We thank all the reviewers for their time and comments. We have made efforts to improve the manuscript accordingly, please find response for corresponding points below.

Reviewer #1

This paper uses empirical methods to derive organic carbon absorption from optical and in situ measurements. The authors apply their results to hundreds of AERONET sites and 10 field sites. Unfortunately, their methodology is not sound, and their results are not consistent with what is known about BrC.

MAIN POINTS

The authors are merely adding a new twist to the AAE method pioneered by Chung (2012) and Bahadur (2012) (although they did not acknowledge the similarity of their work to these previous works). The authors determine the AAE in two spectral regions via Eq 1, and then define the nonlinearity of the AAE as the wavelength dependence of the AAE (WDA) in their Eq 2. Their WDA is presented as a ratio of two exponentials, so mathematically it is equivalent to

WDA = exp(AAE_440/870 - AAE_675-870).

Later, the authors use ln(WDA) in their Eq 4, so they are really just using the difference of AAE measured over two spectral regions to characterize the BC and BrC absorption at 440 nm. This is very similar to Bahadur (2012)!

We respectfully disagree with the reviewer about the similarity of our method with Chung et al. (2012) and Bahadur et al. (2012). We agree that both Chung et al. (2012), Bahadur et al. (2012) and our work focus on the difference of AAE over different spectral wavelengths, however, they are different in many aspects:

1) In Bahadur et al. (2012) (similar for Chung et al., 2012), they describe the total AAOD of carbonaceous aerosols at wavelengths λ as below:

$$AAOD(\lambda) = AAOD_{ref,BC}(\lambda/\lambda_{ref})^{-AAEBC} + AAOD_{ref,OC}(\lambda/\lambda_{ref})^{-AAEOC} \quad (1)$$

They then used observations to find the empirical AAEBC and AAEOC to solve the equation.

In contrast, we do not use the AAE of either BC or OC in our approach. Given the poor understanding of BrC, we do not think it appropriate to assume the properties of BrC (e.g. AAEOC) in a BrC derivation method. However, BC optical properties are better understood, which makes it possible to find a range for BC-WDA. Then the possible range of BC absorption (as well as BrC absorption) can be estimated using the relationship in Figure 2.

The core idea of our method is to find the range of BC absorption in any multiple-wavelengths absorption measurements. We do not try to characterize BrC optical properties in our method. The most important result of our method is the theoretical relationship of BC-WDA and BC-AAE shown in Figure 2, which to our best knowledge, has not been discussed in any previous studies (including Chung et al., 2012 or Bahadur et al., 2012).

2) Our method is primarily based on theoretical modelling (Mie calculations for BC) and also includes empirical information on BC (e.g. size distributions, etc.). In contrast, the studies of Chung et al. (2012) and Bahadur et al. (2012) are purely empirical.

In Bahadur et al. (2012), the values of AAEBC and AAEOC in equation 1 above were determined from AERONET observations. They sorted the AERONET sites into regions by different sources. For example, they assumed the lower end of the AAE values reflects absorption due to BC. By averaging different fractions of the total frequency distribution at the different AERONET sites, they assume $AAE_{440/675nm} \sim 0.55$ and $AAE_{675/870nm} \sim 0.83$ for the pure BC condition. They then assumed $AAE_{440/675nm} \sim 4.55$ for pure OC by using the AAE distributions. It is challenging to evaluate these thresholds numbers that were estimated from a few sites considering that the source of BrC is still under debate (Laskin et al., 2015). To summarize, Bahadur et al. (2012) (same for Chung et al. 2012) both used empirical information from select AERONET observations to derive the BrC absorption in a different set of AERONET observations.

In comparison, we do not apply such empirical information since conditions and properties can vary (e.g. the theoretical BC-AAE shown in our Fig.1). The BC-WDA-AAE relationship was calculated by Mie theory. Unlike Chung et al. (2012) and Bahadur et al. (2012), our method is based on the theoretical calculation using the information provided by the observation itself, not the empirical results from other absorption observations.

3) Because our method does not rely on any previous absorption observations, it is possible to apply it to observations other than AERONET. Our Figure 2 is only an example and our method is not restricted to the 440/675/870 wavelengths pairs. For example, we apply our method to surface measurements to derive BrC absorption at 370nm in Section 4. This is challenging for purely empirical method like Chung et al. (2012) and Bahadur et al. (2012) since there are limited previous observations of absorption at this wavelength.

In summary, our method is different from Bahadur et al. (2012) and Chung et al. (2012) from the initial idea to the applications. Given that 2 of 3 reviewers misunderstand our method, we have included a short discussion to describe the difference between our method and others (Page 6, line 22):

"A number of previous studies have used the AAE to separate the BC (or BrC) contribution from total absorption, however, these analyses typically rely on empirical information from previous observations. For example, Bahadur et al. (2012) and Chung et al. (2012) apply the same

approach where they group AERONET sites by regions and possible source types and by analyzing these groups, they estimate the possible AAE (or SSA or EAE) and the corresponding range for pure BC or pure BrC. They then apply these empirical constraints to estimate the BC or BrC contributions at other sites. In contrast, our method uses the theoretical relationship between AAE and WDA for BC shown in Figure 2 in combination with the observed total AAE, and does not rely on any other data. This also makes our method "wavelength-flexible". Although we use the 440/675/870nm to describe our method, any three wavelengths with one in the near-UV and two at longer wavelengths in the visible spectrum can be used. "

The authors go on to compute WDA for black carbon (BC) using a set of lognormal distributions (p4, lines 22-31), and provide a range of WDA values for their computations as the grey shaded area in their Figure 2. The median of this range of values is the black line, which represents the WDA_0 of Eq 4. Thus, the authors are using theoretical BC AAE calculations as a baseline AAE; differences from this baseline are attributed to BrC. Particle size effects on AAE are not considered, although extreme dust cases are later omitted from the analysis (EAE < 1 and AAE > 1.5).

We must clarify that we are not using any BC-AAE as a baseline AAE. In our Equation 4 and Figure 2, BC-AAE is determined by not only theoretical WDA but also the measured $AAE_{675/870nm}$. For different absorption measurements, different WDA and BC-AAE would be calculated. This method includes the effects of changing properties (e.g. size distribution) in the measurement itself.

In addition, the BrC absorption is not calculated using the BC-AAE from WDA0. It is the median value of the largest possible value (calculated from the lower end of BC-WDA range) and the lowest possible value (or zero, calculated from the higher end of BC-WDA range). We have clarified this point in the text (Page 5, line 30 – Page 6. line 2):

"We calculate the highest and lowest possible BrC absorption at 440nm based on the lowest and highest WDA (WDA_1 and WDA_2) as follows:

$$BC\ AAE_{440/870} = AAE_{675/870} + ln(WDA) \quad (3)$$

$$BrC\ abs(440) = abs(440) - BC\ abs(440) \quad (4)$$

The BrC absorption at 440nm is calculated as the median of these highest and lowest possible absorptions. For those points that fall within the shaded region, the BrC absorption is determined as the median of the highest possible absorption and 0."

There are many problems with this. First of all, the description of the parameters used for their Mie calculations does not include enough details for a reader to understand and duplicate the shaded region of figure 2. For instance, how thick are the coatings when the absorption enhancement factor is 2? What are the optical properties of the coatings? How does varying the optical properties of the coatings affect the size of the shaded area?

We have added a more detailed description of the coating calculation to the text (Page 4, line 26-30):

"The refractive index for coated material is assumed to be 1.55-0.001i, which is the typical value for non-absorbing organic and inorganic (Kopke et al., 1997). We first assume the coating thickness is 10% - 100% of the BC core radius and then only select the calculations with absorption enhancement smaller than a factor of 2."

Which size distributions represent the baseline? Are these 20nm particles, or 300 nm particles? Do the baseline particles have coatings? How thick?

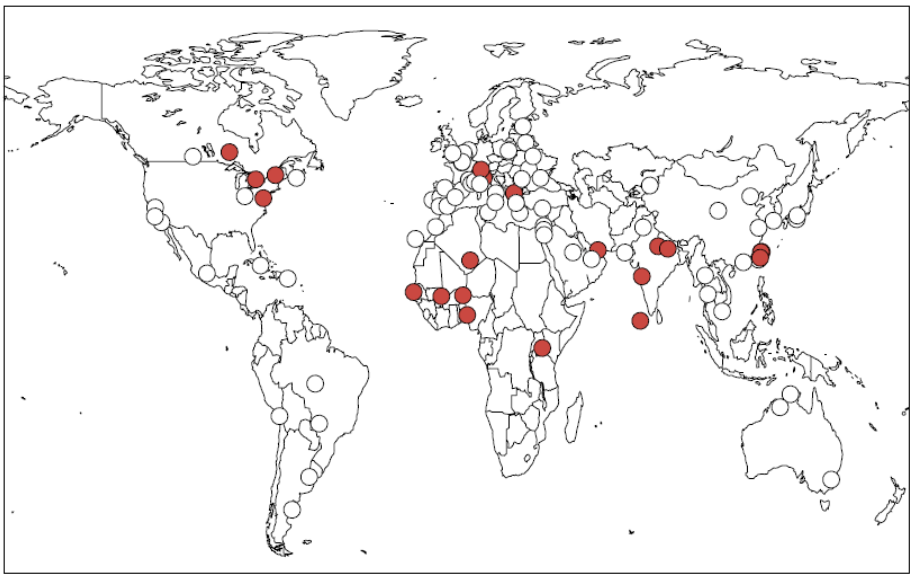
As mentioned above, we do not have any baseline BC-AAE or BrC-AAE in our method. If reviewer means WDA0 here, it is the median BC under a range of size and coating conditions with a certain $AAE_{675/870nm}$. It does not represent a specific size distribution or coating condition.

For example, if $AAE_{675/870nm} \sim 0.5$ is measured, we estimate WDA0 ~ 0.85 for BC from Figure 2. As shown in our Fig. 1, an $AAE_{675/870nm}$ of 0.5 is consistent with BC larger than 100nm. Then the WDA0 reflects the mean of different size/coating conditions for these large particles. In summary, WDA0 depends on observation itself, and does not reflect a specific baseline condition.

Which AERONET sites are falling inside of the shaded region of Fig 2?

Figure 2 is intended as an illustration of our method and we previously showed one year of AERONET data. However, we have modified Figure 2 to correspond with data presented later in the manuscript. Now the points in Figure 2 are the seasonal mean values of 2004-2015 10 years averaged AERONET data in northern hemispheric winter (December, January and February). So the sites in Figure 2 are now the same as those in Figure 3a.

The sites falling inside of the shaded area are painted red in the figure below. This information is not particularly useful since these data points are calculated as 10 years seasonal means and our later analysis is based on the daily data. We have clarified this point at the beginning of section 3.



Urban OC is non-absorbing, so the urban aerosols should fall inside of the shaded region, and the biomass burning aerosols should fall outside of the shaded regions. Looking at Figs 3&4, I don't believe that this is the case.

The source of BrC is still under debate, it is still unclear that whether urban OC is absorbing or not (Laskin et al., 2015). BrC absorption has been observed in US background air (Liu et al., 2015). Furthermore, an urban site can also be impacted by BrC from other sources. Because the availability of AERONET AAOD is not good, many of the 10-years averaged values shown in Figures 3&4 reflect the mean of only several days. To ensure that our means are more representative, we now show only the sites with more than 6 years' measurements available in a single season. We also include a stricter threshold to exclude the data affected by dust. This is described in the paragraph at Page 5, line 15 (shown below). Figure 3 and 4, and the discussion in Section 3 are modified accordingly.

"While AERONET provides global observations of the column-integrated AOD, few of these sites actually have continuous measurements of AAOD throughout the year because the SSA is not always retrieved. For example, more than half of the AERONET sites measured AAOD for only 1 month in 2014. As a result, we use the data from the past decade (2005 – 2014) to enhance our sampling. To reduce the influence of sporadic events in the analysis, when showing the 10 years seasonal average value only sites with data for more than 6 years within a given season are selected. The AAOD from AERONET not only reflects the absorption from BC and BrC, but also that from dust. We use two thresholds to exclude the data possibly affected by dust. First, we use the coarse-mode AOD contribution (at 440nm) provided by AERONET. We assume that dust controls the total extinction of particles larger than 1 μm diameter (coarse-mode), and therefore remove data with a coarse-mode AOD contribution > 20% from our analysis. Second,

the data exhibiting extinction Ångström exponent (EAE) < 1 are also considered to be influenced by dust and are removed prior to our analysis (Russel et al., 2010; Chuang et al., 2012)."

How come the shaded area of Figure 2 does not include the point (1,1), which would represent AAE = 1 for the entire wavelength range? Some of the particles sizes for their calculations are small enough (20 nm GMD) to reach the theoretical limit for small absorbing particles.

Thank you for pointing this out. Our Figure 2 should indeed include the point (1,1).Our original figure was smoothed across the intervals of $AAE_{675/870nm}$. We have modified Figure2 to eliminate this smoothing and more accurately present our results.

Why aren't any coarse mode aerosols included in the computations of the shaded region? There is always a coarse mode in the AERONET retrievals, and this will affect AAE. Sure, the authors eliminate aerosols dominated by dust in Section 3.1 by filtering for EAE < 1, but aerosol systems with 1 < EAE < 2 are mixtures of fine and coarse particles. Thus, there is dust in much (most?) of their analysis.

As described above and in the text, the shaded region in Fig. 2 is for BC only. Our goal here is to separate BC from other absorption. Therefore there is no issue associated with coarse mode aerosols in Fig.2.

We agree with the reviewer that, despite our filtering, the AERONET data may include some influence from dust. It is challenging to completely eliminate dust from these observations. We include a stricter threshold to exclude the data affected by dust. This is described in the paragraph at Page 5, line 15 as shown above.

However, we would like to point out that there is no such problem when applying our method to the fine particle absorption measurements made at the surface.

How well is the AERONET AAOD known? How does the uncertainty in AAOD at each wavelength affect the uncertainty in the AAE in the two spectral regions that you are using? How do the AAE uncertainties affect the BrC AAOD retrieval?

We have added a paragraph to discuss this point (Page 7, line 30):

"One challenge of this analysis is the well-known uncertainties associated with the AERONET observations. The measurement uncertainty is ±0.01 for AOD, ±0.03 for SSA when AOD > 0.2 and could be as large as ±0.07 for SSA when AOD < 0.2. The uncertainty of AAOD depends on the corresponding AOD value, for example, this uncertainty is ±0.015 with AOD = 0.4. Because our method is sensitive to the AAE not the AAOD, the uncertainty could be small for our BrC

contribution analysis if such uncertainties from AERONET are similar at all wavelengths. If the AERONET AAOD uncertainties vary substantially with wavelength, the influence on our analysis could be large and hard to quantify. In addition, the AERONET uncertainties suggest AAOD < 0.01 is certainly below the observed detection limit. In Figure 3 and in the above discussion most sites exhibit derived BrC-AAOD smaller than 0.01. However, all of these values are seasonal means over 10 years, and include both non-BrC detected (BrC-AAOD~0) data and BrC detected data. If instead we replace the BrC-AAOD < 0.01 data points by BrC-AAOD = 0.005 (the median of 0 and 0.01), the results are very similar."

Beyond the theoretical issues with this approach, though, the results do not seem to back up this approach. Figure 3 shows regional maps of seasonal mean BrC AAOD. The biomass burning sites in S. America never show elevated BrC AAOD, and the biomass burning seasons are not detectable over Africa. The Asian sites seem to have elevated BrC in all seasons except for Summer.

Figure 4 provides seasonal mean fractions of (BrC AAOD) / (total AAOD), and those results are even more ambiguous. The colors on these maps are all very random, with dark reds co-mingled with light blues; one would expect neighboring AERONET sites to exhibit similar colors if they were representing similar source regions. The randomness of the colors on these maps, though, seem to indicate noise.

As indicated above, the results of Figs 3&4 are impacted by the quality of AERONET data, which have been extensively discussed in literature. To address this issue, we now show only the sites with more than 6 years' measurements available in a single season. We also include a stricter threshold to exclude the data affected by dust. This is introduced in the paragraph at Page 5, line 15 as shown above. Figure 3, 4, 5 and the discussion in Section 3 are changed accordingly. We note, that this filtering does not significantly impact the distributions shown in Figure 6. We believe that Figures 3 and 4 now better represents salient geographical features, however we note that the biomass burning seasons still exhibits low BrC-AAOD over Africa. This is because such sites/data are removed from the analysis when we filter for dust.

OTHER IMPORTANT POINTS:

Page 3, line 6: AERONET provides *retrievals*, not measurements.

Changed.

Page 3, line 23-25: Chung (2012) and Bahadur (2012) do not assume AAE = 1 for BC, as stated here. The authors need to be more careful when attributing concepts to others.

Here we meant that Chung et al. (2012) and Bahadur et al. (2012) also pointed out the problems associated with assuming AAE = 1 for BC. We did not mean to imply that they assumed BC-AAE = 1 in their studies. We have deleted them from the references here to clarify this point.

Page 5, line 8: Bahadur (2012) has nothing to do with the SSA screening conditions ofAERONET. Cite Dubovik (JGR, 2000), instead.

Changed.

Page 5, line 16: The authors state: "... and cannot be estimated using our method without additional information about the size and coating state of the particles." However, you have information about size in the AERONET retrievals. Why not use them?

The sentence has changed to "... and cannot be estimated using our method without additional information about the size and coating state of BC particles."

The size distribution from AERONET retrievals reflects the entire aerosol in the column which always contains a combination of constituents. However, it is challenging to estimate the BC size distribution from this total size distribution.

Page 5, equations 3&4: These equations should be reversed. WDA_0 is used to compute BC AAE_440/870, which is used to compute BC abs(440), right? So presently, Eq 4 gets used before Eq3.

The reviewer is right, we have reversed the sequence between equation 3 and 4.

Page 6, line 18: The authors state: "In general, BrC-AAOD is smaller than 0.005 at most sites but larger in Asia and occasionally in northern Africa and South America." BrC-AAOD < 0.005 in general? That sounds like a small number. Does the AERONET retrieval really have the sensitivity to quantify this parameter, then? AERONET only claims an accuracy of _0.01 for AOD; the accuracy of AAOD is certainly no better. So all of the BrC-AAODs that you are retrieving seem to be well below AERONET's accuracy threshold.

Note that all the values shown in Section 3 are 10-year seasonal averages, which include days of both with and without BrC. The data with ~0 BrC-AAOD bring down the average. As shown

above, we have added new text to address the uncertainties of AERONET data in paragraph of page 7, starting at line 30.

Page 6, lines 24-26: The authors are quoting numbers for seasonal variability, but they do not specify seasons or sites. The colors on the maps do not obviously correspond to the numbers quoted here. The authors should at least give these "special" sites a different shape, so that the reader can understand which sites are the "African biomass burning sites." Also, the biomass burning season on the Sahel is not July-October. It starts in November and ends in the Spring.

As indicated above, we now use new thresholds to select AERONET data. This part of discussion has been changed as well as Figure 3.

Page 6, line 31: The authors state: "In winter the BrC AAOD contributions in North America (15%), Europe (17%) and East Asia (23%) are larger than in summer (10%, 13% and 14%)." Why should BrC AAOD fractions be higher in the winter in North America? Few Americans use biofuel, and the biomass burning season is in the summer. Also, higher winter BrC AAOD fractions are not obvious from Fig 4.

As indicated above, we now use new thresholds to select AERONET data. This part of discussion has been changed as well as Figure 4.

Equation 6: The authors assume that all OC are BrC for Equation 6. This is not a valid assumption.

Yes, in principle we agree. However, as we do not know what fraction of OC is BrC, nor do we have measurements of BrC mass, it is easiest to estimate the MAC for all OC. As a result, we analyze the optical properties of total OC as a combination of both BrC fraction and BrC optical properties. This is discussed at the end of section 3.2.

Page 12, line 10: The authors state "the correlations between BrC-AAE and BrC absorption contributions are only slight to moderate at these sites." They should still provide the correlations to the readers, though.

Added.

Page 12, lines 22-23. The authors state that OA-MAC is correlated with BC/OC mass ratio, but they do not help the reader to understand why this should be the case (anywhere in the article). Thus, for a given OC mass, the OA-MAC is linearly related to BC mass? This sounds like BC is contaminating their OC-MAC retrieval (rather than proof that their retrieval is working).

This relationship has been shown in the literature (e.g. Saleh et al., 2014) and is likely to relate to the combustion efficiency. We discuss this in section 3.2 and section 5.

Page 13, lines 26-28 and Figs 10b, 10c: Authors find that the OA-MAC for the nonbiomass plumes (Fig 10b) are much much higher than for the BB plumes (Fig 10c). WHY? OA-MAC for urban aerosols should be negligible.

Given the present poor understanding of BrC, it is not clear that OA-MAC is negligible under all urban conditions. OA-MAC is determined not only by OA absorption but also the OA mass. A small OA absorption with high OA-MAC is possible.

A possible explanation for this relates to the BC/OA ratio. As discussed in the text, OA-MAC is found to be positively correlated with BC/OA emission ratio (Saleh et al., 2014). The combustion sources in urban regions typically have much larger BC/OA emission ratios than biomass burning, which could result in higher OA-MAC.

Reviewer #2

This paper attempts to quantify brown carbon absorption using the AERONET data base of aerosol extinction optical depth and single scattering albedo as well as aethalometer measurements at a number of stations. The approach used in this analysis has been used in previous studies [Schuster et al., 2005; Russell et al., 2010, to mention a few], strangely ignored in the literature review. An innovative component of this work is the combined use of surface and space based AAOD retrievals for the estimation of aerosol absorption exponent. There are several issues in the paper that need to be addressed.

We respectfully disagree with the reviewer that our approach has been used in previous studies. The methods in Schuster et al., 2005 and Russell et al., 2010 are very different from ours.

The core idea of our method is to find the range of BC absorption in any multiple-wavelength absorption measurements. This is estimated based on the difference of AAE at different wavelengths pairs (WDA). Our method is primarily based on theoretical modelling (Mie calculations for BC) and does not rely on any information from previous observations. The most important result of our method is the theoretical relationship of BC-WDA and BC-AAE shown

as the shaded region in Figure 2, which, to our knowledge, has not been discussed in any previous studies. Furthermore, our method is "wavelengths-flexible". It is possible to apply it to the observation from not only AERONET but also absorption measurements at other wavelengths.

The work of Schuster et al., 2005 focuses on a completely different topic from this work. They use the Maxwell Garnett effective medium approximation to infer the black carbon concentration and specific absorption of AERONET retrievals. They focus on the AERONET data at 550nm and do not use any information from either the AAE or the WDA. In their analysis, they use a fixed black carbon refractive index of 2-1i and an ammonium sulfate refractive index of $1.53\text{-}10^{-7}$i at 550nm as well as the size distribution of total aerosols provided by the AERONET retrieval. They do not consider BrC as a contributor to absorption, though they estimate the uncertainty from organics by referencing other measurements.

Russell et al., 2010 try to use AAE and EAE to identify the aerosol composition measurements and discuss its future application. By analyzing 11 non-oceanic AERONET sites and other published observations, they found that the AAE values are strongly correlated with aerosol composition. For example, AAE near 1 is dominated by urban-industrial aerosol, larger AAE values are dominated by biomass burning aerosols, and the largest AAE values are indicative of dust aerosols. Their purpose and methods are very different from ours. First, they want to convey the idea of connecting AAE with aerosol composition, but do not attempt to separate BC from BrC as we do. Second, their analysis is based on previous observations. Third, although they use the information from the AAE, they do not address the wavelength dependence of AAE (WDA), which is the core factor in our method.

Since 2 of the 3 reviewers misunderstand our method, we have added a short discussion to the text to describe the difference between our method and others (Page 6, line 22):

"A number of previous studies have used the AAE to separate the BC (or BrC) contribution from total absorption, however, these analyses typically rely on empirical information from previous observations. For example, Bahadur et al. (2012) and Chung et al. (2012) apply the same approach where they group AERONET sites by regions and possible source types and by analyzing these groups, they estimate the possible AAE (or SSA or EAE) and the corresponding range for pure BC or pure BrC. They then apply these empirical constraints to estimate the BC or BrC contributions at other sites. In contrast, our method uses the theoretical relationship between AAE and WDA for BC shown in Figure 2 in combination with the observed total AAE, and does not rely on any other data. This also makes our method "wavelength-flexible". Although we use the 440/675/870nm to describe our method, any three wavelengths with one in the near-UV and two at longer wavelengths in the visible spectrum can be used. "

The use of annual average AAOD values in Figure 2, ignores the seasonal variability of aerosol type. It would be more illustrative to plot instead seasonally averaged data and identify in a table the sites used in the analysis. I suspect the analysis presented in section 2 will change substantially if seasonal AAOD data is used.

We have changed Figure 2 to show the seasonal averaged data. These points now also correspond to those shown in Figure 3a. However, we would like to mention that the purpose of Figure 2 is primarily to illustrate our method.

The seasonal maps showing the 10-year average of seasonal mean BrC-AAOD (as indicated in the caption of Figure 3) look very strange to me. I wonder if that is really what is being plotted. I am very surprised by the number of AERONET sites having 10 years of observations over the 2005-2014 period. Most sites shown on those maps do not have a continuous ten year record. Surprisingly the seasonal maps of derived BrC-AAOD based on AERONET observations do not show the expected AAOD hot spots in the Sahel (DJF), Southern Africa (JJA, SON), and Amazon Basin (SON). The only known hot-spot correctly showing is the Southeast Asia (MAM). I suspect these maps are more a representation of the geographical and temporal bias of AERONET observations over the 2005-2014 period than an actual representation of the AAOD load as intended by the authors. The number of sites in SE Asia and China has increased significantly over the last ten years. On the other hand, the coverage over Southern Africa has gone down significantly. A better description and explanation of this map is needed. I suggest to re-do these maps using a more careful selection of sites making sure that the temporal coverage is similar for all used stations.

Thank you for raising this issue. We have modified our thresholds to be more selective with the AERONET data. Only sites with > 6 years measurements available are used in the analysis. This is described now in the paragraph at page 5, starting at line 15. The poor coverage over Africa is because data that is influenced by dust is removed by the analysis; we comment on this on page 7, line 12:

"In contrast, no significant seasonal variations are found in other regions. The sites in Africa exhibit low BrC-AAOD even during biomass burning seasons. This is because nearly all the data with high AAOD in Africa are excluded from the analysis due to the influence of dust."

Other comments:

Page 6, line 24. Abundant biomass burning also takes place in Northern South-America and the Sahel during NH winter. Also in the NH Spring there is intense biomass burning in Southeast Asia and Central America.

We have changed this part of discussion based on our new AERONET data selection. In our analysis, the influence of biomass burning in South America and Southeast Asia are significant. We agree with the reviewer that the Sahel during NH winter and Central America spring are also characterized by abundant biomass burning, however we do not have enough data points in these regions to characterize this. We comment on this on page 7, line 12 as shown above.

Page 9, line 1. Add the Torres et al [2014] reference that describes the latest algorithm upgrades.

Added.

Page 9, line 8. Nowhere in the quoted references, is it said that OMI and AERONET AAOD have a correlation > 0.8 as stated by the authors. They incorrectly imply that OMI and AERONET AAOD have been directly compared citing papers that refer to AOD validation. Both OMI AOD [Ahn et al. 2014] and SSA [Jethva et al, 2014] have been compared to AERONET produced values. The authors need to read the papers and correctly quote them.

The reviewer is correct, it should be "AOD". This has been clarified in the text now.

The title of the paper is confusing because both AERONET and aethalometer data are obtained by means of surface observations. The title should also reflect the fact that OMI observations are used in the estimation of the reported results. Suggest changing title to 'Deriving brown carbon absorption from multi-wavelength absorption measurements: Method and applications to AERONET, OMI and Aethalometer observations'

Thank you for raising this point. We agree that the "aethalometer" should be used instead of "surface". However, we do not think it is appropriate to include OMI in the title. We use OMI to complement the analysis of the AERONET observations, but do not derive brown carbon from OMI. The title has changed to "Deriving brown carbon absorption from multi-wavelength absorption measurements: Method and applications to AERONET and Aethalometer observations"

Reviewer #3

This work provides an observational constraint for brown carbon aerosols (BrC), which absorb solar radiation, and thus have an important implication for climate. Based on the

previous study for quantifying the brown carbon aerosol absorption, authors used observed absorption angstrom exponents at a pair of wavelengths at AERONET sites to reduce associated uncertainties. I believe that this dataset adds up to the observations of BrC absorptions, which are very sparse globally and thus it will be valuable to evaluate the estimated contributions of BrC to aerosol absorption and radiative forcing. However, the observed quantity derived from the combination of various observations needs additional clarification and the details are listed as follows.

P4, L23 - Authors need to estimate the associated uncertainty with the assumption of spherical BC in their method. For example, Kahnert and Devasthale et al. (2011) estimated the difference of SSA up to 0.05 between spherical vs. aggregate shapes of BC (Figure 5 in their paper).

This is a good point. We have added a short discussion to mention this issue (Page 6, line 18-21) as shown below. However, we do are unable to estimate the uncertainty since there is little information on how aerosol shape might affect AAE and WDA.

"The spherical assumption in the Mie calculations could lead to additional uncertainties, as previous work suggests that the shape of BC can affect both the SSA and the absorption enhancement from coating (Adachi et al., 2010; Kahnert and Devasthale, 2011). However this uncertainty is hard to estimate since it is difficult to quantify how particle shape influences AAE and WDA."

P5, L21-24 - Please elaborate how you obtain 4% uncertainty.

In the text, this 4% is the detection limit, not the uncertainty. We assume the reviewer means the detection limit here.

As described in that paragraph, we estimate the uncertainty of BC absorption by repeating the calculation using the lowest and highest WDA value of the shaded region in Figure 2. The uncertainty of BrC absorption depends on both BC absorption uncertainty and the contribution of BrC to total absorption. If the uncertainty is equal or larger than the BrC absorption, the BrC absorption is below the detection limit. The minimum uncertainty for BC absorption is estimated to be ~4% (based on the smallest variability of WDA). Then we could assume a series of BrC absorption contributions. For example, when BrC contribute 50% of the total absorption, the uncertainty is estimated to be 4.2% based on the minimum ~4% uncertainty for BC. Assuming the total absorption is τ, then the real BrC absorption is 0.5τ and the uncertainty is $0.5 \times 4.2\% = 0.021\tau$. As the BrC contribution decreases, the real BrC absorption decreases. When BrC absorption contribution > 4%, the real BrC absorption is larger than the uncertainty; when BrC absorption contribution < 4%, the real BrC absorption is lower than the uncertainty, thus below the detection limit.

Figure 2 - It would be recommended to remove the dust contribution as shown in Section3.

We have changed Figure 2 to show the 10 years seasonal averaged data for winter instead of the annual averaged data for 2014. The data points are filtered for dust as described in the paragraph on page 5, line 15:

"While AERONET provides global observations of the column-integrated AOD, few of these sites actually have continuous measurements of AAOD throughout the year because the SSA is not always retrieved. For example, more than half of the AERONET sites measured AAOD for only 1 month in 2014. As a result, we use the data from the past decade (2005 – 2014) to enhance our sampling. To reduce the influence of sporadic events in the analysis, when showing the 10 years seasonal average value only sites with data for more than 6 years within a given season are selected. The AAOD from AERONET not only reflects the absorption from BC and BrC, but also that from dust. We use two thresholds to exclude the data possibly affected by dust. First, we use the coarse-mode AOD contribution (at 440nm) provided by AERONET. We assume that dust controls the total extinction of particles larger than 1 μm diameter (coarse-mode), and therefore remove data with a coarse-mode AOD contribution > 20% from our analysis. Second, the data exhibiting extinction Ångström exponent (EAE) < 1 are also considered to be influenced by dust and are removed prior to our analysis (Russel et al., 2010; Chuang et al., 2012). "

P7, L14-15 - Several papers showed that BrC absorption at 675 nm is significant (Alexander et al., 2008; Chung et al., 2012). So I am wondering if you assume an absorption at 879 nm as BC absorption alone, then how would your results differ. Or at least, you many need to discuss those previous papers and the possible effect on your estimates.

Thank you for raising this point. The assumption in our method is that BC dominates the absorption at 675nm regardless of the BrC absorption value. Neither Alexander et al., 2008 nor Chung et al., 2012 show the BC absorption contribution at 675nm. As mentioned in the text, we need at least two wavelengths measurements with negligible BrC contributions. Therefore our method would not work if we only assumed that BrC was negligible at the one wavelength (879 nm). We have added such discussion in the paragraph at page 8, line 7:

"In our method, we assume that the BrC absorption contribution is negligible at 675nm and 870nm. This is supported by the laboratory measurements (Chen and Bond, 2010; Zhang et al., 2013; Yang et al., 2009; Kirchstetter et al., 2004). However, Alexander et al. (2008) find the BrC absorption may be significant at 675nm by examining an electron energy-loss spectrum from a transmission electron microscope. If BrC absorbs significantly at 675nm, our estimate of BrC absorption at 440nm would be underestimated."

P7, L19 - Why did you use GFED3 for 2011 and earlier? If there is no reason for this, you better use GFED4 for the entire period consistently.

We now use GFED4 for the entire period.

P9, L8-9 - Jethva and Torres (2011) and Ahn et al. (2014) conducted an evaluation of AOD alone, not AAOD.

This should be AOD, we have clarified this in the text.

Jethva et al. (2014) did an SSA evaluation and showed that OMI SSA are higher than AERONET SSA. For example, about 50% of total samples showed the difference of 0.03 or higher and 25% showed 0.05 or higher differences. This is a considerable discrepancy between two datasets and may have a huge impact on your estimates. For example, if AERONET SSA is 0.94 and OMI is 0.97, then the estimated AAODs using AERONET versus OMI data differ by 100%. So BrC AAE using OMI and AERONET AAOD together may cause too high uncertainty. Please consider a bias correction for OMI or simply drop out OMI data in your calculation. minor corrections,

Jethva et al. (2014) shows that reasonable agreement is found between the two techniques globally, especially for the "urban/industry" type retrievals. Globally, 49% data at low AOD condition (AOD < 0.7) and 53% data at high AOD condition (AOD > 0.07) fall within the 0.03 uncertainty limit. OMAERUV SSA is either higher or lower than AERONET SSA at different site, not always higher as the reviewer stated. Significant differences between the two datasets are mostly shown at dust-dominated sites, which are excluded in our analysis. In addition, this comparison was made at 440nm. The OMI retrieval at this wavelength is more uncertain than the 388nm data we use in the analysis. It is not possible to directly compare the SSA/AAOD at 388nm since AERONET does not make measurements at this wavelength. Therefore we believe that the comparison between AERONET and OMI is still valid. We have added a discussion on this to the paragraph at page 10, line 31:

"Many studies have evaluated OMAERUV AOD by comparing them with ground-based measurements. The correlation between OMAERUV and AERONET AOD is usually found to be high (R > 0.8) (Jethva and Torres, 2011; Ahn et al., 2014). Jethva et al. (2014) also compare the SSA between OMAERUV and AERONET and find 69% of the data agree within the absolute difference of ±0.05 for all aerosol types. Significant differences between the two datasets are most shown at dust dominated sites. These dust-influenced sites are not included in our analysis. Furthermore, Jethva et al. (2014) compare these products at 440nm. The OMAERUV SSA

estimated at this wavelength relies on a number of assumptions and is more uncertain than that reported at 388nm that we use in our analysis. It is not possible to directly compare the SSA/AAOD at 388nm since AERONET does not make measurements at this wavelength. Therefore we believe that the comparison between AERONET and OMAERUV is still valid. . However, if the OMAERUV SSA is higher or lower than AERONET at 388nm, our estimate of the BrC-AAE$_{388/440nm}$ would be biased low or high."

P2, L28: Forrister et al., (2015) -> Forrister et al. (2015) P7, L15: 675 m -> 675 nm

Changed.