Response to Review 1

We sincerely thank the reviewer for the valuable comments and suggestions. Below we list our point-by-point replies to the comments and the descriptions of the changes we made in the revised manuscript.

This article presents measurements of density, optical properties and chemical composition of biomass particles produced from the combustion of rice straw. In general, I think this study employs an impressive suite of instrumentation to characterize the chemical, optical and physical properties of biomass burn particles. The article does lack a main point and there are some deficiencies in presentation. Moreover, some of the conclusions do not follow from the data as mentioned in the "Detailed Comments" that follow. The main technical problems are:

1) the inference of ammonium nitrate and sulfate as major components of the aerosol without data to back up that claim,

   Response: Ammonium nitrate and sulfate have been proved to exist in the freshly emitted biomass burning particles in many previous works (Silva et al., 1999; Li et al., 2015; Huo et al., 2016). In our work, we have seen ammonium nitrate and sulfate from single particle mass spectra as well, with peaks of *m*/*z* 62 [NO_3^-], 97[HSO_4^-], and 18 [NH_4^+].

   Besides, we did not claim that ammonium nitrate and sulfate as the major components in biomass burning particles as the chemical information obtained from the single particle mass spectra is not quantitative.

2) the use of thermodenuding and bulk optical property measurement to infer things about coatings; I think there are some major assumptions that need to be addressed before confidently reaching this conclusion,

   Response: We are not quite sure about which conclusion the reviewer referred to, but we do have some technique issues about TD measurement to address in the response and the revised manuscript:

   Particle loss in TD
   In this work, we have taken the transport efficiency of TD into account for all the thermo-denuded related measurements including the absorption enhancement calculation. As written in Line 282, *"the measured η were used to correct the particle number concentration in the calculation of all the measurements related to thermal-denuded process"*.

   Particle shrinkage
   As for shrink factor, particles might shrink to smaller sizes after thermal treatment. In this experiment, we heated the particles before the DMA size selection. Thus, the mono-dispersed particles at the fixed size obtained after heating actually are not from the particles with the same original dried-particle diameter. For measurement like absorption enhancement, the particle optical data before and after thermal-denuded process are compared directly. Thus, it is essential to take the particle shrink factor into consideration.

   Here, we accepted the comments of the reviewers and added a new part (2.6) named as

“Shrink factor” (Line 285).

“The thermal-denuded method to separate the coating of particles for absorption enhancement calculation as well as other experiments related to particle volatility has been used in previous work (Nakayama et al., 2014; Chan et al., 2011; Lack et al., 2012). However, the particles might shrink to smaller sizes after thermal treatment. The particle shrinkage should be taken into consideration for size-selected volatility experiments which was neglected in previous work. The major reason could be the extremely low concentration for size-selected particles after thermal-denuded process up to 300 ^oC. The concentration of the size-selected particles was too low to be detected in the following instruments.

Therefore, we developed an approximation of the particle shrinkage calculation. A tandem DMAs (TDMA) was utilized to detect the size change of particles. Here, we used the ratio of the particle diameter after heating (d_{m2}) to the diameter before heating (d_{m1}) as the shrink factor (d_{m2}/ d_{m1}) of particles (shown in Figure S3). An approximation of the peak value for the dominant shrink factor mode was used for each diameter. The selection of particle diameter after thermal-denuded process was based on the original dried-particle diameter multiplied the shrink factor of each diameter (discussed in supplementary). ”

3) the methods utilized to calculate effective densities are not adequately described,

Response: For the calculation of effective density, we have used a large space with pretty detailed information of the method in our manuscript (Line 171-233).

4) there is a body of literature from the FLAME experiments that I think could be used to better interpret these results,

Response: We accepted the reviewer’s advice and added some discussion of our findings and the comparisons with previous studies performed by other groups.

Line 337: *Evidence of external mixing sodium and potassium salts in ambient environment was also observed by single particle mass spectrometry in previous work (Zauscher et al., 2013; Bi et al., 2011).*

Line 339: *A recent work performed by Lee et al. (2016) reported that K^+ was not uniformly mixed in biomass burning particles with less than 20% particles containing high K^+ content.*

Line 344: *The similar results of the externally mixed aerosol population was observed by Moffet et al. (2008) with a wide range of densities (1.1-3.4 g/ cm^3).*

Line 443: *The mass spectra of individual biomass burning particles have been studied in previous work (Silva et al., 1999; Zauscher et al., 2013).*

Line 572: *McMeeking et al. (2014) found that the strongly light-absorbing biomass burning particles tended to have a weak wavelength dependent absorption while the weakly light-absorbing particles tended have a strong wavelength dependent absorption, which is consistent with our results. In this work, the high values of AAE (~ 6.23) and SSA*

(~0.89, at 530 nm) suggested the light absorbing of rice straw burning particles were relatively weak compared to the particles emitted from other types of biofuels.

Line 585: *Previous studies have reported the absorption enhancement values in a range of 1.2-1.6 for biomass burning particles (Moffet and Prather, 2009; McMeeking et al., 2014). However, some other studies suggested that BC absorption enhancement due to lensing is minimal and climate models might overestimate the warming effect by BC (Healy et al., 2015; Cappa et al., 2012).*

Line 605: *Other than coating thickness, absorption enhancement of particles could be related with the mixing state and morphology (Liu et al., 2015).*

5) the composition measurements can be better analyzed and presented,

Response: We revised our manuscript based on the reviewer's advice. Please see the replies to detailed comments 15, 16, 23, 25 and 26.

6) the article needs to be better organized around a main point, and

Response: In this work, we focus on the size dependent mixing states of biomass burning particles and their correlations with the optical properties. To make this main point clear, we rewrote the abstract, revised some parts of the introduction and result discussions. Below is the new abstract:

"Biomass burning aerosol has important impact on the global radiative budget. A better understanding of the correlations between the mixing states of biomass burning particles and their optical properties is the goal of a number of current studies. In this work, effective density, chemical composition, and optical properties of rice straw burning particles in the size range of 50-400 nm were measured using a suite of online methods. We found that the major components of rice straw burning particles included black carbon (BC), organic carbon (OC) and potassium salts, but the mixing states of particles were strongly size-dependent. Particles of 50 nm had the smallest effective density (1.16 g/cm^3), due to a relatively large proportion of aggregate BC. The average effective densities of 100-400 nm particles ranged from 1.35-1.51 g/cm^3 with OC and inorganic salts as dominant components. Both density distribution and single-particle mass spectrometry showed more complex mixing states in larger particles. Upon heating, the separation of the effective density distribution modes testified the external mixing state of less volatile BC or soot and potassium salts. Size-resolved optical properties of biomass burning particles were investigated at two wavelengths (λ=450 & 530 nm). The single scattering albedo (SSA) showed the lowest value for 50 nm particles (0.741±0.007 & 0.889±0.006) because of the larger proportion of BC content. Brown carbon played an important role for the SSA of 100-400 nm particles. The Ångström absorption exponent (AAE) values for all particles were above 1.6, indicating the significant presence of brown carbon in all sizes. Though freshly emitted, the light absorption enhancement (E_{abs}) was observed for particles larger than 200 nm because of the non-BC material coating. Concurrent measurements in our work provide a basis for discussing the physicochemical properties of biomass burning aerosol and its effects on global climate and atmospheric environment."

7) I think the internal mixing of organics influence the effective density and this does not come through very well in the manuscript while I think it can be inferred with the available data.

   Response: See the replies to the detailed comments 16 & 31.

   In many places, the tables and figures can be described better and I have done my best to point that out. I think the article can be better proofread for grammar as well.

   Response: We changed Fig. 3 and combined Fig. 3 and S3 together (detailed comment 22, 26). We revised the title of Table S1 (detailed comment 14).

   In light of this critique, I recommend that the article undergo a major rewrite before it is acceptable for publication in ACP. The article will be a great addition to the literature after a significant transformation.

Detailed Comments:

1) L24: "relative" should be "relatively"

   Response: Changed.

2) L54: awkward sentence

   Response: We deleted this sentence.

3) L59: Surely the authors can provide more current refrences on BC – such as Tami Bond's extensive article published in JGR called "bounding black carbon".

   Response: We accepted the reviewer's advice and added this references in Line 50.

   "BC, which is predominantly produced from the combustion related sources, absorbs solar radiation across the visible spectrum, resulting in a warming effect (Bond et al., 2013)".

4) L60: awkward sentence

   Response: We deleted this sentence.

5) L70: suggest changing "low-visible" to "short wavelength visible"

   Response: Changed.

6) L89: The Tang an Munkelwitz article probably not the best reference here.

   Response: We accepted the reviewer's advice and changed the reference to Schmid et al., 2007.

   Schmid, O., Karg, E., Hagen, D. E., Whitefield, P. D., and Ferron, G. A.: On the effective density of non-spherical particles as derived from combined measurements of aerodynamic and mobility equivalent size, J. Aerosol Sci., 38, 431-443, doi: 10.1016/j.jaerosci.2007.01.002, 2007.

7) L105: This seems to be incorrect – bulk measurements cannot distinguish between

particles in a population.

Response: We changed sentence to *"distinctions among particles might be omitted by bulk measurements"*.

8) L114: grammar

Response: We rewrote the sentence as *"Biomass burning particle is a complex mixture of organic and inorganic species, including strongly light-absorbing BC and BrC. Size-resolved or even single particle level information on the morphology, chemical composition, and optical properties of biomass burning particles are necessary to have a better understanding of the correlations among these physiochemical properties."*.

9) L116: morphology of BB particles has been extensively documented via microscopy measurements. I think the introduction should include some of these. Most notably, Hopkins et al analyzed the optical and morphological properties of rice straw from the Flame experiment: Hopkins, R. J., K. Lewis, Y. Desyaterik, Z. Wang, A. V. Tivanski, W. P. Arnott, A. Laskin, and M. K. Gilles (2007), Correlations between optical, chemical and physical properties of biomass burn aerosols, Geophys. Res. Lett., 34, L18806, doi:10.1029/2007GL030502.

Response: We accepted the reviewer's advice and added the references in Line 77.

10) L119: Grammar

Response: We revised the sentence as *"In this study, laboratory experiments were conducted on rice straw combustion, a main source of biomass burning particles in Southern China"*.

11) L234: The procedures outlined in this section are not clear. Why are densities assumed to be unity? How will these assumptions be cancelled out later? What is being calculated and what procedures (e.g. iterative solutions…etc.) are being used?

Response: We listed the detailed procedures of the shape factor calculation a below:

$$\rho_p = \frac{m_p}{\frac{\pi}{6}{d_{ve}}^3} \tag{1}$$

$$\rho_{eff}^{I} = \frac{m_p}{\frac{\pi}{6}{d_m}^3} \tag{2}$$

$$\rho_{eff}^{II} = \frac{d_{va}}{d_m}\rho_0 \tag{3}$$

$$\frac{d_m}{C_c(d_m)} = \frac{d_{ve}\chi}{C_c(d_{ve})} \tag{4}$$

$$Cc(d) = 1 + \frac{2\lambda}{d}\left[\alpha + \beta\exp\left(-\gamma\frac{d}{2\lambda}\right)\right] \tag{5}$$

$$d_{va} = \frac{\rho_p}{\rho_0}\frac{d_{ve}}{\chi} \tag{6}$$

With assumed particle density (ρ_p) and known particle mass (m_p) measured by an APM, a calculated d_{ve} could be obtained using Equation (1).

$$d_{ve} = \sqrt[3]{\frac{6m_p}{\pi\rho_p}} \tag{7}$$

Though ρ_p was unknown, it would be canceled out later.

Using the same d_{ve} and for any shape factor (χ), a calculated d_m and d_{va} was obtained by Equation (4) and (6), respectively. Iterative solutions might be used in the calculation step of Equation (9)

$$\frac{d_m}{C_c(d_m)} = \frac{\sqrt[3]{\frac{6m_p}{\pi\rho_p}}\chi}{C_c(\sqrt[3]{\frac{6m_p}{\pi\rho_p}})} \tag{8}$$

$$Cc\left(\sqrt[3]{\frac{6m_p}{\pi\rho_p}}d\right) = 1 + \frac{2\lambda}{\sqrt[3]{\frac{6m_p}{\pi\rho_p}}}[\alpha + \beta\exp\left(-\gamma\frac{\sqrt[3]{\frac{6m_p}{\pi\rho_p}}}{2\lambda}\right)] \tag{9}$$

$$d_{va} = \frac{\rho_p}{\rho_0}\frac{\sqrt[3]{\frac{6m_p}{\pi\rho_p}}}{\chi} \tag{10}$$

λ is the mean free path of gas molecules. The empirical constants α, β, and γ are 1.142, 0.558, and 0.999 respectively (Allen and Raabe, 1985).

We could simplify Equation (8) and (10) into d_m = A*m_p* χ, and d_{va} = B*m_p/ χ. A and B were simplified coefficients in the calculation.

Thus, ρ_{eff}^{II} could be obtained by the calculated d_m and d_{va} and an estimated m_p was calculated by replacing ρ_{eff}^{I} by ρ_{eff}^{II} in Equation (2).

$$\rho_{eff}^{II} = \frac{d_{va}}{d_m}\rho_0 = \frac{B}{A\chi^2} = \frac{m_p}{\frac{\pi}{6}{d_m}^3} \tag{11}$$

$$m_{p-estimated} = \frac{\pi}{6}\frac{B}{A}\frac{{d_m}^3}{\chi^2} \tag{12}$$

We then calculated the ratio of the estimated m_p to the exact m_p as a function of d_m and χ (shown in Figure S5, discussed in Section 3.1.5).

We have changed the sentence into "*though* ρ_p *was unknown, nevertheless it would be canceled out later*".

12) L241: This title is too general to be of any use.

    Response: We changed this title to *"Instrumentation for effective density measurements"*.

13) L249: change to Kr-85 (use a dash)

    Response: We accepted the reviewer's advice and changed to "Kr-85".

14) L297 and Table S1: Xc1 and Xc2 need to be defined - I assume these are some indication

the peak positions. With-out knowing this it is difficult to make sense of the table. How did the peak positions change for the different fits? What do the numbers in the Table S1 indicate? Area? Height?

Response: x_{c1} and x_{c2} are the peak values of the effective density modes in Fig. 2.

We accepted the reviewer's advice and changed "x_{c1}" and "x_{c2}" to "Eff_1" and "Eff_2" in Table S1.

Table S1. Peak values and R squares of the average density distribution of 50, 100, 200, and 400 nm particles at 20 °C (room temperature), 150 °C, and 300 °C. Eff_1 and Eff_2 are the peak values of the effective density modes in Fig. 2. R^2 is the square of the correlation coefficient using Gaussian model fitting.

	50 nm			100 nm			200 nm			400 nm		
	Eff_1	Eff_2	R^2	Eff_1	Eff_2	R^2	Eff_1	Eff_2	R^2	Eff_1	Eff_2	R^2
20°C	1.167	\	0.980	0.933	1.454	0.979	0.939	1.519	0.9697	1.344	1.917	0.966
150 °C	0.972	1.642	0.951	0.981	1.691	0.932	0.984	1.746	0.949	1.094	1.798	0.911
300 °C	0.976	1.756	0.851	0.994	1.851	0.864	1.030	1.857	0.850	1.157	2.051	0.779

15) L312-317: What temperature is being referred to? I think the density profile for the room temperature particles is NOT bimodal. The data for (what is presumably) the second mode is noisy.

Response: The temperature in Line 312-317 was room temperature (we added this information in Line 325). The density distribution discussed here was for particles at 100 nm at room temperature. The shoulder peak at ~1.0 g/cm^3 was weak but quite obvious. The existence of this peak as a separate mode had been proved by the thermal desorption experiment as shown in Fig. 2.

Furthermore, figure S1 seems to have some fit parameters that need to be more fully described.

Response: Figure S1 displays the average number size distribution detected by SMPS. No fitting parameter was applied. We added "*detected by the Scanning Mobility Particle Sizer (SMPS)*" in the figure caption of Figure S1.

The bimodal gaussian fits should be used in the discussion. To state that the two components are externally mixed after passing through the thermodenuder is misleading.

Response: We did use the bimodal Gaussian distribution to fit the density modes shown as the dash lines in Figure 2. Here, we only discussed the room temperature measurement. The appearance of the peak at ~1.0 g/cm^3 suggested that BC was partly externally mixed with other components in biomass burning particles.

16) L327: Whos to say that internally mixed OC and potassium salts do not contribute to modes with lower densities? In fact, I think data shown in Figure 3 and S3 support the internal mixing of potassium and organics. The mass spectra shown in Figure S6 also support this. Its not clear why they did not look at the chemical changes as a function of temperature.

Response: We agree with this comment. We reword the statement as following:

"The dominant modes for biomass burning particles in the size range of 50-400 nm (Figure 2) could be a mixture of similar composition (BC, OC, potassium salts and secondary inorganic species) but different proportions."

17) L336: Organic matter can be secondary, so the parenthetic statement is inaccurate.

Response: We accepted the reviewer's advice and revised in the manuscript as "*BC, OC, potassium salts and secondary inorganic species*".

18) L349: What is "deviation range"? It would be useful to mark out suspected doubly charged modes on the figure.

Response: To verify the existence of doubly or multiply charged particles, the equation

$$\frac{d_m}{C} = \frac{2neVL}{3\mu q_{sh} ln\,(\frac{r_2}{r_1})}$$

is used to calculate the particle diameter (d_m) for particles with different number of charges (n) at a set DMA voltage (V), DMA rod length (L), gas viscosity (μ), sheath flow (q_{sh}), inner radius of the DMA annular space (r_1), and outer radius of the DMA annular space (r_2). C is the Cunningham slip factor evaluated at d_p (Knutson and Whitby, 1975).

Thus, the diameters of doubly charged particles should be larger than $d_m = \frac{4eVLC}{3\mu q_{sh} ln\,(\frac{r_2}{r_1})}$.

We deleted the "deviation" to avoid any misleading.

Since the doubly charged particle is not the main topic or finding in our work, we did not show the calculation method or mark the doubly charged mode in the manuscript.

19) L356: "bi-model" should be "bimodal".

Response: Changed.

20) L392: Why is one method for obtaining "effective density" consistently lower than the other? Can other information be extracted from this? I dont understand the value of reporting results from the two methods.

Response: Overall, these two methods had consistent results. The differences between the average values from the two methods were less than 8% for all particle sizes. We noticed that ρ_{eff}^{II} were generally smaller than ρ_{eff}^{I}, which could be due to the systematic error from different measurements.

We added the above discussion in our manuscript.

We didn't mean to use two methods to measure the effective density in this work. Since the SPAMS could obtain the aerodynamic diameters simultaneously, we determined to present results from both calculations.

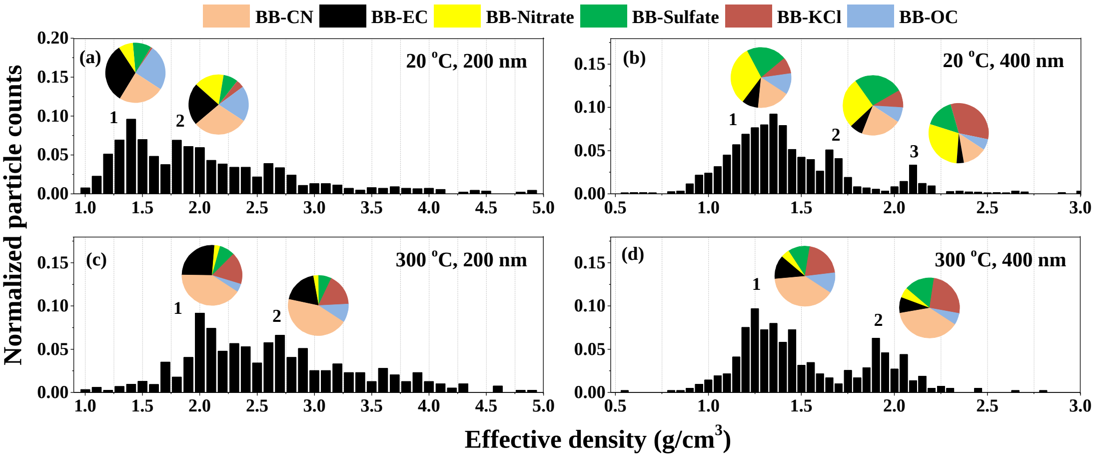
21) L434: I highly doubt the material density of the BB-KCl type is in the CRC handbook – in fact the CRC is referenced repeatedly in weird places.

Response: Here, ~1.99 g/cm^3 is the material density of KCl we found from CRC handbook (Lide, 2008). We revised the sentence to "*For example, the BB-KCl type might have higher effective density compared with others since the dominant composition KCl has a material density of ~1.99 g/cm^3 (Lide, 2008).*"

The CRC was referenced three times in our paper (Line 336, 465, 473). The first two references were used to expound the material density of KCl and the third one was to tell the material density of NH_4NO_3 and $(NH_4)_2SO_4$.

22) L436: Which is the "first" and which is the "second" mode?

Response: We accepted the reviewer's advice and numbered each mode in Fig. 3.



23) L442: I think this should be better referenced. What data suggests ammonium nitrate and sulfate are the dominant composition from rice straw burning? I dont think the measured density is a reliable way to infer chemical composition due to the fact that the material density may be a weighted average of organic and inorganic species. If ammonium salts are so prevalent, one would expect m/z = 18 to be fairly prominent - this is not mentioned in the manuscript.

Response: Firstly, we did not claim that ammonium nitrate and sulfate as the major components in biomass burning particles since the chemical information obtained from the single particle mass spectrometry is not quantitative. Ammonium nitrate and sulfate have been proved to exist in the freshly emitted biomass burning particles in previous works (Silva et al., 1999; Li et al., 2015; Huo et al., 2016). In our work, we have seen ammonium nitrate and sulfate from single particle mass spectra as well, with peaks of *m/z* 62 [NO_3^-], 97[HSO_4^-], and 18 [NH_4^+].

The peak area of each chemical species in single particle mass spectra is mostly determined by its ionization cross section in the laser ablation. The low ion intensity of *m/z* 18 [NH_4^+] is caused by the low ionization cross section of ammonium in the 266 nm laser ablation.

We agree that the particle density may be a weighted average of its organic and inorganic components. Here, we didn't infer the chemical compositions purely based on the density measurement. In this part of the discussion, we tried to combine the mass spectrometry and density measurements to see the connections between the density modes and the mixing of major particle components.

24) L452: Is ammonium nitrate really expected to be amorphous? This needs to be proven (with references), otherwise it is very speculative.

    Response: We added a reference in Line 483 to confirm this statement.

    Audebrand, N., Auffredic, J. P., and Louer, D.: Thermal decomposition of cerous ammonium nitrate tetrahydrate studied with temperature-dependent X-ray powder diffraction and thermal analysis, Thermochim. Acta, 293, 65-76, doi: 10.1016/s0040-6031(97)00064-6, 1997.

25) L457: these modes are hardly discernable – especially the middle mode in figure 3b.

    Response: We agree with this comment. The first two modes of 400 nm particles at 20 °C were quite close and their chemical compositions were also similar. We assume these two modes were derived from one mode. We revised the related discussion in Line 488.

    "For 400 nm mobility selected particles, the pie charts of particle type were almost identical for the first and second modes (as shown in Fig. 3b, 20 °C). Thus, we assume these two modes were derived from one effective density mode."

26) L464-472: I think the differences in SPMS data at the different temperatures is very telling. The drastic decrease in the OC cluster at high temperatures can explain why the effective density increases: these particles loose low density organics and become more dense. I think this point is missing from the discussion of the densities. The loss of organics and other low volatility secondary species is not suprising and can be shown better thorugh the use of difference spectra.

    Response: We accepted this suggestion and added this point in Line 501.

    "The high effective density (>2.0) of biomass burning particles at 300°C could be due to the vaporization of volatile organics with low density since the BB-OC type decreased drastically after thermal treatment."

    The mass spectra of biomass burning particles before and after thermal treatment could be found in our previous work (Zhai et al., 2015). Here, we used the pie charts of particle type to illustrate the changes before and after thermal-denuded process, which is more drastic in terms of the decrease of OC proportion (BB-OC type) comparing with the difference mass spectra.

    Furthermore, I do not see why Figures 3 and S3 are separate. I think it would be much more impactful to show these figures together.

    Response: We accepted this advice and combined Fig. 3 and S3 into one figure.

27) L487: what is offset here? Forcing? This is unclear.

Response: We rewrote the sentence as "*It's worth noting that the optical measurement was based on bulk measurement by CAPSs, which is not sensitive to the diversity of particle mixing state.*"

L560: This sentence is unclear. Furthermore, the procedures referred to in section 2.5 are not really described in that section, so I suggest that the authors give this some detail.

Response: Please see the response to the general comment 2. We added a new part (2.6) named as "Shrink factor" in Line 285 to give the detailed description.

Another more general comment about using thermodenuders to estimate absorption enhancement: how might the TD cause side reactions to affect optical properties?

Response: In our experiment, we used nitrogen gas (99.999%) to dilute the biomass burning smoke in our chamber with a dilution ratio ~ 6. The nitrogen environment could help to avoid the side reactions like thermal oxidation.

28) L567: The above definition (L 553) of absorption enhancement is really a bulk definition. Without knowledge of the exact mixing state of the population, I think it is difficult to attribute "absorption enhancement" to mixing state effects, no?

    Response: Indeed the particle absorption enhancement is a bulk definition. Here, the discussion of mixing state is based on the assumption of core-shell structure of particles which has been widely applied in previous work (Cappa et al., 2012; Lack et al., 2012; Nakayama et al., 2014). The non-BC coating material can enhance light absorption of BC core through "lensing effect". A thicker coating, which equals to a thicker lensing, can induce a larger absorption enhancement.

    In this work, we didn't use the single particle data to over-interpret the absorption enhancement results.

29) L589: "nonspherical" or "fractal" may be a better term to use in place of "aggregate".

    Response: We accepted the suggestion and changed "aggregate" to "fractal".

30) L606: again I think lower densities may be caused by mixing with organics – as described above.

    Response: We agree with this comment and add "low density organics" in the sentence.

31) L613: "acceptable standard" does not seem right here. What is "typical"?

    Response: We changed our wording to "*indicating the significant presence of brown carbon in all sizes*" (Line 651).

32) L616: change "volatile" to "volatility"

    Response: Changed.

33) L619: How can the authors attribute absorption enhancement to coating thickness using bulk measurements? Here the conclusions do not really follow from the data.

    Response: See the response to comment 28.

References:

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