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Interactive comment on "Brown carbon and water-soluble organic aerosols over the southeastern United States" by A. Hecobian et al.

Anonymous Referee #2

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In this paper, the authors investigated the light absorption of water soluble organic carbon (WSOC) both for filter extracts and through online measurements. Three main sources for light-absorbing WSOC were indentified: biomass burning (mainly in winter), secondary organic aerosol formation (mainly in summer) and mobile source emissions (mainly in urban areas). The manuscript has provided information on light absorption of ambient WSOC based on a large dataset and reasonable analysis techniques. The manuscript raises the issue of SOA as a source of light-absorbing WSOC besides biomass burning, which pushes forward what is known about light-absorbing OC in the ambient atmosphere. However, the derivation of one variable - absorption coefficient Abs_{Lambda} has to be verified and corresponding results may need to be changed, although this does not affect the final conclusion. Overall, I recommend the publication of this paper after consideration of the specific comments as stated below.

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Page 7602, I suggest the authors modify the title a little bit. As stated in the last sentence of Introduction, this paper mainly focuses on discussing light absorption of water-soluble organic aerosols (WSOC). Brown carbon was then inferred from the results in the context. Regarding the current title, it seems that brown carbon and WSOC are two separate categories investigated in this paper.

Page 7604, line 23, this sentence is confusing to me. Are the compounds with conjugated systems most absorbing among all types of chemical compounds? Or are they most absorbing near-UV region among the whole UV-vis spectrum?

Page 7604, line 24, incomplete combustion is not just smoldering, and it also includes flaming which produces black carbon.

Page 7606, line 6, 0.6N HCl provides a highly acidic environment. I noticed that the authors also used Milli-Q water to clean the system, which would possibly remove the acids. Did the authors measure the pH value of the system after the cleaning procedure? Will this affect the absorption measurement?

Page 7607, line 1, the filters had been stored at below freezing temperature for approximately one year prior to analysis. To a personal experience, one year is a long period that the properties (such as absorption or mass) of the materials on the filters change to some extent even the filters are stored below freezing temperature. Did the authors investigate the effect of their results caused by the long-term storage?

Page 7610, Eq (1), I think the subscript for C in the term after the third "=" should be i instead of I.

Page 7610, line 17, I suggest the authors just pick one definition of epsilon either molar absorption efficiency or mass absorption efficiency, and stick to it.

Page 7610, line 21, it is reasonable to correct absorbance by removing the absorbance from pure water blanks. Could the authors state how much absorbance is from pure water blanks? How much percentage does the absorbance of pure water contribute to

the total absorbance of a sample?

Page 7611, Eq. 2, I tried to reproduce the derivation of this equation and it seemed that the authors derived this equation by assuming the same mass absorption efficiency for OC in both bulk liquid phase and particulate phase, but they are actually different. I suggest the authors only use the absorption coefficient obtained directly from UV-vis spectrophotometer measurement to avoid the confusion. This will also help the discussion in Sect. 3.2.1 since Abs/WSOC will be the actual absorption per mass of WSOC, indicating how absorbing WSOC is.

Page 7612, line 12, could the authors explain or provide some citations on how the wavelength dependence can be used to infer the imaginary refractive index?

Page 7613, line 6-7, Angstrom exponents were determined by applying linear regression over different ranges for different samples (e.g., 330-600nm for South DeKalb site; 330-475nm for levoglucosan less than 50ng/m3 for Yorkville site). I would suggest the authors to determine Angstrom exponents over the same range for all the samples. In that way, will the conclusion of similar Angstrom exponent (6-8) still be held?

Page 7613, line 18, the authors should add some citations on "biomass burning is a significant source for brown carbon".

Page 7614, line 9, the authors stated that the classification of biomass burning and nonbiomass burning-influenced periods using levoglucosan concentrations of 50ng/m3 was somewhat arbitrary, but this classification has still been used throughout the paper. I would expect to see more explanation or validation on the use of this dividing point.

Page 7618, line 12, could the authors explain why the ratio of WSOC/CO or later in line 22 Abs_{365} /CO are used. Is there a fairly constant OC emission rate in a diurnal cycle? What can these two ratio account for?

The authors used several phrases such as "cool months", "colder periods", "colder months", "winter", "warmer seasons", and "summer" to indicate certain time periods

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throughout the manuscript. It would be wise to use same terms for a certain period.

The authors should pay attention to some terms used to describe optical properties: Page 7610, line 26 and page 7611, line 4, "absorbance coefficient" should be "absorption coefficient". Page 7612, line 6, "absorption cross-section" should be "mass absorption cross-section".

Page 7636, Figure 5, in the caption, A_{365} should be Abs_{365} .

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 7601, 2010.