Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-1060-RC2, 2017

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

Received and published: 6 January 2017

Zhai and coauthors describe a series of experiments used to investigate the chemical, physical and optical properties of particles generated through the combustion of rice straw, chosen to represent local crop residues in Southern China. The motivation is to provide data that will help modelers to better assess regional and global radiative climate impacts of particles from this source. A combination of instrumentation is applied to calculate effective densities in two ways, employing a scanning mobility particle sizer, single particle mass spectrometer and an aerosol particle mass analyzer. Extinction, scattering and calculated absorption coefficients are also derived from a cavity attenuated phase shift spectrometer. These properties are assessed for untreated and thermally denuded rice straw combustion particles. While the experiments

are worthwhile, there are some issues regarding the approaches taken, and the results need to be framed in the context of existing work in a more rigorous way. Suggested revisions are below:

Major comments:

Why is the rice straw dried at 100 °C prior to use? Presumably local farmers do not do this and simply burn the residue with its natural water content. Wouldn't this change the burn conditions, smouldering etc? Would it be more representative to burn untreated fuel?

Why is a relatively low laser desorption ionization energy (0.6 mJ per pulse) selected here? Routinely single particle mass spectrometers of this design employ energies on the order of 1 mJ per pulse. How were the ART-2a cluster parameters chosen- are they simply selected from previous work? If so cite the source and reason for selection.

Is a flow rate of 0.6 L min-1 within the manufacturer's operating range for the thermal denuder?

The transmission efficiency of the thermal denuder was tested using NaCl particles. Why not simply examine the transmission efficiency of size selected biomass combustion particles at room temperature to validate agreement with the NaCl transmission efficiencies at room temperature. Also, for the absorption enhancement calculation, the transmission efficiency needs to be taken into account. The formula on line 553 simply relates babs before and after the denuder. Any losses of BC through diffusion and impaction in the denuder will be incorrectly assigned as absorption reduction through a removal of coating materials (absorption enhancement effect). Furthermore, why is the "shrink factor" used to account for volatilized coating materials? Firstly, the shrink factors shown in Fig S7 demonstrate a wide range of values, with multiple modes, and using a single value does not seem appropriate. In any case accounting for shrinking should not be necessary. Shrinking of the coatings is desirable for this test. The idea is that the bulk BC mass should be conserved through the denuder (applying the

correct transmission efficiency factor should result in BC mass entering the denuder being roughly equivalent to "transmission corrected" BC mass exiting the denuder), but that the coating materials should evaporate. Any reduction in absorption can thus be assigned to either reduced optical "lensing" due to the reduction of coating sizes or evaporation of any non-BC absorbing material. Applying the transmission efficiency correction and removing the shrink factor should be tested.

The x-axis of Figure 3 should show include effective density also. It would also be very useful to show an effective density distribution plot for each of the different particle classes. This will allow the authors to provide better evidence to support their postulations regarding potential composition-density relationships throughout.

Overall, there needs to be much better discussion of the findings here in the context of similar work previously performed by other groups. In terms of single particle mass spectrometry analyses of biomass burning particles (and relating biomass particle composition to optical and physical properties) there is a host of relevant studies that should be discussed. See (Silva et al., 1999; Zauscher et al., 2013; Bi et al., 2011; Guazzotti et al., 2003; Schwarz et al., 2008; Moffet and Prather, 2009; Moffet et al., 2008; Pagels et al., 2013). In terms of efforts examining relationships between biomass burning chemical composition measured through mass spectrometry and optical absorption enhancements see (McMeeking et al., 2014; Healy et al., 2015) and other ambient combustion particle absorption enhancements (Cappa et al., 2012; Liu et al., 2015) among others. One point which is posed as a new conclusion (evidence for external mixing of K and organic aerosol in biomass burning particles using single particle mass spectrometry) has been recently reported by others (Lee et al., 2016).

Minor comments:

Some SI figures should be in the main manuscript (S3 and S6)

Figure S4: the legend does not specify the method used for effective density calculation

Section 3.2: Do all of the particle types need the prefix BB? All particles discussed are from biomass. Also, the order of the particle types does not match Fig S6

Line 392: Test for statistically significant differences

Line 386: Simply remove the intermediate size from the plot if it is not represented in the other plot

Line 453: Is there a way to demonstrate this? See suggestion of effective density plots for each particle class earlier

Line 487: This is not an offset, simply a bulk measurement.

References

Bi, X., Zhang, G., Li, L., Wang, X., Li, M., Sheng, G., Fu, J., Zhou, Z., 2011. Mixing state of biomass burning particles by single particle aerosol mass spectrometer in the urban area of PRD, China. Atmospheric Environment 45, 3447-3453.

Cappa, C.D., Onasch, T.B., Massoli, P., Worsnop, D.R., Bates, T.S., Cross, E.S., Davidovits, P., Hakala, J., Hayden, K.L., Jobson, B.T., Kolesar, K.R., Lack, D.A., Lerner, B.M., Li, S.-M., Mellon, D., Nuaaman, I., Olfert, J.S., Petäjä, T., Quinn, P.K., Song, C., Subramanian, R., Williams, E.J., Zaveri, R.A., 2012. Radiative Absorption Enhancements Due to the Mixing State of Atmospheric Black Carbon. Science 337, 1078-1081.

Guazzotti, S.A., Suess, D.T., Coffee, K.R., Quinn, P.K., Bates, T.S., Wisthaler, A., Hansel, A., Ball, W.P., Dickerson, R.R., Neusüß, C., Crutzen, P.J., Prather, K.A., 2003. Characterization of carbonaceous aerosols outflow from India and Arabia: Biomass/biofuel burning and fossil fuel combustion. J. Geophys. Res. 108.

Healy, R.M., Wang, J.M., Jeong, C.H., Lee, A.K.Y., Willis, M.D., Jaroudi, E., Zimmerman, N., Hilker, N., Murphy, M., Eckhardt, S., Stohl, A., Abbatt, J.P.D., Wenger, J.C., Evans, G.J., 2015. Light-absorbing properties of ambient black carbon and brown carbon from fossil fuel and biomass burning sources. Journal of Geophysical Research:

Atmospheres 120, 6619-6633.

Lee, A.K.Y., Willis, M.D., Healy, R.M., Wang, J.M., Jeong, C.H., Wenger, J.C., Evans, G.J., Abbatt, J.P.D., 2016. Single-particle characterization of biomass burning organic aerosol (BBOA): evidence for non-uniform mixing of high molecular weight organics and potassium. Atmos. Chem. Phys. 16, 5561-5572.

Liu, S., Aiken, A.C., Gorkowski, K., Dubey, M.K., Cappa, C.D., Williams, L.R., Herndon, S.C., Massoli, P., Fortner, E.C., Chhabra, P.S., Brooks, W.A., Onasch, T.B., Jayne, J.T., Worsnop, D.R., China, S., Sharma, N., Mazzoleni, C., Xu, L., Ng, N.L., Liu, D., Allan, J.D., Lee, J.D., Fleming, Z.L., Mohr, C., Zotter, P., Szidat, S., Prevot, A.S.H., 2015. Enhanced light absorption by mixed source black and brown carbon particles in UK winter. Nat Commun 6.

McMeeking, G.R., Fortner, E., Onasch, T.B., Taylor, J.W., Flynn, M., Coe, H., Kreidenweis, S.M., 2014. Impacts of nonrefractory material on light absorption by aerosols emitted from biomass burning. Journal of Geophysical Research: Atmospheres 119, 2014JD021750.

Moffet, R.C., Prather, K.A., 2009. In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates. Proceedings of the National Academy of Sciences 106, 11872-11877.

Moffet, R.C., Qin, X., Rebotier, T., Furutani, H., Prather, K.A., 2008. Chemically segregated optical and microphysical properties of ambient aerosols measured in a single-particle mass spectrometer. J. Geophys. Res. 113, D12213.

Pagels, J., Dutcher, D.D., Stolzenburg, M.R., McMurry, P.H., Gälli, M.E., Gross, D.S., 2013. Fine-particle emissions from solid biofuel combustion studied with single-particle mass spectrometry: Identification of markers for organics, soot, and ash components. Journal of Geophysical Research: Atmospheres.

Schwarz, J.P., Gao, R.S., Spackman, J.R., Watts, L.A., Thomson, D.S., Fahey, D.W.,

Ryerson, T.B., Peischl, J., Holloway, J.S., Trainer, M., Frost, G.J., Baynard, T., Lack, D.A., de Gouw, J.A., Warneke, C., Del Negro, L.A., 2008. Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions. Geophys. Res. Lett. 35, L13810.

Silva, P.J., Liu, D.Y., Noble, C.A., Prather, K.A., 1999. Size and chemical characterization of individual particles resulting from biomass burning of local Southern California species. Environmental Science & Technology 33, 3068-3076.

Zauscher, M.D., Wang, Y., Moore, M.J.K., Gaston, C.J., Prather, K.A., 2013. Air Quality Impact and Physicochemical Aging of Biomass Burning Aerosols during the 2007 San Diego Wildfires. Environmental Science & Technology.

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