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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Dynamic light absorption of biomass burning organic carbon photochemically aged under natural sunlight" *by* M. Zhong and M. Jang

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## Response to Referee 1

We thank Reviewer 1 for valuable comments.

Referee comments:

This manuscript presents nicely the evolution of OC light absorbance as a result of photochemical oxidation of biomass burning throughout the day cycle, a topic of great interest in the field of biomass burning optical properties. It combines laboratory and field measurements, and discusses the effect of RH and NOx on the absorption of

wood smoke OC. The manuscript is well written, and the findings contribute to a better understanding of the different stages of photochemical oxidation on light absorption of wood smoke OC. Therefore, the manuscript is appropriate for ACP after addressing the comments below:

**1. General comments:**

1.1. The authors specifically note that in order to focus on wood burning OC with minimal influence by BC, wood smoke was produced from the smoldering-phase burning; however a short explanation of the characteristics of the smoldering phase is missing (i.e the biomass burning is generally divided into three basic combustion phases: ignition, flaming and smoldering which are all characterized differently). The authors should be more careful comparing the current study to previous studies which may be conducted at different combustion stages (smoldering vs flaming, etc).

**Response:**

A brief description of smoldering combustion has been added into Section 1, paragraph 6.

“The smoldering phase is characterized by a flameless form of combustion, producing low fractions of black carbon (2-5% by mass) in aerosol phase (Reid et al., 2005)."

1.2. formation of SOA highly depends on the combustion stage, temperature and oxygen etc.. The authors state in the Results and discussion P.20791 line 15 : “Saleh et al. (2013) have explained that the increment in aged OC absorption is caused by the formation of SOA, which absorbs more than fresh POA in the short wavelength visible and near-UV regions". And use this explanation in the conclusions to explain the increase in the absorption. The authors should provide evidence that the increase in the absorption was a result SOA formation. e.g SMPS measurements showing an increase in the number of particles , or alternately introduce only gases from the burning to the chamber (filtering the POA) and measure the absorption.



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**Response:**

We measured number concentrations of particles using SMPS. However, number concentrations did not significantly change because SOA formation is predominately controlled by the condensation of semivolatile compounds onto preexisting POA. We have monitored SOA mass using OC data which were corrected for particle wall losses and chamber dilution. As shown in Fig. R1 below, OC gradually increased with time. For example, OC increased by 50% for 10-hour photooxidation. POA mass will more likely decrease as temperature increases. Hence the actual SOA mass increase might be greater than the OC data in Fig. R1.

**Fig. R1.** Comparison of OC mass concentrations with wall-loss correction and without correction

In order to measure SOA only light absorption, SOA was also produced through the photooxidation of wood smoke gas using the outdoor chamber (particles were removed using the specially designed filtering system). However, the loss of gaseous semivolatile compounds to the filter and the injection line was significant. Consequently, SOA yields from the gas only experiment were low compared to those from wood smoke without filtering system. This result indicates that gas only experiment cannot produce the similar SOA which formed from photooxidation of actual wood smoke. Hence, further chamber experiments of this study were conducted using wood smoke without filtering particles matter.

1.3. P.20782 line 1: The authors explain the decreases in $MAC_{OC}$ by the bleaching effect. This sounds like a reasonable explanation; however, the authors need to conduct a control experiment to measure the absorption on a filter containing the POA and SOA in dark condition (after the same number of hours as the original experiment)



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**Response:**

We agree with the reviewer. In order to prove that the color change of OC is mainly via photooxidation processes under the sunlight, an additional outdoor chamber experiment was conducted at night (Sept.27, 2013). Fig. R2 that has been added to the revised supplementary material shows the $MAC_{OC}$ of wood smoke aerosol stood in the outdoor chamber during nighttime. No significant change appears in $MAC_{OC}$ within the error range.

**Fig. R2.** Mass absorption cross section (550nm) of wood smoke OC collected at night time (Sept. 27-28, 2013).

1.4. The authors state in the ï£¡ï£¡Light absorption of ambient aerosolï£¡ï£¡ section p.20789 that the wildfire emission is the main source of the ambient aerosols in UF area during the examined episode based on the elevated OC values, the authors should also check the metrological condition and the back traceries to insure that this is indeed the case during several hours of measurements.

**Response:**

Based on the satellite information (http://www.nrlmry.navy.mil/aerosol/), the scale of optical depth due to smoke concentrations during the County Line Wildfire event was significantly high in Gainesville area, as shown in Fig. R3. Before the wildfire event, no smoke was detected by satellite.

**Fig. R3.** Flambe/MODIS/Cloud/NAAPS data for 09, April, 2012 (County Line Wildfire). The lighter in blue color, the higher the smoke concentration in ambient.



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In addition to the satellite data, meteorological data (http://www.wunderground.com/history/airport/KGNV/) shows that the frequency of the wind directions in Gainesville at nighttime of April 08, 2012 was North and Northwest and the wind was calm during the daytime on April 09, 2012, indicating that the city of Gainesville was significantly affected by the wildfire.

1.5. Also and more importantly, the authors made an assumption that the change in the light absorbance was as a result of photooxidation process. However, an alternative explanation could be changes in the burning phase or ambient temperature (changes the partitioning of semivolatiles) and RH. The authors need to discuss these possibilities

**Response:**

We measured the EC fraction of the total carbon mass using an OC/EC analyzer during the wood fire event. The measured EC fraction ranged between 3.6% and 5%. In general, the EC content during flaming phase is higher than smoldering and varies by over a factor of 5 (Reid et al., 2005). Thus, the wood smoke observed in Gainesville area was more likely produced under smoldering condition.

We agree that the chemical composition of wood smoke aerosol can be influenced by ambient temperature and RH. Our outdoor chamber experiments were also conducted under ambient conditions using nature sunlight, ambient temperature and ambient humidity. The temperature increase in the morning time due to sunrise may result in evaporation of non-colored primary organic compounds. Such condition should decrease the amount of OC and increase light absorption. However, OC significantly increased as shown in Fig. R1 (Response to comment 1.2). This result strongly suggests that the increase of light absorption of wood smoke aerosol mainly originates from either SOA or particle phase reactions through photochemical processes.

Reference: J. S. Reid, R. Koppmann, T. F. Eck, and D. P. Eleuterio, A review of biomass burning emissions part II: intensive physical properties of biomass burning particles, Atmos. Chem. Phys., 5, 799ï£¡C825, 2002.



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**2. Specific comments:**

2.1. P.20789 and throughout the paper: Please be consistent using either BC or EC

**Response:**

BC will be used for the discussion of aerosol light absorption. EC was used to describe data from an OC/EC analyzer.

2.2. P.20792 line 13: Please define ï£¡ï£¡initial total $MAC_{OC}$"

**Response:**

The sentence has been rewritten and reads now (Section 3.3, paragraph 1):

ï£¡ï£¡The relative $MAC_{OC}$ was obtained by normalizing the total $MAC_{OC}$ at a certain time by the total $MAC_{OC}$ of fresh wood smoke.ï£¡ï£¡

2.3. P.20793 “Effect of $NO_x$ on light absorption of OC ", Please add a short discussion about the atmospheric relevance the $NO_x$ concentration use in this measurement

**Response:**

A short discussion has been added into Section 3.4, paragraph 1.

"Controlled dual chamber experiments were also conducted to study the influence of $NO_x$ on $MAC_{OC}$ of wood aerosols: low $NO_x$ (16 ppb) vs. high $NO_x$ (108 ppb) on 30 October, 2012 and low $NO_x$ (16 ppb) vs. middle $NO_x$ (43 ppb) on 11 October, 2012. The low $NO_x$ condition indicates the $NO_x$ concentration originating from wood burning under the smoldering condition. Both the middle $NO_x$ and high $NO_x$ conditions simulate the photooxidation of the wood smoke mixed with the air in urban areas. In the United States, the average urban $NO_x$ concentrations vary between 18ppb and 114 ppb (Baugues, 1986)."

2.4. P. 20795: section 3.5.4 “Hygroscopic properties of wood burning aerosol" Please compare the results to the relevant literature



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**Response:**

We compared our results to that known in literature and reads now (Section 3.5.4, paragraph 1):

“Martin et al. (2013) has also reported the decline of hygroscopicity in photochemically aged wood smoke aerosol by measuring the growth factor of fresh and aged wood smoke particles using a tandem differential mobility analyzer."

2.5. Fig.2: Please add the RH values to the figure legend and add one point before the Sunrise. Fig.3: Please add the $NO_x$ values to the figure legend, add one point before the sunrise

**Response:**

The detailed RH and $NO_x$ values have been added to the legend in Figures 2 and 3, respectively.

The data points shown in Figures 2 and 3 are midpoints for the 20-30 minute sampling duration. The first sample was collected just before or near sunrise where the sunlight intensity is less than 1 watt/m$^2$. In general, the sunlight intensity in midday is about 32-35 watt/m$^2$. Thus, the first point reasonably represents the data without photooxidation.

**3. Technical correction:**

3.1. P.20784 Line1: Define POA

**Response:**

POA has been defined in Abstract.

3.2. p.20785: Line1: Add reference after ï£¡ï£¡solar radiationï£¡ï£¡ Line, Change ï£¡ï£¡blankï£¡ï£¡ to ï£¡ï£¡blackï£¡ï£¡

**Response:**

References have been added and typo has been corrected (Section1, paragraph 1).



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3.3. p.20796 Line 6: Define SVOC

**Response:**

This has been corrected ( Section 4, paragraph 1).

3.4. P. 20794 Line 11: Correct Figs. To Figures

**Response:**

This has been corrected (Section 3.5.1, paragraph 1).

3.5. P.20794 Line 3: Correct Supplement to Supplementary material

**Response:**

This has been corrected (Section 3.5.1, paragraph 1).

3.6. Fig5. : Please be consistent with the colors (e.g. Fresh- blue, Aged- red, in both panels (a and b)

**Response:**

Figure 5 has been updated.



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