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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Reduction in biomass burning aerosol light absorption upon humidification: roles of inorganically-induced hygroscopicity, particle collapse, and photoacoustic heat and mass transfer" *by* K. A. Lewis et al.

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Reply to reviewer 2 comments.

Reviewer comments are given first followed by a discussion of the actions or interpretations of the authors. We thank the reviewer for his/her comments. Many of the comments are particularly useful for the authors and perhaps others that relate to things we could not measure (but will include in later efforts) with enough statistical significance in this work to use in a more quantitative manner.

COMMENT 1: Section 3.3 HTDMA measurements: I am wondering whether the authors can relate humidity growth factors from HTDMA data to the chemical composition presented in Figure 5. The presentation of results in a table is strongly recommended because humidity growth factors of biomass burning aerosol particles as a function of chemical composition are important data.

It is very likely that some of the coauthors will pursue the detailed analysis of HTDMA and chemical composition relationships. Our goal with including both here is to build the evidence that particle chemistry drives hygroscopicity, and that particles restructure as a consequence, with attendant effects on particle absorption.

COMMENT 2: Sections 4.1 and 4.2 Hypotheses on reduction of absorption. 1. On of the key issues addressed in this study is the collapse of particles after humidification and the resulting impact on particle absorption properties. The authors conducted a lot of SEM measurements of particle morphology. Why are the results not presented in a more quantitative manner? For the scientific community, the change in the particle fractal dimension from dry to humidified and again dried particles is a very important topic. I propose to add a table containing data on modification in particle morphology and resulting modifications of aerosol absorption properties, both measured and calculated. In the present form, the reader cannot assess magnitude and statistical significance of the expected effects.

We actually did not perform as much SEM work as we would have liked to, as we noticed in real time that aerosol light absorption was diminishing with humidification, and redirected our SEM resources temporarily towards measuring moistened particle shapes. To do the suggested experiment correctly, it would be highly advantageous to utilize two units for SEM filter exposure, one on the dry side, and one on the moistened side, and to analyze at least 1000 particles to gain statistical significance. Our use of the SEM data is again to build evidence of particle restructuring upon humidification.

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COMMENT 3: Concerning the reduction in aerosol light absorption by morphological changes, the authors cite two figures from other studies without relating their results explicitly to these findings. Referring to Section 4.2.1 it seems more appropriate to discuss the increase of knowledge on the reduction of the particle absorption coefficient as a function of relative humidity instead of discussing Figure 13 from Mikhailov et al. (2006). It would be interesting whether this effect is measured in the presented study with higher statistical significance than shown in Figure 13. Referring to Section 4.2.2 I propose to present own findings and relate them to the work of Liu et al. (2008) instead of discussing the already published work by Liu et al. (2008) with so much detail.

Our use of the Mikhailov and Liu results is to build supporting evidence for the likely values of the changes one would expect when particle restructuring happens. We are attempting to assemble all available related data that helps us understand the basic observation that aerosol light absorption as measured with the photoacoustic method (and by inference photothermal interferometry) is likely to diminish both due to particle restructuring and with mass transfer contributions being added in with a different efficiency than heat transfer contributions.

We added a comment to the conclusion to try to bring things together more completely:

“Three mechanisms altering light absorption measurements were discussed; the magnitude of these are summarized here: Particle collapse within typical particle size range gives a decrease in absorption by as much as 6%, but also shows and increase as fractal dimension approaches three. Modeling efforts from the literature indicate absorption enhancements of around 50% for black carbon cores with weakly absorbing coatings. Particle morphological differences limit the conclusions that can be made from coated sphResere theory. Mass transfer without correction leads to as much as 20% reduction of absorption from the real value for aerosol with large inorganic content. The tentative conclusion is that mass transfer associated with photoacoustic measurement of aerosol light absorption is the mechanism that best explains the observed reduction of aerosol light absorption with RH."

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COMMENT 4: Section 4.3 on mass transfer A model is used to calculate the expected mass transfer from absorbing particles in a photoacoustic resonator. The results are presented in Figure 15. This Figure, however, is difficult to interpret. Please include a quantitative description of the model experiment intercomparison in the text, including the discussion of the statistical significance of the obtained results.

The y-axis label of Fig. 15 has been changed to read ‘aqueous solution fraction’ to dispel confusion. The statistical significance of our model results can be tested against experiments where we measure by 3-D tomography a statistically significant number of particles coated with salt when dry, and/or with an environmental SEM that allows humidification of particles on the machine. Our model is an attempt to show how the effects of mass transfer can be put to use in estimating surface coverage by inorganic matter.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 15247, 2009.