We thank the reviewers for their constructive comments and have made changes, where deemed appropriate. Specific responses to each of the comments are provided below (reviews' comments in black and our responses in red).

Anonymous Referee #3:

This is a nice paper looking at light absorption by water and methanol extracts of collected filter samples. The authors aim to utilize the size-dependent collection to understand how absorption varies as a function of size and, in particular, estimate particulate absorption from these particles based on Mie theory, which requires particle size. I think that this paper will be publishable, but ask that the authors first address the questions/ comments below (given in mostly chronological order, not order of importance).

Line 23: The reference to Gyawali is a bit premature as that reference is (a) only a discussion paper and (b) did not do a sufficient uncertainty analysis in the presented data to justify their conclusions (although this may change in the revised version: : :but currently only the discussion paper is available). There are presumably better references (such as some of the authors own work). Further, the authors seem to say the same thing twice in this paragraph, i.e. the first sentence is redundant with the 2nd and 3rd, so the issue can be alleviated by simply deleting the first sentence.

The first sentence is changed, (see below) to briefly indicate the emphasis of this whole paragraph and the reference to Gyawali has been removed.

The sentence now reads: Brown carbon has both primary and secondary sources.

Line 11: "Exponentially"? Then why do we parameterize as a power law (i.e. the Angstrom relationship)?

"Exponentially" has been changed to "rapidly".

Line 4: In addition to noting that there were 6 complete samples collected, this is a good place to note that 2 were from JST, three from YRK and only one from RS.

The information has been listed in Table 1. We prefer to keep the text simple.

Table 1 lists the EC as "optical". If it is "optical", then it is actually BC, not EC that was measured. Also, this listing as the EC as "optical" seems somewhat inconsistent with the discussion on p18239 where it is stated that OC and EC were determined using the NIOSH protocol, which I understand to measure EC via the thermal method, not BC via the optical method. I also think that, given that a unique temperature split point was not determined for each sample, that non-standard uncertainties apply. The uncertainty in EC (and OC) are determined based on "blank variability" and uncertainties in flow rate. This would not capture any uncertainties due to variations in the split point, which might occur for the different samples. Thus, these uncertainties are undoubtedly lower limits. It would be useful if a more robust uncertainty estimate could be provided.

A good point. Due to operational problems during the sampling period, online thermal EC data were not available, and that's why we used optical EC. We've changed the data in Table 1 to thermal EC determined from the MOUDI measurements and have noted this in the text. (see below)

Regarding to the uncertainty due to the fixed temperature split for OC and EC, we did include this in the uncertainty analysis, but maybe did not state it clearly. The average±stdev of the split time determined from Hi-Vol filters were 500±15.4, and the estimated uncertainty related with the split time is 1.3% for OC and 4.5% for EC, which have been included in the uncertainty analysis. The text (page 9, Lines 190) as well as Table 2 has been modified.

Table 1 (part) now as:

Start time	Stop time	Location	Temperature, °C	RH, %	$PM_{2.5}$, μg/m^3	OC, μgC/m^3	EC*, μgC/m^3	OC/EC* ratio	BC (Avg of all Aeth. λ), μgC/m^3	WSO μgC/
5/17/12 16:45	5/20/12 15:00	JST	22.5±3.5	55.2±13.3	14.63	4.39	0.81	5.4	0.77	2.3
5/25/12 12:00	5/27/12 20:00	JST	28.6±3.4	46.8±11.4	18.96	4.61	0.82	5.6	0.93	2.8
6/15/12 11:35	6/18/12 13:30	YRK	23.8±3.4	55.7±12.6	10.70	4.43	0.38	11.7	0.45	2.4
6/21/12 11:10	6/23/12 11:50	YRK	27.2±3.1	57.8±11.7	16.09	5.24	0.44	11.9	0.44	2.7
6/27/12 12:00	6/29/12 13:55	YRK	28.9±4.5	44.2±15.1	17.73	7.26	0.54	13.4	0.55	4.3
9/26/12 12:00	9/29/12 8:40	RS	24.0±3.7	58.3±16.8	13.36	6.66	1.42	4.7	NAN	2.9

* EC data from MOUDI filter measurements since online thermal EC data were not available due to instrument issues

In general, how were samples treated between collection and analysis? How long did they sit out at room temperature?

The samples were packaged separately using pre-baked aluminum foils and stored in a freezer before analysis. Considering the time needed for packaging, filter punching and preparation for extraction, the filters generally sit out at room temperature for 2 hours.

It is stated on L5, p 18239 that two blanks were included. The uncertainties in the OC/EC and brown carbon absorption and WSOC are apparently derived primarily from "blank variability (1 sigma)". If only 2 blanks were run, how is a 1 sigma value determined and how robust could it possibly be?

2 blanks were included for each set of MOUDI samples, and for 6 MOUDI samples there were 12 blanks in total. The "blank variability (1 sigma)" was determined on the 12 blanks, not only the 2 blanks for each MOUDI sample.

P18242, L20: How was single scatter albedo estimated?

The single scattering albedo was determined as the ratio of scattering/(scattering + absorption) (Page 13, Lines 279-280). As stated in the paper, scattering was estimated from TEOM PM2.5 data, and absorption data were from scattering-corrected Aethalometer measurements by iteration until a converged solution was achieved.

P18242, L22: It would be really nice to see that the size distributions measured by Carrico (2003) were similar to those measured during this study, as this will affect the scattering/mass relationship. If the particle size is not the same, then the relationship will not be the same and the scattering will be under/over estimated. How accurate do they believe this estimate to be? Similarly, I do not understand how it is appropriate to use a scattering Angstrom exponent of 1.4 that derives from a study that measured particles around Asia from a plane. The Angstrom exponent depends on the size distribution. A value of 1.4 implies something specific about the size distribution. Is this value consistent with the particle size during this study? How would the results be changed if they assumed a value of 2 (smaller particles)? Or 0 (bigger particles)?

Unfortunately, we do not have a size distribution for $PM_{2.5}$ measured during this study, corresponding to the PM-scattering relationship in Carrico et al. (2003), but the estimated single scattering albedo of 0.85-0.91 (also applying the scattering Angstrom exponent of 1.4) agrees with previous studies [i.e., *Carrico et al.*, 2003]. From our personal experience in many other studies, a strong and similar relationship between

$PM_{2.5}$ mass and scattering is observed. Moreover, our estimated uncertainty of 36% did include the relevant uncertainty from the scattering estimation, with the absorption Angstrom exponent ranging from 1.0 to 2.0, and a ±10% range in the scattering-PM concentration relationship.

Reference:

Carrico, C. M., M. H. Bergin, J. Xu, K. Baumann, and H. Maring (2003), Urban aerosol radiative properties: Measurements during the 1999 Atlanta Supersite Experiment, Journal of Geophysical Research-Atmospheres, 108(D7).

Also, it is not made particularly clear that the absorption at 670 nm comes from the MAAP. Or at least I assume it comes from the MAAP in determining the SSA, otherwise this would be a problem of circularity. So if the absorption used to determine the SSA comes from the MAAP, then how are the Aeth data corrected at the YRK site (since the MAAP was only deployed at the JST site)? This is not clear. Further, although JST is an urban site, the derived SSA values seem low to me. Can these be justified based on previous measurements? Could this, perhaps, indicate a limitation of the Angstrom scattering coefficient used or the scattering/mass relationship used?

Absorption at 670 nm at three sites came from scattering-corrected measurement by the Aethalometer. MAAP measured BC and Aethalometer-measured BC agrees at JST when they are co-located, and MAAP absorption = BC*6.6 m2/g at 670nm, so we believe it is reasonable to first estimate absorption and SSA using Aethalometer-determined BC * 6.6 m2/g (the MAC of BC), then estimate scattering-corrected Aethalometer absorption by iteration until a converged solution was achieved.

Our derived SSA values agree with *Carrico et al.* [2003], which reported a single scattering albedo in Atlanta of 0.87±0.08. The SSA seems lower than nonurban areas likely due to primary emissions.

Fig. 1: What sort of fit was done? A strict linear fit is not appropriate as there is uncertainty in the x-axis as well as the y. An ODR (or equivalent) fit is more appropriate.

The fit is ODR fit. We have noted this in the text to make it clear.

P. 18243, L18: The authors state that complex refractive indices can be obtained from the solution data. However, they only present results for the imaginary part of the complex refractive index. (The complex refractive index includes both the real and imaginary parts). This should be restated.

We have changed "complex refractive indices" to "the complex part of refractive indices".

P18241/L11: The authors state that the system was cleaned and then baseline zeroed using mill-Q water. They indicate that this was done for both the on and offline systems. I assume that for the methanol extracts the system was actually zeroed using methanol, not water and that this is just not stated? This needs to be stated or, if the system was always zeroed on water, the impacts on the methanol extract measurements needs to be discussed.

The system was zeroed on methanol when doing methanol extracts. The text has been changed (Page 12, Line 246).

Fig. 2: How important is the forcing the intercept to zero?

If not forced through zero, the agreement is better, with all R2 >0.9 and slopes still within 10% of 1 so there is no large difference either way. However, forcing through zero has an important physical meaning; that the data have been blank-corrected appropriately.

P18247/L1: The RS OC is not greater than all of the YRK OC measurements. I also do not understand why it would be "expected" that the OC would be larger at the RS site than the other sites since the road the measurements were made by had limited truck traffic and, in general, the OC/EC for this site is larger than I would expect for very fresh gasoline-vehicle emissions.

Regarding the first part, the southeastern US has a high regional OC level so localized emissions tend to be small relative to this background. Comparing absolute concentrations at different sites, one must note that the measurements were not made simultaneously (as we have noted throughout the paper).

The OC/EC ratios were somewhat larger because no online thermal EC data were available and we used optical EC instead, which is smaller than thermal EC and resulted in a larger OC/EC ratio. In the modified version we've changed the optical EC to thermal EC from MOUDI measurement, and the OC/EC ratio also changed accordingly (Please also refer to the table in the response to a previous comment).

Section 3.3.1: The authors cite Bond and Bergstrom (listed actually as Bond and Bertram) as the source for their EC refractive indices for use in Mie theory calculations. However, if they read this paper closely they will realize that it is well known that spherical particle Mie theory calculations using this refractive index will substantially underestimate the MAC (see Fig. 9 in B&B). Given this, it is somewhat surprising that the agreement between the calculated and observed "BC" absorption is so good. At the same time, though, I am still somewhat confused as to how exactly the "EC" was determined. Is this thermal EC or optical EC? Table 1 suggests optical EC, in which case some MAC must have been applied to determine the EC concentration. What MAC was used to derive optical EC and how does this compare with the calculated MAC?

The citation has been changed to "Bond and Bergstrom".

MOUDI EC were determined using the NIOSH protocol and therefore is thermal EC. The Mie calculation was then applied to size-resolved EC, and the estimated EC absorption was compared to scattering-corrected Aethalometer absorption. Figure 6 shows that at JST, the estimated EC absorption is ~30% lower than Aethalometer absorption, which agrees with Bond and Bergstrom that Mie underestimates MAC. However, the two absorption measurements agreed at YRK, which is probably because that Aethalometer is not sensitive enough to detect the enhancement of coating, given the primary emission at YRK is fairly low.

The authors are also doing calculations assuming spherical particles in the limit where the spherical particle assumption may not apply. In fact, the assumption of an AAE = 1 is inconsistent with their calculations: an AAE = 1 for BC only comes about from consideration of absorption by very small particles (less than 50 nm or so). Above this, the AAE deviates from 1. Further, the calculated MAC falls off dramatically with particle size above some critical (wavelength-dependent) size. This does not happen if absorption is governed by the spherules that make up BC as opposed to the entire particle. Note that all of this will impact the BrC/BC comparison in section 3.3.5 because the particular behavior of BC comes about because it is so strongly absorbing; more weakly absorbing species show different behavior.

We did not simply apply the AAE=1 assumption to BC absorption, but instead calculated BC absorption using Mie theory at different wavelengths. Absorption Ångstrom exponents for EC_bap at JST, YRK and RS, were 0.81±0.02, 0.83±0.02 and 0.79±0.04, respectively.

We are aware that the Mie calculation might under-estimate MAC, which may be explained by the interactions between the spherules that form the aggregate. Given that the agreement between Mie-calculated EC absorption and scattering corrected Aethalometer absorption was well within the uncertainty range, we believe the assumptions applying Mie theory are reasonable.

Section 3.3.4: I must admit that I don't really follow this section. If I understand this correctly, the k values used in the calculations come from analysis of the water/MeOH extracts, which are then used to calculate the absorption by the water/MeOH extracts. This, to me, seems like a circular argument and I am in fact somewhat surprised that the level of agreement is not better in Fig. 5. I believe that some clarification regarding the potential circularity of the analysis at this point is needed. At the same time, I do sort of understand that the authors are making the point that particles can absorb more than the equivalent solution. Another way to state this is simply that the imaginary refractive index is a more descriptive metric than the MAC in that it is independent of particle shape/size.

The analysis is not circular. k comes from the direct measurement of the particle chromophores in the water/MeOH extracts. This solution absorption cannot be directly applied to the atmosphere since it does not include information on how these chromophores are distributed with particle size. (The interaction of light with particles depends on particle size and particle composition). To account for size, we used the size resolved MOUDI data and Mie theory (under a number of assumptions that are stated). The slope shown in Fig.5 is because the chromophores light absorption for a solution (absorption by molecules) is different than if the chromophores are suspended in an aerosol phase.

Section 3.3.5: IF the authors were to add the calculated EC and calculated MeOH BrC absorption together at the different wavelengths, and then were to calculate an AAE based on this combination, what would one obtain? A value that makes sense in the context of the Aeth measurements? I ask because the Aeth AAE seems quite small if BrC really is contributing 30-40% of the total absorption at 350 nm at at least one of the sites. Also, what is the AAE for the calculated EC absorption? I ask because the calculations seem to be done using relatively "large" BC particles, which should give very low AAE values (much lower than 1). For example, 500 nm diameter BC particles would give an AAE closer to -0.3 than to 1. To the extent that the calculated EC absorption is dominated by where the mass is (large particles) I am having some difficulty understanding why the calculated EC absorption visually looks as if there is an AAE close to 1: : :unless smaller particles were used in the calculations. Perhaps I am just missing something in how the calculations are being done or in what exactly is being shown in Fig. 6?

If the calculated EC and calculated MeOH BrnC absorption were added up as the total absorption, the calculated AAE would be 1.3-1.4, which deviates from the ~1 AAE measured by Aethalometer. We do not know the cause of this problem. With this in mind, we use the sum of EC and BrnC absorption as the total absorption to estimate the BrnC fraction. This give a conservative estimate of the BrC contribution since we find the aethalometer tends to under-measure absorption.

As stated in a responses above, the Absorption Ångstrom exponents for EC_bap at JST, YRK and RS, were 0.81±0.02, 0.83±0.02 and 0.79±0.04, respectively. The Mie calculation was done based on size resolution of MOUDI samples over the fine particle size-range, assuming one aerosol size (the geometric mean diameter) for a given stage. It should be noted that the Mie calculation used the Stokes diameter, which is different than the aerodynamic diameter shown in the size distribution plot.