RESPONSES TO REVIEWER #3

The authors thank the reviewer for his detailed and comprehensive comments that will help make the paper stronger. We will address all the comments and suggested corrections as best as we can, below. Note: Figure numbers are consecutive for the new figures provided for the responses. For a figure from the submitted manuscript the same number is used.

General comments:

REVIEWER COMMENT #1: I fail to see what makes this indoor chamber novel, and how this chamber is different from the numerous indoor smog chambers already constructed and are currently being used. The authors need to be explicit when describing the novelty of their chamber.

AUTHOR RESPONSE: The reviewer is right in indicating that there are numerous indoor smog chambers already constructed; however, we do not claim that the chamber itself is novel. What we consider unique and novel is the "Combustion-Chamber System". Given the unique combination of our custom designed and built instruments and combustion aerosol generation system, our entire chamber system is better described as "Unique". The following are unique qualities of our system: BB combustion and aerosol generation using a tube furnace to produce aerosols under controlled burning conditions (temperature, air flow, oxygen content, and amount of fuel burned) and the ability to clearly visually differentiate brown carbon (often formed at around 450-500 °C), Black Carbon (formed around 650 - 750 °C) and other forms (mixed brown/black carbon) at intermediate temperatures. Figure 1 shows two forms of carbon collected on filters. The generation and introduction of the soot particles into the chamber is described in line 194-197. The generation of soot particles and the introduction method, along with filter and impinger sampling and the integration of the chamber to the cavity ringdown/nephelometer system are not common arrangements, and we haven't come across chambers used in this manner. Many chambers are difficult to clean and therefore the characterization for wall loss and light intensity is a moving-target. Our chamber can more easily have each of the FEP panels cleaned and replaced. We believe the entire system is novel, not just the chamber. There is more to say here about the "novel" nature of the chamber system: We have characterized this chamber with particles that are combustion particles from biomass burning and have expended efforts to fully document the characterization with this manuscript. This adds to the "novel" or more rare nature of the chamber in that many chamber characterization efforts are undocumented, which makes the chamber results suspect and uncertain. Other chambers have been characterized with simple experiments, which are incomplete or unsatisfactory for conducting experiments later with different particles, such as from biomass burning. These aspects of our chamber and Figure 1 will be included in the revised manuscript.



Figure 1. Filter samples of Biomass Burning aerosols produced in a tube furnace (a) at 450 °C b) at 800 °C. (Note the 800°C sample is highly concentrated)

REVIEWER COMMENT #2: The authors claim that that this chamber is constructed specifically for aging studies of biomass burning aerosols native to sub-Saharan Africa. However, it seems like this chamber can be used to study any biomass burning aerosol system, not necessarily those native to sub-Saharan Africa. Does the combustion of biomass burning aerosols native to sub-Saharan Africa require special experimental apparatus or setup that is different from those currently being used in other chamber facilities? This is not clear from the manuscript. This also makes the title of the manuscript very deceiving. The title "Construction and characterization of an indoor chamber for the measurement of the optical and physicochemical properties of aging biomass burning aerosols" seems more appropriate.

AUTHOR RESPONSE: We agree with the reviewer that the title gives the impression that a special chamber is needed to study African fuels. Indeed, we don't need a special chamber for African fuels. However, the reason for including that in the title is to emphasize the fact that, to our knowledge, there have not been any laboratory chamber studies of biomass aerosols produced from African biomass fuels. We already have preliminary measurements of optical properties of biomass burning aerosols from fuels native to the region (see below for responses to comment #22). However, we have no objection in changing the title to "Construction and Characterization of an Indoor Smog Chamber for the Measurement of the Optical and Physicochemical Properties of Aging Biomass Burning Aerosol."

REVIEWER COMMENT #3: The amount of results related to the characterization of the indoor chamber presented in this manuscript is insufficient, especially those related to particle wall loss. This makes it hard to judge if the chamber is truly suitable for biomass burning aerosol aging studies as claimed by the authors.

AUTHOR RESPONSE: We have demonstrated that 100 nm particles (those with the shortest chamber lifetime, and far smaller than we plan to use for optical characterization) persist for sufficient periods to enable their collection and/or optical measurement. We have not presented size-dependent wall loss rates constants in this work, nor have many others. The stated goal of

this work is optical property characterization. If we were to determine SOA yield or combustion emission factors, such size-dependent wall loss rate constants would be necessary, and we would perform that level of characterization should we expand to those goals. However, it is not necessary at this time for our purposes. In short, there is no step in our data analysis where a particle wall loss rate constant would be used, so measurement of size-dependant wall loss rate constants are not currently useful. However, the comments may be significant if only the reviewer explained why he thinks the amount of characterization results are insufficient. The reviewer has not specifically stated what he feels is missing from the manuscript.

REVIEWR COMMENT #4: I find the flow of the manuscript meandering and somewhat confusing. There are parts of the manuscript (specifically the results section) where I fail to see how the paragraph(s) is related to the point the authors are trying to make.

AUTHOR RESPONSE: Though it is not clear what the reviewer means by this comment, we will conduct a careful review of the paper to maintain a logical flow of ideas and communicate clearly.

Specific major comments

REVIEWER COMMENT #5: Line 112: How does the lab's temp and RH affect the chamber's temp and RH (when the chamber lights are off)? It would be useful to have a figure showing this. This figure will also help convince readers that the lab's large RH range (30 to 60 %) will not affect the experimental conditions in the chamber.

AUTHOR RESPONSE: As stated in lines 134 - 139: Although the chamber is air tight in theory, in fact there are most likely small leaks. These leaks would allow ambient (room) air into the chamber when positive pressure is not maintained. As a result, chamber humidity increases as the purity of chamber air decreases. Lines 113-117 state that the temperature in the chamber is close to room temperature with UV lights off. While the low rate of sampling and replenishment is maintained, chamber RH is independent of its surroundings, since the air provided by the house compressor that is further purified by the zero-air generator and our conditioning system is extremely dry (\sim 0%).Room temperature varies by only a degree or two during chamber operation.

REVIEWER COMMENT #6: *Line 232: Is the chamber operated as a "batch reactor" or as a "continuous flow reactor"? This should be stated in the manuscript.*

AUTHOR RESPONSE: The chamber is operated as a "batch reactor" having a fixed volume of 9 m³. The Teflon walls are somewhat flexible, allowing for small changes in volume during injections without a change in pressure. This will be stated in the revised manuscript. It is not a "continuous flow reactor", which the authors take to mean a device in which photochemical age is related to the position within the reactor.

REVIEWER COMMENT #7: *Line 254: What are the concentrations of NO and NO2 added into the chamber based on the flow rates and concentrations of certified NO/NO2 gas cylinders?*

Providing these numbers will make it easier to compare these concentrations to those measured by NO or NOx monitors shown in Fig. 3. On a related note, does "elapsed time" in the x-axis of Fig. 3 refer to the time passed since the first gas injection? This should be stated explicitly in the text. Also, please indicate in Fig. 3. when the additional gas injections were made.

AUTHOR RESPONSE:

The concentrations calculated below will be added in the revised paper.

For NO: (NO cylinder concentration) x (flow rate) x (time) / (chamber volume) = (54 ppm)(1 L/min)(20 min) / (9010 L) = 0.12 ppm in chamber

For NO₂: (NO₂ cylinder concentration) x (flow rate) x (time) / (chamber volume) = (80 ppm)(1 L/min)(20 min) / (9010 L) = 0.18 ppm in chamber

Elapsed time of t = 0 does not correspond to the first injection. Vertical lines have been added to Figure 3 to indicate the injection time. A copy of the revised Figure 3 is below and will be in the revised manuscript.



Figure 3: Injections and response (mixing) time for NO and NO₂, done on different days, Vertical lines are injection times for NO and NO₂. Injection time was five minutes each.

REVIEWER COMMENT #8: Line 262: The authors state that particles only become wellmixed in their chamber after 60 to 90 min. A figure showing how the aerosol number/volume concentration changes as a function of elapsed time after aerosol injection into the chamber should be used to back up their claim.

AUTHOR RESPONSE: A heat map, showing size as a function of time has been added to supplementary information and the figure (Figure 2) is included here, below. This is the same data shown in Figure 5 in the manuscript, but including earlier size distribution measurements. The coloring of the heat map, compared to Figure 5, is distorted because the highest concentration during mixing is ~7 times larger than the maximum in Figure 5. The figure below will be included as supplementary information in the revision.



Figure 2- injection of particles at t = 0 fuel sample was pine

REVIEWER COMMENT #9: Line 278: The authors state that O_3 injections took over 3 days. This sentence does not make sense. Assuming that the chamber was run as a "batch reactor" in this experiment, I would expect their 9 m3 chamber to be completely deflated by day 3, even with minimal chamber leaks. If bag deflation did not take place, then what is the volume of the chamber on day 3? On a related note, the initial O3 concentration injected into the chamber can be estimated from the O3 injection time into the chamber and the O3 production rate of the lamp.

AUTHOR RESPONSE: As stated above, the chamber is operated as a "batch reactor" having a fixed volume of 9 m^3 . The ozone generator produces 0.126 ppm of ozone at 1.98 sL/min.

Assuming an average laboratory temperature of 19 °C and 1 atm of pressure, we calculate that the device produces 1.05×10^{14} molecules O₃/sec. This will be added in the revised manuscript.

REVIEWER COMMENT #10: Line 303: The authors claim that biomass burning aerosols from white pine wood was used to enable comparison with other chambers. It is difficult to make direct comparisons when all the chambers used in this comparison use different aerosols to characterize their chambers. For example, the Cocker and Wang papers use ammonium sulfate seed aerosols to characterize particle wall loss rates in the Caltech and Guangzhou chambers, respectively. On a related note, it will be easier for the authors to characterize particle wall loss rates in their chamber if they used inert aerosols such as ammonium sulfate (similar to the Cocker and Wang papers) instead of biomass burning aerosols. This is because inert aerosols will not be subjected to changes in particle size as a result of gas-particle partitioning processes (unlike the biomass burning aerosols used by the 3 authors), thus making the calculation of particle wall loss rates for the different particle size bins easier.

AUTHOR RESPONSE: We disagree with the reviewer. The purpose of the construction of the chamber is for studying BB aerosols and the characterization should involve such particles. We don't care about ammonium sulfate particle wall loss, and would rather not contaminate our chamber with something we won't be using, especially something with such low volatility. What we want to compare is wall loss for combustion aerosols at a specific RH and temperature, which is more relevant to the experiments planned for this chamber. This adds to the "novel" or atypical nature of this work, in that the chamber has been characterized with real biomass burning aerosols instead of some simple experiments which are incomplete or unrelated to relevant particle types. Indeed, quantitative comparisons of wall loss measured using ammonium sulfate may not be as simple as the reviewer indicates.

REVIEWER COMMENT #11: *Line 308: What are the initial aerosol volume and surface area concentrations used in these experiments?*

AUTHOR RESPONSE: The peak total volume occurred at 45 minutes after introduction, and was 1.97×10^{12} nm³/cm³. The peak area occurred at 20 minutes after introduction, and was 4.33×10^{10} nm²/cm³. At 100 minutes, where Figure 5 begins, the concentrations were 1.92×10^{12} nm³/cm³ and 3.32×10^{10} nm²/cm³ for the volume and area concentration, respectively. This will be included in the revised manuscript.

REVIEWER COMMENT #12: Line 317: The sentence "While the rate constant varied over time, it was consistent during the beginning, middle and end of the experiment" does not make sense. I would expect a single rate constant for each particle size bin.

AUTHOR RESPONSE: The authors would like to apologize for any confusion, as this text remained from an earlier draft. The text will be amended to be in line with Comment 17, below.

REVIEWER COMMENT #13: *Line 319: The observation that "The particle wall loss rates decreased with increasing particle diameter..." should be demonstrated using a figure showing how the particle wall loss rates change with particle size (like the one shown in Fig. 1 of Loza et al. (2012)).*

AUTHOR RESPONSE: We refer the reviewer to the response given for comment #3. We agree with the reviewer, in that the phrasing of this sentence seems to indicate that we have performed that analysis. Since we have not, we will adjust the phrasing of the manuscript to make it clear that this observation is from the cited literature, and not our own.

REVIEWER COMMENT #14: How does the particle wall loss rates change with chamber RH and temperature? It will be worth knowing this, especially in the case of temperature since the authors showed in Fig. 8 that the chamber temperature can increase by \sim 9 C during a 5 h experiment when the chamber lights are switched on.

AUTHOR RESPONSE: We thank the reviewer for this suggestion. We have not conducted experiments to determine the change in wall loss rate as a function of temperature and RH. The current planned experiments are focused on studies at near zero RH. RH and temperature dependent wall loss rates are not necessary for our current purposes, and will not impact the optical properties measurements which is the focus of our work. Future work involves measuring changes in optical properties as a function of RH.

REVIEWER COMMENT #15: *Line 321: It seems more appropriate to compare particle wall loss rates at different particle sizes for the different chambers, as opposed to at just one particle size (100 nm).*

AUTHOR RESPONSE: Certainly, it would be great to characterize for a range of sizes, and that can be suggested as future work. For this first manuscript, our reported work should be an adequate start, given the stated goals of the current project. There are more results to be published based on measurements being conducted on fuel sources from Africa. The size we picked is relevant, given the size distribution of combustion particles and availability of data from cited references, which only provide wall loss for 100 nm particles. Comparisons are appropriate for similar sizes. More on this is provided in response to comment #3.

REVIEWER COMMENT #16: Were routine particle wall loss experiments performed (e.g., weekly, monthly experiments)? How reproducible are the particle wall loss rates for the different particle sizes?

AUTHOR RESPONSE: The experiments were performed several times over a period of weeks to months. The rates are consistent and will be explained in the revision. More characterization experiments are planned to be conducted on a routine basis. (See supplemental data to see the consistency of the results.)

REVIEWER COMMENT #17: *Lines 329 to 334: The discussion about modeling particle wall as a second order process should be supported with figures showing the fits.*

AUTHOR RESPONSE: The authors would like to thank the reviewer for demanding this be done. While it bears a resemblance to a second order process, when the inverse of particle concentration was plotted against time, a line was not produced; a curve was. Several fractional rate orders were modeled, but none sufficiently represented the particle number density over time. It was found that an offset double exponential decay did reproduce the observations. This takes the form of

$$y = y_0 + A_1 e^{-(x-x_0)/\tau_1} + A_2 e^{-(x-x_0)/\tau_2}$$

In this, $y_0 = 525.88 \pm 77.5$, $A_1 = (3.9382 \pm 0.0374) \times 10^5$, $\tau_1 = 30.479 \pm 0.29$, $A_2 = (1.1604 \pm 0.0391) \times 10^5$, $\tau_1 = 105.49 \pm 2.07$, and $x_0 = 25$. The fit can be seen below, and seems to well represent the concentration data. It's likely that two different first-order processes are occurring – one being wall loss and the other being coagulation. The revised manuscript (see Comment #12), will be amended to reflect this.



Figure 3. Fitting curve of particle wall loss rate

REVIEWER COMMENT #18: *Line 336: Previous papers have already determined that coagulation plays an important role when large aerosol concentrations are used. See Nah et al. (2017).*

AUTHOR RESPONSE: We thank the reviewer for making us aware of this article. For the concentrations used in our experiments the role of coagulation is evident when the particles are left in the chamber in the dark for two to three days, as seen in the shift of the peak in the size distribution (in Fig 5 of the manuscript).

REVIEWER COMMENT #19: There should be some discussion on how the authors plan to correct for particle wall loss in their experiments. For example, do they plan to use size-

dependent particle wall loss rates (see Loza et al. (2012), Nah et al. (2017)), or the average loss rate of the total aerosol mass or number concentration (see Carter et al. (2005), Pathak et al. (2007), Pierce et al. (2008))?

AUTHOR RESPONSE: The focus of the experiments is to measure extinction and scattering of size selected particles from the chamber. The particle number density is measured with each optical property measurement. The size dependent wall loss will not impact the optical properties, as long as it is slow relative to our measurement time, which it is. That is more of an issue in SOA formation studies as described in the cited references.

REVIEWER COMMENT #20: *Lines 339 to 356: These two paragraphs don't add any value to the discussion. They seem to belong in the intro.*

AUTHOR RESPONSE: We believe it helps to compare our results with how wall loss measurements have been used in other studies.

REVIEWER COMMENT #21: *Line 358: Pierce et al. (2008) is not the only paper that studied particle wall loss rates (see references in point 13). Please cite accordingly.*

AUTHOR RESPONSE: We thank the reviewer for providing these additional references. They will be included in the revision.

REVIEWER COMMENT#22: Line 401: No results were presented in section 3.5 (Soot generation and characterization). I don't see the point of this section.

AUTHOR RESPONSE: We will add preliminary measurements of average values of single scattering albedo (SSA) (Table 1) and SSA as a function of wavelength from 500 to 570 nm (Figure 4a, b, c) of the BB aerosols produced by combusting African fuel samples (eucalyptus combusted in tube furnace at 500 °C) in response to comments and to make this section relevant. The plots show the single scattering albedo calculated from measurement of extinction and scattering cross sections. The measurements were conducted by sampling the particles soon after they were introduced into the chamber, after aging for 48 hours in the chamber in the dark, and after aging for 10 hours in the chamber with the UV lamps on. The measurements were done for three size bins (mobility diameters of 200, 300 and 400 nm).

Table 3. S	SA	Values for	Eucalyntus	BB aerosol	combusted	at 500 °C.
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Particle Diameter (nm)	200	300	400	Peak Mobility Diameter (nm)
Fresh	0.646 ± 0.009	0.660 ± 0.010	0.669 ± 0.011	131
Aged (Dark)	0.729 ± 0.028	0.712 ± 0.021	0.720 ± 0.029	322
Aged (UV)	0.877 ± 0.017	0.923 ± 0.016	0.960 ± 0.020	385





Figure 4. Single Scattering Albedo of Biomass Burning Aerosol obtained by combusting eucalyptus in a tube furnace at 500 °C. Measurement was done soon after introduction into the chamber and aging in the dark and with UV lamps on a) 200 nm particles b) 300 nm particles and c) 400 nm particles.

Minor comments:

1. Line 86: Should be "As opposed to field measurements" CORRECTION will be made in the revised manuscript

- 2. *Line 102: Should be "white pine wood"* CORRECTION will be made in the revised manuscript
- 3. Line 389: There should be a space after (Burkholder, 2015). Also, it should be "Burkholder et al."

CORRECTION will be made in the revised manuscript

4. Fig. 2: The legend is ambiguous. What do "Chamber1, Chamber2, Chamber3 and Chamber4" in the legend refer to?

CORRECTION will be made in the revised manuscript