General comments:

The authors have extensively revised the manuscript and have responded satisfactorily to many of my comments/suggestions. However, some important issues remain which need to be resolved before publication.

We thank the reviewer for really getting after us on the uncertainty analysis. It's been helpful (and educational) to continue to dig into this and we think it's improved the paper. We've now added a figure (new Figure 2) showing the SSA uncertainty dependence on AOD (see response to second general comment below).

Especially regarding Figure 6: Please explain how you estimated the error bar in yellow shading in Figure 6. You had stated previously in this revised manuscript that the in-situ uncertainty in SSA was 0.06 (and gave no AOD dependence for this) while for AERONET it is 0.03 for AOD(440)>0.2 and 0.05-0.07 for lower AOD values (Table 2 of Dubovik et al., 2000). How did you get SSA uncertainty in Figure 6 of 0.05 for AOD>0.20 and 0.08 for lower AOD? In the first draft that I reviewed the in-situ SSA uncertainty was estimated to be 0.04, yet the yellow shaded uncertainty limits in Figure 6 were the same as in the revised manuscript. It appears that the uncertainty shading in Figure 6 needs to be updated.

The yellow shading in Figure 6 (now Figure 7) indicates the combined standard uncertainty of the AERONET and in-situ SSA values. The combined uncertainty is not additive, rather we've utilized the following equation (BIPM, 2008, their eqn 16) to estimate the uncertainty in the difference of the two SSA values based on the reported uncertainties for each:

$Unc^2=(SSA_{is})^2*(unc.SSA_{is})^2 + (SSA_A)^2*(unc.SSA_A)^2+2*SSA_{is}*SSA_A*unc.SSA_{is}*unc.SSA_A*R_{A,is}$

where the subscript 'is' indicates the in-situ data and 'A' indicates AERONET data and R is the covariance between AERONET and in-situ SSA for all the studies (0.1). We used average SSA values for in-situ and AERONET (0.95 and 0.89, respectively) and SSA uncertainties for both AERONET and in-situ of 0.06 for AOD<0.2 and SSA uncertainty of 0.03 for AOD>0.2. The shaded area for the AOD<0.2 cases stays the same (±0.08) while the shaded area for AOD>0.2 decreases to ±0.04. We've updated the plot to reflect this.

The text now has this additional information in the discussion of the figure and the figure caption was slightly modified:

"…, although most of the values are within the combined standard uncertainty of the AERONET and in-situ values indicated by the shading (see BIPM, 2008, their equation 16 for how the combined standard uncertainty was calculated)."

This reference was added.

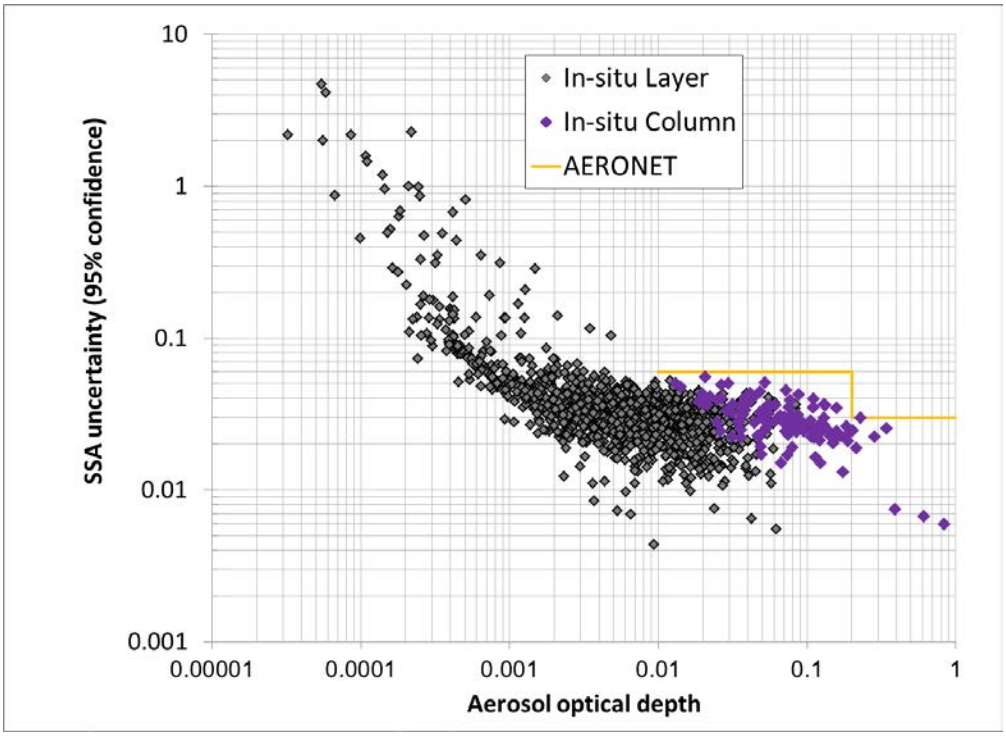
BIPM, "Evaluation of measurement data – Guide to the expression of uncertainty in measurement," Joint committee for guides in metrology (JCGM): 100, http://www.iso.org/sites/JCGM/GUM-JCGM100.htm, 2008.

Some attempt should be made to make an estimate of in-situ uncertainty for extremely low concentrations which occur in winter at mid latitudes (such as at SGP and Bondville), for total column AOD values ranging from ~0.02 to 0.08 at 440 nm (Figure 8). It is not realistic to think that the uncertainty of the in-situ measured/derived SSA will not increase when these very low aerosol loadings occur at all flight levels, especially since the flight segments at lower levels are

presumably still 5 minutes in duration. Concentrations at all altitudes on very low AOD days (<0.08 at 440 nm) would likely be similar or lower than high altitude concentrations on moderate to high AOD days, therefore the overall uncertainty of the in-situ data would likely be significantly greater for these low AOD days. The paper still lacks convincing discussion/analysis regarding in-situ SSA uncertainty on the lowest AOD days (occurring typically in the winter season), as it must be higher uncertainty than on moderate to high AOD days when aerosol concentrations at flight altitudes in the boundary layer can be an order of magnitude (or more) higher. (see Lines 399-400 & Lines 528- 532 in the revised MS for this topic/issue).

We've added the following plot to the paper that shows the calculated uncertainty of the in-situ SSA as a function of AOD. The points on the plot represent all three wavelengths (red/green/blue for the visible) for the in-situ instruments. The larger purple points represent the in-situ 'column' SSA uncertainty as a function of the 'column' in-situ AOD (column is in quotes as the airplane doesn't sample above 4700 m and only samples discretely below that). The small black points represent the SSA uncertainty for each individual flight layer as a function of that layer's AOD. For comparison the orange line represents the AERONET uncertainty values reported in Dubovik et al. (2000, their table 2). They give a range of 0.05-0.07, so we used 0.06 to represent the AERONET SSA uncertainty for AOD<0.2. This figure shows that the in-situ SSA uncertainty of 0.06 is a worst case value, the median in-situ 'column' SSA uncertainty for AOD<0.2 is 0.03. This 0.03 uncertainty is actually consistent with equation 2 in the paper if we use an SSA value of 0.95, which is more representative of the SSA values obtained from the in-situ measurements.

On the AERONET/in-situ comparison plot (formerly figure 5, now figure 6) we left the uncertainty range as 0.06 to represent the worst case scenario.



Specific Comments:

Lines 347-348: Regarding the AERONET Version 3 data, maybe say at the 'time of writing this manuscript' here.

done

Lines 354-355: This should be “if there was a corresponding Level 2 almucantar retrieval available, but AOD (440)<0.4.”

done

Lines 427-429: Please include the reference of Eck et al. (2014; ACP) here, as this paper specifically discusses the phenomenon you are discussing here, of the measurement of higher AOD in the near vicinity of cumulus clouds. This paper also shows that both lidar and in-situ measurements found similar aerosol increases as AERONET measured AOD near to cumulus clouds during this field campaign in Maryland.

done

Line 638: Direct sun AOD are more accurately called measurements, not retrievals (the AERONET retrievals are made with the Dubovik and King algorithm from almucantar sky radiance and spectral AOD input).

Changed ‘retrieval’ to ‘measurement’

Lines 674- 675: That is not possible, as there is always aerosol above 4.2 km asl, such as mid- to upper troposphere plus stratospheric aerosol. Perhaps the word 'significant' needs to be added here, since the exact amount in AOD above the 4.2 km level is often unknown.

Added word significant

Lines 761- 762: Perhaps putting both Hybrid and AERONET on the y-axis label would make this plot (Figure 5) easier to interpret.

Thanks for pointing this out! We’ve changed the axis.

Lines 806- 808: It should be mentioned here in the text that for mineral dust there is significant absorption at 440 nm in the coarse mode from iron oxide content.

The reviewer makes several comments about the importance of mineral dust absorption, suggesting that it could perhaps be a significant contributor to the discrepancies between in-situ and AERONET AAOD and SSA observed here, due to the aircraft inlet 50% sampling cutoff excluding particles with aerodynamic diameter > 5um (equivalent to ~3um once the particle density is accounted for). We respectfully disagree with this suggestion for the following reasons.

(1) While it is true that that dust absorption is dependent on iron oxide content particularly at lower wavelengths, our plots at 675 nm (where dust is less absorbing) don’t look markedly different from the plots at 440 nm (where dust is more absorbing) – see figures 4cd and 5cd in the original manuscript (now figures 5cd and 6cd). We state that this is why the 675 nm plots are included on lines 289-292. We briefly discuss the implications

of these plots in the paragraph starting at line 720 in the first revision of the manuscript (now line 725).

(2) There's variability in how much iron oxide there is in any given dust sample. Recent work by Engelbrecht et al. (2016) show that most dusts and clays have single scattering albedos >0.9 at 405 nm. This suggests that dust will have similar absorption properties to the aerosol we are observing at these two sites. We've added a reference to Engelbrecht in the paragraph discussing the comparisons at 675 nm.

(3) In the paragraph starting on line 792 in the revised manuscript we determine we may be missing ~10% of the aerosol in the in-situ measurements based on the AERONET size distribution retrievals. These particles would have to be very absorbing and not very scattering to account for absorption discrepancies of a factor of ~2.

Engelbrecht et al., "Technical note: Mineralogical, chemical, morphological, and optical interrelationships of mineral dust re-suspensions," Atmos. Chem. Phys., 16, 10809-10830, 2016. http://www.atmos-chem-phys.net/16/10809/2016/

Lines 817- 824: Please add here the underestimation of coarse mode absorption at 440 nm due to dust particles that are under-sampled in the in-situ measurements.
See comment above.

Lines 894-898: This should be stated as greater absorption than in-situ here (rather than over-estimation) since the in-situ data is not recognized (by the scientific community) as truth or gold standard for SSA.
Rephrased sentence:
Most of the SSA comparisons in Table 4 found fairly good agreement between AERONET and in-situ AOD, implying that the discrepancy is associated with the absorption values rather than the scattering values (since scattering is typically 90% of extinction).

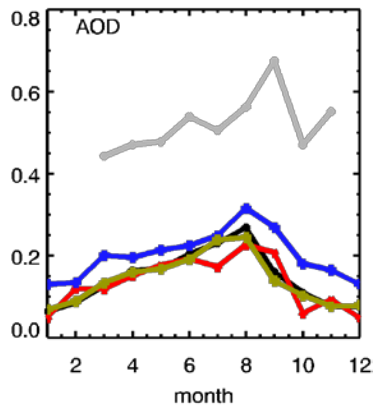
Lines 898-900: In the interest of completeness and unbiased reporting, the number of cases where agreement is excellent (within 0.02), given the in-situ uncertainty of 0.06, should be mentioned here in the text.
We added the following sentence in the previous paragraph – it seemed to fit better there:
"Of the 63 cases depicted in Figure 7, 16 cases (~25%) of the AERONET/in-situ comparisons were within 0.02."

Line 907: Please change 'often' to 'mostly' here since most data shown in Figure 6 for AOD >0.2 at 440 nm are within the yellow shaded uncertainty bounds.
done

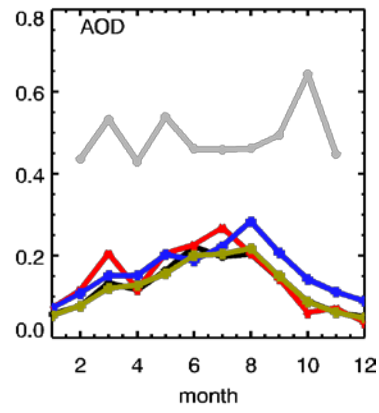
Lines 970 - 971: Not really true, since the almucantar associated measurements (L1.5*) of AOD are biased towards days with lower cloud cover due to sky radiance screening for clouds away from the solar disc. Additionally almucantar retrievals are only valid in the relatively early morning and late afternoon-evening in summer at mid-latitude (SZA>50, which occurs all day in winter) thereby missing the most active fair weather cumulus development times of day in

summer. AOD measurements associated with more cloudy conditions are often higher due to increased particle hygroscopic growth, cloud processing of particles in aqueous phase chemistry, gas-to particle conversion that occurs more rapidly in cloud droplets, and convergence of aerosols associated with larger cloud systems (see Jeong and Li (2010) and Eck et al. (2014)).

We respectfully disagree with this comment – the plot below is the same one we included in our previous response to the reviewers. The mustard colored line represents the AERONET level 2 direct sun climatology. The black line (which is mostly hidden by the mustard colored line) represents the AERONET level 1.5* AOD climatology. We stand by our statement that: *"The AERONET Level-1.5* AOD monthly medians are representative of the direct sun AERONET Level-2 AOD climatology at the two sites."*



BND monthly median AOD values



SGP monthly median AOD values

LEGEND
- AERONET 1.5* AOD
- AERONET Version 2, level 2 direct sky AOD
- Version 2, level 2 AOD for successful Almucantar scans with AAOD and SSA retrievals (i.e., has the AOD440<0.4 restriction)
- INSITU AOD
- AEROCOM (phase II) AOD

Lines 983 - 984: This statement is not rigorously true, as noted in the previous comment.

See response to previous comment.

Lines 986 - 987: These are very large and significant differences ("by up to 50%") between in-situ and AERONET measured AOD. Please note this in the paper and attempt an explanation for the significant low summer season bias of the in-situ AOD measurements.

We agree that it's a pretty significant difference, but remember this figure is comparing climatologies not time-matched comparisons. The AERONET data at BND cover the time period 1996-2011 (but is missing June 2007-April 2008 due to AERONET instrument malfunctions) while the airplane covers several hundred ~3h flights between June 2006-Sept 2009. While we can compare the climatological values in these plots, that was not the primary point of presenting them. Rather, our goal for presenting the climatological information was threefold:

(1) Show that we see similar seasonal patterns amongst different methods of obtaining monthly AOD (in-situ, AERONET level 1.5 (also direct sun level 2.0 as shown above) and model). Our first sentence discussing these figures is "*At both sites, the climatological seasonal patterns for AOD (i.e., high in summer, low in winter) are similar for the three data sets: in-situ measurements, AERONET Level-1.5* retrievals (recall*

that the AERONET 1.5 AOD is representative of the overall AERONET AOD climatology at each site) and AeroCom model output*."

(2) Indicate where there are limitations in the AERONET retrieval climatologies at these two sites (e.g., winter)

(3) Demonstrate that while the level 1.5 values for AOD were at least in the ballpark there could be large discrepancies if just the level 2 almucantar retrievals were used to represent the seasonality AOD and AAOD, while the opposite is true for SSA. (Level 2.0 retrievals of SSA are more representative of the climatological data (in-situ and model) than level 1.5* SSA.)

We've added the following sentences:

"While 50% discrepancy between the AERONET and in-situ climatology may appear significant, it's important to remember that these data sets do not represent the same period of time or measurement conditions (e.g., time of day, cloud cover, aerosol events, ambient humidity, etc). Schutgens et al. (2016) shows there can be large differences when comparing values obtained with different samplings (more than 100% for AOD), particularly when there are high levels of variability in the data."

Lines 1107 - 1113: It would be useful to note that the fine mode volume median radius (and also fine mode effective radius) decrease as AOD decreases at all of these AERONET sites, thereby suggesting one physical mechanism for the lower SSA at lower AOD levels. Sub-micron sized particles scatter less efficiently when they are smaller, therefore even if the particle composition remains constant the smaller sized particles at lower AOD would result in lower SSA. Additionally the coarse mode fraction of AOD increases at lower AOD likely due to a relatively constant background level of coarse mode particles while the fine mode often shows much larger seasonal increases due to stagnation, humidification, cumulus cloud interaction in the summer (opposite seasonality for Fresno due to fog in the winter). The lower Angstrom Exponents seen in low AOD months at most of these sites is indicative of greater coarse mode fraction of AOD when AOD is lower. Since the in-situ instruments under-sample the coarse mode particles this can potentially be a source of bias in the in-situ determination of SSA (440 nm) at lower concentration levels (lower AOD days), since iron oxide absorption in coarse mode dust may be missed. The SGP and Bondville sites are located in rural areas that would be expected to have varying amounts of mineral dust loading, depending on regional soil moisture conditions and transport from drier regions to the west (direction of prevailing winds in many seasons).

This depends on the size distribution of the sub-micron particles relative to the size corresponding to the first peak in the Mie scattering efficiency curve. The Reviewer is correct that particles smaller than the size of the Mie peak will scatter less efficiently when they are smaller, but particles that are larger than the size of the Mie peak will scatter more efficiently when they become smaller (until they shrink below the Mie peak). The Reviewer's assertion may or may not be correct, depending on the size distribution and magnitude of the size change. We have not changed the text. Additionally, as we've noted in the text and in response to other comments (e.g., our response to the reviewers comment about lines 806-808) related to dust absorption, we think it's very unlikely that dust is a dominant culprit in the discrepancy between in-situ and AERONET absorption and SSA at these two sites.

Lines 1180-1182: Suggest changing to "…typically well within the reported uncertainty bounds especially in light of the in-situ value of 0.06"

We've updated our uncertainty analysis and included a plot of the uncertainties in SSA for the in-situ measurements (new figure 3). Because of this we think that the statement is fine as stated.

Lines 1188-1191: It should also be mentioned that a fraction of the 440 nm absorption from coarse mode dust may be underestimated from in-situ measurements due to the large particle under-sampling bias.

See our thoughts about coarse mode dust effects in our response to the reviewers comments about Lines 806-808. We have not added a dust caveat.

Comparison of AOD, AAOD and column single scattering albedo from AERONET retrievals and in-situ profiling measurements

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Abstract

Here we present new results comparing aerosol optical depth (AOD), aerosol absorption optical depth (AAOD) and column single scattering albedo (SSA) obtained from in-situ vertical profile measurements with AERONET ground-based remote sensing from two rural, continental sites in the US. The profiles are closely matched in time (within +/-3 h) and space (within 15 km) with the AERONET retrievals. We have used Level 1.5 inversion retrievals when there was a valid Level 2 almucantar retrieval in order to be able to compare AAOD and column SSA below AERONET's recommended loading constraint (AOD>0.4 at 440 nm). While there is reasonable agreement for the AOD comparisons, the direct comparisons of in-situ-derived to AERONET-retrieved AAOD (or SSA) reveal that AERONET retrievals yield higher aerosol absorption than obtained from the in-situ profiles for the low aerosol optical depth conditions prevalent at the two study sites. However, it should be noted that the majority of SSA comparisons for AOD_{440}>0.2 are, nonetheless, within the reported SSA uncertainty bounds. The observation that, relative to in-situ measurements, AERONET inversions exhibit increased absorption potential at low AOD values is generally consistent with other published AERONET/in-situ comparisons across a range of locations, atmospheric conditions and AOD values. This systematic difference in the comparisons suggests a bias in one or both of the methods, but we can not assess whether the AERONET retrievals are biased towards high absorption or the in-situ measurements are biased low. Based on the discrepancy between the AERONET and in-situ values, we conclude that scaling modelled black carbon concentrations upwards to match AERONET retrievals of AAOD should be approached with caution as it may lead to aerosol absorption overestimates in regions of low AOD. Both AERONET retrievals and in-situ measurements suggest there is a systematic relationship between SSA and aerosol amount (AOD or aerosol light scattering) – specifically that SSA decreases at lower aerosol loading. This implies that the fairly common assumption that AERONET SSA values retrieved at high AOD conditions can be used to obtain AAOD at low AOD conditions may not be valid.

1. Introduction

The amount and location of absorbing aerosol in the atmosphere is critical for understanding climate change (e.g., Hansen et al., 1997; Ramanathan and Carmichael, 2008; Bond et al., 2013; Samset et al., 2013). Ramanathan and Carmichael (2008) note the effects of absorbing aerosol (which they termed black carbon (BC)) on atmospheric heating rates, precipitation and

weather patterns. (Note: The terminology used to refer to absorbing aerosol is imprecise (Petzold et al., 2013, Andreae and Gelencsér, 2006) and encompasses the terms describing chemistry, e.g., 'black carbon' (BC) and terms describing optical effects, e.g., absorption. The measurements reported herein all refer to light absorption.) The vertical distribution of BC can also influence its effect on climate (e.g., Haywood and Ramaswamy, 1998; Samset et al., 2013; Ramanathan and Carmichael, 2008). Single scattering albedo (SSA) is an indicator of the absorbing nature of the aerosol; higher SSA values indicate a more reflective (whiter) aerosol while a more absorbing aerosol will have lower SSA values. SSA is a primary determinant of whether the aerosol will have a warming or cooling effect (e.g., Haywood and Shine, 1995; Hansen et al., 1997; Reid et al., 1998). Uncertainty in the value of SSA due to uncertainties in the amount of absorbing aerosol can even prevent determination of the sign of aerosol forcing on local to regional scales. Bond et al. (2013) assessed BC as the second most important global-average warming species (top-of-atmosphere forcing +1.1 W m^{-2}, 90% bounds: +0.17 to +2.1 W m^{-2}) after CO_2 (in Bond et al. (2013) the direct effect of BC is 0.71, 90% bounds: +0.09 to 1.26 W m^{-2}).

Currently, the only way vertical profiles of aerosol absorption can be obtained is via airborne in-situ measurements. Such flights are expensive and tend to primarily occur during intensive field campaigns, which are usually aimed at studying specific aerosol types (e.g., biomass burning, African dust, urban/industrial pollution). This reliance on short-term campaigns results in profile data sets that are sporadic in both space and time, and not necessarily representative of typical conditions. Additional issues with airborne in-situ measurements include adjustment of measurements to ambient conditions, particle losses in sample lines, and instrument uncertainties. Nonetheless, in-situ vertical profiling of absorbing aerosols has provided useful information to modelers trying to understand climate effects, transport, and lifetimes of these important atmospheric constituents (e.g., Koch et al., 2009; Schwarz et al., 2010; Skeie et al., 2011).

The limited availability of in-situ vertical profile measurements means modelers must rely on globally sparse and/or temporally sporadic airborne measurements to evaluate BC vertical distributions in their models. Alternatively, the column properties retrieved from AERONET measurements and inversions have been widely used to provide a first constraint on modeled vertical aerosol properties (e.g., Sato et al., 2003; Koch et al., 2009; Bond et al., 2013; He et al., 2014; Wang et al., 2014). Use of the AERONET data as an absorption constraint has suggested upscaling of modeled AAOD values by a factor of 2-6 depending on location (e.g., Bond et al., 2013), although Wang et al. (2016) has shown that better spatial resolution of models and emission inventories can reduce some of the previously observed model/AERONET discrepancies.

Ground-based remote sensing of both direct attenuation and sky radiances permit inversions of atmospheric column averaged absorption. By retrieving the complex refractive indices at different solar wavelengths as well as the average aerosol size-distribution, absorption related properties can be determined (e.g., aerosol absorption optical depth (AAOD), single scattering albedo (SSA) and, absorption Ångström exponent (AAE)). The AERONET network has a fairly

wide spatial coverage on land, with long data records at many sites (Holben et al., 1998; Dubovik et al., 2000; Dubovik and King, 2000). One obvious limitation of the AERONET inversion retrievals is that the uncertainty of the derived single scattering albedo (SSA) becomes very large at low values of AOD (Dubovik et al., 2000). To minimize the effects of this uncertainty, the AERONET Level-2 data invalidates all absorption-related values if the AOD at wavelength 440 nm (AOD_{440}) is below 0.4 (Dubovik et al., 2000; Dubovik et al., 2002; Holben et al., 2006). Unfortunately, this restriction greatly reduces the spatial and temporal coverage of absorption-related data that can be obtained from AERONET. Moreover, by excluding low AOD cases, the climatological statistics of AAOD derived from the AERONET Level-2 data may be biased high.

Model analysis of global AOD values suggest that 95% of global AOD_{440} values are below 0.4 (Figure 1), while 89% of the AOD_{440} values over land are below the 0.4 threshold. Five models in the AeroCom suite (GMI-MERRA-v3, GOCART-v4, LMDZ-INCA, OsloCTM2, and SPRINTARS-v385) have reported daily-average values of AOD_{440} (for AeroCom Phase II control experiment), which can be used to develop a cumulative frequency distribution of the percent of the Earth's surface and days where a Level-2 AERONET retrieval of AAOD might be possible (ignoring the presence of clouds and absence of sunlight). Figure 1 indicates that, at best, Level-2 AERONET AAOD retrievals might represent 5% of the days, globally, and less than 11% of the days over land. In other words, the AOD constraint on Level-2 AERONET almucantar inversion retrievals means these retrievals represent only a small fraction of the Earth’s surface and are biased to conditions of high aerosol loading.

The other information that Figure 1 provides is the fractional contribution of regions with different AOD_{440} amounts to the total aerosol and the fossil fuel black carbon (BCFF) radiative budget. These values were derived from monthly data from 4 models in the AeroCom suite. The fractional contribution to the radiative budget can be mathematically described as follows: for each model grid box there are three quantities: (i) the radiative forcing ($W\ m^{-2}$), (ii) the horizontal area of the box (m^2), and (iii) the AOD_{440}. The product of the radiative forcing term and area is the perturbation to Earth's radiative budget due to total aerosol (or BCFF) in the box. The sum of this product over all the boxes is the total perturbation. Figure 1 shows the fraction of the radiative budget perturbation as a function of AOD_{440}. It suggests that approximately 75% of the total aerosol forcing and 83% of BCFF forcing is due to regions of the globe where AOD_{440} <0.4. This highlights the significant contribution of aerosol in these cleaner areas to the total global radiation budget.

It should be noted that there is significant inter-model variation in the AeroCom cumulative AOD_{440} and radiative forcing plots shown in Figure 1. In particular the BCFF cumulative forcing fraction varies with the lifetime of BC predicted by the models. A long BC lifetime results in more dilute AOD and BCFF radiative forcing distributions. Other issues include the fact that global models have limited spatial and temporal resolution, and generally simulate less variability in aerosol properties than is observed in measurements. However, all models used to generate Figure 1 follow the same general trend as is shown in Figure 1 with the take-away point being that AOD_{440} values >0.4 are a relatively rare occurrence.

Because of the potential of the AERONET absorption-related retrievals (e.g., AAOD and SSA) for understanding global distributions of absorbing aerosol, there have been many studies comparing AERONET retrieval values with those obtained from in-situ measurements in order to assess the AERONET retrieval validity. Such comparisons have taken several different forms. There have been direct comparisons where column SSA or AAOD values calculated from individual in-situ vertical profiles have been compared with AERONET retrieved values for retrievals close in time and space (Haywood et al., 2003; Magi et al., 2005; Mallet et al., 2005; Leahy et al., 2007; Corrigan et al., 2008; Osborne et al., 2008; Johnson et al., 2009; Esteve et al., 2012; Schafer et al., 2014). In addition to direct comparisons there have been general, statistical assessments between AERONET and in-situ measurements for both SSA and AAOD including: (a) comparing surface in-situ measurements with AERONET retrievals (e.g., Dubovik et al., 2002; Doran et al., 2007; Mallet et al., 2008; Corr et al., 2009); (b) comparing in-situ SSA (or AAOD) from a few flight segments to the corresponding column SSA (or AAOD) from AERONET (e.g., Kelektsoglou et al., 2012; Müller et al., 2012) and (c) comparison of statistical distributions or averages of AERONET retrievals for a given time period with airborne in-situ measurements (e.g., Ramanathan et al., 2001; Leahy et al., 2007; Andrews et al., 2011a; Ferrero et al., 2011; Johnson et al., 2011). Many of these statistical comparisons have shown good agreement between the AERONET and in-situ values. This increases general confidence in the AERONET retrievals. However, such statistical comparisons are not appropriate for the evaluation of the accuracy of individual retrievals.

The primary scientific question to be addressed in this paper is: *Is there a consistent bias observed between AAOD and column SSA obtained from in-situ profiling flights and AERONET retrievals?* The answer to this question may help determine the validity of adjusting model estimates of AAOD to agree with AERONET retrievals (e.g., Sato et al., 2003; Bond et al., 2013). It should be noted that AERONET does not recommend the use of absorption-related parameters (e.g., single scattering albedo, absorption aerosol optical depth, and complex index of refraction) at AOD_{440} below 0.4. Dubovik et al. (2000) suggests the uncertainty of AERONET SSA values more than doubles for AOD_{440} less than 0.2.

In what follows, we first evaluate how direct AERONET AAOD retrievals compare with those derived from multi-year, in-situ measurements obtained from vertical profiles over two rural continental AERONET sites in the U.S. Second, we create a summary of all direct AAOD or SSA comparisons between in-situ vs. AERONET data previously presented in the literature in order to place our results about AERONET aerosol absorption-related retrievals in a wider context. Finally, we look at the seasonality of in-situ, AERONET, and modelled (AeroCom) SSA and AAOD values to see if the annual cycles can provide any insight into observed discrepancies in the direct comparisons. Because this study focuses on only two low AOD sites in the continental US which are unlikely to be generally representative of other low loading sites around the globe, and because other factors (e.g., Wang et al., 2016) may contribute to reported differences between modelled and AERONET AAOD we do not attempt to suggest implications for global BC forcing.

2. Methods

This study utilizes data from two sites with collocated AERONET measurements and multi-year, in-situ aerosol profiling measurements. The two sites are Bondville (BND, 40.05ºN 88.37ºW, 230 m asl) and Southern Great Plains (SGP, 36.61ºN 97.49ºW, 315 m asl). Surface in-situ measurements and AERONET column measurements have been made at both locations since the mid-1990s (e.g., Delene and Ogren, 2002; Sheridan et al., 2001: Holben et al., 1998). Weekly to twice-weekly flights measuring in-situ vertical profiles of aerosol optical properties over these two sites were made for a subset of the years of ground-based observations. At SGP the in-situ profile flights were centered over the site's central facility where the AERONET sunphotometer is deployed. Due to FAA flight restrictions, the BND in-situ profiling flights took place approximately 15 km to the WNW of the AERONET sunphotometer location at the BND surface site (Sheridan et al., 2012). Additionally, for BND, a low level flight leg (200 m agl) was flown directly over the instrumented BND surface site. The flights at both sites were subject to 'visual flight regulations' which means they took place during daylight hours and the plane did not fly in-cloud.

At BND and SGP, the median AOD_{440} values are 0.14 and 0.11, respectively (based on all AERONET Level-2 data from the start of AERONET measurements at each site). These median values fall right around the 50% mark on the AOD cumulative distribution plot (Figure 1), indicating BND and SGP may be appropriate sites to explore potential discrepancies between AERONET and in-situ AAOD and SSA retrievals at lower AOD conditions.

2.1 IN-SITU

The in-situ aerosol profiles were obtained with dedicated Cessna 206 airplanes flying stair-step profiles one to two times per week over the two sites. Between 2006 and 2009, 365 flights were flown over BND (out of a total of 401 flown in the region (Sheridan et al., 2012)), while 171 aerosol profile flights were flown over SGP in the 2005-2007 time period (Andrews et al., 2011a). The profiles consisted of 10 (at BND) or 12 (at SGP) level flight legs between approximately 450 and 4600 m asl (corresponding to approximately 150 and 4200 m agl). The profiles, which were 'stair-step' descents, took approximately 2 hours to complete as the airplane spent set amounts of time at each level (10 min/flight level for flight legs above ~1600 m asl and 5 min/flight level for flight legs below that altitude) in order to improve measurement statistics at the typically cleaner higher altitude flight levels. Airplane speed was approximately 50 m/s, resulting in the 10 min upper level legs being approximately 30 km long and the 5 min lower level legs approximately half that (15 km) length. This flight pattern means the last 30 min of the profile were typically in the boundary layer for these two sites and encompassed the majority of the aerosol contribution to column aerosol loading. Previous work has shown that the airplane measurements appear to capture the variability in aerosol properties observed by the long-term, continuous measurements at the surface (e.g., Figure 3 in Andrews et al., 2004)

Descriptions of the flight profiles and aircraft package have been described in detail in other papers (Andrews et al., 2011a; Sheridan et al., 2012) so only a brief description is provided

here. The pilot flew within the constraints provided (specifically-defined stairstep profile, vary the time of day, cross wind, over the instrumented field site, during daylight and not within clouds) but without day-to-day scheduling input from scientists. Here, we utilize the same 10 flight levels for both profiling sites: 457, 609, 915, 1219, 1829, 2439, 3050, 3659 and 4575 m asl. Of the 365 flights at BND, 253 flights had complete profiles (all flight levels) with valid scattering, absorption and relative humidity data; at SGP, 132 flights out of 171 were complete. Only complete profiles (all 10 flight levels) were used in this analysis. As is obvious from the vertical range of the flight levels, complete in-situ profiles do not equate to complete atmospheric profiles – this is discussed more in the in-situ uncertainties discussion (Section 2.4.1). The number of flights that could be compared with AERONET measurements is significantly less than this, as discussed in Section 2.3 where the merging of the AERONET and in-situ data sets is described.

The aircraft were equipped with an inlet that sampled particles with aerodynamic diameter D_p<7 μm, and losses in downstream sample lines were estimated to reduce the particle diameter for 50% sampling efficiency to 5 μm (Sheridan et al., 2012). Aerosol light absorption (σ_{ap}) was measured at three wavelengths (467, 530, 660 nm) using a Radiance Research Particle-Soot Absorption Photometer (PSAP) and aerosol light scattering (σ_{sp}) was measured at three similar wavelengths (450, 550, 700 nm) using an integrating nephelometer (TSI model 3563). The measurements of absorption and scattering were made at low relative humidity (RH<40%). Absorption data were corrected for scattering artifacts, flow and spot size calibrations, etc., using the Bond et al. (1999) algorithm, with appropriate modifications for wavelength (Ogren, 2010). The Anderson and Ogren (1998) correction for instrument non-idealities was applied to the nephelometer data.

Ambient temperature (T_{amb}) and RH (RH_{amb}) were measured by a sensor (Vaisala Inc, Model Humicap 50Y) mounted on the aircraft fuselage inside a counterflow inlet shroud, and the nephelometer sample pressure was used as a surrogate for ambient pressure. These measurements of ambient meteorological parameters were used to adjust the in-situ optical data to ambient conditions in order to compare with the AERONET measurements and retrievals, which are made at ambient conditions. Climatological IMPROVE network surface aerosol chemistry measurements of sulfate and organic carbon (Malm et al., 1994) were utilized to determine a value for the hygroscopic growth parameter 'γ' for each site based on the Quinn et al. (2005) parameterization which relates aerosol hygroscopicity to organic mass fraction. For BND γ=0.71±0.08, while for SGP γ=0.65±0.08. At BND the IMPROVE chemistry measurements are co-located at the profile location, while for SGP the measurements at the IMPROVE Cherokee Nation site (approximately 56 km southwest of the profile location) were used. This γ value was then used in conjunction with the airborne RH_{amb} measurements to adjust the in-situ scattering profiles for both SGP and BND.

The equation used to adjust the dry, in-situ scattering to ambient relative humidity (RH_{amb}) is a commonly used aerosol hygroscopic growth parameterization (e.g., Kasten, 1969; Hanel, 1976; Kotchenruther et al., 1999; Carrico et al., 2003; Crumreyrolle et al., 2014):

$$\sigma_{sp}(RH_{amb})/\sigma_{sp}(RH_{dry})=a*(1-(RH_{amb}/100))^{-\gamma}. \quad (1)$$

where $\sigma_{sp}(RH_{amb})$ is the aerosol scattering at ambient RH, $\sigma_{sp}(RH_{dry})$ is the measured scattering at low RH, and γ is the hygroscopic growth parameter derived from the IMPROVE aerosol chemistry. The value of 'a' can be determined using: $a = (1/(1-RH_{dry}/100))^{-\gamma}$ (e.g., Crumreyrolle et al., 2014; Quinn et al., 2005). Here we assume a=0.9 based on the typical RH values measured inside the nephelometer for both profile locations (BND RH_{dry}=12+/-11%; SGP RH_{dry}=14+/-10%). RH_{amb} at BND and SGP averaged 47.4% and 38.6%, respectively, over all flight levels and seasons (56% (BND) and 43% (SGP) below 1500 m asl). The 95th percentile RH_{amb} values (calculated over all flights and flight levels) were 79.3% and 76.6% at BND and SGP, respectively. (Note: scattering-weighted column average RH values were 54% at BND and 43% at SGP). Applying eq. 1 to the observed RH_{amb} and $\sigma_{sp}(RH_{dry})$ profiles, the average enhancement of column-average σ_{sp} due to hygroscopic growth was 1.52 and 1.36 at BND and SGP, respectively. The corresponding 95th percentiles of column average enhancement of scattering were 2.06 and 2.10. While Equation 1 takes into account differences in hygroscopic growth due to RH for each segment of each flight, it does not account for compositional changes that might affect the scattering enhancement due to hygroscopicity. For aerosol events such as biomass burning and dust episodes with significantly different composition than the 'normal' aerosol we would expect to over-predict the aerosol hygroscopicity relative to the normal aerosol. Sheridan et al., (2001) showed that the SGP surface aerosol had lower hygroscopicity when it was influenced by dust or smoke.

The absorption measurements were adjusted to ambient temperature and pressure, but not to ambient RH because the parameterization of the correction and its magnitude are unknown. It is typically assumed that absorbing aerosol is hydrophobic (e.g., Schmid et al., 2003; Reid et al., 2005; Schaefer et al., 2014), i.e., does not take up water. The uncertainties associated with this assumption are discussed in section 2.4.

Both the scattering and absorption in-situ measurements were adjusted to the two nominal Level-2 AERONET wavelengths in the mid-visible spectrum (440 nm and 675 nm). The 440 nm wavelength is of interest as that is the wavelength for which the AOD constraint for retrieving SSA and hence, AAOD, is given; the 675 nm wavelength is also presented because it is less sensitive to NO_2, organics, and dust which could potentially bias the in-situ/AERONET comparison. Also, evaluating data at both wavelengths helps in attributing aerosol absorption to BC versus dust, since at 675 nm absorption is almost entirely caused by BC. The measured scattering Ångström exponent was used to adjust the in-situ scattering measurements to the AERONET wavelengths. For the in-situ aerosol absorption wavelength adjustments we used a constant absorption Ångström exponent of 1.2 to minimize the effects of noise in the measurement. Previous studies have shown that for both BND and SGP the absorption Ångström exponent is ~1.0 in the BL and 1.5 at higher altitudes (Andrews et al., 2011; Sheridan et al., 2012). Using the incorrect absorption Ångström exponent will have a negligible effect on the resulting absorption value because of the small difference between the measured and target

wavelengths; using an absorption Ångström exponent of 1.2 instead of 1.0 will result in a 1% difference in adjusted wavelength while using an Ångström exponent of 1.2 instead of 1.5 will result in a 2% difference in adjusted absorption.

Finally, using these in-situ values adjusted to AERONET wavelengths and ambient conditions the flight profile average properties can be determined. Aerosol extinction ($\sigma_{ep}= \sigma_{sp} + \sigma_{ap}$) was calculated and integrated vertically for the profile to obtain the in-situ AOD. The aerosol absorption for each profile was integrated vertically to obtain the in-situ AAOD. As described in Andrews et al. (2004), the in-situ column SSA (which is compared to the AERONET SSA value in section 3.1) was calculated for each flight level and then extinction-weighted and integrated to determine column SSA. This results in SSA values which are virtually identical to SSA values calculated using: $SSA_{col,in\text{-}situ} = (AOD_{in\text{-}situ} – AAOD_{in\text{-}situ})/AOD_{in\text{-}situ}$) and effectively gives higher weighting to the SSA values at altitudes that had the highest aerosol concentrations. Details of the procedure for calculating the vertical integral are given in Andrews et al. (2004), although, in this study, the in-situ profiles contained two additional high altitude flight levels (at 3659 and 4575 m asl) and the layer at the highest altitude was assumed to extend 457 m above the measurement altitude. Profile statistics for various parameters including SSA are provided in Andrews et al. (2004, 2011a) and Sheridan et al. (2012). Individual flight profiles for various parameters are available online at: http://www.esrl.noaa.gov/gmd/aero/net/iap/iap_profiles.html (for SGP) and https://www.esrl.noaa.gov/gmd/aero/net/aao/aao_prof2007.html (for BND).

2.2 AERONET

AERONET measurements have been made at BND since mid-1995 and at SGP since mid-1994. The AERONET network makes spectral measurements of aerosol optical depth (AOD) using CIMEL sun/sky radiometers (Holben et al., 1998). The measurements are typically made at seven wavelengths, with an eighth wavelength used for water vapor measurements. The AERONET website (http://aeronet.gsfc.nasa.gov) provides links to data from more than 500 sites across the globe. The column extinction Ångström exponent (å) can be directly calculated from the wavelength-dependent AOD measurements (Eck et al., 1999). In addition to AOD and å, algorithms have been developed utilizing both the spectral AOD and the spectral angular distribution of the sky radiances obtained from almucantar scans, which enable retrieval of other column aerosol properties including AAOD, SSA, size distribution, complex refractive index, and fine mode fraction of extinction (FMF_e) (Dubovik and King, 2000; Dubovik et al., 2000; O'Neill et al., 2003; Dubovik et al. 2006). The nominal wavelengths of the almucantar inversion retrievals are 440, 675, 870 and 1020 nm. An additional advantage of the AERONET database is that the retrieval values are obtained consistently – the calibrations, corrections, QC and algorithms are applied identically for each AERONET location.

For Version 2 AERONET data, there are different levels of AERONET data available for download from the AERONET website. Level 1.0 is unscreened data while Level-1.5 undergoes automated cloud-screening (Smirnov et al., 2000). Level-2 represents data with pre-field and post-field calibrations applied, manual inspection, and quality assurance (Smirnov et al., 2000). In addition to the Level-1.5 screening, the criteria for Level-2 almucantar inversion

products include a check of the sky residual error as a function of AOD_{440}, solar zenith angle must be greater than or equal 50 degrees, and almucantars must have a minimum number of measurements in each of the four designated scattering angle bins. Further, for Level-2 absorption-related products (including SSA, AAOD, AAE, and the complex refractive index) the AOD_{440} must be greater than 0.4 to exclude more uncertain aerosol absorption estimates (Holben et al., 2006). Version 3 AOD products are now available but the Version 3 inversion products ~~a~~were not at the time of this writing.

The AAOD values reported in the AERONET almucantar inversion files are obtained using the relationship: AAOD=(1-SSA)*AOD. Schafer et al. (2014) has a nice description of how SSA is obtained from the AERONET measurements. In the present study, in order to maximize the number of AERONET data points available for comparison with the in-situ measurements, Level-1.5 retrievals of AAOD and SSA were included in the analysis if there was a corresponding valid Level-2 AOD value but $AOD_{440}<0.4$ (i.e., the same primary criterion as was used in Bond et al. (2013)). We will refer to these AAOD and SSA values as 1.5* data.

2.3 Merging the IN-SITU and AERONET data sets

Merging of collocated (within 15 km), but temporally disparate data sets can induce discrepancies in the combined data set. Lag-autocorrelation analysis (e.g., Anderson et al., 2003) is used to determine an appropriate time window for comparison of the AERONET and in-situ profile measurements. Figure 2 shows that, at the surface, at both BND and SGP, scattering is well correlated (r(k)>0.8) out to 4-5 hr lag, while absorption is less correlated than scattering (r(k) for absorption is 0.75 at BND and 0.55 at SGP). Based on the correlograms, AERONET retrievals were merged with the in-situ profile data when the retrievals were within +/-3 h of the end of the in-situ profile. This is the same time range constraint used to compare AERONET and PARASOL SSA values (Lacagnina et al., 2015). Additionally, Figure 2 represents the maximum correlation that we can realistically expect to achieve in a comparison of two different instruments with temporally offset measurements and provides context for the AERONET/in-situ comparisons presented in Section 3.

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 way the maximum time difference between the boundary layer portion of the flight and the AERONET retrieval is 3 h; if we’d chosen to match based on the start of the flight the maximum time difference between the boundary layer measurements and the AERONET retrieval could be as large as 5 h. The boundary layer portion (<1800 m asl) of the ~2 h profile takes approximately 30 min. While the +/- 3 h match window was chosen based on the surface in-situ aerosol lag-autocorrelation statistics (Figure 2), other time windows were also examined. For time windows less than +/-3 h (e.g., 1 h and 2 h) the fit coefficients (slope, intercept) did not change significantly although the AOD and AAOD correlation coefficients did improve for those smaller time windows. For time windows longer than +/- 3 h (e.g., 6 h and 12h) there were changes in AOD and AAOD fit parameters and the correlation coefficients decreased significantly. For SSA there appeared to

be no correlation between AERONET retrievals and in-situ calculated values regardless of match window length (highest SSA correlation coefficient was 0.12, but most were less than 0.05 for both sites). The poor correlations for SSA are not surprising given the uncertainties at low loading. The AERONET/in-situ comparisons for the +/-3 h window are discussed in section 3.1 below.

2.4 Uncertainties in IN-SITU and AERONET data

In any study comparing parameters obtained from different instruments and/or methods, an understanding of the uncertainties in each of the parameters being compared is critical. Below we discuss the uncertainties inherent in both the in-situ and AERONET datasets.

2.4.1 IN-SITU uncertainties

Uncertainties for measurements by the in-situ instruments have been described previously (e.g., Sheridan et al., 2002; Formenti et al., 2002; Shinozuka et al., 2011; Sherman et al., 2015) so only an overview is provided here. Sheridan et al. (2002) calculated uncertainties in aerosol light scattering for the TSI nephelometer to be 7-13% for 10 min legs depending on amount of aerosol present – the higher uncertainty value applies to very low aerosol loadings (scattering < 1 Mm^{-1}). We assume that uncertainty in the profile scattering measurements is 13%. 13% is appropriate for the higher altitude flight legs (10 min duration with, typically, low aerosol loading) and is also reasonable for the lower altitude flight legs which are only 5 min in duration but have significantly higher loading. At both BND and SGP the median boundary layer scattering is typically >10 Mm^{-1} while median scattering for the upper altitude flight legs is typically between 1-10 Mm^{-1} (Andrews et al., 2011; Sheridan et al., 2012).

Unfortunately, because profile-specific aerosol hygroscopicity measurements were not available for the in-situ aircraft measurements described here, a single hygroscopic growth parameterization was applied for all profiles at each site as described in Section 2.1 and equation 1. To determine the uncertainty in AOD induced by the uncertainty in the scattering adjustment to ambient RH, AOD values were calculated using different γ values representing the range of hygroscopic growth factors suggested by the aerosol chemistry. Specifically, AOD_{440} was calculated for $\gamma \pm 1$ standard deviation and $\gamma \pm 2$ standard deviations. As described above, γ was calculated from the climatological chemistry measurements made by the IMPROVE network (14 years of data, ~1700 data points at BND; 10 years of data, ~1000 data points at SGP) using the Quinn et al. (2005) parameterization. We calculated the mean and standard deviation of γ based on those climatological chemistry measurements. Using this approach, the uncertainty in AOD due to adjustment to ambient RH was determined to be between 9% and 16%. This uncertainty might seem to be low, but recall that the 95th percentiles of ambient RH values observed throughout the profiles were ~80% but that more typically ambient RH in the boundary layer was less than 70% at BND and less than 60% at SGP. Sum of squares uncertainty analysis suggests the overall uncertainty in the in-situ AOD is approximately 30% for higher ambient humidities (RH_{amb}>70%) and approximately half that at RH_{amb}<50%.

Both Jeong and Li (2010) and Eck et al. (2014) have noted that the presence of nearby clouds may influence AOD values. They've investigated the effect of high RH-halos embedded in aerosol layers that typically exist in the vicinity of non-precipitating cumulus clouds. If the AERONET retrieval went through such a halo it could result in an increased AOD due to the combined effects of hygroscopic growth, cloud processing of aerosols and rapid gas-to-particle conversions. If the aircraft also flew through this RH-halo then the effect would also be accounted for in the RH-corrected in-situ measurements. However, if the high RH layer was between two flight levels then the aircraft measurements would not account for it. Addressing this effect is outside the scope of this paper.

The PSAP measurement of aerosol absorption is more uncertain than the aerosol scattering measurements – PSAP uncertainty is reported to be in the 20-30% range (e.g., Bond et al., 1999; Sheridan et al., 2002; Sherman et al., 2015). It should be noted that the PSAP absorption measurement represents all absorbing aerosol collected on its filter, as opposed to being specific to 'black carbon' absorption. That is actually helpful for this particular study as the AERONET retrieval of AAOD also represents all flavors of absorption (e.g., 'black carbon', 'brown carbon' and dust). Müller et al. (2011) describe detailed experiments to characterize filter-based absorption instruments and describe some additional limitations of the instruments.

There is, however, some question of whether the PSAP (or any filter-based measurement) is able to accurately represent absorption by particles coated with semi-volatile or liquid organics, due to the possibility of such coatings changing the characteristics of the filter substrate (oozing!) after impaction (e.g., Subramanian et el., 2007; Lack et al., 2008). Comparisons of filter-based absorption measurements for denuded and un-denuded particles (e.g., Kanaya et al., 2013; Sinha et al., in revisions, 2017) suggest that the un-denuded particles have absorption enhancements of 5-25% relative to those that have been through a denuder. These comparisons show that stripping off coatings and evaporating the non-absorbing particles reduces the measured absorption, i.e., that the effect of coatings is not completely lost in filter-based measurements. The effect of coatings appears to increase the absorption value reported by the PSAP relative to that reported by a non-filter-based instrument (Lack et al., 2008); in other words the aerosol absorption values obtained from PSAP measurements may have a positive bias. It is worthwhile to explore the potential magnitude of such a bias. The mean mass concentrations of organic aerosol determined from the IMPROVE measurements near BND and SGP (the OCf value in the IMPROVE data set; Malm et al., 1994) are similar for both sites and less than 2 $\mu g/m^3$, putting them firmly in the rural/remote category identified by Lack et al (2008; their figure 4). Depending on whether figure 3 or figure 4 in Lack et al. (2008) is used, Lack et al.'s (2008) results suggest that the PSAP might be overestimating absorption by a factor of 1.1 to 1.5 due to artifacts caused by organic aerosols. However, in a subsequent study, Lack et al. (2012) reported a PSAP overestimate by factors of 1.02-1.06 over Los Angeles, considerably lower then the Lack et al. (2008) results.

The positive bias in absorption related to filter-based measurements is the same order of magnitude and direction of the absorption enhancement factor found by some lab and

theoretical studies for coated absorbing particles suspended in the atmosphere. Absorption enhancement values of 1.3-3 have been predicted for coated particles (e.g., Bond et al., 2006; Lack et al., 2009; Cappa et al., 2012) although enhancements larger than a factor of 2 have not been measured for ambient aerosol (e.g., Lack et al., 2008; Cappa et al., 2012; McMeeking et al., 2014). Wang et al. (2014) suggested that an absorption enhancement factor of 1.1 was appropriate for fossil fuel influenced aerosol and that 1.5 was a more reasonable enhancement factor for biomass burning affected aerosol. Biomass burning does not have a consistent influence on either BND or SGP. Cappa et al. (2012) suggested that the discrepancies between ambient and modelled and/or laboratory results, could be a result of differences in particle morphology and/or chemistry. We have not made any adjustments for the absorption effects of coatings or the potential positive bias in PSAP measurements as the science is still unclear.

In addition to the potential absorption enhancement due to organic coatings, it has been suggested that aerosol water on absorbing particles may also enhance absorption. There have been very few studies where the hygroscopic growth enhancement of absorption was explicitly considered. Redemann et al. (2001) modeled absorption enhancement as a function of RH based on characteristic atmospheric particles and found absorption enhancement values of up to 1.35 at 95% RH; for the 95th percentile RH_{amb} values encountered at BND (78.9%) and SGP (76.6%), the Redemann et al. (2001, their figure 2) study would predict absorption enhancements of ~1.1. Nessler et al. (2005) and Adam et al. (2012) utilized both ambient aerosol measurements and Mie theory to calculate absorption enhancement values due to hygroscopic water uptake. Nessler et al. (2005) does not provide absorption enhancements as a function of RH, but Adam et al. (2012) suggest absorption enhancements due to hygroscopic growth of less than 1.1 at 80% humidity. Brem et al. (2012) report on laboratory studies that show that aerosol absorption was enhanced by a factor of 2.2 to 2.7 at 95% relative humidity relative to absorption at 32% relative humidity, although for RH less than ~80% (i.e., the RH values observed in this study) they show no absorption enhancement (their figure 9). Lewis et al. (2009) actually observe a decrease in absorption with increasing RH for some biomass fuels, but hypothesize the decrease might have been due to their measurement technique and/or a change in the morphology of the particles.

In summary, the positive bias in the PSAP measurements of aerosol light absorption might be as high as a factor of 1.1 to 1.5 due to oozing (e.g., the overestimate of absorption reported by Lack et al., (2008) for filter-based measurements). Atmospheric absorption may be underestimated by PSAP measurements by up to a factor of 1.5 due to not accounting for coating (organic or water) effects. Without additional laboratory and field measurements to quantify the net effect of the possible positive and negative biases in PSAP measurements of aerosol light absorption, it is not possible to estimate the actual uncertainty in the in-situ light absorption measurements reported here due to coating effects. To address this, we double the assumed PSAP uncertainty of ~25% to 50% in the calculations of uncertainty.

One aspect of the in-situ system that will affect both the scattering and absorption measurement is the gentle heating used to dry the particle to RH<40%. The drying process we use (heating of 40 C or less) may remove some volatile components but we believe the removal to be

minimal (<10-20%) based on lab and ambient volatility studies in the literature. Thermal denuder studies suggest little removal of volatile components (<10%) at 40 C (e.g., Mendes et al., 2016; Huffman et al., 2009, Bergin et al., 1997) although thermal denuders results may be limited by short residence times (<20s). However, smog chamber evaporation studies on ambient aerosol over longer time periods (minutes-hours) at ambient temperature also suggest ambient aerosol may be less volatile than previously thought – Vaden et al. (2011) showed that ambient SOA lost just ~20% of its volume after ~4h.

Once the uncertainties in the in-situ aerosol scattering and absorption are known, the uncertainty in SSA (SSA= $\sigma_{sp}/(\sigma_{sp}+\sigma_{ap})$ can also be calculated. Formenti et al. (2002, their equation 5) suggests the uncertainty in single scattering albedo (δSSA/SSA) can be calculated:

$$\delta SSA/SSA = (1-SSA)*[(\delta \sigma_{sp} / \sigma_{sp})^2+(\delta \sigma_{ap} / \sigma_{ap})^2)]^{1/2} \quad (2)$$

As an example, ~~F~~for scattering uncertainties of 30%, (combined nephelometer and f(RH) induced uncertainty), PSAP absorption uncertainties of 50%, and SSA values of 0.95 (typical of the in-situ SSA observations), equation 2 results in an in-situ SSA uncertainty of ~~6~~3% or approximately 0.03~~6~~. For the higher altitude flight segments the loading does tend to be quite a bit lower and thus has higher uncertainty but those upper-level segments contribute little to the overall AOD or AAOD. Because the flight column SSA is calculated using extinction-weighted SSA flight segments, segments with very low aerosol concentrations will have little impact on the column SSA derived from the flight measurements. Figure 3 shows the calculated SSA uncertainties for each flight layer as well for the in-situ column SSA for each individual flight. For high AOD (AOD>0.3) the SSA uncertainty is quite low (less than 0.01), while for lower loading (0.005<AOD<0.2) the SSA uncertainty is less than 0.06 (the median uncertainty in this low AOD range for the in-situ flights in this study is ~0.03).

In addition to instrumental uncertainties there are also uncertainties associated with the aircraft flight patterns, i.e., the presence of aerosols below, between and above the discrete flight levels. Missing aerosol above and below an aircraft profile is a potential issue in all aircraft/column comparisons. Different approaches have been used to assess whether aerosol loading contributions above the highest flight level (4.6 km asl) are important. Andrews et al. (2004) utilized Raman lidar measurements to determine that 80-90% of the aerosol was below 3.7 km asl at SGP (3.7 km was the maximum altitude flown by the original SGP airplane, although all the profile flights utilized here occurred after the maximum flight level was increased to 4.6 km asl). Andrews et al. (2004) also assumed assumed an AOD contribution of 0.005 from stratospheric aerosol which was not done here. At SGP, Turner et al. (2001) segregated lidar aerosol extinction profiles by season and AOD. Their results (their Figure 1) suggest that for the vast majority of cases observed at SGP, 5% or less of the extinction will be found above 4 km. For low AOD cases (AOD_{355}<0.3) their mean extinction profiles suggest little to no aerosol extinction between 4-7 km. At BND, Esteve et al. (2012) noted that CALIPSO data indicated negligible extinction above 4.6 km asl. Regionally, seasonal average profiles from CALIPSO also suggest there is minimal aerosol above the flight's highest level (Ma and Yu, 2014; Yu et al., 2010).

Although statistical profile results (e.g., -Turner et al., 2001; Yu et al., 2010; Ma and Yu, 2014) suggest little contribution from high altitude aerosol layers in the region of these two sites, Schutgens et al. (2016) demonstrates the importance of considering the specifics rather than the statistical. We used the Raman lidar best estimate data product of extinction profiles at SGP to evaluate the presence of aerosol above the highest flight level at the site. For the SGP in-situ profiles that had matches with AERONET inversion retrievals, we identified three lidar profiles that exhibited aerosol layers at high altitudes, but in all three cases the presence of these layers was also hinted at by an increase in the aerosol loading at the highest flight levels of the in-situ measurement. Thus, we further screened in-situ/AERONET comparisons by removing flights at SGP and BND with significant increases in loading at the highest flight levels. There may still be aerosol layers above the level measured by the Raman lidar, but we have no means of assessing that. The AOD comparison presented in Figure ~~3~~ 4 suggests we are unlikely to be missing significant aerosol at high altitudes.

Several papers (Andrews et al., 2004; Esteve et al., 2012; Sheridan et al., 2012) have shown that there is a high correlation ($R^2 > 0.8$) between scattering measured at the surface site (SGP or BND) with scattering measured at the corresponding lowest flight leg, although the slopes of the relationships indicated that the airplane measurements might be missing a fraction (10-20%) of the aerosol below about 150 m agl. Additionally, Esteve et al. (2012) found high correlation (slope=1.01, R^2~0.7) between scattering AOD calculated by assuming the lowest leg represented scattering in the entire layer between surface and that flight leg with scattering AOD calculated from 1-sec data obtained during descent from the lowest flight leg to landing. This result suggested that no consistent bias would result from assuming the lowest flight leg was representative of the aerosol between surface and that altitude. We've looked at the surface/lowest flight leg relationship specifically for the flights with matching AERONET retrievals studied here. We found that at BND the surface and lowest level flight aerosol measurements were virtually identical. At SGP the lowest level leg actually measured slightly higher aerosol loading than was observed at the surface, which could lead to an overestimate of the aerosol optical depth in that layer, depending on the shape of the profile.

Similarly, Esteve et al. (2012) investigated differences in aerosol scattering between and at flight levels by comparing scattering AOD from the airplane descent between layers with that calculated from the individual level legs in the profile. Again they were able to confirm that measurements made during the fixed flight altitudes are representative of the aerosol near those altitudes.

2.4.2 AERONET uncertainties

Uncertainties in AERONET retrievals have been reported in several papers. Eck et al. (1999) indicate that the uncertainty in AOD is approximately 0.01 for a field-deployed AERONET sunphotometer at solar zenith angle = 0 (i.e., sun directly overhead). For the almucantar retrievals (solar zenith angle > 50) used here, the AOD uncertainty will be smaller as the uncertainty in AOD decreases inversely with air mass (Hamonou et al., 1999; their equation 1).

Dubovik et al. (2000) report AERONET retrieved SSA uncertainties in their Table 4. For water soluble aerosol (the predominant aerosol type at both BND and SGP) they report that SSA values are reliable to within ±0.03 when $AOD_{440} > 0.2$, while the uncertainty in SSA increases to (±0.05-0.07) for $AOD_{440} \leq 0.2$. The almucantar retrieval of SSA may be biased by errors in the surface reflectance when the AOD is very low. Another potential issue is that the AERONET retrievals report only one pair of (real, imaginary) refractive index values for the total size distribution (for each wavelength). If there are two or more aerosol modes in the column, this assumption may skew the resulting SSA and AAOD values, although the effect of such skewing would depend on the aerosol properties and cannot be assessed here. Potential impacts in the case of uneven mode absorption in the retrieved size distribution have been found to be minor since the retrieved size distribution is more linked to forward scattering than absorption (pers. comm., O. Dubovik).

Mallet et al. (2013) reports an AAOD uncertainty of 0.01 but does not indicate whether or how the AAOD uncertainty would change with AOD_{440}. Using the sum of squares propagation of errors to calculate the uncertainty in AAOD for both high and low AAOD cases results in an AAOD uncertainty of approximately ±0.015 for both high and low AOD cases (high AOD_{440}=0.5, δAOD=0.01, SSA=0.95, δSSA=0.03, AAOD=0.026; low AOD_{440}=0.2, δAOD=0.01, SSA=0.95, δSSA=0.07, AAOD=0.011). An AAOD uncertainty value of ±0.015 suggests an uncertainty of about 60% in AAOD for AOD_{440}=0.5 and more than 140% uncertainty in AAOD for AOD_{440}<0.2.

3. Results

In this section we first present comparisons of AOD, AAOD and SSA from the in-situ measurements at BND and SGP with AERONET retrievals. This includes (1) direct comparisons of each in-situ profile with contemporaneous AERONET retrievals; the BND and SGP comparisons are then put in the wider context of a literature review of similar direct comparisons of in-situ and AERONET AAOD and SSA; (2) seasonal comparisons of AOD, AAOD and SSA from Phase II AeroCom model results, AERONET retrievals and in-situ measurements for BND and SGP; and finally, (3) we discuss these results in the context of biases in determination of AAOD.

3.1.1 BND and SGP: in-situ vs AERONET – Direct Comparisons

Figures ~~3~~4, ~~4~~5 and ~~5~~6 show the direct comparisons of AOD, AAOD and SSA at both 440 nm and 675 nm. On all 3 plots, the blue points represent the same data set – each point indicates a flight for which there was one or more successful AERONET Level-2 almucantar retrievals within +/-3 hours of the end of the flight profile (if there was more than one retrieval corresponding to a flight, the retrievals were averaged). The thin gray lines on the 440 nm plots indicate the reported (AERONET) or calculated (in-situ) uncertainties in the data. Table 1 provides a comparison of the statistical values (median, mean and standard deviation) at 440 nm for each of the parameters at both of the sites for these direct comparisons (blue points in Figures ~~3~~4, ~~4~~5, and ~~5~~6). The low number of flights for which there are comparisons available (~10% of total number of flights) indicate both the effects of AERONET stringent cloud screening routine and the constraints imposed by the almucantar retrievals. In addition to limiting the number of comparisons available for this study, this limited data availability also has

implications for modellers utilizing AERONET data – for example, Schutgens et al. (2016) has shown the importance of temporal collocation in measurement-model comparisons. Figure ~~3~~ 4 also contains red points – the red data points represent all direct sun AERONET Level-2 AOD ~~retrievals~~ measurements during the +/-3 hours window around the end of each profile. Depending on atmospheric conditions, there may be more than one AERONET ~~retrieval~~ measurement within +/-3 hours of the end of each profile, which is why in Figure ~~3~~ 4 there are more red data points plotted than there are flights. The red points have not been averaged in order to provide an indication of the variability in AOD during the in-situ profiling flight.

The comparison between in-situ and AERONET AOD is important because it can be used to evaluate how well the in-situ and AERONET ~~retrievals~~ data can be expected to agree and, thus, set the context for the AAOD and SSA comparisons. Many studies have investigated the relationship between in-situ and remotely sensed AOD (e.g., Crumreyrolle et al., 2014; Schmid et al., 2009, and references therein). As noted in these studies, the in-situ derived AOD values tend to be slightly lower than the AOD retrieved from remote sensing measurements. Figure ~~3~~ 4 presents the comparison of Level-2 AOD for AERONET and in-situ measurements at 440 nm and 675 nm for two sets of AERONET AOD data. The first comparison (red points on plots) is for all direct sun AERONET Level-2 AOD measurements. The second comparison (blue points on plots) is for flight-averaged AERONET Level-2 AOD measurements where all the criteria required for almucantar retrievals are satisfied. Table 2 summarizes how many points make up each of these data sets.

In general, Figure ~~3~~ 4 shows that AERONET AOD tends to be higher than the in-situ AOD, although there is good correlation between AERONET and in-situ AOD. The uncertainty bars tend to overlap the 1:1 line suggesting that in-situ measurements provide a reasonable proxy of the total column aerosol loading as represented by AERONET AOD. Student t-test evaluation suggests that the AERONET and in-situ AODs are the same at the 95% confidence level. The coefficients of determination (R^2) are within the range we would expect based on the lag-autocorrelation of scattering at these two sites (Figure 2) and the +/-3 h time window. The R^2 values increase when sub-setted for the more restrictive Level-2 almucantar retrievals. The lower in-situ AOD values observed at both sites, compared to AERONET, may be due to the hygroscopicity adjustment from dry in-situ to ambient RH conditions being too low or undersampling of larger particles (e.g., Esteve et al., 2012). Esteve et al. (2012) found slopes closer to 1 when they restricted AERONET/in-situ AOD comparison to low ambient RH (<60%) conditions, although the AERONET AOD values were still larger than the in-situ AOD. The effect of undersampling larger particles or underestimating aerosol hygroscopicity on the AAOD and SSA comparisons are discussed in section 3.1.2. Some of the discrepancy between the in-situ and the AERONET values may also be due to the limited vertical range covered by the airplane (150 – 4200 m asl). We've excluded flights that might have had significant aerosol above the highest flight level, based on Raman lidar comparisons (at SGP) and profile shapes (at BND). The relationships observed between AERONET and in-situ AOD for both sites are very similar to those observed for the recent DISCOVER-AQ campaign (e.g., Crumreyrolle et al., 2014, their figure 3).

One thing to note on Figure ~~3a~~ 4a is the blue point marked BB (the BB stands for biomass burning). This measurement occurred on June 28, 2006 and appears to have been strongly affected by forest fire smoke transported from Canada. We applied the same hygroscopicity adjustment to the measurements of this flight as we did to all of the BND flights and, in this BB case, the hygroscopicity correction was the primary reason the in-situ AOD value is significantly higher than the AERONET AOD value. This point would lie much closer to the 1:1 line if the in-situ BB data were assumed to be hygrophobic. Previous work at the surface site at SGP has shown that dust and smoke aerosol types tend to exhibit lower hygroscopicity than the background aerosol normally observed at the site (Sheridan et al., 2001). This BB point provides an extreme example of the downside of using a constant hygroscopic growth parameter as a function of RH, although without additional information about the aerosol for each profile it is difficult to do otherwise. The light blue dotted line on Figure ~~3~~ 4 represents the relationship between AERONET and in-situ data if the BB point is excluded.

Figure ~~4~~ 5 presents the comparison of AAOD for flight-averaged AERONET and in-situ measurements. As described above, the AERONET AAOD values shown in Figure ~~4~~ 5 are what we have termed Level-1.5* data – i.e., they are from Level-1.5 almucantar retrievals when there was a valid Level-2 almucantar retrieval, but the AOD_{440}>0.4 constraint was not applied. In contrast to the AOD comparison depicted in Figure ~~3~~4, the AERONET Level-1.5* AAOD values are significantly higher than the in-situ AAOD values. Figure ~~4~~ 5 also shows that the correlation between the AERONET and in-situ AAOD is poorer than it was for AOD, particularly at BND (R^2 is 0.49 at BND and 0.68 at SGP for the 440 nm comparison). The lower correlation at BND is somewhat surprising given the lag-autocorrelation results for aerosol absorption (Figure 2a) at the BND surface site. Surprisingly, while 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), the results for BND in Figure ~~4~~ 5 indicate less correlation than at SGP for absorption. Nonetheless, the correlation coefficients for BND in Figure ~~4~~ 5 (R^2=0.49 (blue) and 0.37 (red) correspond to R = 0.70 (blue) and 0.61 (red)) are not that far from the 3 h auto-correlation of r(k=3h)=0.75 for absorption at BND in Figure 2. For AAOD the uncertainty bars, while wider, exhibit significantly less overlap with the 1:1 line (indeed no overlap at SGP) and indeed the student t-test suggests the AERONET and in-situ AAOD values are different at the 95% level at both sites.

Both Figure 5~~4~~ and the median values provided in Table 1 indicate that AERONET Level-1.5* AAOD tends to be larger than the in-situ AAOD, although the scatter in the relationships (particularly at BND) suggests that a multiplicative factor doesn't represent the relationship very well. The purple points in Figure ~~4~~ 5 indicate AAOD retrievals where the flight-averaged AOD_{440}>0.2. There is no obvious improvement of the relationship between in-situ and AERONET AAOD when these points are considered (although there are only 1-4 comparison points above AOD_{440}>0.2 for each site).

The AAOD comparisons at 675 nm at BND (Figure ~~4c~~5c) are quite similar to those at 440 nm, suggesting that there is little contribution to absorbing aerosol from dust, organic carbon and/or NO_2. In contrast, at SGP, there is a change in the relationship between AERONET and in-situ AAOD from 440 to 675 nm indicating that one or more of these components may affect the 440

nm comparisons at that site (Figure ~~4d~~5d). Recent work by Engelbrecht et al. (2016) has suggested that even at 405 nm most dusts have have SSA values > 0.9 meaning they are not much more absorbing than the aerosol typically observed at BND and SGP. Further, ~~-~~Ångström exponent values from the matched AERONET and in-situ profile data do not support the presence of dust, while the rural nature of the site suggests significant levels of NO_2 are unlikely. Thus the most likely explanation is the presence of organic carbon, although the IMPROVE sulfate and organic data used to estimate aerosol hygroscopcity do not support this. The IMPROVE measurements tend to suggest a relatively small contribution of organics to the aerosol mass with the average mass concentration of organics only 40 to 60% that of sulfate aerosol mass concentration for BND and SGP, respectively. In contrast, the Aerosol Chemical Speciation Monitor (ACSM) measurements by Parworth et al. (2015) indicate that, depending on the month, organic aerosol can contribute up to 70% of the total aerosol mass at SGP.

Figure ~~5~~ 6 presents the comparison of column SSA retrieved from flight-averaged AERONET inversions (Level-1.5* data) with the column SSA calculated from in-situ profile measurements of aerosol scattering and absorption at BND and SGP. Consistent with the AOD and AAOD comparisons (Figures ~~3~~ 4 and ~~4~~5) the SSA retrieved from AERONET tends to be much lower than the SSA calculated from the in-situ profile measurements. As with AAOD, the SSA uncertainty bars exhibit little overlap with the 1:1 line and a student t-test suggests the AERONET and in-situ SSA values are different at the 95% level for both BND and SGP. At both sites the range in AERONET-retrieved SSA is much wider than the range in column SSA obtained from the in-situ profiles. Long term, in-situ measurements at the BND and SGP surface sites yield mean SSA values of 0.92 and 0.95 respectively (Delene and Ogren, 2002, based on monthly-averaged data). Delene and Ogren's (2002) surface SSA values are reported at low RH (RH<40%) and 550 nm; adjusting them to ambient conditions and 440 nm would likely cause them to increase making them more comparable to the in-situ column SSA depicted in Figure ~~5~~ 6 but even less like the AERONET Level-1.5* SSA values. As with Figure ~~4~~5, the purple points on Figure ~~5~~ 6 indicate when the flight-averaged AOD_{440}>0.2; although there aren't enough points to draw a robust conclusion, there does not appear to be an improvement in the relationship between in-situ and AERONET SSA when only these purple points are considered.

Figure ~~5~~ 6 also includes a set of 'hybrid SSA' (SSA_{hybrid}) points in yellow. These points have been calculated using the AERONET AOD and the in-situ AAOD:

$$SSA_{hybrid} = (AOD_{AERONET} - AAOD_{PSAP})/AOD_{AERONET} \quad (3)$$

This hybrid approach to SSA eliminates the uncertainty associated with the empirical hygroscopic growth factors applied to the in-situ scattering measurements, and also removes the scattering uncertainty associated with undersampling the coarse mode. It does not, however, eliminate the uncertainties associated with assuming the absorbing aerosol is hygrophobic, that there is little absorption in the potentially undersampled coarse mode, or the unknown contribution from absorption enhancement. SSA_{hybrid} is very similar to the SSA derived from in-situ measurements, suggesting the primary discrepancy between the AERONET SSA and the in-situ SSA is due to the determination of the absorbing nature of the aerosol, either due

to issues with the limitations of the filter-based measurements or to the interpretation of the relative contribution of aerosol absorption from the AERONET inversion retrieval products.

3.1.2 How might in-situ hygroscopicity assumptions and under-sampling of the aerosol affect SSA and AAOD comparisons?

Figure ~~3~~ 4 shows that the AERONET AOD may be slightly larger than the in-situ AOD, while Figures ~~4~~ 5 and ~~5~~ 6 suggest that the AERONET retrievals significantly overestimate the amount of absorbing aerosol (low SSA, high AAOD) relative to the in-situ measurements. The slight deviation between in-situ and AERONET AOD may lead to questions about whether directly comparing other AERONET and in-situ parameters (e.g., SSA, AAOD) is a reasonable thing to do and whether the AAOD and SSA comparisons shown in Figures ~~4~~ 5 and ~~5~~ 6 are related to issues with the AOD comparison. As mentioned above, Esteve et al. (2012) suggested the AOD difference was most likely due to either underestimating the hygroscopic growth correction and/or undersampling of supermicron particles by the aircraft inlet. In this section we evaluate how these two possible causes of the AOD discrepancy might affect the SSA and AAOD comparisons.

Increasing the hygroscopic growth adjustment of the in-situ measurements would enhance the in-situ scattering values used to calculate the in-situ AOD, but would not change the in-situ AAOD because the absorbing particles are assumed to be non-hygroscopic. Consequently, the comparison depicted in Figure ~~4~~ 5 would not change with a different adjustment for hygroscopic growth. Increasing the in-situ AOD, without affecting the in-situ AAOD, would result in higher in-situ SSA values and an even greater discrepancy between AERONET and in-situ SSA values than shown in Figure ~~5~~6. To evaluate the effect of assuming absorbing particles were non-hygroscopic, a sensitivity test was performed assuming the absorption enhancement due to RH was the same as the hygroscopicity scattering enhancement, i.e., $\sigma_{ap}(RH_{amb})/\sigma_{ap}(RH_{dry})=a*(1-(RH_{amb}/100))^{-\gamma}$. While this is likely an extreme assumption, it had minimal effect on the comparisons of AOD, AAOD and SSA.

The other likely candidate to explain the in-situ AOD being slightly lower than the AERONET AOD is aircraft under-sampling of super-micron aerosol particles due to the 5 μm inlet cutoff'. Esteve et al.'s (2012) comparison of column in-situ and AERONET scattering Ångström exponents at BND suggested that the airplane measurements might be under-sampling larger particles. Sheridan et al. (2012) estimated that the aircraft inlet 50% cut-off aerodynamic diameter is approximately 5 μm, so particles larger than that are unlikely to be sampled by the in-situ measurements but will be sensed by the AERONET sunphotometer. If we take into account that atmospheric particles are likely to have a density greater than 1 g cm^{-3}, the actual cut size would be closer to 3 or 4 μm. The AERONET volume size distributions were used to estimate the fraction of column extinction due to particles less than 3 μm. At BND the mean and standard deviation of the 3 μm extinction fraction (extinction(D<3μm)/extinction(D<30μm)) was 0.93 ±0.07, while at SGP the extinction fraction value was 0.88 ± 0.09. At the BND and SGP surface sites, most (80-90%) of the observed sub-10 μm scattering and absorption is also attributed to sub-micron aerosol, with absorption more likely to be in the sub-micron size range

than scattering (Delene and Ogren, 2002; Sherman et al., 2015). This is consistent with the observation that absorbing aerosol tends to be concentrated in sub-micron particles for typical aged continental air masses (e.g., Hinds, 1982). Based on these observations, larger and primarily scattering particles are more likely to be under-sampled by the in-situ measurements than absorbing particles. This is the opposite of what is needed to explain the discrepancies between AERONET and in-situ AOD, AAOD, and SSA shown in Figures ~~3-5~~4-6. The in-situ measurements would need to preferentially under-sample absorbing aerosol relative to scattering aerosol in order to come into line with the AERONET observations. Additionally, Sheridan et al. (2012) calculated particle transmission losses from behind the sample inlet on the airplane to both the nephelometer and PSAP to be similar and to be less than 10% in the particle diameter range 0.01<D<1 μm. This suggests that preferential losses of absorbing aerosol are also unlikely to occur downstream of the aerosol inlet. In summary, we can only see two ways that the in-situ measurements can sample aerosol efficiently enough to represent AERONET AOD fairly well but significantly underestimate AAOD and overestimate SSA: (1) not accounting properly for the effect of coatings (organic or water) on absorption enhancement which we've discussed in detail in the manuscript and (2) not sampling layers of predominantly absorbing aerosol below, between, and/or above the flight layers. We suspect that the SSA required of such layers in order to explain the AAOD and SSA discrepancies is physically impossible.

3.2 Literature survey: in-situ vs AERONET – Direct Comparisons

Direct comparisons at BND and SGP suggest that AERONET retrievals underestimate SSA and, consequently, that AERONET overestimates AAOD relative to in-situ measurements of AAOD for the low AOD conditions typical at these two sites. The next question to address is whether this discrepancy, found for two rural, continental sites in the central US with relatively low aerosol loading, is more widely observed for direct in-situ/AERONET comparisons at a variety of sites/conditions. As in section 3.1, the focus in this section is on direct comparisons of column-averaged SSA (or AAOD) derived from in-situ measurements made during aerosol profiling flights that were flown in close proximity (temporal and spatial) to an AERONET retrieval. Tables 3 and 4 summarize literature results describing the direct comparisons of AERONET retrievals with in-situ aerosol profile measurements for AAOD and column SSA. Figure ~~6~~ 7 provides a graphical overview of the SSA comparisons described in Table 4. Tables 3 and 4 and Figure ~~6~~ 7 also include the BND and SGP comparisons described in this study. With the possible exception some of the profiles reported by Corrigan et al. (2008), the literature comparisons cited in Tables 3 and 4 and shown in Figure ~~6~~ 7 have been made at higher AOD conditions (AOD_{440}>0.3) to reduce retrieval uncertainty. In contrast, the SGP and BND comparisons are more representative of global AOD (Figure 1) with the majority of the comparisons at BND and SGP occurring for AOD_{440}<0.2. Please note that some of the earlier studies shown in Figure ~~6~~ 7 and described in Table 4 used values from Version 1 AERONET Level-2.0 data. Where that was the case, we retrieved Version 2 AERONET Level-2.0 data from the AERONET website and those Version 2 data are what is reported in Table 4 and depicted in Figure ~~6~~7. The comments section of Table 4 mentions the cases where this was done. For some of these references we also retrieved the AOD_{440} values from the AERONET website as the AOD_{440} values weren't reported in all papers.

Tables 3 and 4 have been restricted to studies with direct comparisons of column-averaged AAOD or SSA retrieved from full in-situ vertical profiles flown near (within ~100 km) AERONET sites within a few hours of the AERONET retrieval, i.e., studies that are comparable to the BND and SGP studies described in Section 3.1. For non-plume data sets, Anderson et al. (2003) found autocorrelations $\geq$ 0.8 at 100 km (their figure 6). For plume-influenced data sets they found autocorrelations ~0.6**.** Included in the tables are the field campaign name (if applicable), number of AAOD or SSA comparisons, the primary type of aerosol studied, summary of AOD comparisons (if available), altitude range covered by the airplane, instruments and data processing (e.g., instrument corrections, treatment of hygroscopicity, wavelength adjustment) and a summary of the results of the AAOD comparison. The last column in Tables 3 and 4 includes information on the spatial and temporal differences between the in-situ measurements and AERONET retrievals and comments on treatment of the AERONET and in-situ data. The last column also notes how each campaign dealt with aerosol below and above the in-situ profile if reported. It should be noted that the number of SGP and BND comparisons of AAOD and SSA in Tables 3 and 4 are only possible because we've utilized AERONET retrievals below the recommended threshold of AOD_{440}>0.4. The uncertainty for the BND and SGP comparisons is much higher than for some of the other direct comparisons due to the low AOD conditions observed at these sites.

For the three AAOD closure studies listed in Table 3 (the BND and SGP results presented here, plus results from a field campaign over the Indian Ocean) the AERONET retrievals indicate more absorbing aerosol in the column than is suggested by the corresponding in-situ measurements. The Corrigan et al. (2008) paper mentioned in Table 3 is the sole AERONET/in-situ AAOD comparison cited by Bond et al. (2013), as it was the only published direct AAOD comparison available. Corrigan et al. (2008) present no AOD comparisons that could provide an indication of their sampling system efficiency, and information about the wavelength of the comparisons and profiles specifics are lacking. To our knowledge, no other direct comparisons of in-situ and AERONET AAOD are available in the literature.

The SSA comparison studies listed in Table 4 and visually summarized in Figure ~~6~~ 7 indicate that, even at higher AOD, AERONET retrievals tend to indicate more-absorbing aerosol (lower SSA) relative to in-situ measurements, although most of the values are within the combined standard uncertainty of the AERONET and in-situ values indicated by the shading (see BIPM, 2008, their equation 16 for how the combined standard uncertainty was calculated). Of the 63 cases depicted in Figure 7, 16 cases (~25%) of the AERONET/in-situ comparisons were within 0.02. While much of the observed difference between $SSA_{in\text{-}situ}$ and $SSA_{AERONET}$ may fall within the uncertainty of the SSA values, as noted in Schafer et al. (2014), the fact that the difference ($SSA_{AERONET}$ – $SSA_{in\text{-}situ}$) is predominately negative across all the direct comparisons found in the literature is not what would be expected from random error. Figure ~~6~~ 7 also shows the mean and 2*standard deviation of all of the points (black square and vertical lines) and just the literature value points (black diamond and vertical lines). Based on the characteristics of a normal distribution the standard deviation lines suggest ~80% of the points will be negative – random error would suggest only 50% of the points should be negative. Figure ~~6~~ 7 suggests

that AERONET retrievals of SSA could perhaps be used at AOD_{440}<0.4, perhaps down to AOD_{440}~0.25 or ~0.3 – even at those low AOD values the differences in SSA between AERONET and in-situ still tend to be within the AERONET uncertainty. However, as Figure ~~6~~ 7 shows, there are not a lot of direct comparisons to support such a choice.

Most of the SSA comparisons in Table 4 ~~found~~ reported fairly good agreement between AERONET and in-situ AOD, ~~suggesting~~ implying that ~~the issue is~~ the discrepancy is associated with the absorption values rather than the scattering values (since scattering is typically 90% of extinction~~- an over-estimation of the absorption contribution to AOD rather than an underestimation of the AOD scattering contribution~~. This is consistent with the AERONET AAOD values being greater than those obtained from in-situ measurements presented in Table 3. Out of the 63 profiles compared in Table 4, there are four exceptions, (three from Leahy et al. (2007) and one from this study for the BND site) where $SSA_{AERONET}$ is larger than the corresponding $SSA_{in\text{-}situ}$. Interestingly, the three exceptions from Leahy et al. (2007) were for their high AOD (AOD_{550}>0.6) cases; for their two low AOD (AOD_{550}<0.3) cases the opposite was found, i.e., $SSA_{AERONET}$<$SSA_{in\text{-}situ}$.

In summary, the literature survey featuring measurements across the globe for many aerosol types suggests that even at higher AOD conditions, direct comparisons of AERONET with in-situ aerosol profiles find that AERONET column SSA is consistently lower than the SSA obtained from in-situ measurements (although ~~often~~ mostly within the uncertainty of the AERONET SSA retrieval and in-situ measurements). If there was no consistent bias in the AERONET/in-situ comparison we would expect (AERONET_SSA – INSITU_SSA) to be evenly distributed around zero. Instead, Figure ~~6~~7, which summarizes the literature survey, suggests either that AERONET retrievals are biased towards too much absorption, or that in-situ, filter-based measurements of aerosol absorption are biased low. We note that the results from the literature (e.g., Figure ~~6~~7) indicate that the hypothesized low-bias in in-situ absorption is not associated with a single airplane's measurement system or the atmospheric conditions encountered in a single experiment. That leaves us with possible bias in the in-situ experimental methods (instrument issues (nephelometer, PSAP), treatment of f(RH), vertical coverage, sampling artifacts), all of which we have attempted to address above.

An alternative explanation is that the AERONET SSA uncertainties are non-symmetric. Dubovik et al. (2000) suggest that simulated retrievals of SSA for 'water soluble aerosol' are asymmetric when different 'instrumental offsets' are assumed, particularly at lower AOD values (0.05 and 0.2). Their figure 4 shows a much larger decrease in SSA for some instrumental offsets relative to the increase in SSA observed for an instrumental offset of the same magnitude but opposite sign. Asymmetry is also indicated for 'biomass burning' aerosol (their figure 7) although the asymmetry is in the opposite direction, i.e., the increase in SSA is larger than the decrease for a given pair of instrumental offset values. It is not obvious from their figure 7 whether the retrievals are asymmetric for simulated dust aerosol. Interestingly, at least three of the four points in Figure ~~6~~ 7 with AERONET_SSA>INSITU_SSA represent retrievals of biomass burning aerosol.

3.3 BND and SGP: in-situ vs AERONET and AeroCom model output – Statistical Comparisons

Most of the statistical comparisons between AERONET and in-situ profiles (e.g., Ramanathan et al., 2001; Leahy et al., 2007; Ferrero et al., 2011; Johnson et al., 2011) were for short-term field campaigns with a limited number of in-situ profiles. The advantage of the multi-year, in-situ vertical profiling programs at BND (401 flights) and SGP (302 flights) is that we can compare the statistics for both in-situ and AERONET values as opposed to comparing individual in-situ values to remote retrieval statistics. Figure 1 in Andrews et al. (2011) and Figure 9 in Sheridan et al. (2012) demonstrate that the BND and SGP flight programs captured the multi-year seasonality in aerosol properties at these two sites. Because of the large number of flights over an extended period of time, Skeie et al. (2011) was able to compare the seasonally averaged, in-situ absorbing aerosol profiles from BND and SGP with seasonal vertical profiles of black carbon generated by the Oslo-CTM2 model. Skeie et al. (2011) found that the model underestimated absorbing aerosol relative to the BND and SGP in-situ profiles for most seasons and altitudes, although agreement between the model and measurements tended to be better at higher altitudes.

As mentioned in the introduction, AERONET retrievals of AAOD have been used to suggest upscaling factors for modelled values of absorbing aerosol (e.g., Sato et al., 2003; Bond et al., 2013). These model/AERONET comparison studies are typically based on model and measurement statistics (i.e., properties are averaged over time and region) rather than direct comparisons due to both computational constraints and the discrete nature of the AERONET measurements. Given the statistical nature of some historical AERONET/in-situ comparisons as well as the typical model/AERONET comparison constraints, in this section we compare monthly statistics for in-situ measurements, AERONET retrievals and AeroCom model output. It should be reiterated here that we are comparing asynchronous data and that there are some additional differences amongst the data sets that need to be kept in mind: the AERONET data are rigorously cloud-screened (although cloud halo effects may persist (e.g., Jeong and Li, 2010) and only obtained during daytime; the in-situ measurements are also daytime-only and the airplane did not fly in-cloud due to FAA flight restrictions, but may have flown near clouds; and the model data include day and night with clouds and also represent values over a 1x1 degree grid.

Figure ~~7~~ 8 shows the 440 nm monthly medians of AOD, AAOD and SSA at BND and SGP based on the in-situ profile measurements, and two versions of AERONET retrievals as described below. For the in-situ properties, all profiles were used, regardless of whether there was an AERONET retrieval corresponding to the flight. The AERONET monthly medians in Figure ~~7~~ 8 use the long-term (1996-2013) AERONET data record for each site. As described previously, the lines labeled AERONET 1.5* were calculated from Level-1.5 inversion data with matching Level-2 almucantar retrievals. The lines labeled AERONET 2.0 utilized only Level-2 almucantar retrieval data. In both cases the median AERONET AOD values represent those Level-2 AOD measurements for which there was also an AAOD and SSA retrieval, ensuring that the AERONET AOD medians represent the same set of retrievals as the corresponding AAOD and SSA medians in the figure. The AERONET Level-1.5* AOD monthly medians are representative of the direct sun AERONET Level-2 AOD climatology at the two sites. Figure ~~7~~ 8

also includes the AeroCom Phase II model monthly medians for BND and SGP (Kinne et al., 2006, Myhre et al., 2013) with model emissions, meteorology and other details briefly described in Myhre et al. (2013). The AeroCom values, which were provided at 550 nm, have been adjusted to 440 nm using the reported AeroCom monthly scattering Ångström exponent to adjust AOD wavelength and assuming an absorption Ångström exponent of 1 for the AAOD wavelength adjustment. It should be noted that the three monthly data sets (AERONET, AeroCom, and in-situ) plotted in Figure ~~7~~ 8 are derived from measurements for overlapping, but not identical time periods, i.e., these plots represent climatological comparisons rather than direct comparisons of the data sets.

At both sites, the climatological seasonal patterns for AOD (i.e., high in summer, low in winter) are similar for the three data sets: in-situ measurements, AERONET Level-1.5* retrievals (recall that the AERONET 1.5* AOD is representative of the overall AERONET AOD climatology at each site) and AeroCom model output. At BND the AeroCom model AOD tends to be larger than the in-situ and AERONET 1.5* AOD values by up to a factor of two. AERONET 1.5* AOD is larger than the in-situ AOD in the summer (by up to 50%) but quite close the rest of the year (typically within 20%). While a 50% discrepancy between the AERONET and in-situ climatology may appear significant, it's important to remember that these data sets do not represent the same period of time or measurement conditions (e.g., time of day, cloud cover, aerosol events, ambient humidity, etc). Schutgens et al. (2016) shows there can be large differences when comparing values obtained with different samplings (more than 100% for AOD), particularly when there are high levels of variability in the data. At SGP the AOD monthly medians from in-situ measurements and AERONET Level-1.5* are almost identical for August-December, with slightly more discrepancy among the AOD values in summer and early part of the year. In contrast, AeroCom model median AOD values tend to agree better with AERONET 1.5* and in-situ AOD values from January-July but are noticeably higher (up to a factor of 2) in the later half of the year. At both sites, the median AERONET Level-2 AOD values (corresponding to AAOD and SSA retrievals) are much higher (by a factor of 2 or more) than the Level-1.5* and in-situ climatologies due to the AOD_{440}>0.4 constraint. During the cleanest, lowest humidity, and often cloudiest months of the year (December-February) there are none to few Level-2 almucantar retrievals of SSA and AAOD at either BND or SGP – the gray lines in Figures ~~7ab~~ 8ab are lacking data points for Jan., Feb. and Dec. at BND and Jan. and Dec. at SGP.

For AAOD at BND, the AeroCom model output falls between the AERONET 1.5* and in-situ values, with AERONET 1.5* AAOD being higher than the in-situ data by up to a factor of 8. As with AOD, the AERONET AAOD Level-2 values are much higher than the in-situ or modelled AOD values due to the constraint that they are only retrieved at high loading conditions (AOD_{440}>0.4). The three data sets (AeroCom, in-situ and AERONET 1.5*) agree best in the month of May when the median values of AAOD are within 30%. At SGP there is fairly good agreement between AeroCom model and in-situ AAOD for the first 7 months of the year, while the AERONET 1.5* monthly AAOD values are considerably higher for that same time period. For the latter part of the year the in-situ AAOD values tend to be lower than both AERONET and AeroCom AAOD values.

The AERONET 1.5* SSA values tend to be quite a bit lower at BND, and somewhat lower at SGP, 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). Figure ~~7~~ 8 also shows that the AERONET Level-2 SSA values are similar to the monthly in-situ and AeroCom SSA medians between April and November. There are no AERONET Level-2 almucantar retrievals of SSA in January or December at either site. For the February and March, median Level-2 almucantar retrievals of SSA are based on very few data points resulting in bigger discrepancies between AERONET Level-2 almucantar retrievals of SSA and the in-situ and AeroCom SSA values.

Aside from differences in magnitude, there are also differences in the seasonal patterns of AOD, AAOD and SSA for the three data sets (in-situ, AERONET 1.5* and AeroCom). For example, at BND, the AERONET and in-situ AAOD both have a bi-modal annual distribution with peaks in late spring and early fall, which is not captured by the AeroCom AAOD and which is not seen in the AOD seasonality. The observed seasonal differences may be a result of (a) the different climatology time ranges for each method and/or (b) very little overlap in the measurement times for AERONET and in-situ measurements or (c) in the case of the models, not capturing local emissions near the sites. This highlights the importance of direct (i.e., near in time and space) comparisons in order to understand these seasonal differences. The seasonal cycle plots in Figure ~~7~~ 8 also direct attention to the fact that AOD and AAOD vary independently rather than exhibiting the same seasonal pattern. This suggests that different emission sources and/or atmospheric processes control the variability of absorption and scattering aerosol over the course of the year.

3.4 Discussion

Because AERONET data are readily available and are being widely used as a benchmark data set for evaluating model output of AAOD (e.g., Chung et al., 2012; Bond et al., 2013; He et al., 2014; Wang et al., 2014) as well as for comparison with satellite retrievals and development of AAOD climatologies, we document and discuss some of the previous methods for utilizing existing AERONET retrievals that have been used to estimate AAOD at low AOD (AOD_{440}<0.4) where Level-2 retrievals do not exist. These approaches fall into several categories (1) use only Level-2 data; (2) use Level-2 and Level-1.5 data with acknowledgement of greater uncertainty in the retrievals and potentially additional measurement constraints for the Level-1.5 data; (3) make climatological assumptions about the representativeness of Level-2 SSA for low AOD conditions to obtain AAOD.

Clearly the simplest approach to minimize uncertainty in retrieved AERONET AAOD and SSA is to only use AERONET Level-2 retrievals which include the AOD_{440}>0.4 constraint. This approach has been and continues to be used (e.g., Koch et al., 2009; Bahadur et al., 2010; Chung et al., 2012; Buchard et al., 2015; Pan et al., 2015; Li et al., 2015). However, as shown in Figure 1 the vast majority of the globe has AOD_{440}<0.4, meaning few if any AERONET Level-2 AAOD or SSA retrievals will be available for most locations. This approach is quite useful in regions (or for case studies) with high aerosol loading (high AOD). However, excluding low loading conditions is likely to cause AERONET AAOD statistics to be biased high. This is

particularly important when evaluating models in clean locations such as the Arctic. The AOD_{440}>0.4 constraint may also affect the SSA statistics.

Some studies have utilized AERONET Level-1.5 retrievals of absorption-related aerosol properties in order to avoid being limited to the high AOD levels required by Level-2 data (e.g., Lacagnina et al., 2015; Mallet et al., 2013). These studies note that Level-1.5 data include more relevant AOD values but that there are accompanying higher uncertainties in the retrievals for absorption related properties. Mallet et al. (2013) use Level-1.5 data to evaluate the spectral dependence of aerosol absorption. Lacagnina et al. (2015) utilize both Level-2 and Level-1.5 AERONET data in their comparison with PARASOL satellite retrievals of SSA and AAOD. For the Level-1.5 data they apply the additional requirement that the solar zenith angle must be ≥50°. Lacagnina et al. (2015) find quite good agreement (within +/- 0.03) for AAOD and note that larger differences between PARASOL and AERONET retrieval occur at higher AOD conditions, possibly due to less homogenous aerosol (i.e., plumes).

A more sophisticated approach to deal with SSA (and hence AAOD uncertainties) at low AOD is implemented by Wang et al. (2014). They make the assumption that SSA is independent of AOD (at least as a function of season) and utilize climatological Level-2 SSA values for each season with the measured AOD in order to obtain AAOD. The seasonal climatologies of SSA are based on 12 years of Level-2 AERONET data. For the two US continental sites studied in this paper, the approach of Wang et al. (2014) would likely minimize the potential AERONET tendency towards high AAOD at low AOD conditions as the Level-2 monthly climatological SSA values are quite similar to SSA values obtained by in-situ measurements (Fig. ~~7~~8).

A similar, though statistical, approach was used in Bond et al.'s (2013) bounding BC paper in order to reduce the uncertainty and better represent AERONET SSA and AAOD retrievals at low AOD. Bond et al. (2013) worked with AERONET monthly local statistics for the time period 2000-2010. Monthly values of AAOD and SSA at 550 nm were calculated from size distributions and refractive index when there were at least 10 valid inversion retrievals for that month at that site in the 2000-2010 period (most sites had more than 10 retrievals in a given month over the 11 year period). It was assumed in Bond et al. (2013), based on AERONET reported uncertainties, that the retrieved absorption-related values were more reliable at larger AOD and so they made some adjustments to account for this. For each site, AAOD and SSA values were binned as a function of AOD (there were five AOD bins, with each bin corresponding to 20% of the AOD probability distribution). For lower AOD conditions, the calculated AAOD and SSA values were replaced by values obtained during larger AOD conditions for the same month as follows: (i) the SSA and AAOD values corresponding to AOD_{550} of 0.25 were prescribed for all SSA and AAOD observations at lower AOD and (ii) for locations where all AOD_{550}<0.25, the average SSA and AAOD of the upper 20th percentile of AOD observations at the site was prescribed for all lower AOD bins. Finally, the average of all five bins was used to determine the overall monthly average. In the case of AAOD the bin averages were simply averaged to get the monthly value while for SSA the AOD-weighted bin averages were averaged to get the monthly value. Note: the AOD_{550}=0.25 cutoff point used in Bond et al. (2013) corresponds (approximately) to AOD_{440}=0.35 for smaller particles and AOD_{440}=0.25 when large particles are present. Thus it is less strict than the AERONET recommended constraint of AOD_{440}>0.4, but it

had been suggested that the recommended constraint might be too restrictive (pers. comm., O. Dubovik).

One drawback affecting approaches using climatological values of SSA (e.g., Wang et al., 2014; Bond et al., 2013) is that they may not account for the systematic variability that has been observed between SSA and loading at many sites, although AOD is usually more variable than the composition (or SSA). Still some studies with in-situ data (e.g., Delene and Ogren, 2002; Andrews et al., 2013; Pandolfi et al., 2014; Sherman et al., 2015) indicate that SSA systematically decreases with decreasing aerosol loading. A similar SSA/AOD systematic variability relationship is also observed at some North American AERONET sites. Schafer et al. (2014; their figure 6) shows SSA decreasing at lower loading for the GSFC site near Washington D.C. during the period of their field campaign; they also show similar relationships between SSA and AOD based on the long-term data for three mid-Atlantic AERONET sites. Additionally, a quick survey (not shown) of other long-term North American AERONET sites with good statistics (i.e., lots of points) for Level-1.5 SSA retrievals (e.g., Billerica (Massachusetts), Bratts Lake (Saskatchewan, Canada), COVE (Virginia), Egbert (Ontario, Canada), Fresno (California), Konza (Kansas), SERC (Maryland), and University of Houston (Texas)) indicates this systematic relationship may be observed at a wide range of locations in North America. Such climatological analyses may mask short-lived and/or infrequent aerosol events (e.g., dust or smoke incursions) that may have significantly different optical properties.

Figure 8 9 shows the systematic relationships between SSA_{440} and AOD_{440} for BND and SGP for both the AERONET retrievals and in-situ profile measurements. Consistent with previous figures, we have utilized SSA values for $AOD_{440}<0.4$ when there was a valid Level-2 AOD inversion retrieval, i.e., what we call AERONET Level-1.5*. Also included on the figure is a line showing the SSA_{550} versus scattering ($\sigma_{sp,550}$) relationships for the surface measurements at BND and SGP. The surface measurements are made at low RH conditions (RH<40%) and adjusted to ambient RH using the available meteorological measurements at the site (ambient RH at 2 m at SGP and ambient RH at 10 m at BND); adjustment of the surface measurements from dry to ambient conditions shifts the SSA_{550} values upward (assuming absorption is not affected) and the scattering values to the right.

Figure 8 9 suggests that for all three sets of measurements at both sites, there is a consistent decrease in SSA as aerosol loading decreases below $AOD_{440}=0.2$. This relationship implies that a climatology based on SSA values measured at high AOD may underestimate the AAOD climatology. The AERONET SSA values are lower than the in-situ profile values as would be expected from the results presented in sections 3.1 and 3.3. The AERONET SSA values are also lower than the surface in-situ SSA values – the surface in-situ SSA values adjusted to ambient conditions are quite similar to those obtained from the in-situ vertical profiles. It should however be noted that despite the discrepancy between in-situ and AERONET SSA values, Figure 8 9 shows that the SSA values for all three sets of measurements at SGP are within the reported AERONET SSA uncertainty range of 0.05-0.07 for $AOD_{440}<0.2$ across the narrow and low AOD range shown in the figure. At BND the SSA values are within the AERONET SSA uncertainty range down to AOD_{440}~0.1. At the lowest AOD values ($AOD_{440}<$~0.05) the AERONET SSA values diverge, consistent with very large uncertainties expected in the

AERONET SSA retrievals in the cleanest conditions. Uncertainty in the AERONET AOD retrieval may begin to affect the AERONET SSA retrieval where +/- 0.01 AOD uncertainty is equivalent to a 20% change in AOD for AOD of 0.05. In addition, at such low AOD values, the surface reflectance uncertainties may influence AERONET's retrieval of SSA. Figure ~~8~~ 9 suggests that, in terms of the shape of the systematic variability plot, there are no obvious retrieval issues for AERONET SSA retrievals in the range $0.05<AOD_{440}<0.2$, although this is in the AOD range where high uncertainty in the SSA retrieval is expected (Dubovik et al., 2000).

There are large differences (orders of magnitude) in the number of data points in each of the data sets; the number of points in each bin is indicated by the color-coded histograms shown on Figure ~~8~~9. The mean standard error (MSE) in SSA (MSE=(standard deviation)/(number of points)$^{1/2}$) is indicated by the shading surrounding the solid colored lines. The MSE is quite similar for the AERONET 1.5* and in-situ profile measurements across the AOD range plotted in Figure ~~8~~9, suggesting the observed systematic variability is not merely due to small numbers of data points in each bin, particularly at lower loading. However, the fact that the AERONET MSE is approximately the same as the in-situ profile MSE, despite having approximately an order of magnitude larger number of points/bin, indicates that variability in the retrieved AERONET SSA is larger than the variability in SSA derived from in-situ profile measurements.

This study has utilized a valuable but spatially limited (i.e., two rural continental North American sites) climatological vertical profile dataset to explore AERONET retrievals of AAOD and SSA. Clearly, one way to address the observed discrepancy between in-situ and AERONET AAOD is to pursue a focused measurement program designed to acquire statistically robust in-situ vertical profiles over AERONET sites representing a wide range of conditions and aerosol types. This type of measurement program has been proposed to evaluate satellite retrievals and better characterize atmospheric aerosol (R. Kahn, SAM-CAAM, pers. comm.). Further evaluation and development of in-situ instrumentation for measuring aerosol absorption is also necessary, particularly in assessing the effects of coatings and hygroscopity on the resulting absorption values. Additional evaluation of the AERONET retrieval algorithm may provide insight into a potential SSA and, thus, AAOD bias (e.g., Hashimoto et al., 2012). The discrepancies reported here between in-situ and AERONET values of AAOD and SSA suggest that caution should be used in upscaling model results to match AERONET retrievals of absorbing aerosol as this will have a significant impact on global radiative forcing estimates. The work of Wang et al. (2016) has shown that other factors (e.g., the spatial resolution of models and emissions) may also contribute to the differences observed between model and AERONET retrievals of AAOD. Thus, to really be able to understand and simulate the influence of absorbing aerosol on radiative forcing will require expanded effort on both the measurement and modeling fronts.

4. Conclusion

AERONET retrievals of SSA at low AOD conditions (below the recommended $AOD_{440}<0.4$ constraint) are consistently lower than coincident and co-located in-situ vertical profile observations of SSA (based on detailed comparisons at two rural sites in the US). Correspondingly, AERONET retrievals of AAOD at low AOD are consistently higher than those obtained from in-situ profiles. A survey of the literature suggests that even at higher loading

(AOD_{440}>0.4) AERONET SSA retrievals tend to be lower than SSA values obtained from vertical profiling flights, although discrepancies are within the reported uncertainty bounds down to ~ AOD_{440}>0.3. The tendency of AERONET SSA to be lower suggests either that AERONET retrievals over-estimate absorbing aerosol or that the in-situ measurements under-estimate aerosol absorption. Since the observed discrepancy in SSA can not definitively be attributed to either technique, the idea of scaling modelled black carbon concentrations upwards to match AERONET retrievals of AAOD should be approached with caution. If the AERONET SSA and AAOD retrievals are indeed biased towards higher absorption, such an upscaling may lead to aerosol absorption overestimates, particularly in regions of low AOD. If the discrepancy between the in-situ and AERONET AAOD is due to issues with the in-situ measurements of absorption, the only way we see to increase the in-situ absorption values is a significant enhancement (on the order of a factor of 2 or more) in absorption due to a coating effect. While that level of absorption enhancement factor is within the range suggested by modelling studies, it is significantly higher than many observations of absorption enhancement for ambient aerosol reported in the literature.

The AERONET retrievals of SSA and AAOD have been used as a primary constraint on global model simulations of aerosol absorption. Using only Level-2 retrievals of AAOD (i.e., for AOD_{440}>0.4) on a global scale (e.g., Koch et al., 2009; Bahadur et al., 2010; Chung et al., 2012; Buchard et al., 2015; Pan et al., 2015; Li et al., 2015) is likely to lead to significant over-estimates of absorption in cleaner regions although it may be appropriate for conditions of high loading. Several different approaches of varying complexity have been developed to better represent absorbing aerosol for cleaner conditions. Some of these approaches utilize SSA at high AOD to estimate AAOD at lower AOD conditions (e.g., Bond et al., 2013; Wang et al., 2014), while others utilize Level-1.5 retrievals with the added uncertainty that entails (e.g., Lacagnina et al., 2015; Mallet et al., 2013). Based on the analysis presented here, we cannot say how to best estimate SSA or AAOD from AERONET retrievals for the low AOD conditions prevalent around much of the globe.

Some in-situ measurements suggest that a systematic relationship exists between SSA and AOD, but these measurements are spatially sparse and typically not made at ambient conditions. Nonetheless, systematic relationships between SSA and AOD, similar to those seen in the in-situ data at the two sites, are also observed for multiple North American AERONET sites. The existence of such a systematic relationship may limit the accuracy of AAOD estimates when climatological values for SSA from high AOD retrievals are assumed to apply at low loading conditions. However, for the two mid-continental rural sites studied here, the statistically-based monthly medians of SSA from Level-2.0 inversions (i.e., SSA values derived for AOD_{440}>0.4) appear to be quite consistent with monthly SSA values obtained from in-situ measurements and AEROCOM model simulations. This suggests that, at these two sites, using the Level-2.0 inversion SSA to retrieve monthly AAOD at lower AOD conditions (e.g., AAOD=AOD*SSA) would not bias the resulting monthly AAOD high, as would occur if only AAOD values for high AOD cases are included in the AAOD statistics. . This may not be true for other locations or averaging times. Further, for these two sites, a more complex approach to retrieve monthly AAOD is needed for very clean months when no Level-2.0 inversions are available.

This study points to several areas where additional research would be useful in resolving the observed AERONET/in-situ absorption-related discrepancies. First, continued laboratory, field and modelling efforts are needed to elucidate and unify the current inconsistencies in the literature on the effects of coatings on absorption enhancement reported for field and lab measurements and for model simulations. Second, a more extensive evaluation of the hygroscopicity of ambient (not lab-generated!) absorbing particles would be helpful. Third, better characterization of how filter-based measurements of absorption respond to coated particles would be useful, not just in the context of this study, but also for improving our understanding of the in-situ absorption data acquired by long-term, surface aerosol monitoring networks (e.g., GAW). Finally, the development of a focused measurement program designed to acquire statistically robust in-situ vertical profiles over AERONET sites representing a wide range of conditions and aerosol types could be used to explore the relationships between retrievals of column properties and variable aerosol profiles and to provide further validation of the inversion retrieval data products.

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Tables

Table 1a Statistical values (medians, means and standard deviations) of AERONET versus in-situ comparison where there was an AERONET retrieval within +/-3 h of the end of a 2 h flight profile. AERONET values are for Level-1.5 data when there was a Level-2 AOD value and an almucantar retrieval. (First value in each cell is median; second set of values in each cell are mean± Std.Dev, third row is number of AERONET retrievals corresponding to flights (in AERONET columns) or number of flights (In-situ columns)). These numbers represent the blue points in Figures ~~3~~4-~~5~~6.

	BND		SGP	
	AERONET	In-situ	AERONET	In-situ
AOD	0.118; 0.146±0.099	0.114; 0.135±0.139	0.138; 0.146±0.099	0.137; 0.147±0.077
AAOD	0.013; 0.013±0.007	0.003; 0.005±0.006	0.019; 0.023±0.008	0.004; 0.004±0.003
SSA	0.895; 0.898±0.034	0.961; 0.964±0.020	0.847; 0.839±0.038	0.971; 0.973±0.011
#	51 retrievals[1]	21 flights	23 retrievals[1]	11 flights

[1]retrievals are flight-averaged prior to calculating statistics.

Table 1b Statistical values (medians, means and standard deviations) of AERONET versus in-situ comparison where there was an AERONET retrieval within +/-3 h of the end of a 2 h flight profile and AERONET AOD_{440}>0.2. AERONET values are for Level-1.5 data when there was a Level-2 AOD value and an almucantar retrieval. (First value in each cell is median; second set of values in each cell are mean± Std.Dev, third row is number of AERONET retrievals corresponding to flights (in AERONET columns) or number of flights (In-situ columns)). These numbers represent the purple points in Figures ~~4~~5-~~5~~6.

	BND		SGP	
	AERONET	In-situ	AERONET	In-situ
AOD	0.306; 0.304±0.125	0.299; 0.331±0.230	0.269	0.238
AAOD	0.025; 0.019±0.012	0.010; 0.013±0.012	0.034	0.009
SSA	0.941; 0.942±0.023	0.971; 0.966±0.010	0.875	0.964
#	6 retrievals[1]	4 flights	2 retrievals	1 flights

[1]retrievals are flight-averaged prior to calculating statistics.

Table 2 Number of AERONET/IN-SITU AOD and AAOD flight matches as a function of various AERONET constraints and the +/- 3h time window.

	BND (2006-2009)	SGP(2005-2007)
Total profile flights	402	171
Level-2 AOD	73	37
Level-2 AOD+almucantar retrieval[1]	21	11
Level-2 AOD+almucantar retrieval+AOD_{440}>0.20	2	1
Level-1.5* AAOD	21	11
Level-1.5* AAOD + AOD_{440}>0.20	4	1
Level-2 AAOD	1	0

[1]an almucantar retrieval does not necessarily imply an AAOD retrieval

Table 3 Direct AAOD comparisons – AERONET ("RS") vs In-Situ ("IS")

Study, # profiles Citation(s)	**Location, aerosol type AOD comments Alt. range**	**Instruments corrections size cut**	**AAOD comparison information**	**Comments**
BND 24 profiles This study	Central US Rural, continental AOD_{440} range: 0.04-0.55 AOD comparison See Fig. ~~3a~~4a 150-4200 m agl	PSAP-3wave TSI neph-3wave B1999, O2010, AO1998 f(RH) adjust Dp<5-7 μm	Wavelength=440, 670 nm Ångström interpolation RS AAOD>IS AAOD $AAOD_{440}$ range: 0.001-0.042	Profiles matched within 3 hours of AERONET measurement. Profiles within 15 km of AERONET measurement. Used V2 AERONET Level 1.5 AAOD values for cases with valid V2 AERONET Level 2.0 AOD value. Extrapolated from lowest altitude range to ground to account for aerosol below plane
SGP 14 profiles This study	Central US Rural, continental AOD_{440} range: 0.06-0.43 AOD comparison See Fig. ~~3b~~4b 150-4200 m agl	PSAP-3wave TSI neph-3wave B1999, O2010, AO1998 f(RH) adjust Dp<5-7 μm	Wavelength=440, 670 nm Ångström interpolation RS AAOD>IS AAOD $AAOD_{440}$ range: 0.012-0.052	Profiles matched within 3 hours of AERONET measurement. Profiles within 1 km of AERONET measurement Used V2 AERONET Level 1.5 AAOD values for cases with valid V2 AERONET Level 2.0 AOD value. Extrapolated from lowest altitude range to ground to account for aerosol below plane
MAC 13 profiles Corrigan et al., 2008	Indian Ocean Pollution AOD_{440} range: 0.1-0.6	Aethalometer 3-wave OPC +Mie for scattering A2005	Wavelength not provided RS AAOD>IS AAOD $AAOD_{440}$ range: 0.005-0.033	No details on how profiles matched with retrievals in terms of time or distance. No details on version of AERONET data used; this is relevant, given low AODs in first half of study – not sure if there were comparisons for low AODs.

	No AOD comparison 0-3200 m asl	Dp<5 μm		Note: this study is the one cited by Bond et al. (2013) to support the use of AERONET to scale modeled BC values

In-situ instrument corrections: B1999=Bond et al., 1999; O2010=Ogren, 2010, AO1998=Anderson and Ogren, 1998; A2005=Arnott et al., 2005; Ångström interpolation – indicates in-situ wavelength adjusted to AERONET wavelength using Ångström interpolation; f(RH) adjust – indicates the in-situ measurements were adjusted to ambient humidity conditions for the AOD comparison. IS=In-situ measurements, RS=Remote sensing (AERONET) measurements

Table 4 SSA comparisons – AERONET vs In-situ

Study, # profiles Citation(s)	**Location, aerosol type AOD comments Alt. range**	**Instruments, Corrections, Inlet size cut**	**SSA comparison information**	**Comments**
BND 24 profiles This study	Central US Rural, continental AOD_{440} range: 0.04-0.55 AOD comparison: See Fig. ~~2a~~4a 150-4200 m agl	PSAP-3wave TSI neph-3wave B1999, O2010, AO1998 f(RH) adjust Dp<5-7 μm	Wavelength=440, 670 nm Ångström interpolation RS SSA<IS SSA	Profiles matched within 3 hours of AERONET measurement. Profiles within 15 km of AERONET measurement. Used V2 AERONET Level 1.5 AAOD values for cases with valid V2 AERONET Level 2.0 AOD value. Extrapolated from lowest altitude range to ground to account for aerosol below plane
SGP 14 profiles This study	Central US Rural, continental AOD_{440} range: 0.06-0.43 AOD comparison:	PSAP-3wave TSI neph-3wave B1999, O2010, AO1998 f(RH) adjust	Wavelength=440, 670 nm Ångström interpolation RS SSA<IS SSA	Profiles matched within 3 hours of AERONET measurement. Profiles within 1 km of AERONET measurement. Used V2 AERONET Level 1.5 AAOD values for cases with valid V2 AERONET Level 2.0 AOD value.

	See Fig. ~~2b~~4b 150-4200 m agl	Dp<5-7 μm		Extrapolated from lowest altitude range to ground to account for aerosol below plane
AAO (BND) 1 profile Esteve et al., 2012	Central US Rural, continental AOD_{550} = 0.65 AOD comparison: RS AOD>IS AOD 150-4200 m agll	PSAP-3wave TSI neph-3wave B1999, O2010, AO1998 f(RH) adjust Dp<5-7 μm	Wavelength=550 nm Power law interpolation RS SSA < IS SSA	Profiles matched within 2 hours of AERONET measurement. Profiles within 15 km of AERONET measurement. Used V2 AERONET Level 2.0 AOD value. Extrapolated from lowest altitude range to ground to account for aerosol below plane
DISCOVER-AQ 12 profiles Schafer et al., 2014	East Coast US Polluted air AOD_{440}>0.2 AOD compare: RS AOD > IS AOD (by 23%)* 367-3339 m	PSAP-3wave TSI neph-3wave V2010, AO1998* f(RH) adjust Dp<4 μm*	Wavelength=550 nm AERONET "interpolated" to 550 (no detail provided) In-situ absorption interpolated to 550 using Ångström interpolation RS SSA < IS SSA	Profile matched within 45 min of AERONET measurement. Profile within 1 km of AERONET measurement. Used V2 AERONET Level 2.0 values in paper Altitude range: at least <500 m and >1500 m for column comparisons, min and max altitudes: 367 m and 3339 m Did not specify agl or asl but those are similar for the location.
CLAMS 1 profile Magi et al., 2005	East Coast US Polluted air AOD_{440}=0.60 AOD comparison: RS AOD > IS AOD (by 15%)	PSAP-1wave MSE neph-3wave B1999, AO1998 f(RH) adjust Inlet size cut not reported, Sinha,	Wavelength=550 nm Wave_adj =quadratic polynomial interpolation RS SSA < IS SSA	Profile matched within 1 hour of AERONET measurement. Profile within 3 km of AERONET measurement. Retrieved V2 AERONET Level 2.0 AOD_{440} from http://aeronet.gsfc.nasa.gov/ Also compared campaign AERONET average

	170-1500 m agl	2003 suggests Dp<4 μm		with profile average: SSA's much closer, but profiles weren't necessarily close in time or space to AERONET site
ESCOMPTE 1 profile Mallet et al., 2005	Avignon, France Pollution AOD_{440}>0.55 No AOD comparison 100-2900 m	PSAP-1wave TSI neph-3wave B1999, A1999 No f(RH) adj Inlet Dp not given	Wavelength=550 nm Wave_adj = estimated from visual inspection (spectral dependence is relatively flat) RS SSA < IS SSA	Profile matched within 1 hour of AERONET measurement. Profile within 10 km of AERONET measurement. Used V2 AERONET Level 2.0 AOD_{440} from http://aeronet.gsfc.nasa.gov/, not stated in paper. Did not adjust in-situ measurements for f(RH), so presumably IS SSA would increase so it was even larger than RS SSA. Did not specify agl or asl
SAFARI 5 profiles Leahy et al., 2007 UW plane	Southern Africa Biomass burning AOD_{550}>0.28-1.12 AOD comparison: RS AOD > IS AOD RS=1.12*IS-0.05 R^2=0.99 100-5320 m asl	PSAP-1wave MSE neph-3wave B1999;H2000 f(RH) adjust Dp<4 um	Wavelength=550 nm Wave_adj= 2nd order polynomial For AOD_{550}>0.6 (3 profiles) RS SSA > IS SSA For AOD_{550}<0.3 (2 profiles) RS SSA $\leq$ IS SSA	Profiles matched within 1-4 hours of AERONET measurement. Profiles within 20 km of AERONET measurement. Used V2 AERONETLevel 2.0 Also found: AEROCOM model>insitu Altitude range is min and max over 5 flights – no flights covered that entire range). They used AATS to account for aerosol above plane and extrapolated down to acct for aerosol below plane. (Altitude range from flight info in Magi et al., 2003)

SAFARI 1 profile Haywood et al, 2003 C-130	Southern Africa Biomass burning AOD_{440}=0.71 AOD comparison: RS AOD < IS AOD 330-3420 m agl	PSAP-1wave TSI neph-3wave B1999,AO1998 No f(RH) adj Dp<2-4 μm	Wavelength=native Wave_adj = none RS SSA < IS SSA	Profile matched within 2 hours of AERONET measurement. Profiles within 10 km of AERONET measurement. Used V2 AERONET Level 2.0 data from http://aeronet.gsfc.nasa.gov/ They defend the lack of f(RH) correction because (a) ambient RH values < 56% and (b) previous measurements of f(RH) of BB aerosol suggest minimal hygroscopicity Paper mostly focused on size dist comparison; SSA comparison seems like afterthought. Extrapolated from lowest altitude range to ground to account for aerosol below plane
DABEX 3 profiles Osborne et al., 2008	Africa Dust/BB AOD comparison RS AOD < IS AOD (by up to 40%) AOD_{550}~0.3-0.6 100-5000 m	PSAP-1wave TSI neph-3wave B1999,AO1998 No f(RH) adj Dp<2-4 μm	Wavelength=550 nm Wave_adj=log interpolation RS SSA < IS SSA	No details on how profiles matched with retrievals in terms of time. Profiles within 100 km of AERONET measurement Used V2 AERONET Level 2.0 They defend the lack of f(RH) correction because (a) ambient RH values are mostly low (<60%) and (b) previous measurements of f(RH) of BB aerosol suggest minimal hygroscopicity Jan 21, 23 and 30 profiles IS overpredicts AOD so IS SSA is greater

				than RS SSA Suggest it could be due to large particle correction to IS measurements using PCASP. McConnell et al., (2008) suggests problems with nephelometer sensitivity Did not specify agl or asl Altitude range is min and max over 4 flights – no flights covered that entire range
DABEX 1 profile Johnson et al., 2009	Africa DUST/BB AOD comparison: RS AOD < IS AOD (by ~10%) AOD_{550} > 0.7 150-3000 m	PSAP-1wave TSI neph-3wave B1999,AO1998 No f(RH) adj Dp<2-4 μm	Wavelength=550 nm Wave_adj=log interpolation RS SSA < IS SSA	Profile matched within 1 hour of AERONET measurement. Profile within 100 km of AERONET measurement Used V2 AERONET Level 2.0 They defend the lack of f(RH) correction because ambient RH values are mostly low (<40% with a max of 70%) Jan 19 profile Incorrectly used Mie to adjust σ_{ap} to 550 after B1999 applied Did not specify agl or asl

IS=In-situ measurements, RS=Remote sensing (AERONET) measurements. In-situ instrument corrections: B1999=Bond et al., 1999; V2010=Virkula et al., 2010;O2010=Ogren, 2010; AO1998=Anderson and Ogren, 1998; H2000=Hartley et al., 2000; A2005=Arnott et al., 2005; Ångström interpolation – indicates wavelength adjustment using Ångström exponent interpolation; f(RH) adjust – indicates the in-situ measurements were adjusted to ambient humidity conditions for the AOD and SSA comparison.
*Information about Discover-AQ flights from Crumreyrolle et al. (2014)

Figures

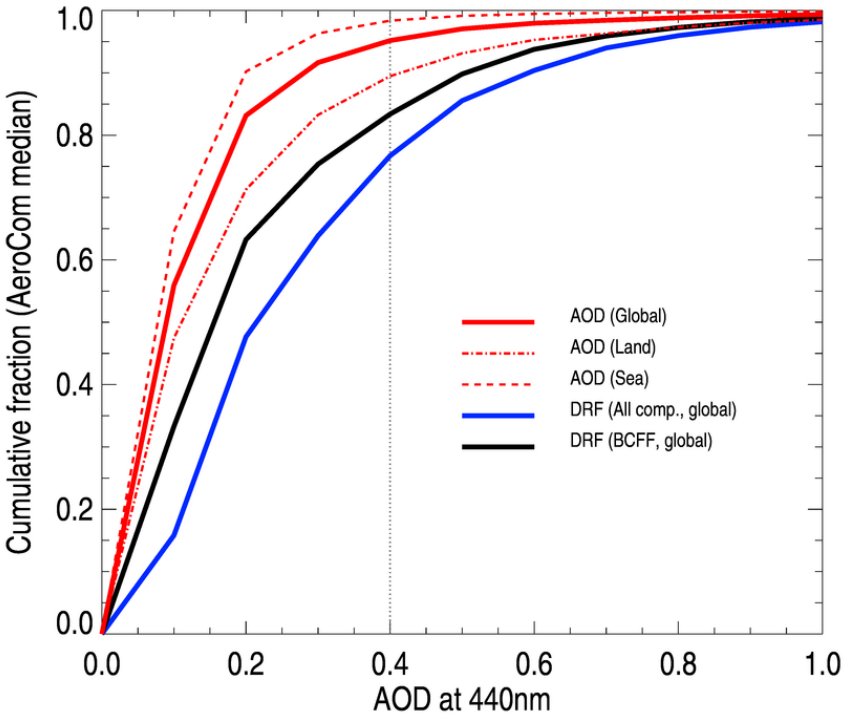


Figure 1. Cumulative AOD_{440} frequency distribution (red lines) based on output from five AeroCom models. Blue and black lines show contribution of total aerosol and fossil fuel black carbon, respectively, to the global radiation budget as a function of AOD_{440}. See text for details. Models used to generate the AOD lines include: GMI-MERRA-v3, GOCART-v4, LMDZ-INCA, OsloCTM2, and SPRINTARS-v385. Models used to generate the radiative forcing lines include all but the GMI-MERRA-v3 model. Model information and references can be found in Myhre et al. (2013).

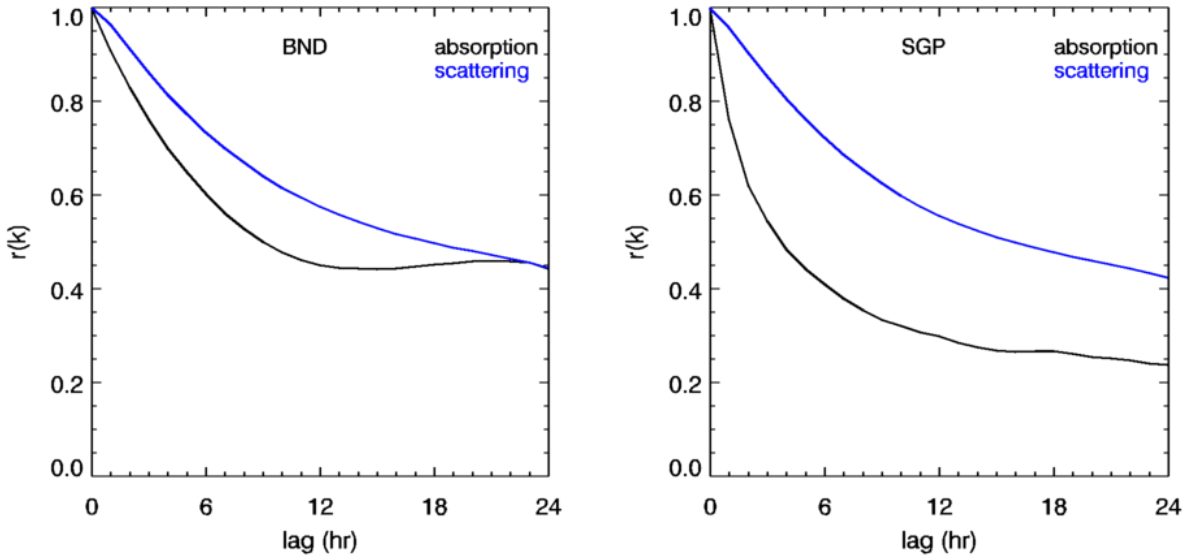


Figure 2. Correlograms for BND and SGP; wavelength = 550 nm, D_p<10 μm, based on hourly averaged surface in-situ data between 1995-2013 (BND) and 1996-2013 (SGP). The value r(k) on the y-axis represents the autocorrelation at lag time 'k'.

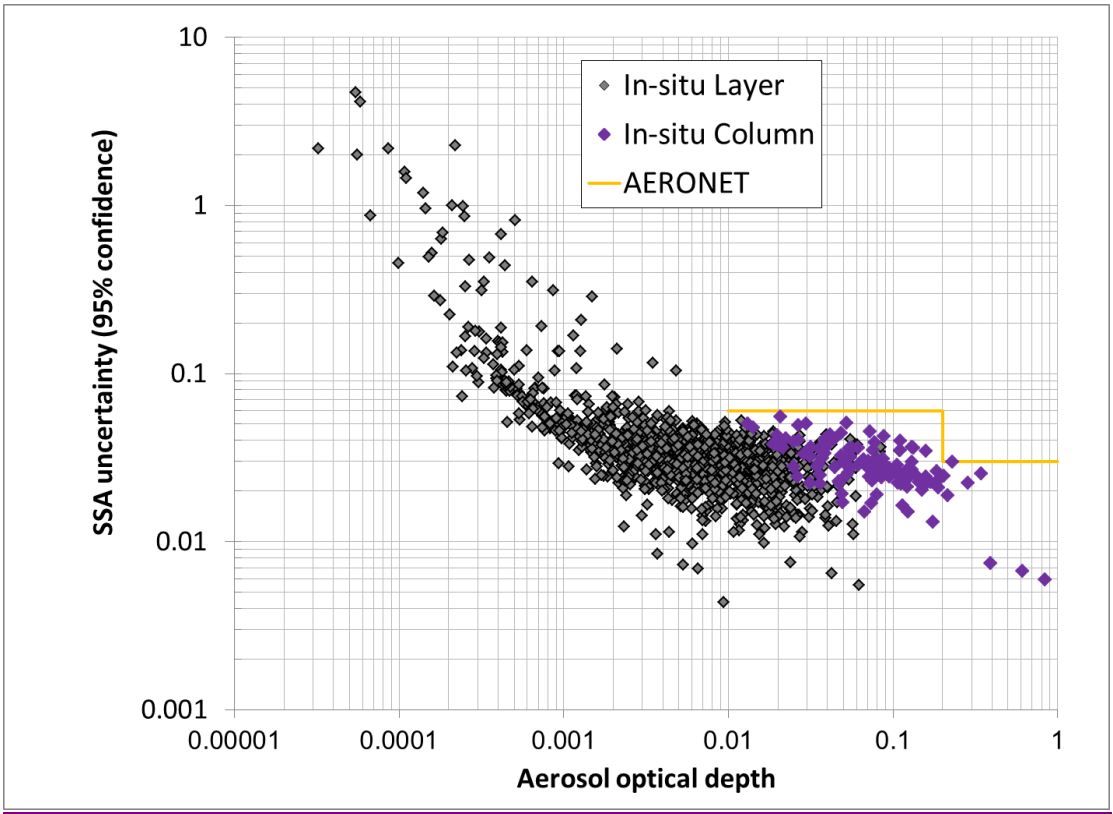
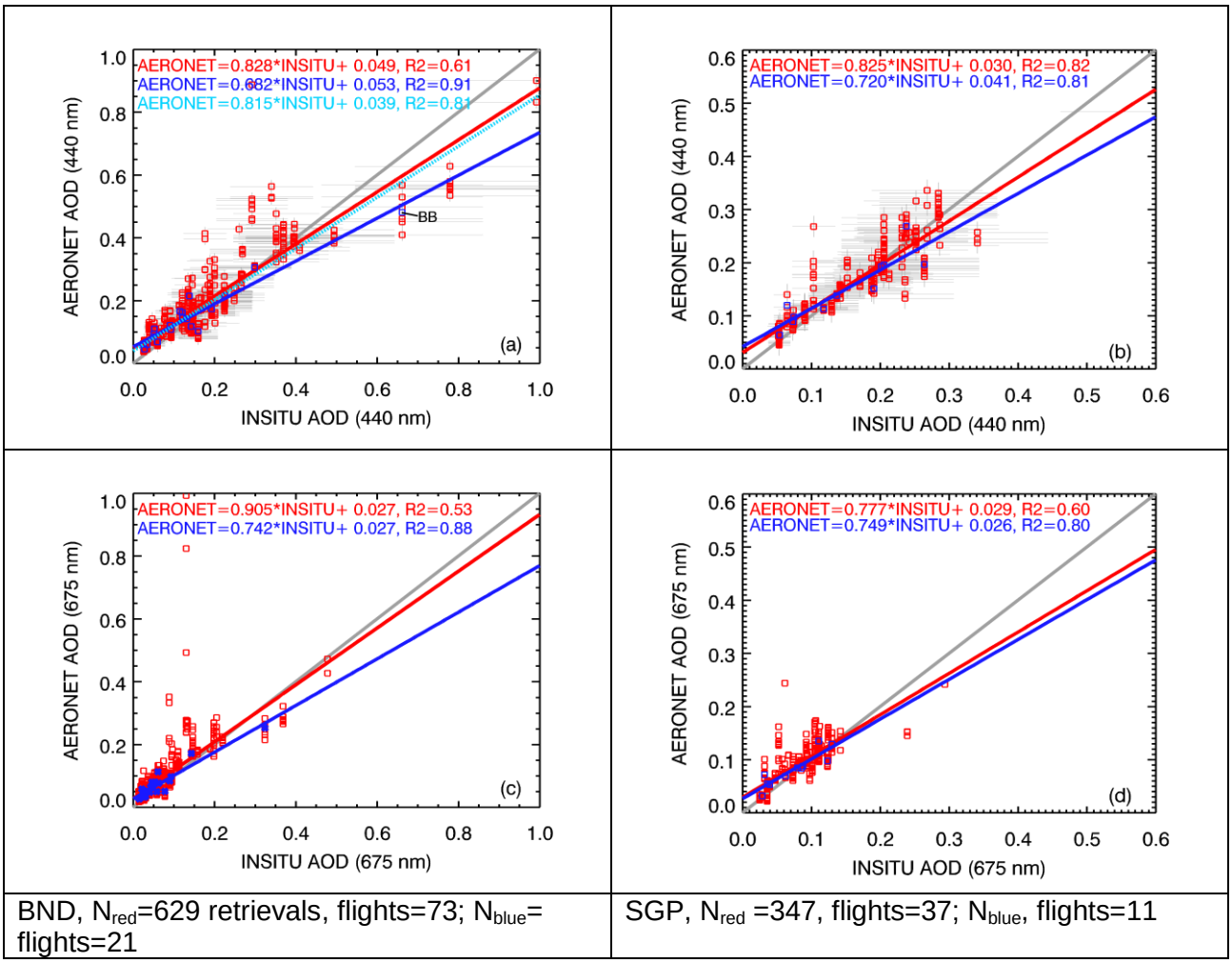
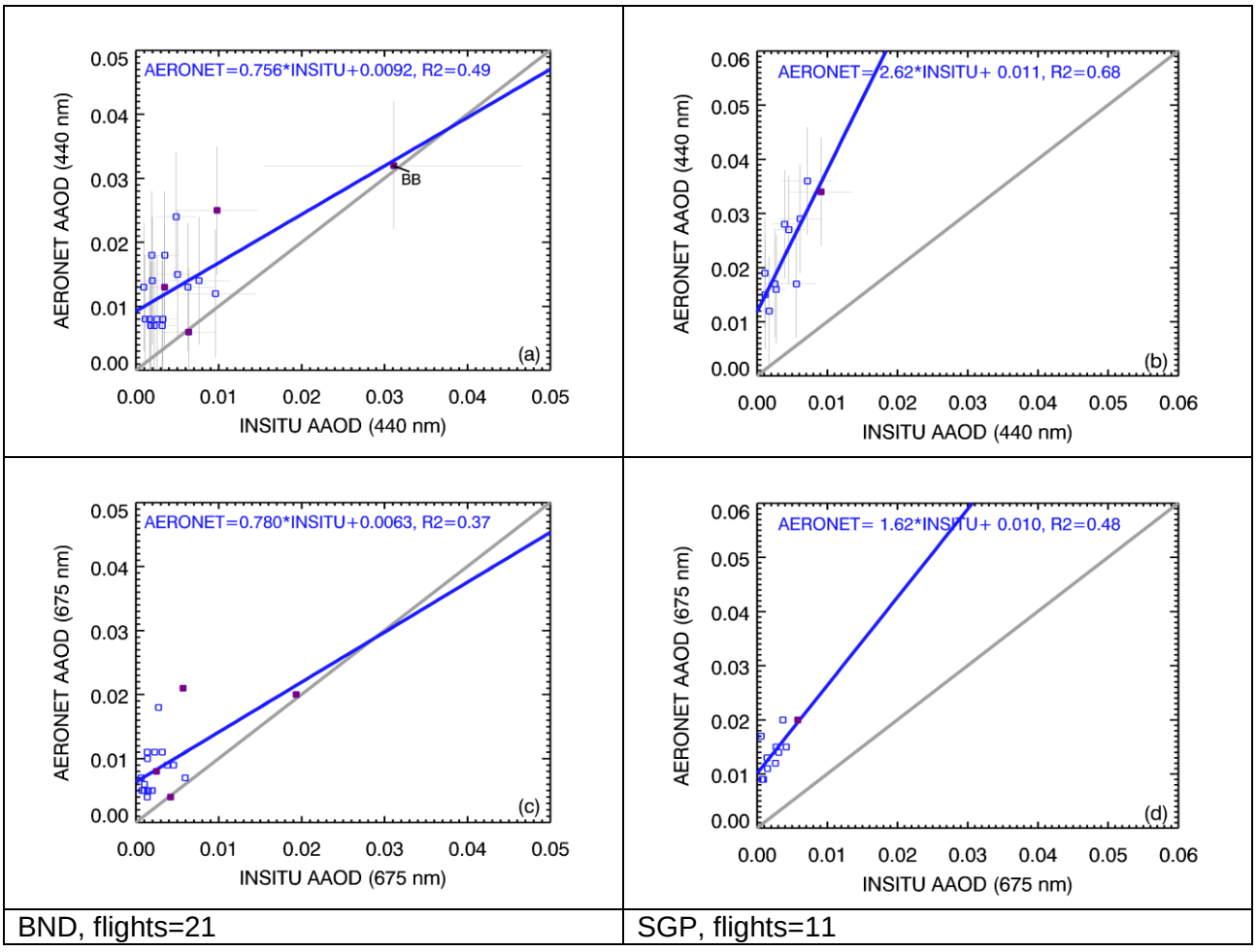


Figure 3. SSA uncertainty as a function of aerosol optical depth. Black points represent the SSA uncertainty (95% confidence) in each flight layer (all three visible wavelengths measured by the PSAP and nephelometer) as a function of the AOD of that flight layer. Purple points represent the in-situ column SSA uncertainty for each flight (again points include all three wavelengths measured by the PSAP and nephelometer) as a function of the in-situ AOD. Orange line represents the uncertainty in SSA reported by Dubovik et al. (2000, their table 2) for two AOD ranges.



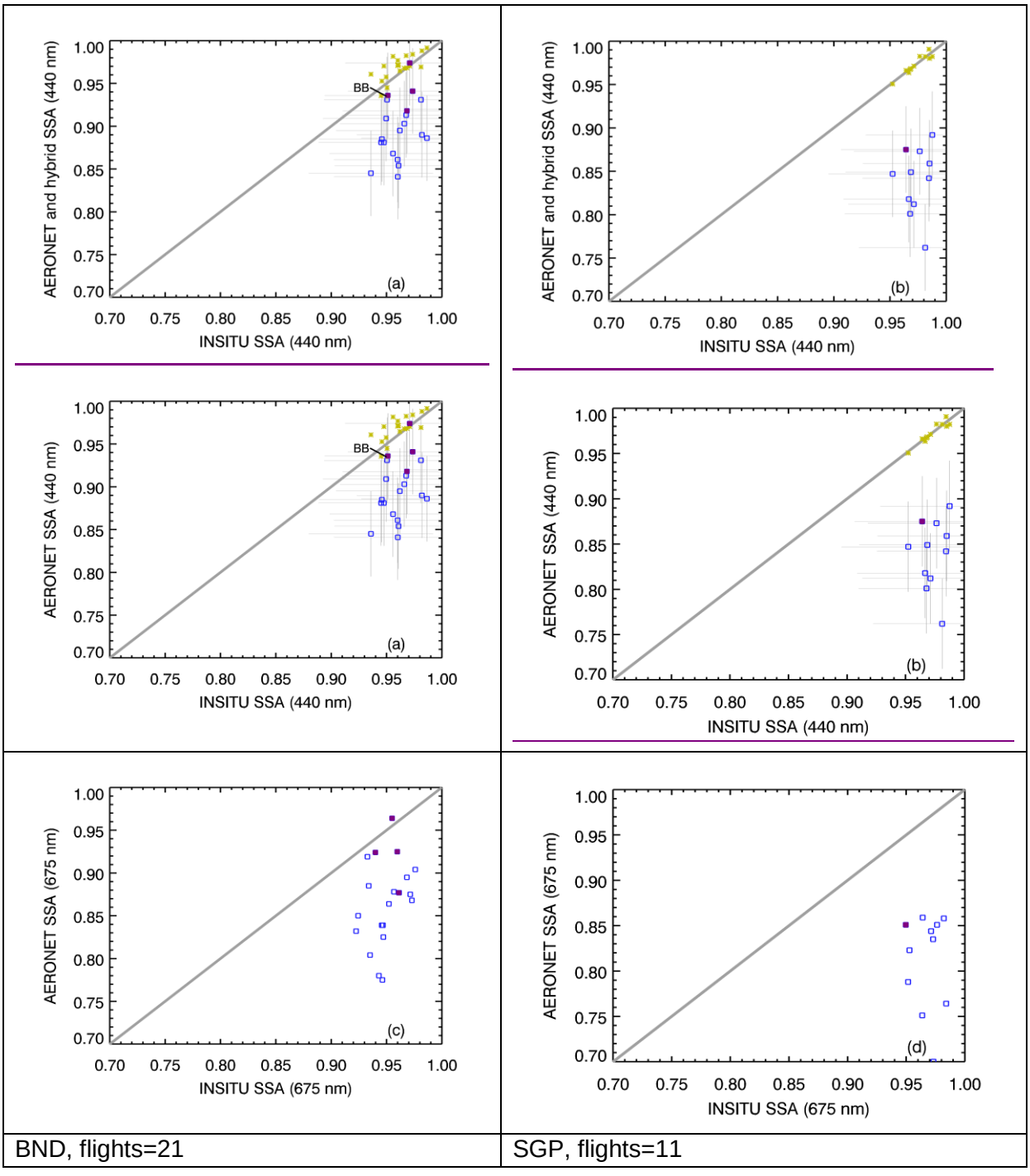
BND, N_{red}=629 retrievals, flights=73; N_{blue}= flights=21	SGP, N_{red} =347, flights=37; N_{blue}, flights=11

Figure ~~3~~4. AOD comparison (a) BND at 440 nm; (b) SGP at 440 nm; (c) BND at 675 nm; and (d) SGP at 675 nm; thick gray line is 1 to 1 line. Thin gray lines associated with each data point represent measurement uncertainties. Red points and fit line represent all AERONET direct sun Level-2 AOD measurements within +/-3 hours of end of profile. Blue points and fit line represent the average of AERONET Level-2 AOD measurements with successful almucantar retrievals within +/-3 hours of end of profile. The light blue dashed line is the fit if the BB point is excluded. Note: two BND direct sun AOD440 points corresponding to the two highest AOD675 points in the figure below are off the scale of the plot and not shown. The third high AOD440 point is partly obscured by the legend.



BND, flights=21	SGP, flights=11

Figure 45. AAOD comparison, (a) BND at 440 nm; (b) SGP at 440 nm ; (c) BND at 675 nm; and (d) SGP at 675 nm. Blue line is linear fit for all points shown; gray line is 1 to 1 line. Thin gray lines associated with each data point represent measurement uncertainties. Points show the average of AERONET Level-1.5 AAOD retrievals for which there was a successful AERONET Level-2 almucantar retrieval within +/-3 hours of end of profile. Purple points indicate the few comparisons points for which there are AERONET Level-2 almucantar retrievals and where the average AERONET AOD_{440} for those retrievals was great than 0.2.



BND, flights=21	SGP, flights=11

Figure ~~5~~6. SSA comparison, (a) BND at 440 nm (b) SGP at 440 nm; (c) BND at 675 nm; and (d) SGP at 675 nm. Blue line is linear fit for all points shown; gray line is 1 to 1 line. Thin gray lines associated with each data point represent measurement uncertainties. Blue points show the average of all AERONET Level-1.5 AAOD retrievals for which there was a successful AERONET Level-2 almucantar retrieval within +/-3 hours of end of profile. Purple points indicate the few points for which there are AERONET Level-2 almucantar retrievals and where where the average AERONET $AOD_{440}$for those retrievals was great than 0.2. The yellow points

represent the ‘hybrid SSA’ which utilizes the AERONET AOD and the in-situ AAOD to derive SSA as described in the text. The in-situ uncertainty lines here represent SSA uncertainty of 0.06 which is the worst case uncertainty.

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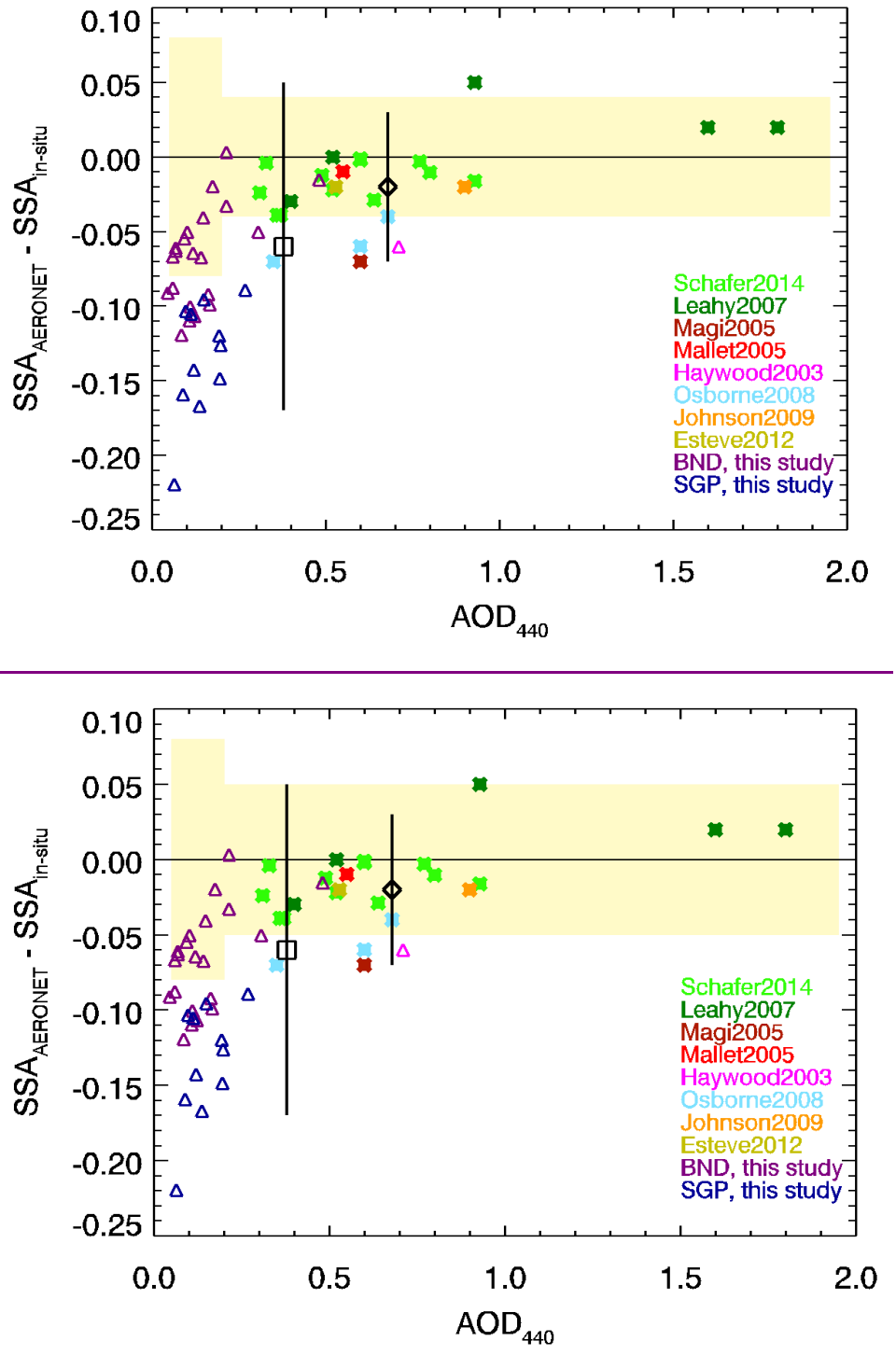

Figure ~~67~~. AOD_{440} vs [$SSA_{AERONET}$-SSA_{INSITU}] for direct comparisons studies listed in Table 4. Open symbols are for SSA_{440} difference; filled symbols are for SSA_{550} difference. AOD_{440} values for Leahy2007, Osborne2008, Johnson2009 use the Level-2 values reported on the AERONET webpage for the locations and dates of the specific profile. Shading indicates the combined

standard uncertainty of AERONET SSA values as function of AOD as reported in Table 4 of Dubovik et al. (2000) and uncertainty in the in-situ SSA ~~calculated using equation 2~~based on Figure 2. The black square and black diamond with vertical black lines represent, respectively, the mean and 2*standard deviation for all direct comparisons (including BND and SGP) and for literature direct comparisons only.

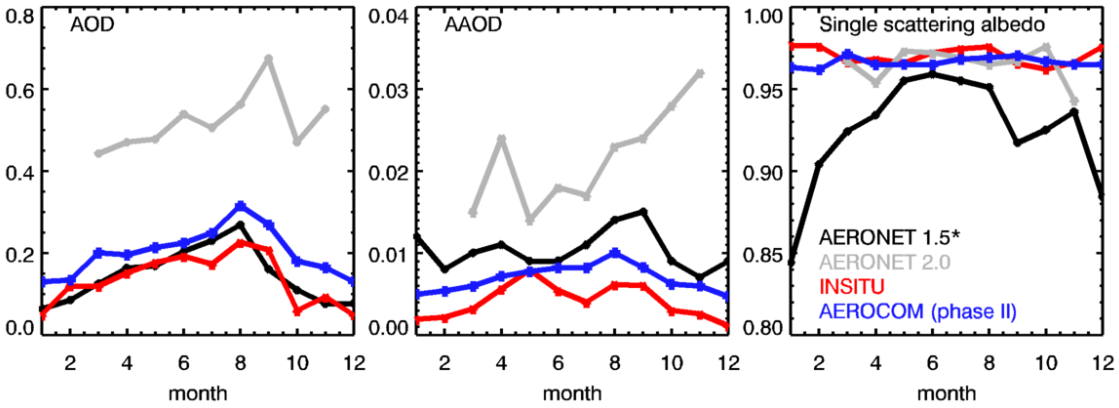

Figure ~~7a~~8a. Monthly medians of BND aerosol optical properties at 440 nm. AERONET medians are for 1996-2011. AERONET AOD medians are for observations with Level-2 almucantar retrievals, with corresponding AAOD and SSA retrievals at Level-1.5 (black) or Level-2 (gray). In-situ data are for June 2006-September 2009. AERONET Level-2.0 almucantar AOD and AAOD values are biased high by definition, because of the AOD_{440}>0.4 constraint. AERONET 2.0 direct sun retrievals (not shown) are similar to the AERONET 1.5 AOD values. In-situ values are derived from 365 flights over BND. AeroCom Phase II median model results cover various time periods (depending on the model) and are reported at 550 nm.

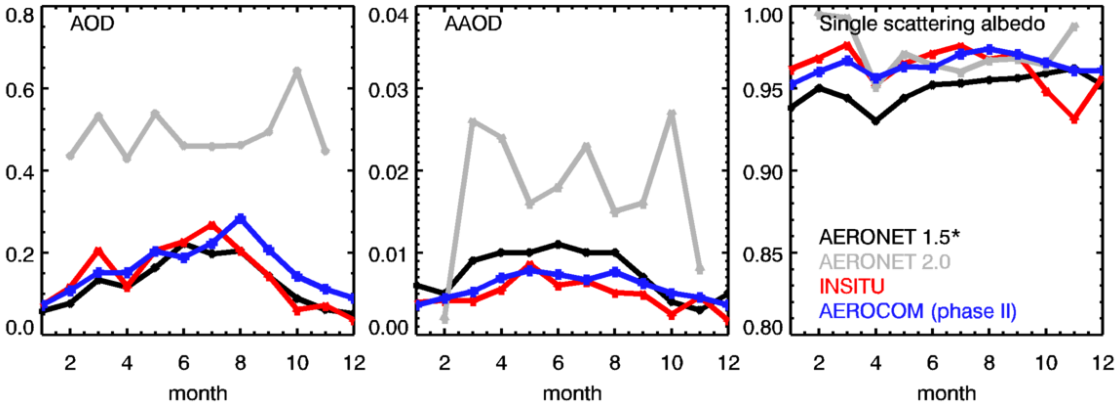

Figure ~~7b~~8b. Monthly medians of SGP aerosol optical properties at 440 nm. AERONET medians are for 1996-2011. In-situ data are for September 2005-December 2007. AERONET AOD medians are for observations with Level-2 almucantar retrievals, with corresponding AAOD and SSA retrievals at Level-1.5 (black) or Level-2 (gray). AERONET Level-2.0 almucantar AOD and AAOD values are biased high by definition, because of the AOD_{440}>0.4 constraint. AERONET 2.0 direct sun retrievals (not shown) are similar to the AERONET 1.5 AOD values. In-situ values are derived from 322 flights over SGP. AeroCom Phase II median model results cover various time periods (depending on the model) and are reported at 550 nm.

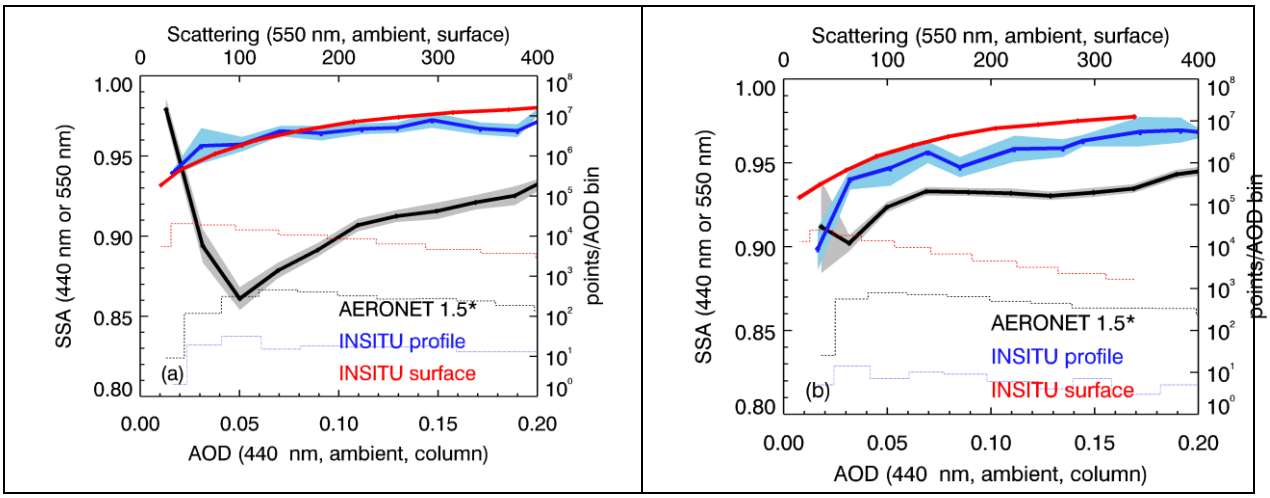

Figure ~~8~~9. Systematic variability of SSA as a function of loading for (a) BND and (b) SGP for AERONET 1.5* AOD and SSA (black lines), AOD and SSA from in-situ profiles (blue lines) and in-situ scattering and SSA from surface measurements (red lines). Solid lines indicate mean values of SSA and AOD for each 0.05 AOD bin (10 Mm^{-1} scattering bin). Shaded areas represent mean standard error (mean standard error for surface data is within thickness of red line). Histograms indicate the number of points in each AOD (or scattering) bin. Plot based on BND and SGP AERONET data (date range: 1996-2012) and BND INSITU profile data (date range: 2006-2012); SGP INSITU profile data (date range: 2006-2007). Surface data (orange lines) are for 550 nm, low RH, hourly in-situ data from the surface sites at BND (date range: 1997-2013) and SGP (date range: 1998-2013). AERONET 1.5* is from Level-1.5 retrievals with a corresponding Level-2 almucantar retrieval.