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 #2:

This paper reported the light absorption properties of BC and OC at three sites (urban, rural, and road side) at Atlanta, USA. Evaluations of contributions of light absorption by brown carbon to total light absorption at UV and shorter visible wavelength as well as understandings of possible sources of brown carbons are important to estimate the impacts of aerosols on climate change and photochemistry in the atmosphere. This paper provides the valuable information on these issues under conditions where impact of biomass burning was small. Especially, the findings of possible contributions of vehicle related emission to the light absorbing organics are interesting. The manuscript is logically written and the topics and results are relevant to this journal. I therefore recommend publication once the comments and questions below are addressed.

Major Comments:

1) In this work, particles were sampled by a MOUDI for 2-3 days before analysis. How the authors consider the artifacts due to possible changes in concentrations and chemical compositions of organics during the sampling? Especially removal of semi-volatile organic compositions from aerosol phase and oxidation and nitrifications of reactions of organic aerosol with gas phase compositions such as O3 and NOx on the filter may influence to the results in this work.

Chemical changes due to the long sampling time, including reactions on MOUDI impactor filters and the removal of semi-volatile organic components, are possible, but do not seem to change overall bulk properties since there was good agreement with online data (Figure 2), which are less prone to these artifacts. However, there was no online-data for methanol extractable brown carbon, and since our analysis suggested that light-absorbing materials became less water-soluble with age, artifacts may exist for the methanol extracts.

2) Page 18239, line 10: =>For the data obtained by the Sunset OCEC analyzer, the authors used a specific split time (150 sec). Because the correct split time changed with the concentrations of OC and EC ratio as well as chemical composition of OC, the specific split time lead to errors in the obtained EC and OC. How did the authors estimate the errors?

Due to the uneven loading of aerosols onto MOUDI filters, the laser correction in OC/EC split could not be applied and thus a fixed split time, determined from simultaneously sampled Hi-Vol filters (evenly loaded), was applied. The average±stdev of the split time determined from Hi-Vol filters were 500±15.4. The estimated uncertainty related with the split time is 1.3% for OC and 4.5% for EC, which is already included in the uncertainty of reported OC/EC concentrations. The text (page 9, Lines 190) and Table 2 was modified to include more details in the uncertainty estimation.

3) Page 18240, line 26: => In the offline measurements of OCEC, the authors used the ther-EC. Why did the authors use the opt-EC in the online measurements instead of ther-EC?

Operational problems during the sampling period resulted in loss of online thermal EC data. We've changed the data in Table 1 from online measurements of optical EC to thermal EC from MOUDI measurements, and have stated it clearly.

4) Page 18241, line 9: => The authors reported the insoluble particles larger than 0.2 micron were removed from PILS-generated liquid sample. How did the authors check the contributions of EC-containing particles smaller than 0.2 micron to light absorption measurements?

As in our response to comment #2 of referee #1, although pure BC is not soluble in either water or methanol, it is possible that when BC is coated with some soluble organics, or for aged BC particles as they become slightly hydrophilic, a small fraction of BC could be dissolved. In our study, the contribution of BC, or EC-containing particles, is examined by the absorbance at 700 nm in the solution spectrum in both water and methanol extracts. All MOUDI samples showed a zero-absorbance at 700 nm, indicating the contributions are negligible. Furthermore, the spectra never show evidence of pure absorbing particles (i.e., uniform absorption over all wavelengths).

5) Page 18249, lines 6-9: "This may suggest that the chromophores become less water soluble with age, possibly due to chemical aging. However, because measurements at various sites were not made simultaneously, these contrasts are somewhat uncertain." => I think the mixing with other organic components with less water soluble properties may also contribute to the difference in the greater fraction of water soluble chromophores at the RS site.

It is possible that some non-absorbing organic components also become less water-soluble with age, and mix with chromophores to result in to a greater fraction of water-insoluble at JST and YRK. However, we measure BrnC in solutions, and the mixing in aerosols would not actually impact the determination of BrC by our method.

6) Page 18250, lines 3-6: "The results suggest that it is not necessary to apply more complex internal mixtures and/or core/shell assumptions for particles to accurately estimate the light absorption coefficient based on size distributions measurements of EC at longer visible wavelengths." => I think the results may also be explained if Aethalometer could not correctly detect enhancement of BC light absorption due to coating.

That is possible, but we are not aware of studies that show this. Indeed MAC's based on aethalometer measurements are often observed to be greater than what is expected for pure BC and thought to be due to coatings.

7) Page 18250, lines 13-14: "leads to an overall uncertainty in bap,EC (and bap,H2O and bap,MeOH) of 30%" => I think the uncertainties in bap,H2O and bap,MeOH are different with bap,EC, because the uncertainties in particle density, refractive index, and mass of WSOC and OC are different with those of EC.

The uncertainty of 30% shown here is the relative uncertainty associated with variables used in the Mie calculation. This includes: that aerosols in each stage were assumed to have the same size, the selection of organic mass vs. particle mass, real part of the refractive index, and sensitivity to density (ρ), both in Mie calculation and conversion from MOUDI aerodynamic diameters to physical sizes. The details are provided in footnote #8 below the table. Uncertainties related to different variables in the Mie calculation, such as the mass of WSOC and OC, are included in the estimation of b_{ap_EC}, $b_{ap,H2O}$ and $b_{ap,MeOH}$, with details given in footnotes 10-12.

8) Section 3.2.1 and 3.3.3 (Fig.3 and Table 4): => At JST and YRK sites, light absorptions (k-values) of OC extracted by MeOH were larger than those of OC extracted by water (Table 4). The results indicate the significant contributions of water insoluble organic compounds to the light absorption. However, the size distributions of light absorption of MeOH extracted OC were similar with size distributions of WSOC rather than OC (Figure 3). How do the authors explain the results?

This is an interesting point and can be explained by the light absorbing components comprising only a small mass fraction of the WSOC or OC, and at least some components of the organic aerosol are externally mixed. For example, at the Roadside site where the OC geometric mean diameter is smaller than that for the methanol soluble chromophores, it would suggest a large fraction of the fresh smaller-size particle emissions are not light absorbing, whereas larger particles from this source, although not as prevalent, contain most of the absorbing material.

9) Page 18256, lines 12-23: => I agree with the suggestions of authors about the needs of further studies. I think the ability of Aethalometer to measure light absorption of brown carbon may also be depend on the phase (liquid, semi-liquid, solid) of brown carbon.

We agree that the ability of an Aethalometer to detect brown carbon might be limited.

Minor Comments:

1) Page 18238, line 22: "roughly 48 h" => I think the sampling periods listed in Table 1 seem to be roughly "60 h".

The text has been changed (page 8, line 169).

2) Page 18244, line 6: "normalizing mass concentrations" => What does the "normalizing" means?

“Normalizing” has no physical meaning in here, has been deleted (Page 15, Line 317, also applies to page 16, Line 327, and page 40, line 902).

3) Table 5: => Explanation of "Noise" should be added.

The “Noise” should be “NAN”. Text has been changed.

4) Figure 4, caption: => "log Dp" may be "log lambda"

The figure caption has been modified.

5) Figure 6, caption: => "Table 1" may be "Table 2"

The text has been changed.