

Interactive comment on “Size-resolved measurements of brown carbon and estimates of their contribution to ambient fine particle light absorption based on water and methanol extracts” by J. Liu et al.

Anonymous Referee #2

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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

C6561

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 NO_x on the filter may influence to the results in this work.
- 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?
- 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?
- 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?
- 5) Page 18249, lines 6-9: "This may suggest that the chromophores become less watersoluble 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

C6562

chromophores at the RS site.

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.

7) Page 18250, lines 13-14: "leads to an overall uncertainty in $b_{ap,EC}$ (and b_{ap,H_2O} and $b_{ap,MeOH}$) of 30 %" => I think the uncertainties in b_{ap,H_2O} and $b_{ap,MeOH}$ are different with $b_{ap,EC}$, because the uncertainties in particle density, refractive index, and mass of WSOC and OC are different with those of EC.

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?

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.

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".

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

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

C6563

4) Figure 4, caption: => "log D_p " may be "log λ "

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

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C6564