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Interactive comment on "Laboratory studies of fresh and aged biomass burning aerosols emitted from east African biomass fuels – Part 1 – Optical properties" *by* Damon M. Smith et al.

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The full response is attached as a pdf file with figures. RESPONSES to REFEREE #1

The paper summarizes results on optical properties of aerosols generated by burning of 3 common fuels in Africa. Some measurements were carried out on fresh BBOA, some on BBOA aged in the dark, some on photooxidized BBOA in the presence or absence of additional VOCs (typical urban aromatics). Fuels were burned at low (500 C) and high (800 C) temperatures. Non-refractory chemical composition of the aerosol was also measured, although not really discussed here. Single scattering albedo data, determined on size-selected aerosols, at mid-visible confirmed production of more absorbing aerosols under high temperatures. There was not a strong wavelength dependence to SSA between 500-570 nm. Dark aging of the BBOA resulted in increase in SSA possibly due to condensation of SOA and increase in the scattering cross section. The novelty of the experiment is in the selection of the fuels. However, I see some flaws in the approach (contribution of multiply charged particles was not corrected for in Mie calculations); the paper does not seem complete without the description of the compositional measurements (some conclusions are drawn on the composition of SOA without providing any support; there were no measurements of BC, yet overall refractive indices were estimated to use in Mie calculations); last the chamber oxidation experiments in the presence of additional VOCs don't seem to have worked. There were also several sentences that were confusing and need to be rephrased/clarified. Overall, I don't find the quality of this paper appropriate for ACP and cannot support any revisions.

AUTHORS RESPONSE: We thank the reviewer for valuable and helpful suggestions. Embarrassing grammatical errors and sentences that seemed confusing will be corrected in the revision and changes made to the manuscript are provided here. Detailed responses are provided on the scientific questions raised. The authors feel that the reviewer either misunderstood our descriptions and overlooked some aspects of the paper. We will show this is indeed true in our detailed responses with relevant references to each question and comment. We feel that there is rush to dismiss and devalue the work and undermine the paper instead of giving us a chance to respond and clarify our claims. We hope our responses and explanations will convince the referee to make a different final determination. Three main issues raised by the reviewer in his/her the first paragraph is discussed below:

A. “Non-refractory chemical composition of the aerosol was also measured, although not really discussed here”

We would like to point out to the reviewer that this work is presented and submitted as a two-part paper, where part 1 was focused on optical properties and part II focused

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on chemical composition and characterization published in ACPD (Smith et al., 2020). Further integration of the two manuscripts will be done, and summaries from Part 2 will be incorporated into the revised manuscript. Following the paragraph contrasting eucalyptus and acacia combusted at 800o C, the following text will be added to line 393:

“In the companion paper to this (Part 2), methanol extracts from BBA collected on Teflon filters were analyzed by ultra-performance liquid chromatography interfaced to both a diode array detector and an electrospray ionization high-resolution quadrupole time-of-flight mass spectrometer (UPLC/DAD-ESI-HR-QTOFMS) in negative ion mode. This was used to determine the relative abundance and light-absorption properties of biomass burning organic aerosol constituents. MS analysis of BBA extracts from combustion at 800 °C revealed very little difference between the two fuel types, suggesting that there are either very few BrC species produced for either fuel under these combustion conditions, or there are numerous species that are essentially the same between the samples. However, given that Eucalyptus has a higher SSA than Acacia, this would suggest that Eucalyptus has more non-absorbing OA, or at least less absorbing than BC. Since it is Acacia that appears to have many more low-abundant organic constituents, several possibilities exist to explain these differences in SSA, as explored in more depth in Part 2. It is likely that Eucalyptus combustion products are not captured by some aspect of the extraction and UPLC/DAD-ESI-HR-QTOFMS analyses, that the observed differences in SSA are due to morphology differences, or some combination thereof. One potential explanation would be the presence of significant amounts of eucalyptol in the BBA, which is a large fraction of Eucalyptus oil, and is a cyclic ether that lacks any basic functionality amiable for negative ion mode analysis, has good solubility in alcohols, and does not absorb in the UV and visible. An examination of the UV-Visible spectra from the DAD shows no absorbing species in either region.”

The following paragraph will be included in manuscript following the discussion of fresh emissions produced by combusted at 500o C, added to line 410:

“Chemical analysis revealed that, when combusted at 500 °C, eucalyptus and acacia had a variety of compounds in common, such as lignin pyrolysis products, distillation products, and cellulose breakdown products. Several lignin pyrolysis products and distillation products are more prevalent in Eucalyptus than Acacia, while pyrolysis products of cellulose and at least one nitroaromatic species were more prevalent in Acacia. Given that these lignin pyrolysis and distillation products are known chromophores and are more prevalent in Eucalyptus than Acacia, while Acacia has a higher abundance of non-chromophores derived from sugars and cellulose, one would assume that Eucalyptus would be more absorbing in the visible (i.e. have a lower SSA) than Acacia. Despite the chemical analysis not capturing absolute amounts of OA, Acacia was found to have an SSA that is higher than Eucalyptus by 0.1 to 0.2, which is consistent with chemical measurements. This suggests that Acacia has either larger absolute amounts of non-chromophore compounds or Eucalyptus has a greater quantity of chromophores whose absorptive properties extend to the 500 – 570 nm region of the visible spectrum. An analysis of the chromatographically-integrated UV/Visible spectrum shows that there are chromophores whose absorption features peak $\sim$290 nm and extend into the 500 – 570 nm region, though a normalized spectrum does not appear to show drastic differences between species.”

Later, starting at line 504 with the sentence “We attempted. . .”, the remainder of this paragraph and the next (after Figure 7) will be replaced with the following text:

“Despite the use of anthropogenic VOCs a concentrations larger than those average values found in urban and suburban regions of South Africa, no distinct effect was observed for SSA values of BBA produced during combustion at 500 °C. While it is possible that the relatively long aging time could obscure some of the effects due to the presence of VOCs, it is also possible that combustion products dominate molecular species and the effects of additional VOCs are insignificant. The later would suggest that anthropogenic pollution does not seem to affect the optical properties of BBA. Indeed, in examining the effects of aging on the chemical composition of BBA shows very

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few species that could be attributed to anthropogenic VOCs; specifically, only dihydroxyphthalic acid produced from xylene. For both Eucalyptus and Acacia, an isomer of dihydroxybenzene, such as resorcinol or catechol, was removed to the highest degree from the fresh BB aerosol upon photochemical aging. Generally, very few compounds were produced to a significant extent and both fuels were dominated by loss of chromatophoric lignin pyrolysis and distillation products. Not surprisingly, the associated absorbance from these chromophores, mostly from 200 – 350 nm, also attenuated with respect to age. This may be caused in part by the photo-bleaching effect created by irradiation of UV light for 12 hours, heterogeneous OH oxidation, and SOA formation of non-chromophores. This fact suggests that a study with a higher temporal resolution is needed to simulate the impact of the VOCs on aerosol SSA, where continuous or much more frequent measurement are needed to determine impact of urban pollution on aerosol single scattering albedo. Such a study, using continuous measurements, is not possible for our setup when particles are also size selected. Like dark aged conditions, we were not able to estimate SSA for combustion at 800 oC due to the low particle concentration and highly absorbing nature of the aerosol. A chemical analysis of aged BBA produced at this temperature revealed very few changes, suggesting there are few molecular species produced by combustion at this temperature."

B. "However, I see some flaws in the approach (contribution of multiply charged particles was not corrected for in Mie calculations"

There seems to be a misunderstanding regarding Mie calculation. As stated on P 11, L 356-358, we estimated the SSA of size-selected aerosol using the refractive indices estimated by Bond and Bergstrom, (2006) and Levin et al. (2010) for BC and bulk aerosol. The whole purpose of this effort was to explore if our estimated SSA for 200 nm size particles was impacted by the presence of multiply charge particles. The impact of multiply charged particles on SSA for 300 and 400 nm size particles was minimal. We only run the Mie models to make a qualitative comparison of Mie result with our measurements, focusing on the SSA dependence on particle size. The disclaimer “Neither line is a fit to the data.” Will be added to the caption of Figure 3. Further discussion on multiply charged particles is provided below in response to reviewer comments on L361-362.

C. “Some conclusions are drawn on the composition of SOA without providing any support”

The focus of the work is not SOA formation. However, as we observe changes in SSA we looked at the data more closely as stated on P 16, Line 460, we hypothesized that there could be SOA production during dark aging, and we cited previous work showing nitrogen-containing SOA production under nighttime conditions as potential chemical basis for this observation. As clearly stated in our discussion and in the abstract section, we hypothesized SOA production during dark aging but never definitively concluded about composition of SOA, as we do not have measurements for these experiments. We were inferring to work that was previously reported and focused on SOA formation during dark aging. (Tiitta et al., 2016;Li et al., 2015;Hartikainen et al., 2018). It seems that our statement regarding this was misunderstood and needs further clarification, which will be included in the revised draft. Furthermore, the statement “there were no measurements of BC, yet overall refractive indices were estimated to use in Mie calculations” is not valid in our work given we never stated that we estimated refractive indices nor do we present any refractive indices of our own at any point. We have provided more details about this in response to the comment on L357. The reviewer is mischaracterizing things as “additional VOCs don’t seem to have worked”. Just because something didn’t significantly change, doesn’t mean it didn’t work. It just means that it doesn’t seem to have had an impact.

Other specific comments are listed below: - L 20-21: Bad grammar – Agreed. It will now read as “This work represents the first such study of the optical and chemical properties of three wood fuel samples used commonly for domestic use in east Africa.” L 23-24: bad grammar – Agreed: “values are in the range between 0.287 and 0.439 while the SSA for fuels combusted at 500o C, the range is between 0.66 and 0.769”

L32: “injecting” and not “ejecting” – Agreed: ejecting will be replaced by injecting L33: it’s unclear why measurements after 12 hrs of injecting VOCs and BB aerosol in the chamber were not useful. – We believe that measurements should have been made soon after injecting the VOC’s L 43: “measuring the. . .” – “of” will be removed L47: “partial evaporation. . .” – Agreed: partial evolution should be partial evaporation L85: “authors’. . .” – Will be replaced with “To our knowledge”

L112: The sentence related to drying of the fuels should be moved earlier, before the weighing discussion. –

AUTHORS RESPONSE: We agree with the reviewer. Sentence related to drying of the fuels will be moved earlier, before the weighing discussion.

L115: the EF mentioned is for tropical forests, which is not really representative of the African fuel’s studies here. Please provide your justification for using this value. –

AUTHORS RESPONSE: We agree with the reviewer. We will delete the whole sentence talking about the EF and will add the following text:

“During all the experiments, we normally burned 0.5 grams of fuel which produced about 600 to 800 μg m-3 of mass loading in the chamber. The mass loading was estimated by determining the total aerosol volume, based on measuring the volume distribution with an SMPS and assuming a density of 1 g cm-3 for fresh aerosol.”

L164-165: It’s unclear what is meant here “SSA and AAE were derived from these relationships using observations of CO, CO2, OA, and BC.” –

AUTHORS RESPONSE: We agree with the reviewer. We will delete the whole sentence.

L187: Temperature change even if very small should be included since it can impact gas-aerosol partitioning and evaporation of BB-POA. –

AUTHORS RESPONSE: Chamber temperature started at room temperature (around

20 °C or slightly above) and increased to a maximum of 30 °C after 5 hours of use when all the UV lights were turned on, with most of the increase happening within the first hour (Smith et al., 2019). The figure below is the temperature profile for different runs.

Fig1. Temperature profile of the chamber. Runs 1b and 4b were done on the same day as runs 1a and 4a, respectively, with initial temperatures higher for 1b and 4b than 1a and 4a.

L216-217: Consider indicating what the aerosol number concentrations, and absorption and scattering coefficients typically were after 24-hr flushing and before start of a new expt. –

AUTHORS RESPONSE: Number concentrations were measured and must be below threshold values before a new experiment could begin. These values were typically around 25 – 40 particles cm-3 as measured by the CPC. Absorption and scattering at these low number concentrations were not measured and gives a typical background mass loading of 1ïĄ■gm-3.

L221: what does it mean that “The experiments were repeated after keeping the BB aerosol in the chamber overnight (24 hours) without the UV lights”? –

AUTHORS RESPONSE: We are sorry for the typo. Now the sentence will read “For dark aging, measurements were repeated after 12 hours without the UV lights”.

L231: indicate manufacturer and level of purity –

AUTHORS RESPONSE: These mixtures were composed of benzene (≥99.9%, Sigma-Aldrich), toluene (99.99%, Acros Organics), and ortho-xylene (99%, Alfa Aersar). This information will be added in the revised manuscript.

L245-251: Typically, urban concentrations of VOCs are expressed as volumetric mixing ratios (ppmv or ppbv, etc). Is that not the case for the aromatics indicated here? If not, to clarify, you need to include ppmm or ppm by mass. If indeed the mixing ratios of

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5:14:6 was ppmv based, then one expects different volumes (and different masses of them based on their density) of each to be injected and the calculation here is not correct. –

AUTHORS RESPONSE: The reviewer is correct. There seems to have been some miscommunication among investigators during the experimental design of this particular experiment. See our response to the next comment, where both will be addressed.

L254-256: what VOC mixing ratios were achieved in the bag so readers can compare them with the typical urban mixing ratios of these VOCs in Africa? –

AUTHORS RESPONSE: The two paragraphs and the first table, from lines 239 – 263, will be replaced with the following text:

“To represent a polluted urban environment, we used emission inventory for urban environments from South Africa. This does not necessarily represent the east African emission inventory, but this serves as a baseline, since this is the only available data to us for the continent. This was obtained from South African Air Quality Information System (SAAQIS) and included concentrations of NOx, NO, NO2, CO, O3, benzene, toluene, ortho-xylene, and ethylbenzene for several South African Sites (Diepkloof, Kliprivier, Three Rivers, Sharpeville, Sebokeng, Zamdela, Thabazimbi, Lephalale, Phalaborwa, and Mokopane). The VOC data was obtained from the two weeks (M-F) of July 11 – 15 and July 18 – 22, which was in the middle of the peak burning season for South Africa for the year 2016. The urban areas (Diepkloof and Kliprivier) had combined average mixing ratios of 1.16, 3.48, and 1.44 ppbv for benzene, toluene, and o-xylene, respectively. Suburban areas (Three Rivers, Sebokeng, and Zamdela) had combined average mixing ratios of 1.69, 4.02, and 0.70 ppbv for the aforementioned gases, respectively. Interestingly, suburban regions had somewhat higher average benzene and toluene mixing ratios, though o-xylene was only half the average urban concentration. A mixture was prepared using equal by volumes of benzene, toluene, and o-xylene, and 2.5 mg was injected by syringe into a U-shaped glass tube attached to the chamber. This tube was then flushed by zero air into the chamber. This resulted in a mixing ratio of 29.7, 24.9, and 21.9 ppbv for benzene, toluene, and o-xylene, respectively. The concentration injected into the chamber was approximately 7 – 26 times more concentrated than values found from urban South African emissions and 6 – 18 times more concentrated than suburban values. The reason for these elevated levels was mostly due to sample preparation constraints, since the amounts needed for an exact match were too small for our scale to weigh appropriately. Concentrations in the chamber were intentionally higher than atmospheric conditions, in order to age the BB aerosol faster and accentuate the potential effect of SOA."

L267: what does "710 μm impactor inlet" mean? It seems 710 um is not related to the size cut. – AUTHORS RESPONSE: That is the diameter of the impactor used right before our DMA. We will rewrite the sentence as "BB aerosol was size selected for optical property measurements by passing the sample through an impactor inlet with a 710 μm nozzle (3.8 ïĄ■m diameter cut point), charge neutralizer (TSI model 3081), and a long differential mobility analyzer (DMA) (TSI model 3080).

L269- 270: how was the concentration of multiply charged particles accounted for? Those can significantly impact the measured optical coefficients given their larger physical size, even if their number concentration is not high. This is mentioned in lines 318-320, but still no consideration is given for correcting 200 nm particle concentrations.

AUTHORS RESPONSE: As mentioned in the manuscript, concentration of multiply charged particles was not considered in this study. We completely agree with the reviewer that this will significantly impact the measured optical coefficients and cross sections. However, the impact of multiply charge particle on SSA is low (ratio of scattering to extinction coefficients/cross-section). We recently acquired an aerosol particle mass analyzer (APM) and after this comment was posted, we did an experiment using the APM in-line after the DMA and performed optical property calculation for freshly emitted BBA. Detail about the measurement strategy is given by Radney and Zangmeister, 2016). It was found that, due to the presence of multiply charged particles, our

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reported SSA values were overestimated by a maximum of 8% for 200 nm &300 nm particles. This information will be included in the revised manuscript.

- Equation 2: need to define tau and tau0, sigma_ext and N(CRD). –

AUTHORS RESPONSE: We thank reviewer for pointing this out. We will define those terms in Equation 2 in the revision.

L294: Since the CPC is placed after the optical instrument, what is the fractionall loss of particles in the CRD and nephelometer? –

AUTHORS RESPONSE: We thank reviewer for pointing this out. We used PSL sphere size standards to estimate the losses in CRDS and Nephelometer. The estimated particle losses in CRDS are 14.2, 14.7, and 11.4 percent for 200, 300, and 400 nm sizes, respectively, whereas for Nephelometer those losses are 8.6, 7.1, and 6.3 percent for those respective sizes. We accounted those losses in our final calculations of extinction and scattering coefficients, as discussed in depth in our previous publication (Singh et al., 2014) and this work is cited in the paper.

L316: it is worth indicating what each of these uncertainties are. –

AUTHORS RESPONSE: We will report those uncertainties. The sentence will now read "The calculation flow determining average extinction cross section (σext), absorption cross section (σabs), and single scattering albedo (ω), was already described (Singh et al., 2014)(Singh et al., 2016). The estimated uncertainties are 11%, 15%, and 2.1% for σext, σabs, and ω".

L330-331: It's unclear what the CE value was and how it was determined for this one species. What assumptions need to be made to justify using the same CE for all the species? –

AUTHORS RESPONSE: We appreciate reviewer for mentioning this. This sentence will now read as "Since this work does not use mass loading in a quantitative way, we chose a collection efficiency of 1 for all species, similar to a previous study (Ng et al., 2011)".

L340: this sentence needs to be rephrased "The optical properties were also measured as a function of different forms of aging: dark aged, photochemically and photochemically aged with added VOC's lights on in the presence of VOCs injected into the chamber before particles were introduced at both temperatures."

AUTHORS RESPONSE: We agree with reviewer. We will rephrase the sentence as "The optical properties of dark and photochemically aged aerosol were also measured".

L351 and 363: section 2.3.1 is not in the paper.

AUTHORS RESPONSE: We thank reviewer for pointing out this typo. We will correct that as section 2.4 now.

L357: How was concentration of BC taken into account to calculate the SSA? And how were the aerosols treated? Internal or external mixtures? These details need to be fully explained.

AUTHORS RESPONSE: We think there is a misunderstanding of what we did. We ran Mie model based on the RI of BC from Bond and Bergstrom, (2006) and RI of bulk aerosol from Levin et al, (2010). Based on these model result, we estimated the size selected SSA at 532 nm and did comparison with the SSA calculated from our measurements. The whole purpose of this was to point out two facts: 1. Aerosol formed during 800o C burning are very absorbing (equivalent to pure BC) and 2. Due to the presence of multiply charge particles, our estimated SSA are biased, especially for lower size particles.

L361-362: why would the impact of multiply charged particles not be present in lower combustion temperature samples? How different were the chamber size distributions under these conditions? The geometric mean for all fresh aerosols seems to be the same and 50 nm (Fig 1). - Figure 3. The legend needs to include the imaginary number

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indicator “i”.

AUTHORS RESPONSE: The typical particle size distribution (corrected for multiple charging) of two different burning temperatures is shown in Figure a. As depicted in the distributions, the higher temperature burn would be more impacted by multiply charged particles because the relative particle concentrations at 200 nm are lower compared to equivalent mobility particles with charges of +2 (314 nm) and +3 (418 nm). On the other hand, for lower temperature burn, concentration at 200 nm size is about 3.5 time larger than that of +2 (314 nm) and about 9 times larger than +3 (418 nm) size. All these facts suggest that there will be more impact due to multiply charged particles for the higher temperature burn than that of lower temperature burn as depicted in Figure a.

Figure 2: Normalized particle size distribution for 500o C and 800o C burning cases.

We will re-plot Figure 1 with x-axis on log scale (shown below as Figure b), which shows the difference in the geometric mean diameter (GMD) of two different burning conditions. For the lower temperature case, it is typically about 85-90 nm whereas for higher temperature burn it is below 40 nm. We will update the legend for Figure 3.

Figure 3: Same as Figure 1 in the manuscript (x axis in log scale).

L380: Do the dashed lines show the uncertainties in SSA or the variability of the measurements, e.g., standard deviation of the average? Please be specific. –

AUTHORS RESPONSE: The dashed lines represent the propagated uncertainty (1 standard deviation) of the SSA, based on extinction and scattering coefficients. The scattering coefficient error is mainly produced by measurement variability from the nephelometer (∼1%), while extinction errors from the CRDS are mainly influenced by variability in the ring-down time. This has been thoroughly described in our system characterization paper (Singh et al., 2014) cited in this paper.

L391-392: Unlike what’s mentioned here, SSA for eucalyptus aerosol is not uniformly higher or lower than other fuels; please correct this statement. This needs to be corrected in the conclusions as well. – L382-400: The discussion on burn temperature and BC vs BrC emissions gets repeated; consider describing this dependence more concisely and to the point only once. -

AUTHOR RESPONSE: We agree with the reviewer. We will rewrite the two paragraphs (L382-409) and it will read as:

“The range of SSA for different fuels combusted at 800o C is 0.287 to 0.439, whereas the range for the same fuels combusted at 500o C is 0.66 to 0.769. The average MCE for 800o C burn cases is 0.974 ± 0.015 and that for 500o C burn cases is 0.878 ± 0.008. These MCE values suggests that 800o C burns are dominated by flaming phase and 500o C burn cases are dominated by the smoldering phase of the burn. The flaming stage of the combustion produces more black carbon and smoldering stage produces more organic carbon (Christian et al., 2003; Ward et al., 1992), which explains the lower SSA values at higher temperature combustion. The impact of combustion temperature on aerosol can also be separated visually by looking at the color of the collected filter samples, as shown in Figure S1. As evident from Figure S1, aerosol emitted from the 800o C combustion looks black, whereas that from 500o C combustion looks brownish, indicating a visual difference between black carbon dominated and organic carbon dominated emissions from same fuel under different combustion temperatures. Although the variation in SSA of the different fuels is dominated by the burning conditions, there occurs a clear but small dependence of SSA on fuel type under the similar burn conditions.

The range of SSA for combustion at 500o C is comparable to previous studies with similar MCE values (Liu et al., 2014; Pokhrel et al., 2016). On comparing the SSA of the three different fuels under two different combustion temperatures, it is apparent that SSA is controlled more by the combustion conditions rather than the fuel types. There is a larger variation in SSA for the same fuel under two different combustion conditions, compared to the variation due to the inter fuel variability under the same combustion temperature. This result is consistent with a previous study, which showed that SSA is highly correlated with the EC/TC (proxy for the combustion conditions), even for a wide variety of fuels (Pokhrel et al., 2016). A complete list of sizes selected SSA of different fuels measured at two combustion temperatures and under different aging conditions is provided in Table S1."

Figure 5: why is the SSA for 400 nm included here whereas in other plots 300 nm observations were included? What is the reason for having much larger error bar on one of the data points? –

AUTHORS RESPONSE: The reason that this data point seems to be an outlier in terms of its error is because, for that particular experiment, the standard deviation for the ring-down time of the blank was about double what it normally is, even compared to other wavelengths in the same experiment. This propagated to an SSA error of ±0.113. The standard deviation of repeat SSA measurements was only ±0.008, as measurements during this experiment (Olive combusted at 500o C) was extremely reproducible.

L414: The sentence related to the higher uncertainty needs to be rephrased. –

AUTHORS RESPONSE- We thank the reviewer again for pointing out this poorly constructed sentence. It will be rephrased as "This outlier could also be because of the higher error associated with the scattering measured by the nephelometer."

L419-421: I don't think the example with actual values of SSA are needed. Also, the last sentence is stating something that has been known in the community for a long time; therefore, it's not worth reiterating or at least provide proper literature reference. –

AUTHORS RESPONSE: We agree with the reviewer. The sentence with the example is removed and the reference is provided in the last sentence and will read as "This suggests that by simply varying the combustion temperature, we can generate aerosols with very different optical properties and combustion efficiencies (Saleh et al., 2018;Liu et al., 2014)” in the revision.

L431: Why is particle dynamic expected to be different at night compared to daytime such that it would influence nighttime oxidation differently? Please clarify.

AUTHORS RESPONSE: The particle dynamics is different at night since there is, a pronounced increase of particle size and density which was also observed in previous laboratory and ambient measurements (Reid et al., 1998;Zhang et al., 2011). Even though the RH remains the same in our experiments, it was also shown that RH may facilitate heterogeneous reactions during the night (Li et al., 2015).

L461: there’s no basis for suggesting nitrogen-containing OA as opposed to other types of SOA were formed under these dark aging conditions since no information on composition was provided. Please rephrase/remove this in the Conclusions as well. –

AUTHORS RESPONSE: We thank reviewer for pointing this. To make this clear, we are not suggesting nitrogen-containing OA as opposed to other types of SOA. On Line 460 we wrote that “We hypothesized secondary organic aerosol formation as a potential phenomenon happening during dark aging.” Then at Line 461 we refer to the previous studies which concludes higher production of nitrogen containing OA and then we hypothesized this could be a possibility in our case. This was explained in more detail at the beginning of our response.

L487: Section 2.2.1 includes description of photooxidation without additional VOCs –

AUTHORS RESPONSE: We thank reviewer for pointing out the typo. This will be corrected to be 2.2.2

L505-507: the explanation doesn’t seem to be valid. Why can’t it be that the SOA form these VOCs has the same optical characteristic as the SOA formed in the absence of the VOCs?

AUTHORS RESPONSE: We agree that the wording here is strange. We will rephrase it in the revision as: “This is most likely because we took our measurements after 12 hours of aging, which was enough time for the scattering nature of the SOA produced during aging to drive measured SSA values to unity, regardless of the chemical pathway taken due to the addition of VOCs"

L539: it appears that there was insignificant additional SOA formed from oxidation of aromatics that were added to the chamber. What was the NOx level in the experiments? Perhaps the high NOx conditions of the burns lead to low SOA yield from these precursors and therefore no significant SOA is observed. If results from aging in the presence of additional VOC were not conclusive, I suggest removing all the discussion related to it throughout the paper. –

AUTHORS RESPONSE: NOx concentrations were only available during the chamber characterization experiments used for our previous paper. Equipment malfunctions prevented us from measuring this during chamber experiments. However, no additional NOx was added to the chamber during these experiments, only what was available due to combustion.

L 516-517: I'm not following why continuous size-selection was not possible during these aging experiments

AUTHORS RESPONSE: Unfortunately, our current setup does not allow continuous monitoring when particle sizes are being selected. Mainly, the volume of the nephelometer is too large compared with the relatively small flow rate provided by the WCPC. We have found that a flush time of at least 15 – 20 minutes is required to completely replace a sample within the nephelometer with a new one (when changing particle sizes, for example). At this rate, it takes about 4 hours to collect measurements for three different mobility diameters, including blank measurements, repeat measurements, and the time to flush between samples.

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Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-1156/acp-2019-1156-AC1-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-1156, 2020.

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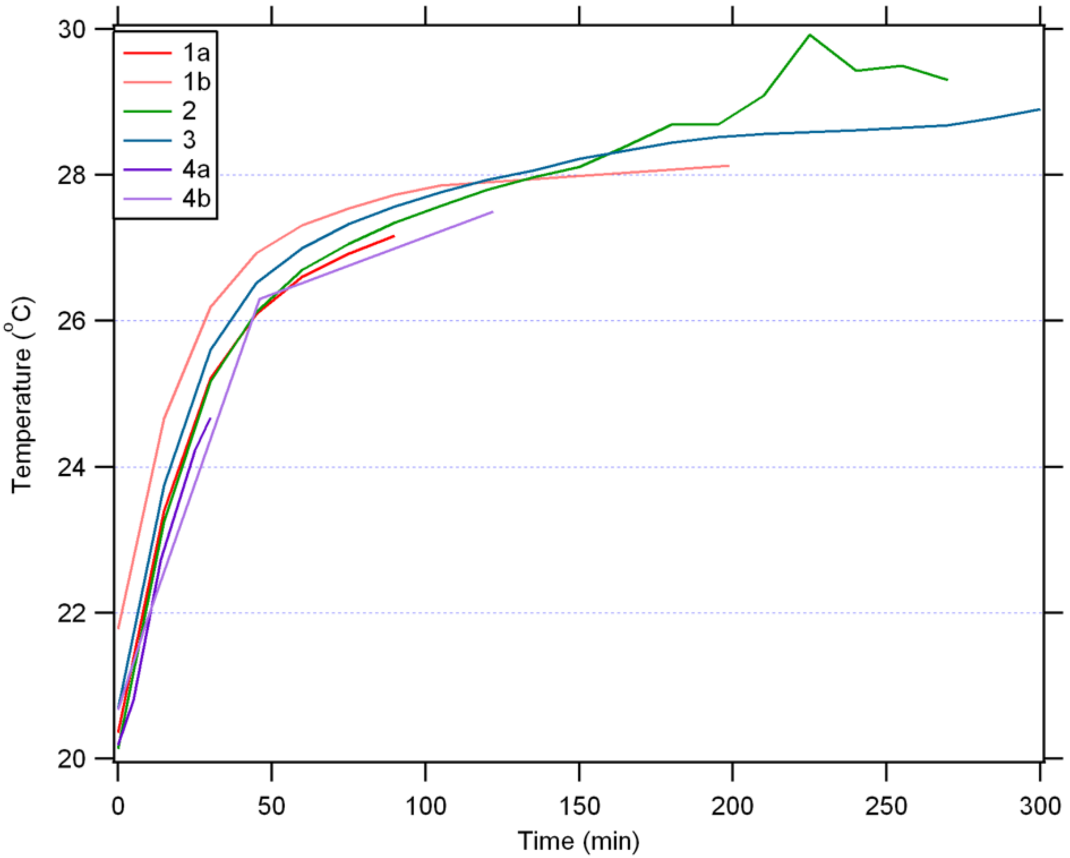

Fig. 1. Temperature profile of the chamber. Runs 1b and 4b were done on the same day as runs 1a and 4a, respectively, with initial temperatures higher for 1b and 4b than 1a and 4a.

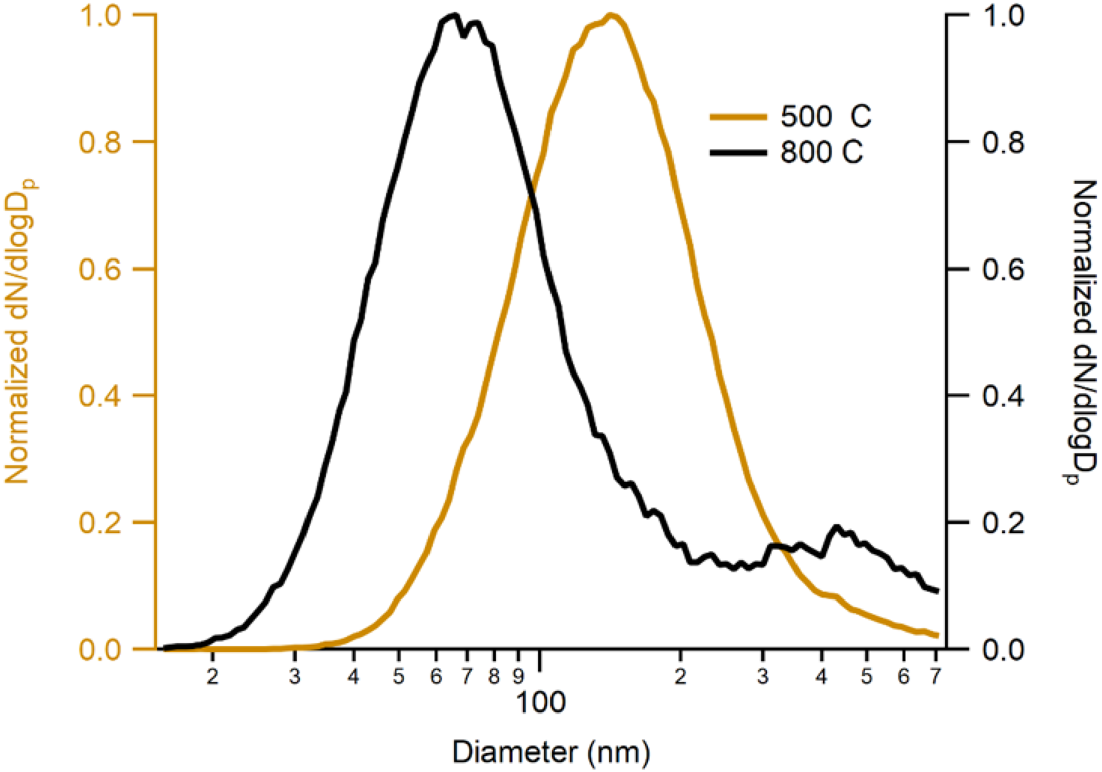

Fig. 2. Normalized particle size distribution for 500o C and 800o C burning cases

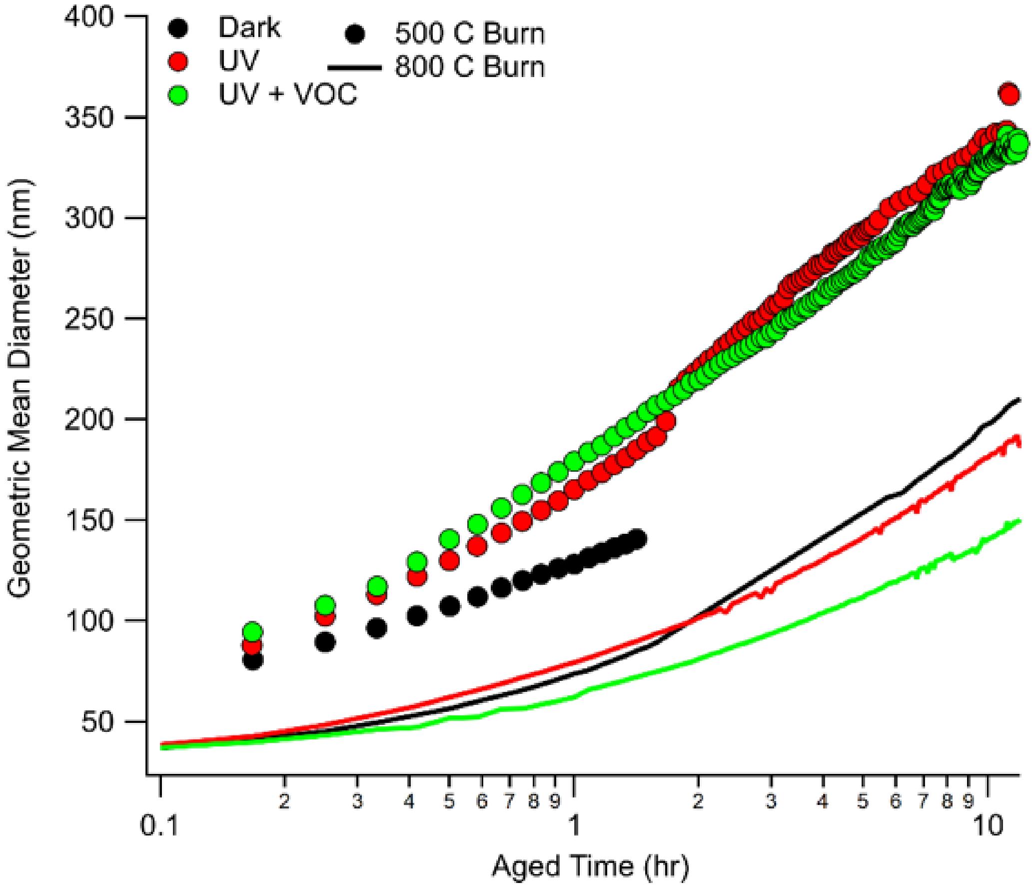

Fig. 3. Same as Figure 1 in the manuscript (x axis in log scale).

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