

We thank Dr. Tom Eck for his many helpful and insightful comments and suggestions. We have responded to his comments, point-by-point, below, and have revised the manuscript accordingly.

General Comments:

The sections of this paper that focus on the trends in AOD are reasonable since these measured data are highly accurate, although it needs to be clearly stated that only Level 2 AOD were utilized in the paper. Currently, there is insufficient discussion in the manuscript concerning the uncertainty of all of the various measurements and retrieved parameters provided by AERONET. The use of Level 1.5 data for absorption parameters is extensive (94% of sites analyzed; page 14356 lines 26-28) due to the analysis of data where AOD at 440 nm is less than 0.4. At the minimum, if L1.5 retrievals data are used to analyze lower AOD observations then the data must have solar zenith angle > 50 degrees (this ensures sufficient scattering angle range of input data; larger airmass increases sensitivity to absorption) and also sky error (residual of computed versus observed sky radiances) less than 5% to ensure a robust retrieval. These are the main data quality controls of L2 retrievals in addition to the $AOD(440\text{ nm}) > 0.4$. Only L1.5 data should be analyzed that have L2 AOD data and also subsequently a L2 retrieval (but with $AOD(440\text{ nm}) < 0.4$) to ensure high quality of input AOD and sky radiance data. Additionally, the authors should still impose some lower limit on the $AOD(440\text{ nm})$ for analysis of absorption parameters since the uncertainty of SSA increases exponentially as the product of optical airmass and AOD decreases (Sinyuk, personal communication). A reduction of the lower limit of $AOD(440\text{ nm})$ to 0.20 or 0.15 would result in much less data to analyze than in the current paper, but eliminate observations where there is little real sensitivity to actually measure an absorption signal. For example the annual average AOD at Birdsville, Australia is only 0.06, therefore sky radiance calibration uncertainty and assumed input surface reflectance uncertainty (as a function of SZA) would dominate any real ability to actually measure the aerosol absorption at that particular site and other sites with low AOD.

Thank you for this very helpful information and suggestions. We have completely updated the data selection criteria for the Level 1.5 inversion products according to your suggestions. Now solar zenith angle > 50 and sky error <5% requirements are applied. Also only data with a coincident Level 2.0 AOD retrieval are selected. For the AOD threshold, we tested a few thresholds and found that $AOD > 0.2$ will result in only 12 stations for analysis and $AOD > 0.1$ results in 24 stations. The bulk of these stations are located in North Africa or Asia (with only a few exceptions in Europe). And the pattern of the trends does not differ significantly from that of Level 2.0 inversion products. Moreover, many regions such as North America, South America, Europe and Australia, are not covered. Therefore, we have kept the Level 1.5 data without AOD thresholds (the other quality control criteria are still applied) for a reference for global results. We also showed the results for the 12 stations for $AOD > 0.2$ as a comparison. In the revised paper, we have separated the discussion and presentation of Level 2.0 and Level 1.5 results for the absorption parameters. However, in the Level 1.5 results, we noticed some spatial coherency, e.g., uniform reduction in absorption and increase in SSA for Europe and North America, and some agreement with the trends inferred from in-situ

measurements (Collaud Coen et al., 2013), which lend more credibility to these Level 1.5 results. However we did add the statement that the Level 1.5 data are subject to larger uncertainty and the most reliable results are those based on the Level 2.0 retrievals.

Section 3.4 of the manuscript compares L1.5 with L2.0 retrievals, but only for sites that have extremely high AOD, and these are the sites with the highest annual and monthly average AODs in the entire AERONET network (typically having monthly average AOD(440 nm) > 0.4). As a result of the very high AOD it is expected that L1.5 would still have relatively small uncertainties since the very large aerosol signal at these selected sites overwhelms any biases in calibration or incomplete scan angle range of the sky radiance data. For example, at the Beijing site the average AOD at 440 nm for Level 1.5 retrievals for the years 2001 through 2012 is 0.62 (from 11487 retrievals) while the average AOD(440 nm) for L2 retrievals with AOD>0.4 at 440 nm at the same site is 1.04 (3209 retrievals). Therefore the AOD of the L1.5 retrieval data at Beijing is higher than the average AOD at nearly all AERONET sites. Please also add a discussion in the manuscript text that L2 retrievals are a subset of L1.5 retrievals, with only those retrievals that pass quality control checks (primarily for sky radiance error) and also AOD >0.4 for the absorption parameters of SSA and refractive indices, reaching Level 2. The main problem in the current paper is with using L1.5 retrieval data for absorption parameters when AOD is low, and this results in very large uncertainties. Therefore the analysis presented in this section does not address this major issue at all due to the high AOD levels of all sites mentioned in this section, and therefore it remains unaddressed in the entire manuscript. As a result the last sentence of this section is a completely false assumption, since the large uncertainty in absorption parameters at Level 1.5 for sites with low average AOD can have a significant influence on trend analysis at those sites.

We agree that the comparison between Level 1.5 and Level 2.0 trends for Beijing does not address the problem for low AOD sites. However, for the low AOD sites, it is not possible to make the comparison, as AOD>0.4 will eliminate the bulk of the data and there will be too few data for a meaningful trend analysis. We therefore remove this comparison between Level 1.5 and Level 2.0 data. In the revised manuscript, we separately present and discuss Level 1.5 and Level 2.0 results, emphasizing that Level 2.0 results are the most reliable while Level 1.5 data have larger uncertainty. Meanwhile, the consistency of the trends at different data Levels for the large AOD stations (those with sufficient Level 2.0 data) can still be observed by comparing the trend maps for Level 2.0 and Level 1.5 (Figure 8 and Figure 11 in the revised manuscript).

Additionally some key sites seem to be missing from the analysis presented here, such as the Solar Village site in Saudi Arabia for which other studies (both Hsu et al. (2012) and Yoon et al. (2012)) have found a large trend in AOD (in fact the largest AOD trend in the entire network)). The Solar Village site has data from 11 to 14 different years of data for each of the 12 months. Additionally there are some sites in Brazil, notably Alta Floresta (seasonal biomass burning site) that are also not analyzed in the current paper even though they have large and long-term data records. The longest and most complete data record of any AERONET site is the GSFC site yet it is also missing. Perhaps the data

section criteria should be revised somewhat to accommodate these important data sets that have been omitted from your study.

Thank you for noting this. In looking into it we found a bug in our selection routine that accidentally excluded the long-term stations whose first measurement was made before 2000. This mistake has been corrected and the results have been updated. Another factor contributing to missing stations is that in the original submission, the trends are selected based on Level 1.5 inversion products only (but Level 2.0 AOD and AE are used for those stations). Therefore, some stations with good Level 2.0 direct sun measurements were not selected due to insufficiency in their Level 1.5 inversion products. In the revised version, we made separate selections for different data products and levels, which is more reasonable as Level 2.0 AOD and AE products are highly accurate. With these corrections, our study now includes: (1) 90 Level 2.0 AOD and AE sites; (2) 7 Level 2.0 SSA, ABS and AAE sites; and (3) 44 additional Level 1.5 SSA, ABS and AAE sites.

Specific Comments:

Page 14355-14356, Section 2. AERONET data: The authors should be clear in this section that the AOD that were analyzed are all Level 2 direct sun measured data and that the accuracy of the AOD data for the channels studied in this manuscript is very high at 0.01 (Eck et al., 1999). This is also very important for the retrievals since the a priori assumption of the Dubovik and King algorithm almucantar retrievals is an accuracy of 0.01 for the input AOD data at 440, 675, 870 and 1020 nm. The resultant retrieved size distribution and refractive indices are consistent with the measured AOD to within 0.01 at all four wavelengths, due to the assumed high accuracy of AOD. Additionally in section 2 it should be explained whether the Angstrom Exponent was computed by using all 4 wavelengths in the 440 to 870 nm wavelength interval by linear regression or whether just the AOD at two wavelengths 440 nm and 870 nm were utilized.

We apologize for the confusion. The accuracy of AOD has been added to the text. The AE and AAE parameters used are directly from the AERONET standard product, which are derived using all four wavelengths within the 440 to 870 nm interval. We have added the detailed description to the text as follows: “The AE and AAE parameters are from the standard AERONET product, which are derived using AOD and ABS measurements at all four wavelengths in the [440, 870] nm interval, respectively, to provide information in aerosol size and composition.”

On page 14356 lines 6-10, the claim that there would likely be no biases in the L1.5 retrievals (even at low AOD) is misleading since it is well known that the AERONET sky radiance calibration is accurate to 5% (Holben et al, 1998). Calibration uncertainty is not a random error in a given year, and therefore would bias retrievals of absorption parameters (imaginary index and subsequently SSA). Trend analysis with data having different biases in differing years is therefore problematic in detecting true trends. Additionally, surface reflectance used by AERONET is based on MODIS satellite climatology and generic global ecosystem BRDF models. These estimates would also

introduce small biases (not random variations) in the retrievals, and become an increasingly important contribution to retrieval bias as AOD decreases. Additionally, the Kaufman (2002) paper is a poor reference to use to claim lack of bias in absorption, especially since they did not analyze SSA directly in that paper, and their results are primarily constrained by the highly accurate AOD measurements made by AERONET.

Thank you for this information. We mean that there is no bias associated with the assumptions of the aerosol model used in the retrieval, which is the focus of Dubovik et al. (2000) study. We have added “due to assumptions in the retrieval model” to the end of this sentence. The Kaufman reference has been removed. In the revised manuscript, we weakened the arguments that Level 1.5 data are accurate, and stated that these results are subject to larger uncertainties.

On Page 14356, lines 15-17, the authors say that SSA data is screened when values are less than 0.5 due to data quality limitations, however the lower limit of 0.5 is very low. There are no published papers in the literature to support such low column integrated SSA values (less than 0.7) in homogenous aerosol haze (required for an almucantar retrieval). In fact, columnar averaged SSA is very rarely reported as lower than 0.75 and even these values are quite uncommon. AERONET Level 2 SSA retrievals are rarely less than 0.80 and many of those lower values have data quality issues that will be screened in the upcoming Version 3 database with improved quality controls. The authors need to discuss why Level 1.5 SSA retrievals that are so low (<0.5) exist in the AERONET database, since this is inconsistent with their claims of high accuracy SSA even for low AOD. The percentage of SSA retrievals with values <0.5 and also <0.7 should be given in the text of the manuscript. Additionally, truncating a data set of retrievals at some cutoff (such as 0.5 SSA) is also statistically problematic as it biases the dataset, especially since the maximum SSA is constrained by the AERONET retrieval algorithm to be slightly less than 1.0, therefore the data set can only be truncated in the low extreme and not the high extreme.

The reason for screening SSA is that these extremely low SSA values do not seem realistic but sometimes may bias the trend. Overall the percentage of SSA<0.5 is ~1%. However, the results have almost no change with or without the truncation. Therefore, in the revised manuscript, we choose not to apply any threshold on the Level 1.5 SSA data. The only quality assurances are solar zenith angle and sky error criteria.

Page 14339, line 22: The reference of Ignatov et al. (2000) is an error, as it should be O'Neill et al. (2000). You have truncated the first author from this citation in both the reference list and in the text of the paper (cut and paste type of error).

The reference error has been corrected.

Page 14362, section 4.1: It should be noted in the text here that the AOD trends are the only robust trends in the entire paper for the majority of stations, due to the very high accuracy of the measured AOD in the AERONET database.

OK. We have added “The AERONET AOD measurements are highly accurate, therefore the AOD trends are the most robust among the parameters analyzed.”

Page 14364, lines 8-10: Please note that the uncertainty in SSA and ABS is generally very high in Europe due to AOD magnitude, except for the summer season when AOD is much higher. Some of the sites in Spain show positive trends while other sites show negative trends and this suggests possible non-physical reasons (relatively low AOD signal leading to high retrieval uncertainty as a result of both radiance calibration bias and surface reflectance biases) for these spatially variable trends.

The results for Europe have been moved to the Level 1.5 results (Section 4.3 in the revised manuscript) and the uncertainty associated with these data has been emphasized throughout the manuscript.

Page 14364, section 4.3: This is currently an inadequate and non-rigorous description of the reasons for noisy AE data. It is well known that AE has very large uncertainties at low AOD that increase as AOD decreases. Some discussion should be added about how AE error increases as AOD decreases, using some calculations from Equation 6 of Kato et al. (2000; JGR) to estimate the uncertainty in AE (Kato calls it the Lundholm exponent although it is equivalent to the Angstrom exponent).

We have revised the discussion of AE uncertainty, and used the equation by Kato et al. (2000) to give an estimation of AE uncertainty during winter (low AOD) and summer (high AOD) conditions using measurements at GSFC station.

Page 14365-14366, Section 4.4: In this section it should be noted that the AAE has very large uncertainty (even larger uncertainty than AE) since ABS has smaller values and larger uncertainties than AOD. See Giles et al. (2012) for a discussion of the uncertainty of AAE. Please include some discussion and analysis of uncertainty in AAE in your manuscript. On page 14366 Lines 25-26, please mention that AAE uncertainty is very large at low AOD levels and the majority of the data you have analyzed are at low AOD.

A discussion of AAE uncertainties has been added to the text as the following: Note that the AAE parameter has even larger uncertainty levels than the AE, owing to the smaller ABS values and large uncertainties at low aerosol loading. Giles et al. (2012) performed a series of sensitivity studies on the AAE parameter by perturbing SSA using AERONET measurements, and found that AAE can vary by ± 0.6 for dust sites with ± 0.03 perturbation in SSA which is the uncertainty level associated with this parameter in Level 2.0 data. Therefore, the AAE trends alone are not sufficient to infer aerosol composition changes and need to be evaluated in the context of other information such as AE and ABS.

Page 14367, lines 19-21: You seem to imply here that the satellite studies of Zhang and Reid (2010) and Hsu (2012) validate your trends of absorption (ABS). However these

papers have analyzed AOD trends only and not aerosol absorption. Please explain your reasoning/justification better regarding this issue in the text.

We are sorry for the confusion. Here we meant the statement only to apply to the AOD trends and have revised the text accordingly.

Page 14370, line 13-14: You suggest that “: : :uncertainties in individual measurements largely cancel out in the monthly medians: : :”. This is not true since the AERONET retrieval uncertainties at low to moderate AOD levels are mainly caused by biases in radiance calibrations and/or biases of input surface BRDF and therefore are not random and do not cancel out with time interval statistics on a monthly or even yearly time scale.

Thank you for the information. We removed the claim that uncertainties will cancel out.

Page 14371, Conclusions: It is important to mention in the Conclusions section that there are large uncertainties in all parameters analyzed except AOD and SCT (since SCT is dominated by AOD). The exceptions are sites with very high AOD such as Beijing, Kanpur, XiangHe, IER Cinzana, Hong_Kong_PolyU and Agoufou where very high AOD levels allow for accurate retrievals of all parameters analyzed (including SSA and ABS).

We have added the statement “only the results at stations with consistently high aerosol loading, i.e., those having sufficient Level 2.0 inversion retrievals, are the most reliable” in the conclusion section.