplane is in the boundary layer and thus, typically, sampling the highest aerosol concentrations." This is inconsistent with other statements in the paper. The flight takes 2 hours and the authors are matching AERONET retrievals within +/- 3 hours of the end of the profile. Hence, the actual retrieval could occur up to 1 hour before the flight profile begins, and anytime during the flight. Thus, the AERONET retrievals are not necessarily “more closely aligned when the airplane is in the boundary layer.

Line 359: I don’t understand how the authors can test $\gamma \pm 1$ standard deviation when they do not measure γ.

Lines 360-361: Authors estimate the uncertainty in AOD by varying γ by up to 2 S.D., but there are hundreds of sunphotometer AODs available during their flights (per figure 3). Why not just compare their in situ AODs to AERONET AODs?

Line 379: The Lack 2008 citation is not included in the references.

Lines 434-435: Authors state that it is not possible to estimate the uncertainty in measured coatings due to coating effects. Consequently, they seem to be choosing zero uncertainty associated with coatings. Many readers will not agree with this choice.

Line 516: Authors state that AERONET AOD tends to be higher than in situ AOD in Figure 3; however, the slope is only greater than 1 in one of those figures, although the offset is always positive. So AERONET $>$ in situ AOD is not obvious, especially since many of the regression lines in Figure 3 fall below the 1:1 line. Additionally, AERONET median and average AODs are almost always less than the corresponding in situ AODs in Table 1, indicating that line 516 is not correct for the medians or the averages.

Line 519: Should be changed to "The correlations improve **when subsetted** for the more restrictive Level-2 almucantar retrievals." Otherwise, it sounds like the almucantar scans are incorporated into the AOD measurements, which is not the case.

Lines 541-543:
Authors state: "although the scatter in the relationships (particularly at BND) suggests that a multiplicative factor doesn't represent the relationship very well." I agree that the BND data looks quite scattered, but the SGP values do not look scattered. The R2 values are 0.76-0.77, which is excellent compared to the autocorrelation values for absorption provided in Fig 2 (r_{abs} = 0.45-0.55). Additionally, the scatter shown for AERONET AAOD is deceiving. There are approximately 2 AERONET retrievals for each in situ flight. The in situ flight represents a 2-hour average, but each AERONET retrieval occurs over the period of a 10-minute almucantar scan. The AERONET scans could occur up to 6 hours apart, and up to 3 hours after the flight is complete. Finally, R^2 = 0.3 implies that R = 0.55. Thus, the correlation in Figs 4 and Fig 2 at BND are not very different.

Lines 574-576: Well... you don't really have enough data to draw this conclusion, in my opinion. Why not include the averages for the purple points in Table 1, though?

Line 579: "Figure 3 shows that the AERONET AOD may be slightly larger than the

in-situ AOD. . . " Again, this is inconsistent with Table 1.

Line 644: It should be noted that 100 km is a long ways away! What kind of auto-correlations did Anderson (1998) find at those distances? That value should be noted here.

Line 673: Authors state that SSA differences are greater than would be expected from random error, even when AOD $>$ 0.4. Why not quantify that? That is, compute the mean and 2xSDOM of all independent measurements in that figure to see if the null hypothesis is valid; add this point with the resulting errorbar to the figure. It is important to isolate the independent measurements, though – multiple values that are obtained within 4-5 hours are not independent measurements (as you argued in Fig 2).

Page 10+: Since the authors are discussing non-synchronized data here, I think that they should note that the models and the in situ flights include cloudy periods, whereas the AERONET data are stringently cloud-screened.

Lines 830-837: This is a nice approach in some regions, but it is virtually impossible to capture seasonal variability over North America with this approach. For instance, from 4/1994 through 10/2012 the Cart Site had 3 lev2 retrievals in DJF, 22 Lev2 retrievals in MAM, 122 lev2 retrievals in JJA, and 51 retrievals in SON. Thus, results would be skewed to the high humidity summer periods.

Lines 840-844: This passage needs some massaging, as it makes no sense to me.

Line 872: Emphasize that this comment only pertains to AOD $<$ 0.2.

Line 1125: Müller (2012) is not included in the text. This is an important citation that needs to be presented in the text, as it discusses the difficulties associated with using in situ instruments to measure absorption.

References

Andreae, M. O., and A. Gelencsér (2006), Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys., 6, 3131-3148.

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Discussion paper