

Interactive comment on “Size-resolved chemical composition, effective density, and optical properties of biomass burning particles” by Jinghao Zhai et al.

Anonymous Referee #1

Received and published: 5 January 2017

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, 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

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confidently reaching this conclusion, 3) the methods utilized to calculate effective densities are not adequately described, 4) there is a body of literature from the FLAME experiments that I think could be used to better interpret these results, 5) the composition measurements can be better analyzed and presented, 6) the article needs to be better organized around a main point, and 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. 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. 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: L 24: “relative” should be “relatively” L 54: awkward sentence L 59: Surely the authors can provide more current references on BC – such as Tami Bond’s extensive article published in JGR called “bounding black carbon”. L 60: awkward sentence L 70: suggest changing “low-visible” to “short wavelength visible” L 89: The Tang and Munkelwitz article probably not the best reference here. L 105: This seems to be incorrect – bulk measurements cannot distinguish between particles in a population. L 114: grammar L 116: 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. L 119: Grammar L 234: 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? L 241: This title is too general to be of any use. L 249: change to Kr-85 (use a dash) L 297 and Table S1: Xc1 and Xc2 need to be defined - I assume these are some indication the peak positions. With-

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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? L 312-317: 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. Furthermore, figure S1 seems to have some fit parameters that need to be more fully described. 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. L 327: 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. L 336: Organic matter can be secondary, so the parenthetic statement is inaccurate. L 349: What is "deviation range"? It would be useful to mark out suspected doubly charged modes on the figure.

L 356: "bi-model" should be "bimodal". L 392: 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. L 434: 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. L 436: Which is the "first" and which is the "second" mode? L 442: 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. L 452: Is ammonium nitrate really expected to be amorphous? This needs to be proven (with references), otherwise it is very speculative. L 457: these modes are hardly discernable - especially the middle mode in figure 3b. L 464-472: 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 lose 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 surprising and can be shown better through the use of difference 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. L 487: what is offset here? Forcing? This is unclear. L 560: 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. Another more general comment about using thermodenuders to estimate absorption enhancement: how might the TD cause side reactions to affect optical properties? L 567: 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? L 589: "nonspherical" or "fractal" may be a better term to use in place of "aggregate". L 606: again I think lower densities may be caused by mixing with organics – as described above. L 613: "acceptable standard" does not seem right here. What is "typical"? L 616: change "volatile" to "volatility" L 619: How can the authors attribute absorption enhancement to coating thickness using bulk measurements? Here the conclusions do not really follow from the data.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1060, 2016.

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