Author response to referee's comments for acp-2018-159: Production of particulate brown carbon during atmospheric aging of wood-burning emissions by Kumar et al.

Dear editor,

We thank both reviewers for their constructive comments, which significantly enhanced the quality of our manuscript. Below, we provide a point-by-point response (regular typeset) to the comments (blue font) of both reviewers. The modifications made to the manuscript are in grey font, indented and *italicized*. Please note that all references to line numbers are to the submitted manuscript (the ACPD file) and not the revised manuscript.

Anonymous referee 1.

GENERAL COMMENT

The paper presents an analysis of black carbon (BC) and organic aerosol absorption properties upon aging. The experiments were done in a laboratory using a smog chamber where aerosols were exposed to OH radical and UV radiation. Several optical and chemical properties were measured online and filter samples were collected to be analyzed later offline by a multiple wavelength absorbance measurement technique and an EC/OC analyzer. Methanol and water filter extracts were also analyzed and absorbance was measured. The data is very valuable and the paper is well written and presented. I would recommend its publication after addressing the comments I present below.

My main concerns are related to the following aspects:

- The offline techniques used in this study suffer of different artifacts and they are not sufficiently discussed in the manuscript.

We had carefully considered such artifacts, but could have made more reference to them in the manuscript. We consider the reviewer to be referring to unquantifiable uncertainties. Based on both reviewers' comments, we have added a section discussing quantifiable and unquantifiable uncertainties in the method section. This section reads as follows:

> *Uncertainty analysis. It is important to draw a clear distinction between uncertainties related to measurement precision and accuracy and those related with experimental variability. In this section we discuss the quantifiable and unquantifiable uncertainties related with the different measurements. In the result section, we will present our confidence levels on the average parameters determined based on the experimental variability, which we judge to be the main source of variance in the data.*

1

➔ *Quantifiable uncertainties:*

The estimated uncertainty in the AMS-derived OA mass concentrations is ~25%, which includes both potential biases and precision. This estimate is based on the variation in the AMS calibration factors and estimated uncertainties in the SMPS used for the AMS calibration (Bruns et al., 2015, 2016). Uncertainties related to particle transmission efficiency in the AMS are considered negligible for the particles sampled here (Liu et al., 2007), whose volume size distribution falls within the range transmitted efficiently by the AMS aerodynamic lens (see Fig. S4). The bounce-related collection efficiency (CE) of the AMS was concluded to be unity for wood-burning OA in the literature reviewed by Corbin et al. (2015b; in their Section S1.2). For the present data, the comparison between the SMPS mass (predicted from fitted volume distributions using a density of 1.5 g cm^{-3}) and the total PM predicted as AMS-OA+eBC, suggest a CE value between 0.7 and 1.0 (19% relative uncertainty), consistent with average literature values and the uncertainties estimates. The uncertainty in EC mass concentration, estimated from measurement repeats based on the EUSAAR2 protocol only, is within 7% in our case. The precision uncertainty in the aethalometer attenuation measurements was estimated as 15 Mm^{-1} based on the standard deviation of its signals prior to aerosol being injected into the smog chamber. The MWAA data have an estimated noise level and precision of 12 /Mm and 10% respectively, and these uncertainties have been added in quadrature to provide the overall uncertainties shown, for example, as error bars in Fig. 1 below. To compare the MWAA and aethalometer measurements, we determined $b_{abs,MWAA,880nm}$ by extrapolating the absorption coefficients measured at 850 nm to 880 nm using an α-value determined from the ratio between the absorption coefficients at 850 nm and 635nm. The uncertainty associated with this extrapolation is considered negligible relative to the overall MWAA uncertainty.

➔ *Possible unquantified uncertainties:*

There are significant uncertainties in the measurement of aerosol absorption using filter-based techniques (e.g., Collaud Coen et al., 2010). Here, we have used MWAA measurements as a reference to scale the aethalometer data, using a single C value. The correction factor C, which accounts for scattering effects within the filter matrix (Drinovec et al., 2015), may depend on the aerosol sample (Collaud Coen et al., 2010). In this study, we evaluated the variability in this factor for our primary and aged samples, by directly comparing the aethalometer to MWAA measurements, as discussed below. The MWAA has been previously validated against a polar nephelometer and a MAAP (Massabo et al., 2013), which, in turn, has been validated against numerous in situ methods (e.g., Slowik et al., 2007). The excellent correlation between MWAA and EC in our study (discussed below) supports the high confidence in the MWAA filter based absorption measurements conducted here. Another significant source of uncertainty in filter-based absorption measurements is the

possible sorption (or evaporation) of volatile organics on (or from) the filter material. This may lead to an overestimation (or underestimation) of OA absorption. However, we have minimized sorption artefacts by utilizing a charcoal denuder. We have obtained an excellent correlation between OA absorption measurements derived from the MWAA-calibrated aethalometer and from quartz filter samples (see discussion below, Fig. 6 in the main text and S13 in the supplementary information). Although both of these techniques involved filter sampling, their sampling timescale is an order of magnitude different, and a difference is therefore expected if sorption (or evaporation) caused a substantial bias in our results. We therefore conclude that it is unlikely that artifacts associated with filter sampling have biased the absorption measurements. Finally, uncertainties related to pyrolysis during thermo-optical analysis may bias EC measurements. Such uncertainties arise from unstable organic compounds, and can be significant for biomass-burning samples, leading to biases on the order of 20% for EC (e.g. Schauer et al., 2003; Yang and Yu., 2007). To minimize these biases we applied the EUSAAR2 protocol. The optical properties of such organics are generally different from BC; therefore, the excellent correlation between MWAA and EC data in Fig. 1A suggest that pyrolysis effects were not a major source of uncertainty in our data set.

We have added the following abbreviation to the corrected text:

L146. *multi-angle absorption photometer (MAAP, Petzold and Schönlinner, 2004).*

We have also expanded the text in some places to reflect the considerations presented above, as shown in the response to the next comment.

New references added:

Schauer, J. J., Mader, B. T., Deminter, J. T., Heidemann, G., Bae, M. S., Seinfeld, J. H., Flagan, R. C., Cary, R. A., Smith, D., Huebert, B. J., Bertram, T., Howell, S., Kline, J. T., Quinn, P., Bates, T., Turpin, B., Lim, H. J., Yu, J. Z., Yang, H., and Keywood, M. D.: ACE-Asia intercomparison of a thermaloptical method for the determination of particle-phase organic and elemental carbon, Environ. Sci. Technol., 37, 993–1001, https://doi.org/10.1021/es020622f, 2003.

Slowik, J. G., E. S. Cross, J.-H. Han, P. Davidovits, T. B. Onasch, J. T. Jayne, L. R. Williams, M. R. Canagaratna, D. R. Worsnop, R. K. Chakrabarty, H. Moosmüller, W. P. Arnott, J. P. Schwarz, R. S. Gao, D. W. Fahey, G. L. Kok and A. Petzold, An inter-comparison of instruments measuring black carbon content of soot particles, Aerosol Sci. Technol. 41, 3, 295-314, 2007.

Collaud Coen, M., Weingartner, E., Apituley, A., Ceburnis, D., Fierz-Schmidhauser, R., Flentje, H., Henzing, J. S., Jennings, S. G., Moerman, M., Petzold, A., Schmid, O., and Baltensperger, U.: Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms, Atmos. Meas. Tech., 3, 457-474, https://doi.org/10.5194/amt-3-457-2010, 2010.

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122   *Yang, H. and Yu, J. Z.: Uncertainties in charring correction in the analysis of*
123   *elemental and organic carbon in atmospheric particles by thermal/optical methods,*
124   *Environ. Sci. Technol., 36 (23), 5199–5204, 2002.*

125

126   - In this study the BC particles were not observed to be coated with other kind of particles
127   (i.e., no internal mixing but external mixing was observed). However, ambient studies have
128   shown that BC particles in the atmosphere are usually coated and this coating causes an
129   enhancement of BC absorption. Given that, how representative is this study of "atmospheric
130   aging"? Were the experiments not long enough to "age" the BC particles?

131   The reviewer raised two points, to which we reply separately.

132   The reviewer questions how representative our study was of atmospheric aging. The
133   emissions studied here are representative of flaming wood in stoves typically used in Western
134   Europe, while aging is equivalent to ~2 days of OH-driven photochemistry, under
135   atmospheric winter day time conditions in the mid-latitude.

136   The reviewer questions the reasons for the lack of lensing with aging, when such effect had
137   been observed in the field. We have clarified our language here. Our aim has been to assert
138   that our measurements are poorly represented by a pure core-shell conceptual model of
139   internal mixing. This assertion is based on our measured absorption coefficients, and we have
140   modified the text to explain this in more detail as quoted immediately below.

141   The AMS measurements showed that the amount of OA generated during aging was
142   substantial. Likewise, the SMPS showed a considerable growth of the primary particles with
143   aging. If BC and OA are naively treated as core-shell mixtures, an absorption enhancement of
144   ~1.8 would have been predicted, with an average increase in the coating mass by a factor of 3
145   (see Bond et al., 2006). However, our absorption-coefficient measurements in Fig. 1a showed
146   that we did not observe any absorption enhancement. Therefore, we do not conclude that "BC
147   particles were not coated" but rather than "the particles studied could not be represented by a
148   core–shell description of coatings that envelop the central BC core". The particles may be
149   internally mixed, but of a morphology more complex than core-shell − e.g. off-centered
150   coatings with complexities due to the aggregated morphology of BC, see e.g. the microscopy
151   images of biomass-burning particles by China et al. 2013. Modelling or even accurately
152   describing such morphologies is well beyond the scope of our experimental study. Current
153   literature reports for the lensing effect are conflicting, showing that absorption enhancements
154   upon significant BC coating can be less than 5% (Cappa et al., 2012) or as large as 150% (Liu
155   et al., 2015). Recent experimental work suggests that such discrepancies are related to the
156   complex black carbon morphology and a core–shell description does not adequately capture
157   mixed-BC optical properties and may considerably overestimate the observed absorption
158   values (Liu et al., 2017). As lensing effect was negligible in our case, we have assumed that
159   the aerosol optically behaves as an external mixture between BC and BrC. We note that while
160   this assumption is important for estimating the BC absorption, the BrC absorption is not very

161 sensitive to the assumed morphology. Based on both reviewers comments and to avoid
162 confusion we have modified the Results section 4.1 as follows:

163

164 *Section 4.1*.

165 *We have independently determined the $MAC_{BC}(880nm)$ and the aethalometer C values*
166 *under our conditions, as follows. We determined $MAC_{BC}(880nm)$ from the regression*
167 *between the absorption coefficients at 880 nm obtained from the MWAA and the EC*
168 *mass measured by the Sunset analyzer (Fig. 1A). The slope of this regression may be*
169 *used to estimate the $MAC_{BC}(880nm)$, which we retrieved as 4.7 ± 0.3 $m^2 g^{-1}$ by an*
170 *uncertainty-weighted linear least-squares fit . The corresponding intercept was not*
171 *significantly different from zero (-3 $\pm$ 3 /Mm). Our $MAC_{BC}(880nm)$ is not statistically*
172 *significantly different from the value recommended by Bond et al., (2006) for*
173 *externally-mixed BC (extrapolating their $MAC_{BC}(550nm)$ to 880 nm by assuming α=1*
174 *provides $MAC_{BC}(880nm)= 4.7 \pm 0.7$ $m^2 g^{-1}$). The strong correlation between*
175 *$b_{abs,MWAA,880nm}$ and EC in Fig. 1A shows that $MAC_{BC}(880nm)$ did not vary with aging*
176 *during our study (see also Fig. S2-a). It also indicates that measurement artefacts for*
177 *both instruments were negligible, as the fundamental differences between the two*
178 *techniques mean that any artefacts are unlikely to be similar between them (charring*
179 *for EC vs. adsorption artefacts for MWAA). Our absorption coefficient measurements*
180 *also provide insights into particle mixing state in this study. Since a single MAC*
181 *adequately described our samples at all levels of aging (Fig. 1A and Fig. S2-a), in*
182 *spite of a factor of 3.3 average increase in the aerosol mass, our samples cannot be*
183 *adequately described by a core-shell Mie model. Such a core-shell model would*
184 *predict an absorption enhancement by a factor of ~1.8 (Bond et al., 2006) for the*
185 *observed OA mass increase with aging, which was not observed in our case. This*
186 *observation is also supported by the time resolved attenuation measurements at 880*
187 *nm using the aethalometer (Fig. S3), suggesting that little (<10%) to no increase in*
188 *the attenuation coefficients upon SOA formation. We emphasize that this conclusion*
189 *does not indicate that no internal mixing occurred, but rather that the simplified*
190 *concept of negligible mixing better describes our data than the equally simplified*
191 *concept of a core-shell description of coatings that completely envelop the central BC*
192 *core. This may be due to the complex morphology of internally-mixed BC, which has*
193 *been previously observed for wood burning particles (e.g., China et al., 2013; Liu et*
194 *al., 2015; Liu et al., 2017). It may also be related to the fact that OA and BC are*
195 *emitted during separate phases of combustion. OA rich particles are emitted during*
196 *the pre-flaming pyrolysis stage of combustion, whereas most BC is emitted during*
197 *flaming combustion (Corbin et al., 2015a, 2015b; Haslett et al., 2018; Heringa et al.,*
198 *2011). These two stages of combustion may coexist in different regions of the stove,*
199 *particularly during simulated real-world usage. As lensing effect was negligible in*
200 *our case, we have assumed that the aerosol optically behaves as an external mix*

between BC and BrC during Mie calculation (see section 3.4). We note that while this assumption is important for estimating the BC absorption, the conclusion drawn about the BrC absorption are not very sensitive to the assumed morphology.

We determined time-resolved wavelength-dependent absorption coefficients as follows. We used the aethalometer to obtain filter attenuation coefficients with high time resolution, which were then calibrated to obtain absorption coefficients by deriving the factor C (Equation 1) using the MWAA measurements of filter samples. C was obtained from an uncertainty-weighted linear least-squares fit as 3.0 ± 0.2 (Fig. 1B); the intercept of the fit was not significantly different from zero, within two standard deviations (-17 ± 14). A very strong correlation could be observed between MWAA and aethalometer (Fig. 1B), implying that C is independent of the type of the aerosol sampled (see also Fig. S2-B). Therefore, we used a single C value to obtain time-resolved wavelength-dependent absorption coefficients from the aethalometer attenuation measurements at the different wavelengths for primary and aged aerosols.

Note that the manufacturer's default values, which were not applied in our case, are 1.57 for C (using TFE-coated glass fiber filters) and 12.2 m^2 g^{-1} for σ_{ATN} at 880 nm (Gundel et al., 1984 , Drinovec et al., 2015). The C value determined here is larger than the manufacturer-default value for the AE33, resulting in smaller absorption coefficients. However, the calculated σATN at 880 nm (13.8 m^2 g^{-1}), which can be retrieved as the product of the C value and $MAC_{BC}(880nm)$ (Eq. (3)), is similar to the factory-default σATN. Therefore, our calibrated M_{eBC} concentrations, calculated from the attenuation coefficients using σATN (Eq. (2)), are similar to the factory-default M_{eBC}. We note that M_{eBC} has not been used for MAC_{OA} calculations, and is only used for the calculation of the mass fractions of BC and OA for display purposes (Fig. 2, 3, 7 and 8).
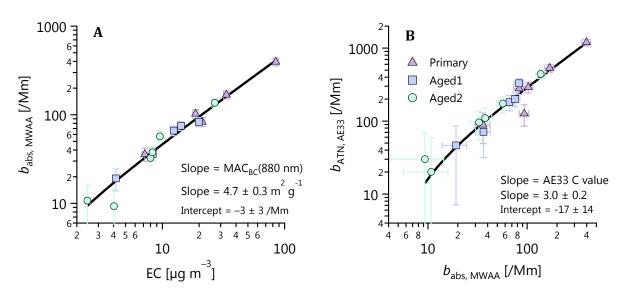
References:

China, S., Mazzoleni, C., Gorkowski, K., Aiken, A. C., and Dubey, M. K.: Morphology and mixing state of individual freshly emitted wildfire carbonaceous particles, Nat. Commun., 4, 1–7, doi:10.1038/ncomms-3122-2013, 2013.

Liu, S., Aiken, A. C., Gorkowski, K., Dubey, M. K., Cappa, C. D., Williams, L. R., Herndon, S. C., Massoli, P., Fortner, E. C., Chhabra, P. S., Brooks, W. A., Onasch, T. B., Jayne, J. T., Worsnop, D. R., China, S., Sharma, N., Mazzoleni, C., Xu, L., Ng, N. L., Liu, D., Allan, J. D., Lee, J. D., Fleming, Z. L., Mohr, C., Zotter, P., Szidat, S. and Prévôt, A. S. H.: Enhanced light absorption by mixed source black and brown carbon particles in UK winter, Nat. Commun., 8435, doi:10.1038/ncomms9435, 2015b.

Liu, D. T., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, D. V., Reddington, C. L., Kong, S. F., Williams, P. I., Ting, Y. C., Haslett, S., Taylor, J. W., Flynn, M. J., Morgan, W. T., McFiggans, G., Coe, H., and Allan, J. D.: Black-carbon
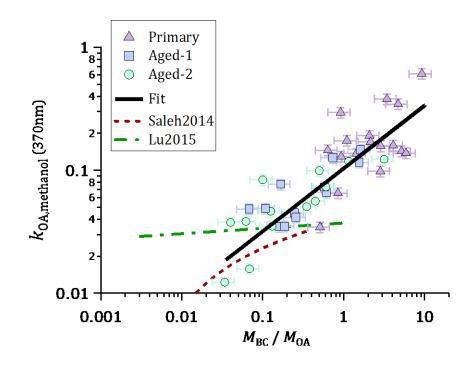
absorption enhancement in the atmosphere determined by particle mixing state, Nat. Geosci., 10, 184–188, doi:10.1038/ngeo2901, 2017.

Haslett, S. L.; Thomas, J. C.; Morgan, W. T.; Hadden, R.; Liu, D.; Allan, J. D.; Williams, P. I.; Keita, S.; Liousse, C. and Coe, H. Highly controlled, reproducible measurements of aerosol emissions from combustion of a common African biofuel source, Atmos. Chem. Phys, 385-403, doi:10.5194/acp-18-385-2018 , 2018.

We slightly modified Fig. 1A to reflect these changes. We included error bars in Fig. 1 which had been missing previously.

We also included error bars in Fig. 7:

SPECIFIC COMMENTS

1.   Lines 107-108: How can you guarantee the correction factor C is wavelength independent?

This is a good question which has often been neglected in the literature. Recently, Corbin et al. (2018), in their Section S3.2, presented a very detailed discussion and reanalysis of the wavelength dependence of the C-value.

In that publication, the authors described the wavelength dependence of the C-value as separated into a filter dependence and a scattering cross-sensitivity measurement, and presented four different arguments for its wavelength dependence being negligible:

1. They compared their wavelength-dependent absorption coefficient measurements with MWAA measurements (which do not rely on a C-value-like correction) and found good agreement between the two techniques. We have also verified our AE33 data with MWAA data in the present study.
2. They pointed out that the measured AAE would be biased by a wavelength-dependent C value, so that their measurements of an AAE of 1.0 for samples dominated by BC (in agreement with extensive literature) indicates a negligible wavelength dependence of C.
3. They combined measurements of aerosol SSA (ranging from 0.5 to 0.9) with size-dependent scattering cross-sensitivity measurements to quantitatively estimate the influence of scattering cross-sensitivity as negligible. SSA measurements were not available in our study, but our measured size distributions indicate that our particles were generally small enough that their conclusions can be extrapolated to our samples.
4. They described previous work where different filter materials were compared, with no significant effect on the wavelength dependence of the C value (Drinovec et al., 2015).

In conclusion, the C value is known to depend on the filter material but its wavelength dependence has been shown to be negligible for samples such as those studied in the manuscript presently under review.

We have updated the manuscript as follow:

L111. *As discussed in detail by Corbin et al. (2018), the wavelength-dependence of C can be expected to be negligible*

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References:

Corbin, J. C.; Pieber, S. M.; Czech, H.; Zanatta, M.; Jakobi, G.; Massabò, D.; Orasche, J.; El Haddad, I.; Mensah, A. A.; Stengel, B.; Drinovec, L.; Mocnik, G.; Zimmermann, R.; Prévôt, A. S. H. and Gysel, M. Brown and black carbon emitted by a marine engine operated on heavy fuel oil and distillate fuels: optical properties, size distributions and emission factors, J. Geophys. Res. Atmos., 123, 6175-6195, doi:10.1029/2017JD027818, 2018.

Drinovec, L., Močnik, G., Zotter, P., Prévôt, A. S. H., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T., Wiedensohler, A. and Hansen, A. D. A.: The"dual-spot" aethalometer: an improved measurement of aerosol black carbon with real-time loading compensation, Atmos. Meas. Tech., 8(5), 1965–1979, doi:10.5194/amt-8-1965-2015, 2015.

2.   Line 122: "1.57 for C". Does this mean your Aethalometer was using TFE-coated glass fiber filters? Please mention the filter material.

Our Aethalometer was using TFE-coated glass fiber filters. We have added some clarifications in the text as follows:

*The manufacturer default values are 1.57 for C (**TFE-coated glass fiber filters**) and 12.2 m^2 g^{-1} for σ_{ATN} at 880 nm, which corresponds to a MAC_{BC}(880 nm) of 7.77 m^2 g^{-1} (Gundel et al., 1984 , Drinovec et al., 2015).*

3.   Line 128: It should be necessary to add some more discussion about possible artifacts that affect both techniques (MWAA and Sunset analyser).

We have added text, cited and quoted in the preceding responses, to better explain how the MWAA functions and has been validated. We have also added a discussion of artifacts affecting the Sunset analyzer in the new subsubsection quoted above. The relevant text (Section 4.1, as cited at L128) has been updated following this comment and was quoted in our response to this reviewer's overall comments. For clarity, we will not quote the text again here.

References:

Cavalli, F., Viana, M., Yttri, K. E., Genberg, J. and Putaud, J.-P.: Toward a standardised thermal-optical protocol for measuring atmospheric organic and elemental carbon: the EUSAAR protocol, Atmos. Meas. Tech., 3(1), 79–89, doi:doi:10.5194/amt-3-79-2010, 2010.

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Massabò, D., Bernardoni, V., Bove, M. C., Brunengo, A., Cuccia, E., Piazzalunga, A., Prati, P., Valli, G. and Vecchi, R.: A multi-wavelength optical set-up for the characterization of carbonaceous particulate matter, J. Aerosol Sci., 60, 34–46, doi:10.1016/j.jaerosci.2013.02.006, 2013.

4.   Line 136. MWAA measurements. Which artifacts are to be considered when using this technique in comparison to the Aethalometer? Can you provide more evidence on the comparison of this technique to other absorption measurements like MAAP or PAS?

This good comment has been addressed in our response to this reviewer's first major comment. The MWAA has been validated against the MAAP by Massabo et al. (2013).

5.   Line 171: What do you mean with the online kOA? How was this measured?

We apologize for this confusion; we have not measured the k_{OA} online. We corrected this error and rephrased the sentence for clarity:

If the OA extraction efficiency was less than unity, then the absorption (or MAC) predicted from our solvent-extraction measurements would be less than that measured (or calculated) using our real-time measurements (MWAA-calibrated aethalometer).

6.   Lines 403-411: How does this result compares to other studies? Ambient measurements have shown quick oxidation of brown carbon chromophores. Please comment about it.

The results of Forrister et al., 2015 suggest that BrC emitted from wildfires has very poor atmospheric stability with most of the aerosol being lost within a day, either due to chemical loss or evaporation. However, other ambient and chamber studies clearly (Zhong et al., 2014, Lee et al., 2014) imply that the optical properties of BrC are strongly dependent on the complex process of aging. Photo-chemical destruction and formation of chromophores is a topic of current research and may strongly depend on the gas phase composition.
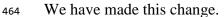
References:

Forrister, H., Liu, J., Scheuer, E., Dibb, J., Ziemba, L., Thornhill, L. K., Anderson, B., Diskin, G., Perring, A. E., Schwarz, J. P., Campuzan-Jost, P., Day, D. A., Palm, B. B., Jimenez, J. L., Nenes, A., Weber, R. J.: Evolution of brown carbon in wildfire plumes, Gephys. Res. Lett.,42, 4623-4630, doi: 10.1002/2015GL063897, 2015.
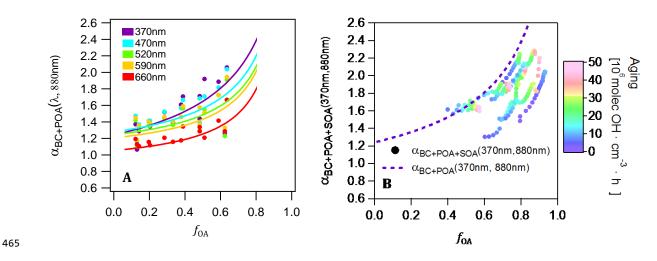
Zhong, M. and Jang, M.: Dynamic light absorption of biomass-burning organic carbon photochemically aged under natural sunlight, Atmos. Chem. Phys., 14, 1517-1525, doi:10.5194/acp-14-1517-2014, 2014.

Lee, H. J., Aiona, P. K., Laskin, A., Laskin, J. and Nizkorodov, S. A.: Effects of solar radiation on the optical properties and molecular composition of laboratory proxies of atmospheric brown carbon, Environ. Sci. Technol. Lett., 48(17), 10217-10226, doi:10.1021/es502515r, 2014.

Technical Corrections:

7.   Line 101: It would be convenient to add numbers to these headlines across the manuscript for the sake of readability (e.g., "2.2.1 Aethalometer").

We agree, however, as our manuscript still needs to be converted from Word to LaTeX during typesetting, we will leave this decision to the journal staff.

8.   Line 123: at?

The 'at' was removed from the sentence in line 123.

9.   Line 141: Which angles?

We modified the text as follows:

> L140. *The first photodiode is placed behind the filter for transmittance measurements (0° relative to the incident light, 1.5 cm from the sample), while the other two photodiodes are positioned at 125º and 165º (11 cm from the sample) to collect the back scattered light.*

10.   Lines 294-295: Please rephrase this sentence to improve understanding.

We have rewritten the entire section for improved clarity, as quoted at the beginning of this response.

This was not the case, the two variables for the MAC_{BC} came from the MWAA and EC. The aethalometer provided high time resolution attenuation coefficients which could be calibrated to give absorption coefficients needed for the MAC_{OA} retrieval. eBC was not needed for MAC_{OA} calculations as now clarified in the text, but for display purposes (Fig. 2, 3, 7 and 8). In the revised manuscript we have clarified the text (see new Section 4.1 quoted at the beginning of this response).

This has been done in the corrected version of the manuscript.

This has been done in the corrected version of the manuscript:

*The mean $MAC_{SOA}(370nm)$ was 2.2 m^2 g^{-1} (one-sigma variability: 1.6 – 3.1 m^2 g^{-1} according to a **GSD = 1.39**) under our experimental conditions, 2.3 times lower than the mean $MAC_{POA}(370nm)$ but approximately an order of magnitude higher than MAC values estimated for ambient oxygenated aerosols or reported for SOA from biogenic and traditional anthropogenic precursors.*
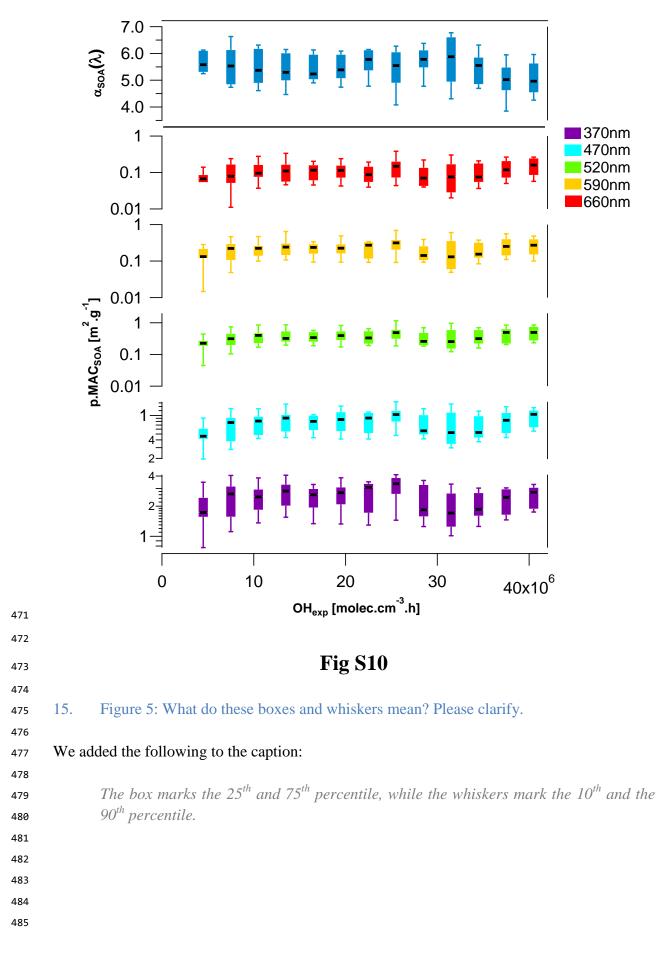
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461   14.   Figures 3A, S6, and S10: The data is presented using discrete colors for each
462   wavelength. Please make the legend discrete too.

463

464   We have made this change.

465

466   **Fig 3A**

467

468   **Fig S6**

469

470

Fig S10

15.   Figure 5: What do these boxes and whiskers mean? Please clarify.

We added the following to the caption:

The box marks the 25th and 75th percentile, while the whiskers mark the 10th and the 90th percentile.

14

486

487 16.   Figure 6: Could you please add the correlation coefficients to the figure?

488

489 We are not sure if the reviewer is referring to a Pearson correlation coefficient r or to the
490 coefficient of determination (R^2) of the fit. Although these two quantities are numerically
491 related by a square root function, r is used to describe a data set and R^2 is used to describe a
492 fit. But R^2 is not appropriate to define our fit, as we performed an orthogonal regression. We
493 have reported the standard error of the fit parameters as appropriate diagnostic statistics. We
494 have followed the reviewer's request to include a correlation coefficient in the paper, but, to
495 avoid confusing the reader, we have mentioned the Pearson correlation coefficient in the text
496 but not added it to the figure, as we expect most readers to misinterpret it as reflecting
497 goodness of fit:

498

499   Line 421. *Fig. 6B shows excellent correlation between the $MAC_{OA}(370nm)$ values*
500   *obtained from the kOA of the solvent-extracted OA with the in-situ method described*
501   *above. The Pearson correlation coefficient was 0.8, for both solvents.*

502

503 17.   Figure S6: I guess you meant α as a function of λ or do you mean only the
504 wavelength pair 370-880 nm?

505

506 We apologize for the confusion; we modified the figure caption as follows:

507

508   *Figure S6: Relationship of $\boldsymbol{\alpha_{BC+POA+SOA}}(\lambda, \boldsymbol{880nm})$ to $\boldsymbol{f_{OA}}$ for seven wavelengths,*
509   *with symbol sizes indicating OH exposure.*

510

511 18.   Figures S4, S7, and S9: Please stick to journal guidelines and avoid the use of the
512 jet (or rainbow) color map: "For maps and charts, please keep colour blindness in
513 mind and avoid the parallel usage of green and red. For a list of colour scales that are
514 illegible to a significant number of readers, please visit ColorBrewer 2.0".

515

516 Based on the referee comment, we have changed the three figures as follows:
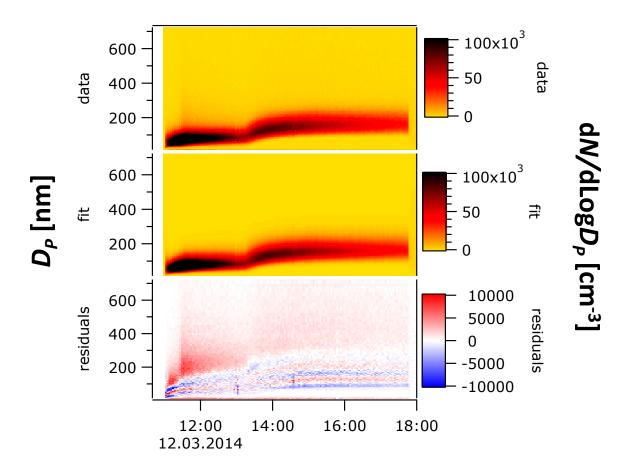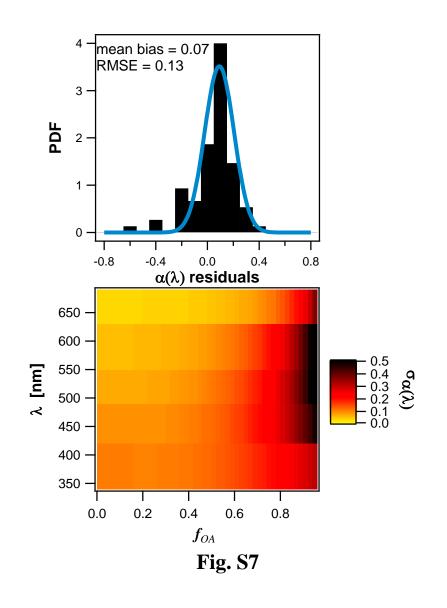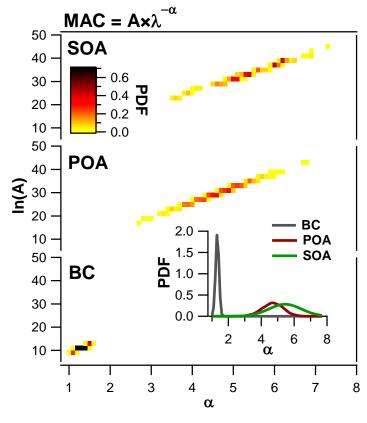
517

Fig. S4

Fig. S7

Fig. S9

Anonymous referee 2.

GENERAL COMMENT

Overall, I find that the authors have presented novel and interesting results on the influence of photochemical aging on absorption by residential wood combustion emissions. They have done a better job than I often see in assessing the performance of the aethelometer for their specific situation, but should include additional details regarding measurement uncertainties, and how these measurement uncertainties propogate to their final atmospheric implications. I have numerous comments, mostly just asking for clarification. I believe this paper should be publishable, with revisions.

We thank the referee for her/ his constructive comments, which we address below.

1.   Title: It would be good to state "residential wood-burning emissions."

The title has been modified in the corrected version of the manuscript:

> *Production of particulate brown carbon during atmospheric aging of residential wood-burning emissions*

2.   L14: It might be good to indicate this was under (likely) high NOx conditions.

The experiments were conducted at estimated $NO_x/NMOG$ ratios of ~ 0.035 – 0.35 ppm ppm C^{-1} (Bruns et al., 2016). These conditions can be considered as high NO_X, where most of the RO_2 radicals react with NO, rather with RO_2/HO_2. This information has been added to the corrected version of the manuscript in section 2.1.

> *Section 2.1.*
> *Laboratory measurements were conducted in an 8 m^3 Teflon smog chamber (Bruns et al., 2015; Platt et al., 2013) installed within a temperature-controlled housing. Conditions in the chamber were maintained to represent winter time in Europe, i.e. relative humidity ranging between 50 – 90%, at 263 K (Bruns et al., 2015, 2016). Beech wood was combusted in a residential wood stove. Primary emissions were sampled through heated lines at 413 K, diluted by a factor of ~14 using an ejector diluter (DI-1000, Dekati Ltd.), then sampled into the chamber, which provided an additional ten-fold dilution. The overall dilution was a factor of 100 to 200. As we aimed to sample only flaming-phase emissions into the chamber, samples were taken when the modified combustion efficiency (ratio of CO_2 to the sum of CO and CO_2) was > 0.90. Despite maintaining the same combustion conditions, the resulting organic fraction to the total carbonaceous aerosols in the different samples was*

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highly variable, indicating that these samples are representative of a mixture of pre-ignition and flaming emissions (with varying contributions of each combustion stage). Finally, the resulting NOx/NMOG ratios, which dramatically influence SOA formation through influencing the fate of peroxy radicals, RO_2, were estimated to be between 0.035 – 0.35 ppm ppm C^{-1} (Bruns et al., 2016). These conditions can be considered as high NO_X representative of urban/sub-urban conditions, where most of the RO_2 radicals react with NO, rather with RO_2/HO_2.

3.   L15: It would be good to clarify what wavelength, or if this is integrated in some way.

This has been clarified in the corrected version of the manuscript:

At shorter wavelengths (370 – 470nm), light absorption by brown carbon from primary organic aerosol (POA) and secondary organic aerosol (SOA) formed during aging was around 10 % and 20 %, respectively, of the total aerosol absorption (BrC plus BC).

4.   L17: Reporting the GSD is informative, but an actual uncertainty estimate would be better.

Based on this comment and others below, the reviewer is asking for providing uncertainty propagation based on measurement precision and accuracy, instead of a GSD which represents the experimental variability. As we discussed in the reply to the first reviewer, we have added in the corrected version of the manuscript a new section in the Method, discussing quantifiable and unquantifiable uncertainties. However, in the result section we still present the variability in the parameters determined as GSD, as these represent our confidence levels in these average parameters. We believe that such information is relevant if these parameters were to be used for future predictions. We also consider that for most of the parameters experimental variability is much more important than measurement uncertainties and biases.

5.   L54: It would be good to also cite the work from the Georgia Tech group (*Forrister et al.*, 2015).

As suggested by the reviewer the work of Forrister et al., 2015 was cited.

Forrister, H., Liu, J., Scheuer, E., Dibb, J., Ziemba, L., Thornhill, L. K., Anderson, B., Diskin, G., Perring, A. E., Schwarz, J. P., Campuzan-Jost, P., Day, D. A., Palm, B. B., Jimenez, J. L., Nenes, A., Weber, R. J.: Evolution of brown carbon in wildfire plumes, Gephys. Res. Lett.,42, 4623-4630, doi: 10.1002/2015GL063897, 2015.

6.   L229: It is not clear to me why the MAC for POA would be unaffected. The authors write for Eqn. 13 what amounts to:

$$\alpha(\lambda, 880) = \frac{-1}{\ln\left(\frac{\lambda}{880}\right)} \ln\left(\frac{[EC]MAC_{BC_\lambda}}{[EC]MAC_{BC_{880}}} + \frac{[OA]MAC_{OA_\lambda}}{[EC]MAC_{BC_{880}}}\right)$$

$$= \frac{-1}{\ln\left(\frac{\lambda}{880}\right)} \ln\left(\frac{[EC]MAC_{BC_\lambda}}{[EC]MAC_{BC_{880}}} + \frac{[OA]MAC_{OA_\lambda}}{b_{abs,880}}\right)$$

and where they have only included the second line, not the first. The $b_{abs,880}$ value, while seemingly independent of the EC measurements as the authors have written them, actually do depend on the EC measurements because everything has been referenced to the EC measurement. Perhaps I am simply missing something, but I think that the authors statement must be further justified. Yes, the MAC of OA is fundamentally independent of the MAC of BC. But I am not certain that these are practically separated to the extent indicated by the authors. This is the same challenge that all AAE extrapolation methods face in quantitatively determining OA absorption in the presence of BC. See e.g. the cited Moosmuller paper or (*Lack and Langridge*, 2013). Also, the statement on L231 is self-evident. Of course the AAE at a given wavelength depends on the relative contributions of BC and OA.

The reviewer raises two separate points. The first is whether the $MAC_{OA,\lambda}$ directly depends on the EC mass. The second is whether the BC absorption at a given λ affects the estimation of $MAC_{OA,\lambda}$. Below, we address these points separately.

1. We consider it inaccurate to say that OA MAC has been referenced to EC. This reflects how we have expressed and applied Equation 13. The intermediate steps leading to equation 13 were intentionally omitted, as they tend to mislead the reader. Equation 13 clearly shows that:
   - EC mass concentration is not explicitly required.
   - A potential bias in $MAC_{BC,880nm}$ due to a bias in EC mass would directly affect the resulting $MAC_{BC,\lambda}$ in a proportional manner, whereas $MAC_{OA,\lambda}$ would remain completely unaffected.
   - The resulting $MAC_{OA,\lambda}$ depends on the input parameter M_{OA}, thus being affected by potential AMS calibration bias.
   - A potential bias in absorption coefficients measured by the MWAA would propagate to a proportional bias in $MAC_{OA,\lambda}$, as aethalometer measurements of b_{abs} are referenced against the MWAA. Such bias in absorption coefficient would also propagate to a proportional bias in $MAC_{BC,\lambda}$, which would happen through a corresponding bias in $MAC_{BC,880}$
   - The resulting $MAC_{OA,\lambda}$ also depends on the input parameter $b_{abs,880nm}$, which is referenced to the MWAA measurement, whereas the EC data are by no means

blended into the b_{abs} data. Such bias in absorption coefficient would also propagate to a proportional bias in $MAC_{BC,\lambda}$, which would happen through a corresponding bias in $MAC_{BC,880nm}$.

2. We acknowledge that the determination of $MAC_{OA,\lambda}$ depends on the estimated absorption of BC at a given λ, which in turn depends on the estimated $MAC_{BC,\lambda}$. This is an issue on any multivariate fitting, where the theoretically independent fitted quantities are not independently determined. We note that such uncertainties are taken into account by the fitting errors presented as GSDs in the manuscript. The $MAC_{BC,\lambda}$ is similar to an extrapolation of the absorption measurements at $f_{OA}= 0$. We note our experiment covered BC rich particles allowing for an accurate determination of $MAC_{BC,\lambda}$.

Reference:

Saleh, R.; Adams, P. J.; Donahue, N. M. & Robinson, A. L. The interplay between assumed morphology and the direct radiative effect of light-absorbing organic aerosol. Geophys. Res. Lett., 43, 8735-8743, doi:10.1002/2016GL069786, 2016.

We have added the statements below to the manuscript:

L222. [...] Equation 10. We have intentionally formulated of Equation 13 as such to highlight that the retrieved $MAC_{OA}(t,\lambda)$ depends mainly on the input $M_{OA.}$ Correspondingly, the retrieved $MAC_{OA}(t,\lambda)$ is mainly sensitive to potential AMS calibration biases.

7.   L264: What about chemically induced changes of POA mass, as opposed to just absorption?

We have mentioned the changes in POA mass and MAC in the sentence before, in L263: "Equation 19 is based on the assumption that POA is "chemically inert", i.e. no chemically induced changes of M_{POA} and MAC_{POA} occur". We have adjusted the text based on the Reviewer's comment to avoid confusion:

*Such chemically induced changes of absorption coefficient by POA**, through a change of M_{POA} or MAC_{POA},** if they occur, are assigned to the absorption by SOA, thus resulting in a corresponding adjustment of the inferred MAC_{SOA}.*

8.   L287: It is unclear what is meant by "a higher than measured lensing effect."

This text has been rewritten following the comments above. The revised text clarifies this meaning and, to avoid confusion, we will simply refer to the revised text in response to Reviewer #1's first comment. The corrected text reads as follows:

Section 4.1.

We have independently determined the $MAC_{BC}(880nm)$ and the aethalometer C values under our conditions, as follows. We determined $MAC_{BC}(880nm)$ from the regression between the absorption coefficients at 880 nm obtained from the MWAA and the EC mass measured by the Sunset analyzer (Figure 1A). The slope of this regression may be used to estimate the $MAC_{BC}(880nm)$, which we retrieved as $4.7 \pm 0.3\ m^2g^{-1}$ by an uncertainty-weighted linear least-squares fit . The corresponding intercept was not significantly different from zero (-3 ± 3 /Mm). Our $MAC_{BC}(880nm)$ is not statistically significantly different from the value recommended by Bond et al., (2006) for externally-mixed BC (extrapolating their $MAC_{BC}(550nm)$ to 880 nm by assuming α=1 provides $MAC_{BC}(880nm)= 4.7 \pm 0.7\ m^2g^{-1}$). The strong correlation between $b_{abs,MWAA,880nm}$ and EC in Fig. 1A shows that $MAC_{BC}(880nm)$ did not vary with aging during our study (see also Fig. S2-a). It also indicates that measurement artefacts for both instruments were negligible, as the fundamental differences between the two techniques mean that any artefacts are unlikely to be similar between them (charring for EC vs. adsorption artefacts for MWAA). Our absorption coefficient measurements also provide insights into particle mixing state in this study. Since a single MAC adequately described our samples at all levels of aging (Fig. 1A and Fig. S2-a), in spite of a factor of 3.3 average increase in the aerosol mass, our samples cannot be adequately described by a core-shell Mie model. Such a core-shell model would predict an absorption enhancement by a factor of ~1.8 (Bond et al., 2006) for the observed OA mass increase with aging, which was not observed in our case. This observation is also supported by the time resolved attenuation measurements at 880 nm using the aethalometer (Fig. S3), suggesting that little (<10%) to no increase in the attenuation coefficients upon SOA formation. We emphasize that this conclusion does not indicate that no internal mixing occurred, but rather that the simplified concept of negligible mixing better describes our data than the equally simplified concept of a core-shell description of coatings that completely envelop the central BC core. This may be due to the complex morphology of internally-mixed BC, which has been previously observed for wood burning particles (e.g., China et al., 2013; Liu et al., 2015; Liu et al., 2017). It may also be related to the fact that OA and BC are emitted during separate phases of combustion. OA rich particles are emitted during the pre-flaming pyrolysis stage of combustion, whereas most BC is emitted during flaming combustion (Corbin et al., 2015a, 2015b; Haslett et al., 2018; Heringa et al., 2011). These two stages of combustion may coexist in different regions of the stove,

particularly during simulated real-world usage. As lensing effect was negligible in our case, we have assumed that the aerosol optically behaves as an external mix between BC and BrC during Mie calculation (see section 3.4). We note that while this assumption is important for estimating the BC absorption, the conclusion drawn about the BrC absorption are not very sensitive to the assumed morphology.

We determined time-resolved wavelength-dependent absorption coefficients as follows. We used the aethalometer to obtain filter attenuation coefficients with high time resolution, which were then calibrated to obtain absorption coefficients by deriving the factor C (Equation 1) using the MWAA measurements of filter samples. C was obtained from an uncertainty-weighted linear least-squares fit as 3.0 ± 0.2 (Fig. 1B); the intercept of the fit was not significantly different from zero, within two standard deviations (-17 ± 14). A very strong correlation could be observed between MWAA and aethalometer (Fig. 1B), implying that C is independent of the type of the aerosol sampled (see also Fig. S2-B). Therefore, we used a single C value to obtain time-resolved wavelength-dependent absorption coefficients from the aethalometer attenuation measurements at the different wavelengths for primary and aged aerosols.

Note that the manufacturer's default values, which were not applied in our case, are 1.57 for C (using TFE-coated glass fiber filters) and $12.2 \ m^2 \ g^{-1}$ for σ_{ATN} at 880 nm (Gundel et al., 1984 , Drinovec et al., 2015). The C value determined here is larger than the manufacturer-default value for the AE33, resulting in smaller absorption coefficients. However, the calculated σ_{ATN} at 880 nm ($13.8 \ m^2 \ g^{-1}$ see Fig. 1C), which can be retrieved from Fig. 1-C or as the product of the C and $MAC_{BC}(880nm)$ (Equation 3), is almost equal to the factory default σ_{ATN}. Therefore, our M_{eBC}, which can be directly calculated from the attenuation coefficients using σ_{ATN} (Equation 2), would be consistent with factory default M_{eBC}. We note that M_{eBC} has not been used for MAC calculations. However for displaying purposes, we have calculated time resolved M_{eBC} using σ_{ATN} and b_{ATN} to estimate fraction of BC and OA in the aerosol (Fig. 2, 3, 7 and 8).

9.   L290: The authors mention uncertainties here for the UV-Vis measurements. But what about for all of the in situ or other measurements? This includes [EC], [OA], absorption. Further discussion of uncertainties is necessary.

We thank the reviewer for emphasizing the importance of uncertainties. In the previous version, uncertainties related with the different measurements were discussed very briefly in sections related with the description of the measurement techniques. As this comment was raised by both reviewers, we have added a new section discussing quantifiable and non-quantifiable uncertainties, which is referenced in the corresponding response and will not be duplicated here for clarity.

10.   L305: It is not clear how this uncertainty estimate was arrived at. Also, this differs from the figure. Finally, it is not clear whether this fit has been forced to zero or not.

The reviewer is right, the uncertainty value in the figure and the text are not the same and the correct value is the one indicated in the figure (this has been changed accordingly). The uncertainty is the standard error of the slope of the uncertainty-weighted linear least-squares fit. The fit has not been forced through zero. Rather, we have fit the full linear model and test for the significance of the intercept term. The intercept was at -3 ± 3 m^2/g, not statistically significantly different than 0. This intercept was missing from the submitted manuscript and has now been reported, together with further modifications to the text as quoted in our modified Section 4.1 above.

11.   L307: Bond et al. did not "report" a value at 880 nm. They reported at a shorter wavelength. This value is inferred assuming an AAE = 1. It should be noted as such.

The reviewer is right and the text has been modified accordingly, as specified in the quoted response to Reviewer #1's first comment.

12.   L308: While the authors obtain a value of the MAC in good agreement with extrapolated values from Bond et al., it is not clear to me how this definitely indicates no lensing effect is present. An uncertainty analysis is necessary. What if, hypothetically, the EC was biased high and the absorption biased low? The obtained MAC might appear in agreement with literature, but only within the bounds of the measurement uncertainty. This statement should be quantified.

The reviewer raises a valid point that unidentified measurement biases in opposing directions may have led to the illusion of agreement between our measured MAC_{BC}(880nm) values and the literature values for bare BC. The major issue here is that of referencing our absorption data and EC measurements to reliable and calibrated technique. Absorption measurements were obtained using MWAA, which has been validated as described in the text, and biases are expected to be within 10%. Measurement biases related to total carbon measurement are negigible (within 5%). The high correlation between absorption and EC measurements also indicates that unquatifiable uncertainties and artefacts (e.g. charring for EC and vapor adsorption artefacts for MWAA) are negligible, as the fundamental differences between the two measurements mean that any artefacts are unlikely to be similar between them. Therefore, measurement biases and unquatifiable artefacts are unlikely to affect the estimated values for MAC_{BC}(880nm) and our conclusions about the lack of lensing. We also note that such conclusions are also supported by independent time resolved attenuation measurements by the aethalometer, suggesting that little (<10%) to no increase in the attenuation coefficients upon SOA formation.

Our analysis has combined multiple analytical techniques and found good agreement between all of them. This good agreement reduces the likelihood that opposing measurement biases led, by chance, to our measurements being in agreement with literature. While it remains

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theoretically possible that unknown uncertainties and biases existed in our analysis, it is by Occam's razor more probable that our measurements were in fact accurate and that our cross-validation by employing different techniques was successful.

In the corrected version of the manuscript, we have added a new section discussing the measurement biases estimated for the different measurements. We have additionally discussed potential unquantifiable uncertainties. For clarity, we do not duplicate the mofified text here, but we quote the response to reviewer 1.

13.   L315: It is clear that the distribution is reasonably log-normal with a single mode when considered in number space. But what about in surface area or in volume space, which is important for the calculation of the MAC_OA_bulk (Eqn. 20)? Also, to what extent does the volume-weighted distribution exceed the SMPS bounds?

The absolute SMPS volume is not as important for calculating $MAC_{OA,bulk}$ as the reviewer understood. As can be seen from Equation 20, we did not use the SMPS data to calculate total OA volume. We measured total OA mass with an AMS, converted mass into volume using an assumed density (the assumed density has no impact on the results as we use the same density to calculate MAC), and then "distributed" this volume across the size distribution measured by SMPS. That is, the SMPS data provide only a weighting factor for the size dependence of absorption, which means that uncertainties in these data do not have a major effect on our results. We performed the calculations in this way to minimize associated uncertainties, but acknowledge that substantial uncertainties may result. We have estimated that these may be on the order of 20%, based on the magnitude with which particulate absorption varies as a function of size (according to Mie theory).
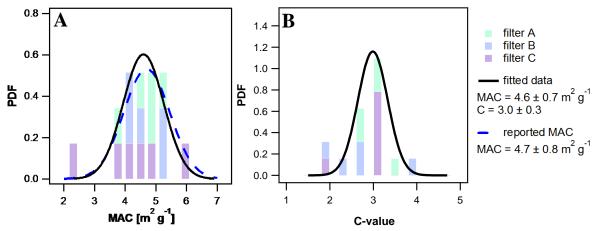
14.   L325 and Fig. S2: I do not understand why in Fig. S2 it says the "reported value" is 2.6 while here 3.0 is given. This should be clarified.

The reported C value is calculated using a $\sigma_{ATN} = 12.2$ m^{2}g^{-1}, as given by the manufacturer, and $MAC_{BC}(880nm) = 4.7$ m^2 g^{-1}. The C value = 3 is determined from our attenuation and absorption measurements which is used in our calculations. This has been discussed in lines 330-336.

For better clarity, we have now omitted the reported C-value from Fig. S2:

Figure S2: (A) Probability density function (PDF) comparing the MAC values determined by normalizing MWAA absorption measurements of offline primary (filter A), slightly aged (filter B: Aged1) and aged (filter C: Aged2) samples to EC (EUSAAR2) measurements of the same samples (bold line). A literature value for pure BC is also shown (Bond et al., 2006) (dashed blue line). (B) PDF comparing aethalometer attenutation measurements at 880 nm and MWAA absorption measurements at 850 nm to retrieve the aethalometer C value.

15.　L332: Not only wood combustion, but really any aerosol if it is assumed that the true MAC for BC at 880 nm is ~4.7 m2/g.

We agree with the reviewer. We specify wood burning because it is the focus of our study. This section has been substantially modified and this sentence was removed.

16.　L336: This seems circular to me, if the authors are using C = 3 in coming to this conclusion. This is demanded through the various inequalities.

We do not think that the math here is circular. Below, we present in bullet points the approach followed:

- $\sigma_{ATN} = MAC_{BC} * C$
- The Aethalometer provides b_{ATN}
- We determine a C value of 3 from Equation 1 of the paper:
$$C = b_{ATN,AE33} / b_{abs,MWAA}$$
- We determine a MAC_{BC} of 4.6 m^2 g^{-1} by a fit through $b_{abs,MWAA}$ and EC thermo-optical measurements for primary and aged filter samples.
- We have therefore determined the two variables required to calculate σ_{ATN} and determined it as 13.8 m^2 g^{-1}.
- We then compare the σ_{ATN} determined by us to the manufacturer value of 12.2 m^{2}g^{-1}.
- We then concluded that while the factory default $b_{abs}(\lambda)$ has a substantial bias the eBC mass determined using default σ_{ATN} is consistent with the EC mass. We have rewritten Section 4.1, as quoted at the beginning of this response, to clarify this reasoning.

While we agree with the reviewer here, one reason that we have not cited e.g. Lewis et al (2008) is that those authors did not experiment on log-wood burning in a modern stove but rather simulated wildfires. Considering the very low emphasis placed on Fig. S5 in our manuscript, and the importance of avoiding confusion between log-wood burning and wildfire burns, we have simply removed the α ranges in Fig. S5 is only for primary biomass burning emissions. We have also modified the legend to show the two wavelengths of α.

Fig. 3A refers to primary emissions and Fig. 3B refers to secondary emissions (as indicated in the caption), so yes, the presumption here was correct, but only partially. More data are included in Fig. 3A because not all burn experiments were aged. This information has been added to the figure caption. In Fig. 3A each point represents an experiment; therefore the variability in f_{OA} is due to the variability in emission composition between experiments. Meanwhile, in Fig. 3B the variability in f_{OA} is due to SOA formation with aging; data from several experiments are shown which explains the wide range of f_{OA} at low OH exposures. Based on the reviewer comment, we have added the following clarifications to the Fig. 3 caption:

> *Figure 3: (A) Relationship of α_{BC+POA} (λ, 880nm) to f_{OA} for seven wavelengths for primary emissions. Data are colored by the wavelength. Curves are fits of Equation 13 to the data. Each point represents the average of one experiment and therefore the variability in f_{OA} is related to the variability in the emission composition between experiments. (B) Relationship of $\alpha_{BC+POA+SOA}$(370nm, 880nm) to f_{OA} for several experiments for aged aerosols. Data are color coded by the OH exposure. The variability in f_{OA} is due to SOA formation with aging; data from several experiments are shown which explains the wide range of f_{OA} at low OH exposures. Note that more data are included in A than B, as primary emissions for some experiments were not aged.*
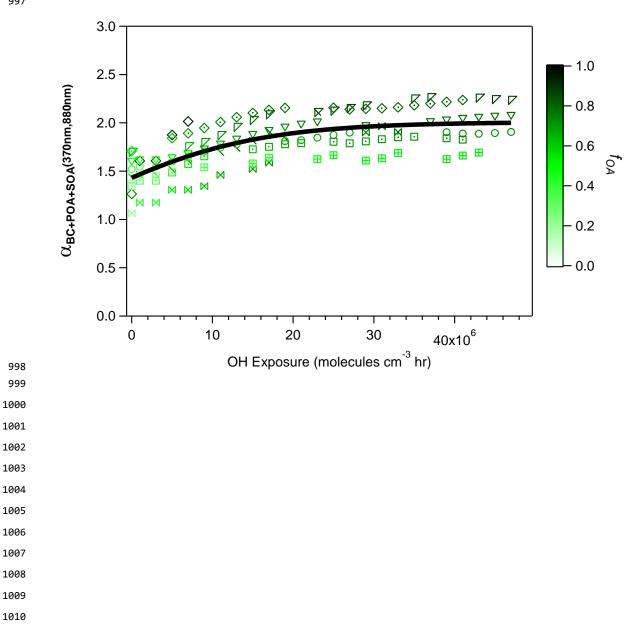
19.   L344: I do not dispute that the AAE values increase with f_OA. However, it is evident that as wavelength decreases the difference from AAE = 0.9-1.1 and the observations increases. This is not clear from the discussion here.

We have stated that the AAE values at low f_{POA} are close to those reported for pure BC. We note that for λ = 660 nm, the AAE value extrapolated at f_{POA} = 0, is equal to 1.04, while for all other wavelengths the value is statistically similar, equal to ~1.2. It can be seen from Fig. 4 that $MAC_{BC}(\lambda)$ profile can be clearly described by a power law, consistent with a constant AAE. To avoid confusion, we have modified the text as follows:

> L344. *The $\alpha(\lambda, 880nm)$ is slightly higher than that of pure BC (~1.2; Bond et al., 2013; Zotter et al., 2017) for small f_{POA}, while increasing f_{POA} corresponded to a distinct increase of $\alpha(\lambda, 880nm)$.*

20.   L346: The range reported is inconsistent with what is shown in the graph.

The range presented before denoted the P10 and P90; we apologize that we have forgotten to mention this in the text. Based on the reviewer comment and to avoid confusions we have replaced the [P10-P90] by the total range:

> *The f_{POA} ranges from **0.12 to 0.63**, which is lower than f_{POA} reported for open burning emissions (e.g., f_{POA}~0.75, Ulevicius et al (2016)), because our wood-stove emissions feature a more efficient combustion.*

21.   L350: this could be strengthened simply by showing a graph of the observations as a function of wavelength, and showing that a single AAE value does not provide for a good fit.

This is shown in Fig. S5 of the SI. We have added in the corrected version of the manuscript a reference to Fig. S5 and modified the figure caption to highlight the point raised by the reviewer as follows:

> *Figure S5: Absorption coefficients of fresh wood burning emissions measured using an aethalometer normalized to the eBC mass as a function of wavelength. In the legend each color denotes the $\alpha_{BC+POA}(370nm,880nm)$ for an individual experiment. The dashed lines mark the absorption profiles calculated using the literature range of α values obtained for primary biomass burning emissions. The observed absorption spectra have steeper gradients with decreasing wavelength compared to the lines of constant alpha. The systematic decrease in $\alpha(\lambda, 880nm)$ with increasing λ reflects the more-efficient light absorption by BrC at shorter wavelengths (Moosmüller et al., 2011), and shows that the power law wavelength dependence is an inaccurate oversimplification for this mixed aerosol.*

984   We have added the following in the text in L350:

985

986   *As illustrated in Fig. S5, the observed absorption spectra have steeper gradients with*

987   *decreasing wavelength compared to the lines of constant alpha. Such systematic*

988   *increase in $\alpha(\lambda, 880nm)$ with decreasing λ reflects the more-efficient light absorption*

989   *by BrC at shorter wavelengths (Moosmüller et al., 2011), and shows that the power*

990   *law wavelength dependence is an inaccurate oversimplification for this mixed*

991   *aerosol.*

992

993   22.   Fig. 2: For consistency, it would be helpful if the color scale were labeled as f_OA,

994 similar to Fig. 3 and the text.

995

996   This has been modified in the corrected version of the manuscript:

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23.   L356: Looking at Fig. 3B, it is not clear that this is generally the case. The highest f_OA in Fig. 3B does not have the highest AAE. Perhaps the authors mean this just for the high OH exposures. If so, they might consider plotting AAE vs. f_OA for subsets of data binned according to OH exposure. But, as presented, it is not evident that this is a fully general conclusion.

We do mean at higher OH exposures. This has been added in the corrected version of the manuscript:

> *Also, note in Fig. 2 that at highest OH exposures, the highest $\alpha_{BC+POA+SOA}$(370nm, 880nm) were reached, on average 1.8, during experiments where the fOA was highest.*

24.   L362: While I don't necessarily disagree with this point, I will reiterate that the relationship between Fig. 3A and 3B is not clear. The authors give a dashed curve, but it is not clear how, for example, the data in Fig. 3B that start at such low AAE values at high f_OA values come from Fig. 3A. A stronger connection needs to be made to make this clearer

We think that the misunderstanding comes from the fact that we had not adequately highlighted that not all experiments in Fig. A are shown in Fig. B, as for some of the experiments the emissions were not aged. We chose to represent use all the data available in Fig. A to increase our statistics and expand the f_{OA} range. As mentioned above this information has been added and the Figure caption now reads as follows:

> *Figure 3: (A) Relationship of α_{BC+POA} (λ, 880nm) to f_{OA} for seven wavelengths for primary emissions. Data are colored by the wavelength. Curves are fits of Equation 13 to the data. Each point represents the average of one experiment and therefore the variability in f_{OA} is related to the variability in the emission composition between experiments. (B) Relationship of $\alpha_{BC+POA+SOA}$(370nm, 880nm) to f_{OA} for several experiments for aged aerosols. Data are color coded by the OH exposure. The variability in f_{OA} is due to SOA formation with aging; data from several experiments are shown which explains the wide range of f_{OA} at low OH exposures. Note that more data are included in A than B, as primary emissions for some experiments were not aged.*

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25. L364: For the data in Fig. 3B, extrapolation to f_OA -> 0 for wavelengths < 600 nm suggests an AAE ~1.2- 1.3, larger than the 0.9-1.1 range the authors have taken for BC. This is consistent with the derived MAC(370) = 13.7 m2/g for BC, given the value at 880 nm. There is, however, a bit of an inconsistency with how the authors compare with Bond. They state that 13.7 is within the 95% confidence interval of the 11.1 m2/g value reported by Bond. But, they have also stated that the AAE = 0.9-1.1. If this is the case, then isn't the range actually narrower? Really, my question here is about the consistency of the statistical interpretation/uncertainty representation.

We thank the reviewer once again for raising opportunities for clarificating the text, where some of the information were missing. The range of $MAC_{BC}(370)$ we calculate is based on an error propagation calculation considering not only the range of AAE reported (0.9-1.1) but also the errors on the absolute $MAC_{BC}(520)$. This has now been clarified in the text:

> *The obtained fit value of $MAC_{BC}(370nm)$ was 13.7 m^2 g^{-1} (GSD 1.1, one-sigma uncertainty 12.4—15.1 m^2/g), higher but not statistically significantly different from the range estimated based on Bond et al. (2013), considering the uncertainties on both the α_{BC} values and the $MAC_{BC}(520nm)$.*

26. Fig. 7: I find the legend to be incomplete in that it leaves the reader thinking that the Lu and Saleh measurements are from methanol extraction, which they are not. Also, for Saleh (2014), the authors do not at all make clear that the Saleh measurements are at 550 nm, not 370. This is not a fair comparison. Neither is which fuel type of Saleh's has been considered. The authors should provide a fuller picture. Also, the Saleh reference is missing from the bibliography.

Thank you for pointing out the missing reference. The Saleh et al. (2014) data in Fig. 7 were extrapolated to 370 nm using the wavelength dependence of k_{OA} i.e. $k_{OA} = k_{OA,550}$ x $(550 / \lambda)^w$, given by those authors. Likewise, the Lu et al., 2015 data have also been determined at 370 nm using the wavelength dependence of k_{OA} provided by the authors ($k_{OA} = 0.017$ x $(550 / \lambda)^{1.62}$) and the parameterization of k_{OA} against BC/OA ratio. We have updated the figure legend and caption according to the reviewer comment:
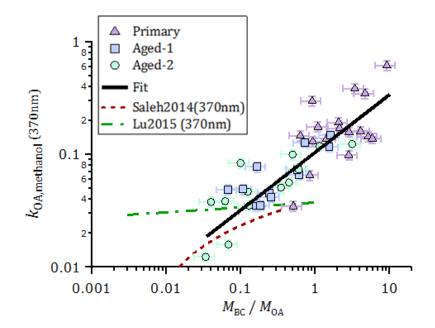
Figure 7: Imaginary part of the OA refractive index at 370 nm, obtained from offline UV/vis spectroscopy of methanol OA extracts, plotted as a function of **fOA**. The data could be empirically represented by a linear function in the log-log space, in the measurement range. The ordinary least-squares fit is ($k_{OA,nm}$) = $log(M_{BC}/M_{OA})(0.51\pm0.07) + (-0.98\pm0.05)$. Also shown are parameterizations of $k_{OA}(370nm)$ for open burning against M_{BC}/M_{OA} estimated based on the online $k_{OA}(550nm)$ measurements in Saleh et al. (2014) and Lu et al. (2015), using the k_{OA} wavelength dependence reported by the respective authors.

Reference added:

Saleh, R., Robinson, E. S., Tkacik, D. S., Ahern, A. T., Liu,S., Aiken, A. C., Sullivan, R. C., Presto, A. A., Dubey, M. K., Yokelson, R. J., Donahue, N. M., and Robinson, A.L.:Brownness of organics in aerosols from biomass burning linked to their black carbon content, Nat. Geosci., 7, 2–5, doi:10.1038/ngeo2220, 2014.

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1116   27.   Fig. S7: It is not clear why the propagated uncertainty in the AAE increases with
1117   wavelength or f_OA. The AAE is a measured quantity that depends only on the measured
1118   absorption at two wavelengths. . Why would uncertainty in absorption depend on f_OA? And
1119   are the authors saying that the uncertainty in absorption increases with wavelength?
1120   Uncertainty in the AAE should directly propagate from Eqn. 10, which is independent of
1121   f_OA. Perhaps I am misunderstanding?

1122

1123   We believe that there is a misunderstanding. Fig. S7A is obtained from the error propagation
1124   of equation 13 solved for different wavelengths, using the geometric mean and standard
1125   deviation of $MAC_{POA}(\lambda)$ and $MAC_{BC}(\lambda)$. The resulting error term represents the variability
1126   in/ the confidence level on the $\alpha(t_0, \lambda, 880nm)$ at different wavelengths. Equation 13 is
1127   expressed below:

1128

1129 $\alpha(t_0, \lambda, 880nm) = \alpha_{BC+POA}(t_0, \lambda, 880nm)$

$$= \frac{1}{\ln(880\text{nm}/\lambda)} \ln\left(\frac{MAC_{BC}(t_0, \lambda)}{MAC_{BC}(t_0, 880nm)} + \frac{M_{OA}(t_0)MAC_{POA}(t_0, \lambda)}{b_{abs}(t_0, 880nm)} \right)$$

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1131   As $\alpha(t_0, \lambda, 880nm)$ depends on $M_{OA}/b_{abs}(t_0, 880nm)$ $\sigma_{\alpha(t_0,\lambda,880nm)}$ also does. We expressed
1132 $M_{OA}/b_{abs}(880nm)$ as f_{OA}, using σ_{ATN} to estimate EC mass from $b_{ATN}(880nm)$.
1133   The image plot in panel B shows that at short wavelengths and low fractions of OA, the
1134   confidence level on α is within 0.1. However, with increasing f_{OA}, and at longer wavelength
1135   the uncertainty in predicting α increases. The idea behind this figure is to provide an error on
1136   the predicted α when the f_{OA} is extrapolated to values higher than measured here (typical of
1137   open burning).

1138   We have updated the figure caption adding the explanations above:

1139   *Figure S7: Analysis of the fitting errors of $\alpha(\lambda, 880nm)$ of primary emissions as a*
1140   *function of f_{OA}. Panel A shows the α residual as a probability density function. Panel*
1141   *B is an image plot of the $\alpha(\lambda, 880nm)$ error, $\sigma_{\alpha(t_0,\lambda,880nm)}$, as a function of f_{OA} at*
1142   *different wavelengths. $\sigma_{\alpha(t_0,\lambda,880nm)}$ is obtained from the error propagation of Eq.*
1143   *(13) solved for different wavelengths, using the geometric mean and standard*
1144   *deviation of $MAC_{POA}(\lambda)$ and $MAC_{BC}(\lambda)$. This error term represents the variability in*
1145   *or the confidence level on the $\alpha(t_0, \lambda, 880nm)$ at different wavelengths. As*
1146   *$\alpha(t_0, \lambda, 880nm)$ depends on $M_{OA}/b_{abs}(t_0, 880nm)$ in Equation 13, $\sigma_{\alpha(t_0,\lambda,880nm)}$ also*
1147   *does. We expressed $M_{OA}/b_{abs}(t_0, 880nm)$ as f_{OA}, using σ_{ATN} to estimate EC mass*
1148   *from $b_{ATN}(880nm)$. At short wavelengths and low OA fractions, the confidence level*
1149   *on α is within 0.1. However, with increasing f_{OA}, and at longer wavelength the*
1150   *uncertainty in predicting α increases.*

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28.   L384: A larger GSD does not necessarily mean a larger uncertainty. This could be variability that is independent of uncertainty. I do not regard this as a true assessment of "uncertainty." It is only an assessment of variability. The authors should, however, consider uncertainties explicitly.

The reviewer is correct, the word "variability" rather than "uncertainty" should have been used in this sentence. That is, the GSD values relate to variability in the MAC values that is not explained by the variability in f_{OA}. Overall, our data show that this variability is related to a real change in the chemical nature of the compounds present and their intrinsic absorptivity, as online MAC values correlate well with k_{OA} values independently measured offline after methanol extraction. As mentioned above, we have now added a new section discussing the quantifiable and unquantifiable uncertainties. We have also updated the related to the variability in the determined parameters as follows:

> ***Uncertainties and variability in MAC_{BC}, MAC_{POA} and MAC_{SOA}.*** *Table 1 shows the fitting errors related with $MAC_{BC}(\lambda)$, $MAC_{POA}(\lambda)$ and $MAC_{SOA}(\lambda)$, arising from our measurement precision and experimental variability. These fitting errors are greater than our estimated uncertainties in the absorption coefficients measured by MWAA (10%), and comparable to our estimated uncertainty in OA mass measured by AMS (30%). The residuals in the fitted $MAC_{BC}(\lambda)$ are relatively low (< 10%), increasing with decreasing λ. By contrast, the uncertainties in the fitted $MAC_{POA}(\lambda)$ are much higher (GSD = 1.2–1.5) and increase with increasing λ. The relative residuals between the measured and fitted $\alpha(\lambda,880nm)$ for primary emissions showed a mean bias and RMSE of 0.07 and 0.13, respectively (Fig. S7), indicating that our fitted MAC results provide a good description of the data set. $MAC_{SOA}(\lambda)$ values determined were highly variable between experiments with a GSD = 1.39 and 2.42 for λ=370 nm and 660 nm, respectively. In Fig. S10, we show the distribution of $MAC_{SOA}(\lambda)$ values as box and whiskers against OH exposure, showing no particular dependence of these values with aging as it will be discussed below. Therefore, we expect the fitting errors in MAC_{SOA} and of MAC_{POA} to be mainly related to true changes in the organic aerosol chemical composition between different burns, since the variability of $MAC_{BC}(\lambda)$ was relatively small. In Section 4.3, we discuss this variability further using the results of an additional and independent analysis.*

The MACs of primary species (POA and BC) at different wavelengths are obtained through fitting equation 13, which used the AAE as dependent variable. Therefore, we have assessed the model goodness of fit by showing the residuals in the AAE values (Fig. S7). While we cannot show the residuals distributions for MAC_{POA} and MAC_{BC} resulting from the fit, the obtained fitting errors (GSD) can be used to represent these distributions. The normal fit of the AAE residuals in Fig. S7A serves only to illustrate the distribution of fitting errors and is not essential to our analysis. We note that we have never claimed that MAC values to be normally distributed and hence we performed our fits in log-transformed space to constrain the MAC to be greater than zero. MAC_{SOA} is the only unknown parameter in equation 19. Therefore, we have shown the distributions of MAC_{SOA} as a function of wavelength and OH exposure, in Fig. 4 and 5, respectively.

The ranges for AAE values have been already provided in Fig. S9 and Table S1, and following the reviewer's comment we have added the following to the main text:

"This yielded αBC = 1.2, [...], with corresponding uncertainties of approximately 20% (complete details of the uncertainties are provided in Table S1)."

The new sentence reads as follows:

The high α values obtained for the organic fractions are consistent with previous measurements for BrC containing POA (e.g. Chakrabarty et al., 2010, 2013).
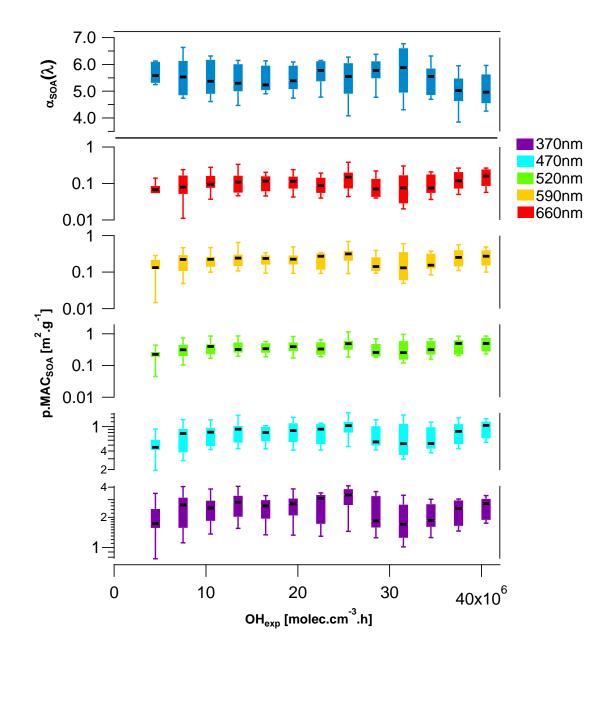
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32.   Fig. S10: I find that the use of the log scale for the y-axis makes it difficult for the reader to see what sort of changes did/did not occur. Variability in the AAE over so many orders of magnitude is not expected, but a factor of 2 would be reasonable. Thus, a linear scale should be used.

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In response to the reviewer comment, we have modified the figure to better illustrate the variability in our data.

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33.   L424: In Fig. S13, and Fig. 6, it is unclear why the authors fit only the "aged" data.
Why exclude the primary, especially in Fig. S13? Because the relationship is visibly much
worse? This goes to the statement about sensitivity to Mie calculations.

1261

1262 We apologize; this information went missing during revisions of an earlier manuscript draft.
1263 We excluded the primary because our assumption of particle sphericity inherent in the Mie
1264 calculations is generally more likely to be violated for fresh than for aged combustion
1265 particles. Similarly, our interpretation of the SMPS-measured mobility diameter as
1266 representative of a physical particle diameter is violated in the presence of fractal-like soot
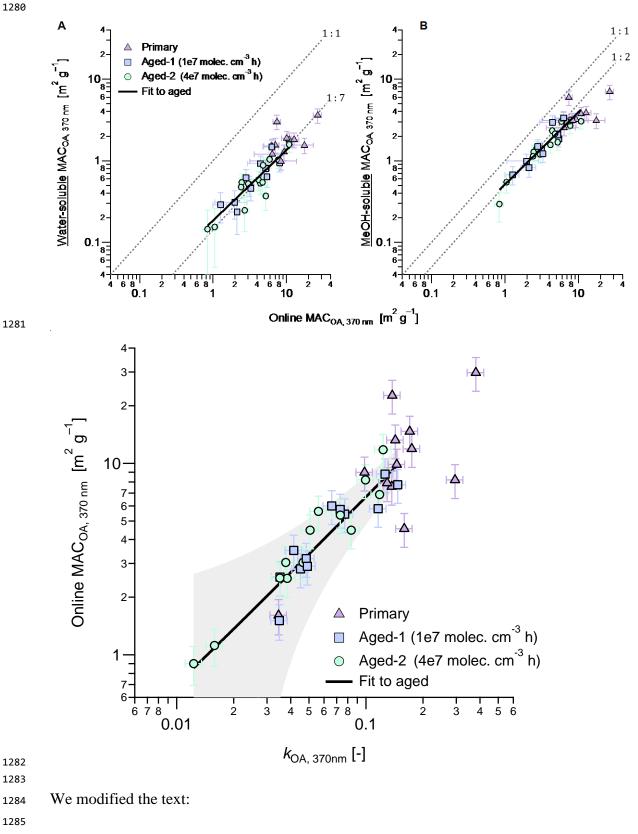1267 particles, which have a shape factor significantly different from unity.

1268

1269 The variability in our primary results is interpreted as illustrating directly the impact of this
1270 variability on our analysis. This variability will reflect differences in burn conditions as well
1271 as the chaotic impacts of the combustion process (for example, uncontrolled differences
1272 between time spent in pre-ignition [where most OA is emitted] versus flaming phases [where
1273 most BC is emitted], between the physical distribution of flames during each burn, etc.).

1274

1275 In spite of these simplifications, the fresh data in Fig. 6 generally fall on the best-fit line for
1276 the aged data. The fact that they fall on the best-fit line is a direct demonstration of the
1277 magnitude of the uncertainties in our retrieved OA MAC.

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1279 We have revised the figures and captions for clarity. The revised figures are:

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1284   We modified the text:

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1286   L431. *The data in Fig. 6B show that the methanol extracts correspond to a MAC*
1287   *about 50% smaller than the online data. The scatter in the data is significantly*
1288   *reduced for the aged data (note that, in this analysis, aged OA refers to the sum of*
1289   *POA and SOA, since the reported values represent all OA after aging). This reduced*

scatter is expected, considering that aging is likely to result in more-spherical
particles. We have assumed particle sphericity when interpreting the SMPS data and
performing the Mie analysis. While the propagation of quantifiable uncertainties
leads to an error estimate of ~25%, considering the simplifiations that were necessary
for the Mie analysis, we consider a 50% closure to be an adequate agreement.
Despite this, we cannot exclude additional methanol insoluble brown carbon.
Conversely, the fit in Figure 6A indicates that the apparent MAC of water-soluble
species was a fourth of the respective methanol MAC, according to the slope of only
12 ± 3%. Only the aged data have been fit to illustrate this point. This strong
disagreement shows that the BrC in our samples was hardly water soluble, even for
the most aged samples. As we expect that the majority of OA in our samples formed by
wood pyrolysis (Di Blasi, 2008; Corbin et al., 2015b; Shafizadeh, 1984), we can
compare our results directly to those of Chen and Bond (2010), who also found that
primary wood-pyrolysis BrC was water insoluble. Moreover, the water-insoluble
nature of the light absorbing components of SOA is in line with the results by Bruns et
al. (2016) who showed that SOA precursors during these experiments were
predominantly aromatic compounds.

We modified the Figure 6 caption:

Figure 6: Comparison of the MAC_{OA}(370nm) of aged aerosols determined from online
and offline absorption measurements. The offline filter-extraction method directly
quantified properties of total OA (ordinate), while the average of MAC_{SOA} and
MAC_{POA} weighted with respective mass concentrations is shown on the abscissa. The
panels show offline measurements of (A) water-soluble OA, (B) methanol-soluble OA.
Fits are to aged data only due to the significantly smaller scatter of those data,
although primary data on average follow similar trends. The fitted slopes and
intercepts are, respectively, (A) 0.13 ± 0.02 and 0.05 ± 0.06 m^2g^{-1} and (B) 0.12 ± 0.1
and 0.38 ± 0.03 m^2g^{-1}.

We modified the Figure S13 caption:

Figure S13: MAC_{OA} at λ = 370 nm calculated from aethalometer measurements vs.
k_{OA} at λ = 370 nm from the UV/visible measurements of the methanol extracts. The
shaded region shows the 90% confidence interval of a weighted orthogonal
regression (slope 66 ± 9 m^2g^{-1}, intercept 0.0 ± 0.3 m^2g^{-1}) to illustrate the relatively
small range of variability in the data for aged samples.

34.   L427: Fig. 6 normalizes out any uncertainty/variability in the measured [OA], because both absorption values are normalized by this. Fig. 4, in contrast, does not. How can the authors rule out the possibility that there is not some variability in the measurement of OA between burns, perhaps dependent on particle shape or variability in bounce in the AMS (which can differ between POA and SOA)?

Fig. 4 actually does normalize the absorption by [OA], so we are not sure which figure the reviewer had in mind. Our goal with Fig. 6 is to relate the offline and online absorption measurements, in such a way that any unknown uncertainties would influence scatter in the plot.

The bounce-related collection efficiency of the AMS was concluded as close to 1.0 for wood-burning OA in the literature reviewed by Corbin et al. (2015b; in their Section S1.2). From recent results from our group using a similar setup, we have measured with an AMS primary organic aerosol rich emissions from smoldering biomass and found the POA collection efficiency to be close to 1.

We reanalyzed our SMPS, AMS, and eBC (MWAA-calibrated AE33) data for the present study by fitting the SMPS mass (predicted with a density of 1.5 g/cm3) against the total PM mass predicted as AMS OA + eBC. The 95% CI of the slope of this fit corresponds to a CE of 0.7-1.0 (relative 19% relative uncertainty), consistent with the literature cited above. Combining this 19% uncertainty (slope uncertainty) with the 30% uncertainty already assigned to the AMS OA (largely reflecting uncertainties in RIE) in quadrature results in a 36% uncertainty in AMS OA, which we have updated in the text.

Shape-related collection efficiency issues in the AMS are unlikely as such issues mainly come into play when transmission through the AMS lens is considered. That is, shape itself is not an issue in the AMS except as it affects aerodynamic diameters (DeCarlo et al., 2014). In our study, particles were large enough that transmission and therefore shape-related issues were minor.

We modified the text:

> L94. *Uncertainties related to particle collection efficiency in the AMS are considered negligible for the relatively-large particles sampled here, which in terms of volume are within the size range transmitted efficiently by the AMS aerodynamic lens (Liu et al., 2007). The collection efficiency of wood-combustion OA is expected to be unity (Corbin et al., 2015b).*

References:

Corbin, J. C., Keller, A., Lohmann, U., Burtscher, H., Sierau, B. and Mensah, A. A.: Organic emissions from a wood stove and a pellet stove before and after simulated atmospheric aging, Aerosol Sci. Technol., 49(11), 1037–1050, doi:10.1080/02786826.2015.1079586, 2015b.

DeCarlo, P. F., Slowik, J. G., Worsnop, D. R., Davidovits, P., and Jimenez, J. L.: Particle morphology and density characterization by combined mobility and aerodynamic diameter measurements. Part 1: Theory, Aerosol Sci. Technol., 38, 1185– 1205, 2004.

Liu, P. S. K., Deng, R., Smith, K. A., Jayne, J. T., Williams, L.R., Canagaratna, M. R., Moore, K., Onasch, T. B., Worsnop, D.R., and Deshler, T.: Transmission efficiency of an aerodynamic focusing lens system: comparison of model calculations and laboratory measurements for the aerodyne aerosol mass spectrometer,Aerosol Sci. Tech., 41, 721–733, 2007.

35. L432: This 46% must state that it is for aged OA only. It remains unclear to me why the primary is excluded.

This statement was removed following comment #33.

36. L440: Is this a fair comparison, given that the authors have focused on the aged OA?

This comment is deprecated following our response to comment #33, but we would also like to point out that we did point out in the text that Chen and Bond studied primary and not aged OA.

37. L431: Are these fits forced through zero?

No fits in this work were forced through zero, but all intercepts were not significantly different from zero. We acknowledge that it is our responsibility to report those intercepts clearly and have updated the text where fits are still reported (the present fits are no longer discussed, see response to comment 33).

38. L441: The authors seem to be implying that SOA formed from oxidation of aromatic precursors is not especially water soluble, or at least less soluble than in methanol. The authors might consider citing e.g. (*Zhang et al.*, 2011), to strengthen this argument.

Thank you, we have added this citation in the corrected version of the manuscript.

Reference:

Zhang, X., Lin, Y. –H., Surratt, J. D., Zotter, P. and Prévôt, A. S. H.: Light-absorbing soluble organic aerosol in Los-Angeles and Atlanta: A contrast in secondary organic aerosol, Geophys. Res. Lett, 38, 2011.

39.   Fig. 7: As already noted above, I find the comparison here insufficient. Saleh et al. (2014) and Lu et al. (2015) report values not at 370 nm. This is unclear. Also, the line shown for Lu et al. (2015) appears to be incorrect. See their Fig. 1D. Further, and importantly, the Lu et al. (2015) data are largely, although not entirely, derived from the Saleh measurements. Thus, they are not really an independent assessment.

We have replied to comment #26 about the data in Fig. 7 and we think we have addressed all the points raised by the reviewer in this question. We acknowledge that the Saleh and Lu data sets are not entirely independent, but have treated the corresponding parameterizations as independent for lack of any method to disentangle their interdependence. We expect other studies to cite Saleh and Lu's separately. Note that we have not focused on the more comprehensive Lu data set because the Saleh dataset represents biomass burning specifically, which we have also studied in the present work.

Figure 7: Imaginary part of the OA refractive index at 370 nm, obtained from offline UV/vis spectroscopy of methanol OA extracts, plotted as a function of fOA. The data could be empirically represented by a linear function in the log-log space, in the measurement range. The ordinary least-squares fit is $(k_{OA,nm})$ = $log(M_{BC}/M_{OA})(0.51\pm0.07) + (-0.98\pm0.05)$. Also shown are parameterizations of $k_{OA}(370nm)$ for open burning against M_{BC}/M_{OA} estimated based on the $k_{OA}(550nm)$ measurements in Saleh et al. (2014) and Lu et al. (2015), using the k_{OA} wavelength dependence reported by the respective authors.

We have also updated the text at line 460:

The parameterizations reported by these authors are included in Fig. 7, where the wavelength dependence reported by those authors has been used to adjust their parameterizations to 370nm.

1465

We do agree with the reviewer that k_{OA} will likely tend towards k_{POA} and k_{SOA} when M_{BC}/M_{OA} is very large and very small, respectively. However, within the range covered, a line is the simplest model which can describe our data adequately and the linear fit used is simply empirical. We do not aim to include a complete physical interpretation in this simple fit With our fit in Fig. 7, we mainly wish to emphasize that k_{OA} can be described as a function of M_{BC}/M_{OA} only. For clarification we have modified the figure caption as follows:

> *Figure 7: Imaginary part of the OA refractive index at 370 nm, obtained from offline UV/vis spectroscopy of methanol OA extracts, plotted as a function of **fOA**. The ordinary least-squares fit is $\mathbf{log(k_{OA,370nm})} = \mathbf{log(M_{BC}/M_{OA})(0.51\pm0.07)} + (\mathbf{-0.98\pm0.05})$ and illustrates that the observed k_{OA} can be described as a function of M_{BC}/M_{OA} with reasonable accuracy, regardless of the degree of aging. Also shown are parameterizations of $\mathbf{k_{OA}(370}nm)$ for open burning against M_{BC}/M_{OA} estimated based on the online $\mathbf{k_{OA}(550}nm)$ measurements in Saleh et al. (2014) and Lu et al. (2015), using the $\mathbf{k_{OA}}$ wavelength dependence reported by the respective authors.*

We agree, however, we do not think that this calculation will shed any new insights into our data set. In Fig. 7, we have only chosen to use M_{BC}/M_{OA} as an abscissa because previous studies have used this quantity; our goal in Fig. 7 is a comparison of our results with related literature. We do not find this ratio to be particularly meaningful or interesting physically, but we acknowledge that future studies are likely to measure it as well and so it provides a useful basis for comparison.

42.   The origin of these "uncertainties" is unclear. They are explained later for f_OA, but for the MAC values it is not abundantly clear.

We apologize but we do not see which part of the manuscript the reviewer is referring to. We assume that the reviewer is referring to the fitted MAC values, which we have commented on above and adjusted the manuscript to include.

43. L486: This statement regarding mass yields of SOA requires much further detail.

The analysis of SOA gas phase precursors has been thoroughly presented in Bruns et al. (2016) and the discussion about SOA yields is beyond the scope of this study. As this statement is not required for the understanding of the paper we have removed it in the corrected version of the manuscript. The section now reads as follows:

> The $M_{SOAP,WLC}/M_{POA,WLC}$ was on average equal to 7.8 (GSD = 1.4) and kOH was estimated as 2.7×10^{-11} molecule^{-1} cm^3 (GSD = 1.4), consistent with the SOA precursors chemical nature measured (e.g. PAH and phenol derivatives) by a proton-transfer-reaction mass spectrometer (PTR-MS) (Bruns et al., 2016, 2017). These high rates and enhancement ratios indicate the rapid production of SOA.

44. L512: A note about terminology. I am not certain that "error analysis" is appropriate here. Variance in the POA fraction is not "error." It is variability. A substantial aspect of this "error analysis" is really just a "sensitivity analysis." I suggest that the authors limit the term "error analysis" to when they are truly considering errors, and use some other term when they are considering variability. This is true here and elsewhere.

We agree with the reviewer and have changed the word "error" to "sensitivity".

We have also made the requested modifications related with the section entitled "*Uncertainties and variability in MAC$_{BC}$, MAC$_{POA}$ and MAC$_{SOA}$*".

45.   L499: The authors should clarify the origin of the solar irradiance data that they have used.

We have now added the reference on which the solar irradiance data are based:

> Gueymard, C.; Myers, D.; Emery, K. "Proposed Reference Irradiance Spectra for Solar Energy Systems Testing," Solar Energy, 73, 6, 443–467, 2002.