

# **Reply to Reviewer Comments on “Evolution of multispectral aerosol optical properties in a biogenically-influenced urban environment during the CARES campaign” by Gyawali et al., acpd-13, 7113-2013**

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## **Response to all Reviewers**

We would like to thank the reviewers for their time reading and commenting on our manuscript. In almost all cases, we revised our manuscript based on their comments.

In the following, we reply the reviewers point by point.

## **Anonymous Referee #2**

### **Reviewer Comment:**

Gyawali et al. present measurements from the 2010 CARES study that took place around Sacramento, CA. They focus on differences in aerosol optical properties observed between two sites: one located in the city and one located further downwind. Through this, they aim to understand how photochemical processing affects the particle optical properties, with a particular emphasis on the formation of so-called brown carbon. They also focus on contributions from supermicron particles to the observed scattering. The latter story is mostly disconnected from the former. My main concern at this point and that actually limits my ability to decide whether this paper is potentially publishable or not, is that I find the consideration of uncertainties to be exceptionally weak, almost nonexistent. In a study that aims to investigate differences between measurements made at two sites using a multitude of different instrumentation, explicit characterization and description of the uncertainties involved for all of the methods, and appropriate application and consideration of such uncertainties is required to draw meaningful conclusions. I believe that this manuscript should be reconsidered (i.e. re-reviewed) after the authors are given an opportunity to address this primary issue. Many of my specific comments stem from this concern.

### **Author Reply:**

We agree with the reviewer that the measurement uncertainties must be reported explicitly with the results. Further, the manuscript is revised to include the instruments' uncertainty and an error propagation estimate as suggested by the reviewer. The following section, “2.4 Error Analysis”, has been added to the revised manuscript.

## 2.4 Error Analysis

We have used basic uncertainty propagation theory to compute the error bars for derived aerosol optical properties,  $f(\beta_{abs}, \beta_{sca}, BC_{mass})$ , such as SSA, AEA, and MAC. Assuming that  $\beta_{abs}$ ,  $\beta_{sca}$ , and  $BC_{mass}$  are uncorrelated, the error,  $\delta f$ , in  $f$  follows from the method of propagation of errors ( e.g. Bevington, 1969)

$$\delta f(\beta_{abs}, \beta_{sca}, BC_{mass}) = \sqrt{\left(\frac{\partial f}{\partial \beta_{abs}}\right)^2 (\Delta\beta_{abs})^2 + \left(\frac{\partial f}{\partial \beta_{sca}}\right)^2 (\Delta\beta_{sca})^2 + \left(\frac{\partial f}{\partial BC_{mass}}\right)^2 (\Delta BC_{mass})^2} . \quad (4)$$

The three partial derivatives appearing on the right hand side of this expression can be evaluated using equations (1), (2), and using the expression for  $MAC$  ( $MAC = \beta_{abs}/BC_{mass}$ ). Any attempt at assigning uncertainties would be arbitrary (Corrigan et al., 2006), in our study both instrument calibration uncertainties and statistical variations are considered to estimate the error bars for calculated aerosol optical properties. The terms  $\Delta\beta_{abs}$ ,  $\Delta\beta_{sca}$  and  $\Delta BC_{mass}$  correspond to the root squared-sum of the squares of the uncertainties due to instrument calibrations and the statistical errors. The calibration uncertainties associated with PA absorption coefficient measurements and RN scattering coefficient measurements are 5% and 15%, respectively. Gyawali et al., (2012), Lewis et al., (2008), Lewis et al., (2007) and Arnott et al., (2000) have discussed these uncertainties. Similarly, the uncertainties in BC mass concentrations are about 22% (Subramanian et al., 2010). The statistical errors in MAC were obtained from linear fits (Regression analysis is provided in the supplement). For SSA and AEA the statistical errors are one standard deviation around the mean. The error bars on MAC, AEA, and SSA reflect both uncertainty developed due to the calibration of the instruments and ambient variability in the aerosol parameters; 12 hour averaging periods for AEA and SSA (Fig. 3-d,e and 5-d,e) , and 7 days averaging periods for MAC (Fig. 7). The error bars on the measured aerosol optical ( $\beta_{abs}$  and  $\beta_{sca}$ ) chemical (mass concentrations) and physical (PM1 particle mean diameter) properties are one standard deviation of the measurements collected during the time period.

### Reviewer Comment:

Section 2.2: Given the many different wavelengths involved, and the multiple instruments involved, the authors should provide information on the basic calibration procedures used and must state explicitly the absolute % uncertainty at every wavelength for both scattering and absorption. Only finally on P7131 do I find any mention of uncertainties (stated as 5% for absorption and 15% for scattering), but it is not clear whether these are appropriate for this study at all wavelengths since no information on calibration methods is provided.

### Author Reply:

As stated above we addressed this issue in the revised manuscript. The 5% absorption and 15% scattering uncertainty are applicable for all instruments as previously discussed (e.g., Lewis, 2007; Lewis et al., 2008; Gyawali et al., 2012; Arnott et al., 2000). Briefly, scattering is calibrated by comparison with extinction measurements for high concentrations of nearly non absorbing aerosol (various salts are used.) The repeatability of this calibration pertains to the 15% uncertainty. Absorption is calibrated by use of calibrated microphones and laser power

meters and use of the ab initio model for instrument response, as well as the difference of extinction and scattering for strongly absorbing aerosol (kerosene soot). The manuscript was revised to add these references wherein we discuss uncertainties and calibration in detail. The manuscript was revised to include the following sentence: “The absorption and scattering calibration procedure shown and discussed in and around Figure 1 of Gyawali et al 2012 was performed for all of the photoacoustic instruments.”

**Reviewer Comment:**

Section 2.2: The authors indicate that absorption and scattering measurements were made at many wavelengths, but then only 3 wavelengths are considered for SSA determination at each site (and not the same 3). What is the reason for this? Similarly, why are similar pairs of wavelengths not used at the two sites for AEA determination? I understand they say that they want to emphasize light absorption by organics, but by at least comparing results between the two sites with the same wavelength pairs, one would be potentially able to more easily assess differences. The consideration of a non-matched wavelength pair could then be used to provide more in depth understanding.

**Author Reply:**

We agree to compare similar wavelengths of PAs measurements if they are available. Although the PA instruments were measuring at several of the same wavelengths at both T0 and T1, there were technical problems which constrained PAs of different wavelengths to operate successfully at different times. For example, at T0 all PAs were operating successfully after 12<sup>th</sup> of June instead of 2<sup>nd</sup> of June (as in T1) while only the 532 nm UNR instrument was operating successfully for the entire month at T0 [the problem was caused by another instrument that had a leaky inlet system before June 12<sup>th</sup>; this compromised the data for everyone on that 1/2 of the sampling inlet system]. Additionally, at T1, two of the PAs were not operating successfully from the beginning of the campaign: the PAs operating at wavelengths 405 and 375 nm were measuring consistently only after 21<sup>st</sup> of June. Therefore, SSA and AEA calculations (Fig. 3 and Fig. 5) were reported for the longest available PAs measurements at each site. The 870 nm PA at each site was successfully operating and is shown in Figures 3 and 5. Figure 7 represents the longest available consistent measurements for the PAs and SP2, which correspond to the 22<sup>nd</sup> to 28<sup>th</sup> June.

**Reviewer Comment:**

I do not believe it is appropriate to quantitatively compare the AEA values determined for different wavelength pairs. Similarly, if fits are performed (as in Fig. 7) using different wavelengths, then the results are not comparable.

**Author Reply:**

We agree. The AEA was re-calculated by performing the fits for the same wavelengths (405, 532, and 870 nm for T0 and T1) as suggested by the reviewer. Figure 7 was revised to show this. The revised figure shows error bars calculated additionally using the relative uncertainty (22%) of the SP2 instrument (Subramanian et al., 2010) and 5% for the photoacoustic instrument measurement of light absorption, in addition to the fit error. The size of the error bars is strongly influenced by the SP2 uncertainty. However, for T0 we used the same SP2 instrument for all measurements; therefore, the relative uncertainty for the various wavelengths

are affected the same way (and similarly for T1). The mean values of MAC at 870 nm at T0 and T1 agree well with each other as expected for sites affected by black carbon light absorption.

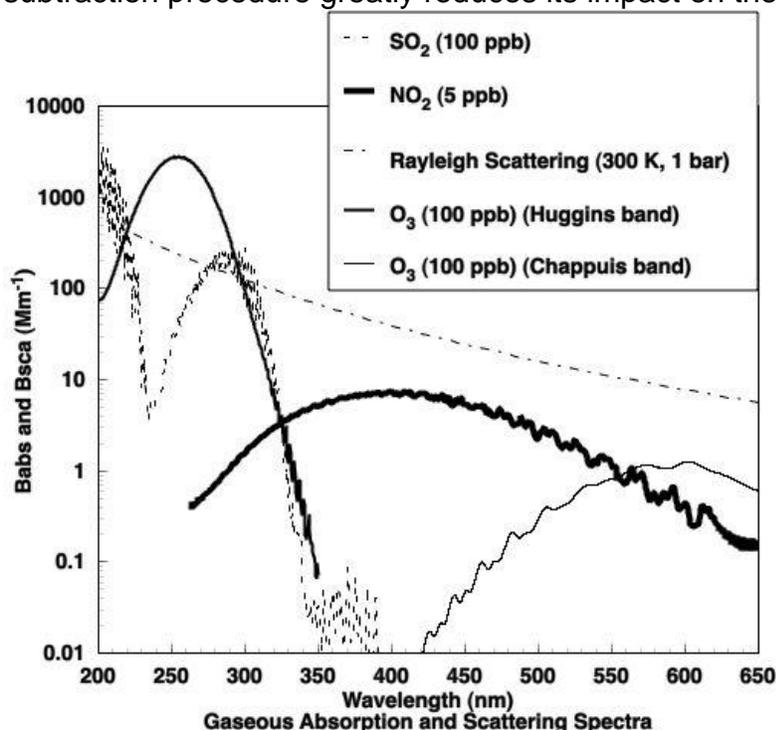
The message of figure 7 is that “the MAC is similar at 532 nm and 870 nm at T0 and T1. However, at 405 nm, 375 nm, and 355 nm, T1 shows some excess absorption that may be attributable to brown carbon.”

**Reviewer Comment:**

Is there a potential for absorption by gas-phase species at UV wavelength

**Author Reply:**

NO<sub>2</sub> is the primary absorber to worry about with regard to the photoacoustic measurements at 355 nm, 375 nm, 405 nm, and 532 nm, see the figure below. However, we used an annular denuder for removing NO<sub>2</sub>; in addition, our zeroing system operates by passing gas plus particles during the online measurement phase while passing only gas during the zeroing phase. As long as the gas concentration is constant over periods of 5 to 10 minutes the subtraction procedure greatly reduces its impact on the particle light absorption measurement.



Caption: Empirical gaseous absorption and scattering in the wavelength range of the study.

**Reviewer Comment:**

Section 2.3: Regarding the correction of the [rBC] to account for the use of Aquadag as a calibration standard, the authors multiply by 1.6, but if one looks at the “Slope of linear fit” from Moteki and Kondo (2010) for Aquadag and ambient soot, the ratio should actually be 1.32. The ratio between Aquadag and Fullerene soot is 1.42. Both are smaller than the factor of 1.6 applied here. Laborde et al. (2012) similarly find a smaller ratio than 1.6 (they find the ratio is size dependent, with a value of 1.3 for a particle of 10 fg rBC).

**Author Reply:**

Thanks for pointing this out. We noticed that the use of 1.6 as a calibration standard is incorrect. The manuscript is revised to reduce the BC mass concentration by 10% ( $1.6 \times 0.9 = 1.44$ ) to get a better value.

**Reviewer Comment:**

P7122: Technically, the APS aerodynamic diameter to spherical-equivalent diameter adjustment should also include the change in the particle [Cunningham] slip correction [factor] which is size dependent.

**Author Reply:**

Agree. The manuscript “DeCarlo, P.F., J.G. Slowik, D.R. Worsnop, P. Davidovits, and J.L. Jimenez (2004). Particle morphology and density characterization by combined mobility and aerodynamic diameter measurements. Part 1: Theory. *Aerosol Science and Technology*, 38(12): 1185-1205.” shows that considerable uncertainty is associated with particle morphology. Complete closure of optics calculated from size distribution measurements requires more knowledge of size dependent morphology than is available to us in this study.

**Reviewer Comment:**

Figure 7: I find that this figure does not properly account for/present the uncertainties associated with the measurements. The authors plot their derived mass absorption coefficients (MAC) vs. wavelength to assess the influence of wavelength on light absorption. The error bars they include are derived from the fit error to a plot of babs vs. [BC]. These error bars do not present a true indication of the actual uncertainty in the measurements made at each individual wavelength, and in fact substantially underestimate the actual uncertainty. Consider that the apparent uncertainty on the 870 nm MAC at T1 is 0.08 out of 5.57 Mm<sup>-1</sup>, which is a percent uncertainty of 1.43%. Neither the absorption nor the [BC] measurements are good to within 1.43%. However, recognizing that all of the absorption measurements are effectively divided by a “constant” (i.e. the [BC] time series), uncertainty in [BC] can, for the time being, be ignored. Then the actual uncertainties should simply be the absolute uncertainty associated with the absorption measurement at each wavelength. These will be wavelength specific. This is particularly important because the AEA determination depends on the accuracy of the measurements at the different wavelengths, not their precision. I believe that it is important that this figure indicate the absolute uncertainty, and not the fit uncertainty. Errors should be propagated accordingly. Further, given that [BC] is a constant (by which I mean all the absorption time series are divided by the same BC time series), there is not necessarily any benefit to presenting results here as MAC as opposed to absolute absorption.

**Author Reply:**

We revised the manuscript to address this issue. Now the uncertainties on the plots were calculated by using error propagation by including the 5 % uncertainty in PA absorption coefficient measurements and 22% uncertainty in SP2 BC mass measurements and the fit error. We believe that presenting MAC is important here, since BC itself is not necessarily constant from T0 to T1. We have discussed above other modifications of Figure 7.

**Reviewer Comment:**

Figure 8: I do not understand the particular utility of this figure unless the authors are going to compare these calculations to their observations. It does not add any information that Fig. 9 does not already contain.

**Author Reply:**

Figure 8 presents the time series of sub-micron and super-micron aerosol light scattering. It's important to note here the variation of sub-micron and super-micron aerosol light scattering with NW and SW flow, which cannot be studied from Fig.9 alone.

**Reviewer Comment:**

Fits: Are all of the linear fits shown in the manuscript one sided? If so, this is not appropriate since there is error in both the x and y axes. I recommend that the authors use a two-sided fit or use an ODR method.

**Author Reply:**

We perform the fits for linear regressions of Babs and BC to obtain the MAC reported in figure 7 (now in the supplemental section). The systematic error of the SP2 (with regard to calibration) was much larger than the standard deviation. In other words, the SP2 measurements were less 'noisy' than the photoacoustic absorption measurements (as the SP2 uses a much higher laser power irradiance than does the photoacoustic instruments.)

There are many methods for linear regressions; we chose to use the simplest one.

**Reviewer Comment:**

P7128: A small R2 does not, by itself, indicate anything about the contribution of absorption by organic species. It may simply be an indication of measurement uncertainty. (Consider that I could theoretically do an experiment where I know that parameter x and y have a perfectly linear relationship, but my measurement methods are so noisy so as to hide this relationship. I am not saying this is the case here, only that low R2 values do not by themselves imply a fundamental lack of correlation.) However, since the authors do not show any correlation plots for wavelengths besides 870 nm (for which the best correlation coefficient is obtained), it is not possible for the readers to decide for themselves whether they agree with the presented interpretation or not. This is related to the broader issue of a lack of appropriate uncertainty analysis. I believe that scatter plots of absorption vs. [BC] should be shown for every wavelength as supplementary material.

**Author Reply:**

The appropriate uncertainty calculations are performed in the revised manuscript. The scatter plots of absorption vs BC will be presented as supplementary material as suggested by the reviewer.

**Reviewer Comment:**

The statement on P7128 that "the rather small increases in absorption for 870 and 532nm wavelengths at T1 compared to T0 suggests that the additional coating on the aged BC particles at T1 may not have produced an appreciable lensing effect" must be justified through

rigorous consideration of absolute uncertainties involved in both the absorption and BC measurements.

**Author Reply:**

We have amplified the uncertainty discussion and calculation for Figure 7. The relative performances of SP2 instruments at T0 and T1 have an influence on this issue.

**Reviewer Comment:**

In the abstract, the authors indicate that the MAC increases by 60% in going from T0 to T1. I do not believe that this is justified by the observations. They are, presumably, determining this by extrapolating the T0 results to shorter wavelengths and then comparing with the T1 results at 355 nm. This cannot be done because, as seems to be the case at T1, the absorption by brown carbon could take off below 375 nm at T0: the actual behavior is unconstrained and unknown. At best, the authors can compare the 405 nm MAC values between the two sites, from which they might conclude there was an increase of 30% between the two sites. However, any such comparison must also account for the absolute uncertainties in these measurements, which is not reported. A standard deviation here would not be appropriate, since that simply tells about the variance in the measurements.

**Author Reply:**

We agree with the reviewer. As explain above, the manuscript is revised to implement the appropriate uncertainty calculations. In the revised manuscript, we will state 30% increase of MAC values instead of 60%.

**Reviewer Comment:**

On P7129, the authors state: “a more detailed analysis of the absorption data with the core-shell Mie theory, constrained with the observed BC coating thickness (from SP2 measurements) and morphological information (based on SEM images), is needed to estimate the relative contributions of the lensing effect”. I would urge caution here, because one can easily end up in a circular argument if SP2 LEO analysis is used to derive coating thicknesses since LEO analysis relies explicitly on core-shell assumptions and Mie theory. Thus, it cannot provide an independent measurement of coating thickness that can be used in Mie theory calculations.

**Author Reply:**

The caution mentioned by the reviewer is well-founded. Thanks for the comment.

**Reviewer Comment:**

P7126: As written, I find that the discussion of the Moffet and Prather work implies that their measurements were made concurrent with the measurements in this study. I suggest revising. Further, It should be clarified that Moffet and Prather did not observe an absorption enhancement of 1.6. They calculated this value based on Mie theory, constrained by their BC/coating ratio.

**Author Reply:**

We find that this paragraph is out of context in the submitted manuscript. It was meant to be a discussion of what sort of absorption effects might occur due to aerosol morphology and composition. This paragraph was revised to start with a clarification sentence at its start. “Previous laboratory and field studies employing direct measurements and inference from use of Mie theory have reported absorption enhancement.” This paragraph is more appropriate for the introduction. It was moved to be the third to the last paragraph of the introduction

**Reviewer Comment:**

I am not certain I see the utility in the comparison between calculated scattering and observed scattering since loss of supermicron particles to the optical instruments is not well constrained. As they note: “the more than double overestimation of the scattering at 1064nm could have been caused by coarse mode particle loss in the inlet system.

**Author Reply:**

We believe that the Mie calculation of aerosol light scattering and its comparison with the measured scattering values is important for this study. It’s almost matching for all wavelengths except at 1064 nm at T0. From this comparison, we came to the conclusion that there is loss of coarse mode particle in the inlet system at T0. Aerosol scattering closure is an important topic; in this study we find that a relatively simple model for aerosol composition, as reflected through use of a single refractive index, does a good job of reproducing the measured values.

**Reviewer Comment:**

Minor comments

P7117, L23: This is awkwardly stated. “85% of time. . .”)

**Author Reply:**

We have revised this sentence to read “In another study, Kassianov et al (2012) showed that coarse mode aerosols contributed between 50% and 85% of the total aerosol volume observed at T0 and T1 during CARES.”

**Reviewer Comment:**

P7119: I suggest “Since these PA instruments were designed” to measure both absorption and scattering. Really, in the context here, PA is a bit of a misnomer (or incomplete-nomer) since PA + photoacoustic and the scattering measurements are independent of the photoacoustic technique. Perhaps PA/RN (to indicate reciprocal nephelometer)?

**Author Reply:**

We agree with the reviewer that the terminology gets confused below the definition of “PA” given at the beginning of Section 2.2. We prefer to keep the simpler notation “PA” for these instruments.

**Reviewer Comment:**

Fig. 3 Caption: “particle mean diameter” should be “particle mean number-weighted diameter”

**Author Reply:**

We have added a sentence in the revision to state that the particle mean diameter reported in this paper is the particle mean number-weighted diameter.

**Reviewer Comment:**

P7123, line 20 and throughout, as appropriate: I encourage the authors to use “larger than” or “greater than” as opposed to “enhanced” (larger is an adjective, enhanced is a verb)

**Author Reply:**

We prefer to stay with “Enhanced” as an adjective (Enhance is the verb). “Enhanced” describes “formation”.

**Anonymous Referee #3****Reviewer Comment:**

Gyawali et al. investigated aerosol optical properties in a biogenically-influenced urban environment during the CARES campaign using the photoacoustic spectrometer as well as other instruments. An advantage of the photoacoustic spectrometer is that it can avoid the artifacts associated with the filter-based method (e.g., Aethalometer). Therefore, this kind of optical measurement, especially in combination with the chemical composition information, is useful for a better understanding of the optical properties of black carbon (or elemental carbon). My overall assessment is that the information presented in the study is a useful addition to the literature. Thus, this manuscript should not be rejected. However, it is not acceptable in its present form and re-review is necessary.

1. A substantial concern is the measurement uncertainties, which have been pointed out by both reviewer 1 and reviewer 2. This question cannot be avoided. The authors should add a detailed discussion on the absolute uncertainties on all of the measurements mentioned in the manuscript.

**Author Reply:**

We agree with the reviewer. The manuscript is revised to report the results based on the uncertainty of the measurements for different instruments.

**Reviewer Comment:**

2. I also noticed that different wavelength pairs are used to calculate the AEA values for the two sites (Page 7120). As the authors mentioned in their response to reviewer 1, “the choice of wavelength pairs was driven by the best measurements over the longest time period during the campaign”. To my understanding, the so-called “best measurements” depend on the measurement uncertainty. However, this kind of description is too cursory for a scientific paper. In other words, the uncertain issues should be presented quantitatively.

**Author Reply:**

The best measurements over the longest period mean the successful and consistent measurements over the period of time. We have explained this further in the response to reviewer 2. We had measurements that were compromised by malfunctions of the inlet

system for part of the campaign, making data unavailable for certain wavelengths for this part of the campaign. It was not an issue of measurement uncertainty, but an issue of measurement availability.

**Reviewer Comment:**

3. In addition, I am very disappointed that only OA results from the AMS measurement are presented, although OOA and HOA results are mentioned occasionally. HOA and OOA should be presented separately in Figure 3 and Figure 5. Influences of the abundance of OOA on the AEA values should also be investigated.

**Author Reply:**

We appreciate the reviewer for these important suggestions. We had separately investigated the influence of OOA, HOA including biogenic SOA marker, cooking OOA marker on the AEA. Although these species show some influence on AEA, but we didn't find that certain species that have a particularly dominant influence on AEA. So, we only presented OA results from the AMS measurements.

**Reviewer Comment:**

4. At both sites, babs (Mm<sup>-1</sup>) correlated well with BC concentration (Figure 7). However, I am not sure whether these results necessarily mean that the MAC values would exhibit little variations. Thus, I suggest that MAC values should also be calculated for each data point, rather than only relying on the slope of the linear regression of babs on BC concentration. Moreover, the influences of OA, OOA and sulphate (e.g., their concentrations as well as abundance) on the MAC values should also be added in the revised manuscript.

**Author Reply:**

The use of the aggregated MAC versus Babs values were needed to obtain sufficient averaging for a broad look at the data set. There may very well be certain events or episodes of a short time duration that deviate from the average values, and these may be the subject of future investigation.

With regard to the OA OOA, etc question, we did look at the speciated OA and absorption values. OA dominated the non black carbon mass.

**Reviewer Comment:**

5. Some minor comments. Page 7122, Line 1~2, please reword the sentence. Page 7128, Line 9, I am confused about the use of T1 and T2. Please clarify or reword the sentence.

**Author Reply:**

We agree with the reviewer. The following revisions were made to manuscript in response to the suggestions.

Page 7122, Line 1~2: "where  $Q_{sca}$  is the Mie scattering efficiency and is a function of refractive index  $m$ , wavelength  $\lambda$ , and particle diameter  $D_p$ ."

Page 7128, Line 9: at sites T1 (north of the Mexico City) and T2 (~35 km farther to the northeast) downwind of Mexico City"

## **Interactive comment on “Evolution of multispectral aerosol optical properties in a biogenically-influenced urban environment during the CARES campaign” by M. Gyawali et al**

### **Anonymous Referee #2**

#### **Reviewer Comment:**

I believe that in this first response, the authors have failed to directly answer a number of Reviewer #1's (R1) questions. First, R1 specifically states "There is no discussion on the uncertainty in PASS measurements and given a range of approximately 20% uncertainty in other similar measurements, this difference of 10% seems fairly unremarkable and within the uncertainty range of the instrument." Given this comment, I would have thought that the author's response might make some concrete statements about instrument uncertainty, which, as R1 notes, is not discussed. Unfortunately, the authors seem to avoid this question, instead giving some answer about the economic downturn with only a cursory mention of uncertainties that does not answer the question. The authors absolutely did not present their "results in the context of instrument uncertainties." The instrumental description provides no discussion of absolute uncertainties on any of the measurements. Further, the error bars that are provided are not the appropriate error bars for consideration. For example, (and as I've noted in my review) the uncertainty determined from a linear fit to a plot of absorption vs. [rBC] is NOT the appropriate uncertainty to consider as this only says something about the precision of the measurements and the level to which the data conform to a linear fit. It provides no information about the absolute uncertainties involved and is not appropriate when measurements from different instruments are being compared between different sites. The minimum percent uncertainty in the MAC is actually the square root of the sum of the squares of the percent uncertainties for each of the measurements involved, which I imagine is actually wavelength dependent and may even be different at the two sites since two sets of independent instruments were used. I therefore strongly disagree with the authors contention that they "presented their results in the context of instrument uncertainties."

#### **Author Reply:**

We believe all of these issues and concerns are properly addressed in the revised manuscript.

#### **Reviewer Comment:**

Second, the authors miss the entire point of R1's question regarding the use of different wavelength pairs for calculating AEA at T1 and T0. As the authors note just a few responses above, "The AEA of course does depend on wavelength choice." Given this, comparing AEA values that are derived at different wavelengths, and then aiming to draw conclusions based

on differences between these AEA values, is not justifiable. Further, the authors must propagate their uncertainties to determine an absolute uncertainty in the AEA values. I will note that if the authors were simply looking at the variability in the AEA as measured at one site by one set of instruments, then it may be possible to identify differences/changes in the AEA that are smaller than the absolute propagated uncertainty because the measurements can then be looked at in a relative sense (which then depends on instrument precision and not accuracy). However, because they are comparing between multiple sites with independent instruments absolute uncertainties cannot be ignored and fit uncertainties are, for the most part, not a useful metric.

**Author Reply:**

Please refer to the reply to Reviewer#2

**Reviewer Comment:**

I make these additional comments here in the hopes that the authors are able to provide more direct answers to my questions and concerns raised in my review, as opposed to what I view as very cursory responses provided here for many of R1's most important points.

**Author Reply:**

Please refer to the reply to Reviewer#2

**References:**

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