

We thank the referees for their valuable input. Below is a point by point response to the comments (the comments are italicized).

Referee # 1

1) In this paper a unique rather convoluted method to infer the complex component of the biomass burning aerosol refractive index is used. The method makes many assumptions, however, the results seem reasonable when compared to published values (although as noted below, I believe some of the comparisons are incorrect).

We do not agree that the method is convoluted. We perform optical closure, which is similar to what previous studies have done (Chakrabarty et al., 2010; Lack et al., 2012). It is true that we make several assumptions in this work. We state the assumptions clearly in the manuscript so that the reader is aware of them. As stated in the manuscript, the main assumptions are either conservative from the perspective of OA absorption or represent limiting cases. The assumption that OA absorbs negligibly at the wavelengths of 950 nm (p. 11517, L. 6) minimizes the retrieved k_{OA} values. In other words, if this assumption is invalid, the conclusions of this work will be even stronger (i.e. OA is more absorptive). The other main assumption concerns the mixing state of OA with BC. We consider the limiting cases described in the paragraph on p. 11517 line 19 and Figure 1. The results (Figure 6) show that although the limiting cases result in a wide range of k_{OA} values, the main conclusion (POA and SOA are absorptive) holds for both cases.

2) It would be useful if the authors discuss if the method could be used for ambient aerosol. Example, over what concentrations of smoke would the method work.

The method works for ambient level concentrations. In fact, the concentrations in our experiments are close to ambient ($\sim 10 \mu\text{g}/\text{m}^3$). Of course, for low concentrations, the method is limited by the signal-to-noise ratios of the instruments used (SMPS, SP2, and aethalometer), and averaging over longer periods of time might become necessary.

3) These measurements were made in what would represent fairly fresh (max 1 hr old) plumes in the ambient atmosphere. One major issue with this paper relates to how useful the data presented really is given the complexity of biomass burning emissions and the rather limited data presented; this is discussed more below.

Most previous laboratory studies investigating optical properties of biomass burning emissions have examined only fresh emissions (POA), while some field studies have investigated plumes from biomass-burning in the atmosphere and reported optical properties of the aged OA (POA + SOA), without distinguishing between the two. Here, we investigate the emissions both when fresh, and after aging, which enables us to constrain optical properties of both POA and SOA, independently. To our knowledge no one has done this before for biomass burning emissions. We show that SOA is absorptive. This is useful. Of course, we acknowledge the complexity of biomass burning emissions, and by no means claim that our data represent the whole range of possibilities. However, our work is an important step in the right direction.

In the revised manuscript, we add in section 2.1 that “the amount of aging in the experiments is relatively modest, however we were able to generate appreciable amounts of SOA.”

4) Secondly, it is not clear how the problem is constrained when there is no actual information on the size distribution of the brown carbon. The implicit assumption is that the brown carbon distribution is the same as the OA distribution, which there is no evidence for. It is highly possible that the mass concentrations of light absorbing components are very small relative to the POA or SOA. Thus, size distributions and chemical properties of the bulk OA, as measured by an AMS in this case, could be of little value. This limitation is not considered in the uncertainty discussion.

This is an interesting point. We do believe that the BrC size distribution is similar to that of OA. POA forms by rapid quenching of organic vapors in the cooling plume and subsequent condensation on available seed particles. There is no reason for BrC (or other OA components) to exhibit preferential condensation on certain particles because when the hot vapors are quenched; they are highly supersaturated and will condense on any surface available. Thus, we expect the POA composition to be fairly uniform among all particle sizes.

When SOA forms by oxidation of organic vapors, it condenses on all particles to maintain similar mass fraction (activity) in all particle sizes. Using AMS pTOF data, it has been shown that SOA composition is uniform among all particles sizes for both biomass-burning (Grieshop et al., 2009) and diesel (Donahue et al., 2013) emissions.

We include this discussion to the end of section 2.2.1 by adding the following:

“Another implicit assumption is that the BrC size distribution in the emissions is similar to that of OA. This is justified because POA forms by rapid quenching of organic vapors in the cooling plume and subsequent condensation on available seed particles. There is no reason for BrC (or other OA components) to exhibit preferential condensation on certain particles because when

the hot vapors are quenched; they are highly supersaturated and will condense on any surface available. Thus, we expect the POA composition to be fairly uniform among all particle sizes. SOA is also expected to be uniformly distributed among all particles. When SOA forms by oxidation of organic vapors, it condenses on all particles to maintain similar mass fraction (activity) in all particle sizes. Using AMS pTOF data, it has been shown that SOA composition is uniform among all particles sizes for both biomass-burning (Grieshop et al., 2009) and diesel (Donahue et al., 2013) emissions.”

5) The background and references are not complete. There a number of missing references to papers that have specifically investigated the prevalence of brown carbon from biomass burning, however, more importantly, recent papers have investigated the chemical composition of brown carbon, one specifically focusing on smoke, which likely came out following submission of this work (Desyaterik et al, JGR, 2013). This is relevant to the note above and also to the authors claim that linking light absorption and composition is a future goal (pg 11524 lines 9 -12); others researchers have done it and this should be noted.

We would appreciate it if the referee can provide the missing references that he/she refers to.

Interestingly, since we submitted this manuscript to ACPD, 3 papers (that we are aware of) on chemical composition of BrC came out, including the work of Desyaterik et al. We address the findings of these papers in the revised manuscript by adding the following:

“For both oak and pocosin pine, SOA absorbs light less efficiently than POA in the long visible wavelengths, however it exhibits a stronger wavelength dependence and is more absorptive than POA in the short visible and near-UV. This might be due to differences in chemical composition, namely higher aromaticity in POA, but the true cause is not known. This hypothesis is supported by the recent findings of Lambe et al. (2013) who reported that SOA from aromatic precursors can retain light-absorbing conjugated double bonds, while light-absorption in SOA from non-aromatic precursors is due to carboxylic and carbonyl functional groups. Thus, BrC in POA and SOA in biomass-burning emissions may have overlapping as well as different chromophores. Desyaterik et al. (2013) investigated composition of BrC in cloud water droplets that were heavily affected by biomass-burning. They found that most of BrC absorption is due to aromatic carbonyls, which exist in fresh biomass-burning emissions, and nitrated phenols, which form through gas phase and aqueous chemistry. Nitrated phenols were also observed by Mohr et al. (2013) in biomass-burning plumes over Delting, United Kingdom.”

6) Pg 11511 lines 5-10. *This seems to imply the mass weighting is due to brown OA and black BC, but the OA is composed of many compounds, all of varying optical characteristics, thus the overall color of the OA depends on the mass weighted optical properties of the various chromophoric OA components and their size distributions. It is more complex than OA to BC ratios.*

This is true. However, the goal of this study, and many other studies in the literature, is to obtain effective absorptivity of OA in biomass-burning emissions, where all organic species are lumped together. Given the limitations of the organic speciation data (only a small fraction of the OA can be speciated), this seems like the only path forward at the moment. It is not perfect, but it is a major improvement over lumping OA and BC together and obtaining a single MAC to represent absorptivity of biomass burning emissions. Since BC constitutes a small fraction of the emissions (usually < 10%) but is 1-2 orders of magnitude more absorptive than OA, a slight change in OA-to-BC ratio can alter MAC of biomass burning emissions significantly. So, MAC values obtained from source measurements where OA-to-BC ratios are high might be quite different from MAC values for the same aerosol when diluted in the atmosphere.

7) Pg 11511 line 16-17. *Data on atm OA k as a function of size are even more rare, and are what is needed.*

This statement is not valid. The referee assumes that atmospheric OA composition is variable across particle sizes but uniform for each particle size, which is not necessarily true. As we point out in response #4, OA from a single source exhibits fairly uniform composition across different particle sizes. Atmospheric OA is more complex. If, for example, a biomass-burning plume mixes with background aerosol dominated by biogenic SOA, then this atmospheric OA will be heterogeneous both across sizes and within the same size (since the size distributions of biomass-burning emissions and biogenic SOA will surely overlap). The semi-volatiles in the two particle populations may or may not mix through the gas phase over time, depending on whether mixing is thermodynamically favorable or not. If they mix, this will happen for all sizes, creating a homogeneous aerosol.

A realistic goal is to obtain, with good constraints, k_{OA} values for different atmospheric OA types that are tracked in regional and global models, which enables us to estimate their climate forcing. This paper is an effort in this direction.

8) Pg 11511, lines 19-23. *There are assumptions made that influence the magnitude of k calculated from solvent extracts. Generally, there is no particle size information so the k represents an average for some unknown size distribution, which will depend on the emission and subsequent processing. Furthermore,*

the k calculation requires a measure of the mass concentration of the absorbers in solution. Clearly not all extracted components absorb light so use of some bulk mass concentration, such as organic carbon mass, is only representative of that extract (eg, a different extract may have different proportions of organic carbon that are absorbers or non-absorbers). Some studies have used carbon mass, but different measures of mass could be used. This use of a rather arbitrary mass concentration for absorbers is not a significant issue if in the subsequent Mie calculation the same mass concentration is used. However, the reported k values do depend on mass concentration and so comparing k from these bulk measurement is not straight forward and the direct comparisons shown in Fig 6 may be incorrect.

This is a valid point. We know that k obtained from optical closure and from solvent extract are not “the same thing”. However, Chen and Bond (2010) and Kirchstetter et al. (2004) – the studies cited in this paper which used solvent extract method – report their best estimates of absorptivity of OA in biomass-burning emissions based on their method. And we report our best estimates based on our method. Each method, as any other experimental method, has its limitations, but it does not mean that results from both methods cannot be compared. In fact, the comparison can be informative.

9) Page 11512, Lines 5-14. The argument is that biomass burning BC gets coated with OA whereas in urban plumes this does not happen? Urban measurements of BC size distributions show rapid shifts to the accumulation mode do not support this.

As described in the paragraph referred to by the referee, when OA forms in biomass-burning plumes, it HAS to coat the existing BC particles. This is most likely the case for any system with emissions containing BC and significant amount of OA, and should apply to urban plumes as well. However, the study of Cappa et al. (2012) indicates, rather surprisingly, otherwise. Maybe in that study the BC-containing emissions were lean in OA. We do not analyze the findings of Cappa et al. (2012) in this paper, but we just point out that the Cappa et al. (2012) conclusions regarding mixing state most likely do not apply to our experiments.

10) Page 11516 line 10-12, This statement seems to strong, all that can be said is that the aethalometer and 4 wavelength PAS give similar AAE, not that they are the true values for the ambient aerosol.

A discussion on the sensitivity of the predicted absorption coefficient on AAE would be informative.

The argument here is that collecting the particles on the filter does not alter AAE relative to the AAE measured for the suspended particles by the PAS. We changed the sentence in the revised manuscript to better reflect this. It now reads: “Thus, the agreement between the two

instruments indicates that $b_{\text{abs,norm}}$ (or AAE) derived from aethalometer measurements is not altered due to collection on the filter.”

As discussed in section 4.1, the sensitivity of k_{OA} to measured AAE is very small relative to the major uncertainty induced by mixing state.

11) Figure 3: The mass distributions would also be of interest since it would provide some insight on coating mass. Also note Fig. 3c typo in legend.

We added figures of mass distributions to the SI.

12) Page 11523. These arguments attempting to explain the wide differences in the various results shown in Fig 6 seem to demonstrate the great limitations when attempting to apply these results to make predictions for ambient smoke plumes. Factors of 10 or so between k_{OA} at low wavelengths (300nm), possibly due to different types of burning, as argued here, seem to make the idea of reliable model predictions highly unlikely. The apparent high degree of variability in the optical properties of biomass burning aerosols means that many ambient measurements will be required to link burning type, conditions etc, to refractive index. This means a fairly simple robust method will be required. The authors might want to discuss if their technique could provide this type of data.

Biomass burning emissions are very heterogeneous. This applies to emission rates, composition, and, as our and other results demonstrate, optical properties. This is not surprising given the complexity and poorly controlled nature of biomass combustion. Therefore, the referee’s concern applies to biomass burning in general, not just the topic of this manuscript.

As the referee points out, the absorptivity of OA in biomass-burning emissions is a complex problem. More careful investigation of this problem is needed in order to be able to represent OA absorption reliably in models. This is ongoing work in our research group, and others for sure. We do not agree that it is necessary or even sufficient to perform many ambient measurements as the referee argues (it might be). Insight can be gained from well-designed experiments and theoretical analysis as well. Thus, we do not agree that we need to boil down the discussion to the need for endless ambient measurements, and what technique best fits this criterion.

13) Page 11524, lines 12-14. It should also be pointed out that there are studies that show very little SOA formation in biomass burning plumes.

This does not alter the conclusions of this work. We are not extrapolating the findings of the cited studies to obtain global estimates of SOA absorption. We just use them as evidence that SOA from biomass-burning is “potentially” important contributor to radiative forcing. In the revised manuscript, we change the sentence to start with “Some field studies..” to avoid giving the impression that all field studies have observed significant SOA formation.

Referee # 2

The main point the referee makes concerns the uncertainty in the Absorption Ångström Exponent (AAE) of the aethalometer measurements, namely that AAE decreases with loading. He/she writes:

1) Care was taken in mentioning that Aethelometer measurements are biased during periods when the filter is heavily loaded, however, the authors did not attempt to remove the data just prior to the filter change which would potentially diminish this issue. The authors use the aethelometer data in a relativistic sense due to the uncertainties in the measurement; however, it seems that could be avoided by removing the data just prior to the filter change. It was also not clear how scaling the absorption data removed the bias in the aethelometer data, which appears to affect the absorption measurement to a greater extent for shorter wavelengths, effectively changing AAE in addition to the magnitude of absorption. In SI F2 the authors show that the data before the filter change biases the AAE, therefore, scaling the absorption data would not take into account the change in slope of the absorption signal versus wavelength. For this reason, I think some of the analysis and interpretation in this manuscript needs to be redone in order to minimize the decreased AAE that results from using the aethelometer data when the filters are heavily loaded. One way to do this would be to eliminate the data just prior to the filter change, while another would be to at least quantify the uncertainties, which would affect AAE and refractive index calculations. The real magnitude of the absorption measurement is discarded, and the focus is redirected to AAE's, yet this limits the level of interpretation that can be done with the data. I would suggest trying to remove or understand the uncertainties with the data taken while the filters were heavily loaded and using the real magnitude of the absorption data to help bound the different assumptions being used in the manuscript. This could be added to the discussion section, along with some other situations/issues that should be addressed that are discussed below.

As pointed out by the referee and described in section 2.2.2 and 2.2.3 in the manuscript, we use the aethalometer to constrain only the wavelength-dependence of absorption (i.e. AAE) and not the magnitude. The magnitude is scaled by BC concentration measured using the SP2. The reason we do so is due to potential uncertainties in the magnitude of aethalometer measurements due to organic loading on the filter (Cappa et al., 2008; Lack et al., 2008; Subramanian et al., 2007). These studies pointed out a potential bias in magnitude of filter-based measurements, but did not discuss its wavelength-dependence.

Here, we show that AAE is NOT biased by organic loading. Figure S2 shows that the maximum possible decrease in AAE due to loading is 10% for these experiments, which corresponds to the measurement immediately before filter change. The idea behind Figure S2 is not to show that AAE in our analysis is biased by 10%, but to say that the bias is small, and the upper bound is 10%. In fact, the data used in our analysis were measured well before filter change, when the organic loading effect is minimal.

We made this point clearer in the revised manuscript by adding the following sentence to section 2.2.2: "The data used in the analysis were taken from measurements well before the end of the aethalometer filter cycle, thus the bias in AAE is considerably less than 10%."

On the other hand, we cannot quantify the bias in magnitude of the aethalometer measurements, and so we scale it with BC concentration measured with the SP2. As discussed in section 2.2.3, this analysis is based on the assumption that OA absorbs negligibly at the wavelength of 950 nm, which is the most widely accepted assumption in the literature. In any case, this results in a conservative estimate (lower bound) of OA absorptivity.

2) Data from 3 aging BB experiments are discussed; however, aged gallberry data is not shown since the concentrations were determined to be too low for reliable absorption data. However, in Fig. S1(a) the AMS data is shown. It appears that the experiment did not produce appreciable SOA, however, this was not discussed in the draft. Either in the experimental or discussion session, whichever is relevant, it should be mentioned whether no conclusions can be drawn from this or whether it appears that gallberry does not produce SOA under similar conditions that do produce SOA for the pine and oak.

Gallberry does produce SOA, but, as the referee points out, less efficiently than pocosin pine and oak. It is true that we might not be able constrain optical properties of gallberry SOA because of the relatively small amount of SOA produced making it difficult to separate from the POA. However, the main reason we could not analyze the gallberry aged data was that aethalometer measurements were noisy.

3) The authors attempt to bound the k_{OA} values with two different scenarios based on the mixing state of the aerosol. Case 1 assumes an external mixture of BC and OA, while case 2 assumes non-absorbing coatings on the BC. The magnitude of the absorption measurements are scaled based on these two scenarios, Case 1 being the lower bound and Case 2 being the upper bound for the imaginary part of the refractive index of the OA. However, it is mentioned in the discussion section that “if the magnitude of the absorption coefficient measurements were assumed to be true rather than scaled by BC concentration, limiting case 1 would yield larger k_{OA} than limiting case 2. Rather than scale the aethalometer absorption measurements due to uncertainties when the filters have high loading, why not remove the data before the filter changes and use the SP2 data to determine whether coatings are present on the BC in order to constrain the data.

As discussed in response #1, we cannot quantify the bias in the magnitude of the aethalometer measurements, which is why we scale them by BC concentration measured using the SP2.

Previous studies have used SP2 data to investigate mixing state of BC, however this type of analysis is qualitative, and is subject to debate. In this work, we opted to perform our analysis using the two bounding cases for mixing states.

The referee seems to suggest that we assume that the coating is non-absorbing at all wavelengths for case 2. This is not correct. We assume that it is negligibly absorbing only in the near-IR, which is the most accepted assumption in the literature. We make this clearer in the manuscript by adding: “The assumption that OA absorbs negligibly at the long visible wavelengths is justified based on numerous findings reported in the literature (e.g. Kirchstetter et al., 2004; Chen and Bond, 2010; Chakrabarty et al., 2010; Lack et al., 2012).”

4) Despite using the 2 cases to bound the real scenario, the authors do not discuss which case they believe to be most likely based on comparison with other data sets, etc. A section on this would be a nice addition to the discussion section.

As we point out in page 11512 line 10: “BC in biomass-burning emissions is most likely coated by OA. During combustion, BC particles form at high temperatures, and act as condensation sites for OA.” So, we believe that reality resembles case 2 (coating). We added the following to section 4.1:

“For these experiments, where substantial amount of OA are generated (as in biomass-burning emissions in general), limiting case 2 (core-shell) is more likely to resemble the mixing state of BC. OA condenses onto the BC particles when the emissions are cooled, creating a core-shell morphology.”

5) It would be interesting to see a case 3 where the BC coatings of POA are assumed to be absorbing. What happens with this case/why was it not mentioned/addressed, what would it's assumptions require of the data presented here?

In fact, the POA coating is assumed to be absorbing in case 2. The POA is assumed to be negligibly absorbing only at the wavelength of 950 nm in order to scale the aethalometer measurements using the SP2 data, as discussed in section 2.2.3. This assumption is justified, since most previous studies have reported negligible OA absorption at long visible wavelengths.

6) The authors deduce that BB SOA is absorbing, however, they do not consider that the increased absorption over the primary emissions could also be due to aging of the original POA/BC mixture. This issue should be addressed in the text and discussion.

We cannot completely discount the possibility that chemical transformation of POA contribute to the BrC. The POA will be transformed through heterogeneous reactions with OH. However, the extent of this processing is relatively modest for these experiments given the relatively modest OH exposure. The much larger changes to the OA is the formation of substantial SOA. However, the mass of SOA used by this study is a lower bound, which we address in page 11520 line 18: “The SOA-to-OA ratios reported here are a lower bound, because in our analysis we assume that POA is conserved upon aging. However, as shown by Hennigan et al. (2011), a fraction of the POA likely evaporates as semi-volatile vapors are oxidized.”

7) Why is there no discussion section of the relative amount of SOA formed versus the initial POA/BC present in the samples?

The relative amount of SOA formed is shown in Figure 4, and the initial POA concentrations are shown in Figure S1. We don't think that additional discussion on this issue is needed in the paper.

8) P11512 L2: *A little more detail is needed about the reference to this work. What is meant by "no BC"? The sentence seems to imply that there was no BC from the smoldering data.*

It simply means that in the experiments of Chakrabarty et al. (2010) the emissions from smoldering of duff contained no BC. This is what is reported by the authors. We change the sentence to read: "Chakrabarty et al. (2010) applied this method to determine refractive indices of PM in duff-burning emissions. Due to the smoldering burning conditions, the emissions were dominated by OA and contained negligible amounts of BC."

9) L9: *Cappa et al. shows that the enhanced absorption of ambient BC is less than what was predicted from MIE calculations and laboratory data, however, to say that it shows core-shell morphology as "unlikely for aged urban aerosol" is a bit speculative.*

The major conclusion of Cappa et al. (2012) is that core-shell morphology does not represent the mixing state of BC in urban aerosol. We do not necessarily indorse this finding. We replace "showed" with "proposed".

10) P11516: L25: *Rather than ignoring the issues from OA loading, is it not possible to remove the data just prior to the filter change? If this is too difficult to do or, since the bias is only expected to be 10% of the AAE, perhaps this could be used and reported in terms of the uncertainty in the extracted kOA values. It seems like this would have been simple to just remove the data though, and eliminate the issue.*

As we point out in response #1, the 10% bias in AAE is not the major issue with aethalometer measurements. The major issue is potential bias in the magnitude of absorption, which we cannot quantify.

Again, we repeat (see response to comment #1) that the data used in our analysis is from measurements well before filter change, thus the uncertainty in AAE is much less than 10%.

11) P11517: L27: *I suggest changing the reference to "details can be found in Sect. 3.2" since referring ahead is generally not recommended.*

Done.

12) P11519: *How was the SP2 calibrated and with what? What density was assumed for the experimental data? Please provide details of how this was done so that future comparisons can be referenced to the results discussed here. If aquadag calibrations were done, they should be converted to fullerene soot values as is referenced in Baumgardner et al., 2012 as this is coming to be the new standard.*

The SP2 was calibrated using aquadag. We added a paragraph in section 2.1 in the revised manuscript on the calibration of the SP2. We appreciate the referee pointing out that fullerene soot is the new recommended standard. We will certainly use it for future work; however, for this paper we will stick to the original aquadag calibration. We will state this clearly in the revised manuscript, as the referee recommended, for the reader to be cautioned. We assume a density of 1.8 g/cm^3 for BC in the analysis (Bond et al., 2005). This will also be mentioned in the revised manuscript.

13) P11520: L16: *There is no discussion about how the oak has higher SOA/OA values than the pine. Was this expected? Is this relevant or perhaps is not discussed since is thought to be due to differences between the experimental setups?*

Although a very interesting subject, investigating variation in SOA formation across different fuels is not the focus of this study. This paper is just focused on the optical properties. We do not know what drives the variability in the SOA formation. In fact, this is an ongoing research work in our group.

14) P11521: L15: *Does “investigated fuels” mean the two samples? The number should be stated in the text.*

We change the sentence to read “... across the 3 fuel samples investigated in this study.”

15) P11522: L4-6: *How does the fact that limiting case 1, which is used as the lower limit for k_{OA} , could be larger than case 2 if the absorption measurements were not scaled with BC mass not undermine using case 1 and 2 to bound the derived k_{OA} values? Please describe this in detail since it seems to unsubstantiated the whole analytical method used in this manuscript. What range of k_{OA} values would this increase the uncertainty to/or why is this not a good assumption?*

The problem has 2 degrees of freedom: 1) scaling aethalometer measurements, and 2) mixing state. As described in section 2.2, by assuming that OA absorbs negligibly at 950 nm and scaling aethalometer measurements using the SP2 data, we are setting a LOWER bound on the derived k_{OA} values in this degree of freedom. Case 1 and case 2 represent the limiting cases within the mixing state realm. And as shown graphically in Figure 1, because we pinned down the aethalometer measurements using negligible OA absorptivity at 950 nm assumption, case 1 yields a lower bound and case 2 an upper bound (in the mixing state degree of freedom).

This does not undermine the method at all. Simply put, case 1 and case2 represent lower and upper bounds for k_{OA} values, respectively, if OA is negligibly absorbing at 950 nm. We believe that case 2 (core-shell morphology) is the best estimate.

If we had more reliable measurements of the magnitude of absorption, we would not need to pin the aetholometer data using the assumption that OA absorbs negligibly at 950 nm, so the magnitude of absorption would not depend on mixing state assumption, but would be fixed. In this case, it follows that:

- Case 1 (external mixing): total absorption (measured) = BC absorption + OA absorption.
- Case 2 (core-shell): total absorption (measured) = BC absorption + coating enhancement of BC absorption + OA absorption

It is obvious that case 2 will yield the lower bound of OA absorption (k_{OA}).

In any case, we opted to omit this sentence from the manuscript, since it causes confusion and does not affect the conclusions of this work.

16) L22: Why not remove the points with heavily loaded filters and then use the magnitude of the absorption coefficients? Are there other uncertainties associated with this that would make this not possible?

As we point out in response #1, the 10% bias in AAE is not the major issue with aethalometer measurements. The major issue is potential bias in the magnitude of absorption, which we cannot quantify.

Again, we repeat (see response to comment #1) that the data used in our analysis is from measurements well before filter change, thus the uncertainty in AAE is much less than 10%.

17) P11524: L1: How do you know that the increased absorption over the original POA emitted and BC is due to SOA? Could the increased absorption be from aging of the POA?

We cannot completely discount the possibility that chemical transformation of POA contribute to the BrC. The POA will be transformed through heterogeneous reactions with OH. However, the extent of this processing is relatively modest for these experiments given the relatively modest OH exposure. The much larger changes to the OA is the formation of substantial SOA. However, the mass of SOA used by this study is a lower bound, which we address in page 11520 line 18: "The SOA-to-OA ratios reported here are a lower bound, because in our analysis we assume that POA is conserved upon aging. However, as shown by Hennigan et al. (2011), a fraction of the POA likely evaporates as semi-volatile vapors are oxidized."

18) L6-9: The statement does not appear to be supported by the data shown in Figure 2. What is meant by "SOA absorbs light less efficiently than POA in the long visible wavelengths"? It is also mentioned that the SOA has a higher AAE, but in Fig. 2 this appears to be true for the pine sample, but only marginally true for the oak. Why should the reader focus on this difference, which is significant for picosin pine, with

an aged AAE of 1.73 vs 1.48 for fresh, but is not very large for oak, 1.42 vs 1.38, respectively. Is a difference of ~3% AAE for aged versus fresh oak larger than the uncertainty in determining the AAE? How is this small relative difference AAE constrained when it was mentioned earlier that the AAE's have at least a 10% error associated with them due to using the data when the aethelometer filters were heavily loaded?

Figure 2 shows total absorption coefficients of fresh and aged aerosol. The fresh aerosol contains BC and POA, and the aged aerosol contains BC, POA, and SOA. The distinction between POA and SOA absorption is obscured by the presence of other absorbing components. Therefore one cannot gain insight into SOA absorptivity from Figure 2.

The comparison between POA and SOA absorption (basically between k_{POA} and k_{SOA}) can be read in light of Figure 6, not Figure 2. Figure 6 clearly shows that “SOA absorbs light less efficiently than POA in the long visible wavelengths...”

19) Table 1: The k_{OA} 's and w 's should have uncertainties or error measurements associated with them in order for the reader to understand the level of uncertainty in the methods used for the two cases.

In page 11523 line 5, we mention that the uncertainty in k_{OA} values is approximately 50%, and we show representative error bars in Figure 6. We added this information to the caption of Table 1 in the revised manuscript.

20) Figure 5: Line 5: Change (a) to (d).

Done.

Referee # 3

1. P. 11511 Lines 12, 33 should read “light absorption of homogeneous, spherical particles can be calculated using Mie theory: : :” instead of “light absorption can be calculated using Mie-theory: : :”.

Done.

2. P. 11513 Lines 22-23: “A 7-wavelength Aethalometer (Magee Scientific, model AE- 31) was used to obtain the wavelength-dependence of the absorption coefficients: : :” The Aethalometer per se doesn’t measure absorption coefficients or their wavelength dependence. However, different publications [e.g., Weingartner et al., 2003; Arnott et al., 2005] have suggested methods to derive absorption coefficients from Aethalometer measurements. On p. 11515 Eq. 1, it is questionable if the assumed $1/\lambda$ dependence of MAC holds; see Arnott et al., 2005. This needs to be discussed and these two references need to be cited.

We thank the referee for pointing this out. We actually use the empirical method of Kirchstetter and Novakov (2007) to convert $b_{\text{aethalometer}}$ to b_{abs} . We added a section in the SI to describe this procedure, and sentence in section 2.2.2 to refer to SI.

3. Figures plotting absorption coefficients as function of wavelength would greatly benefit from the use of a log-log scale enabling the reader to judge how well a power law describes the wavelength dependence.

We added Figures in the SI showing the same plots in Figure 5 but on a log-log scale.

References:

Donahue, N.M. (2012). *Topics in Current Chemistry* DOI 10.1007/128_2012_355.

Grieshop, A.P. et al. (2009). *Atmospheric Chemistry and Physics*, 9: 2227-2240.

Kirchstetter TW, Novakov T (2007). *Atmospheric Environment* 41: 1874-1888.