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## ***Interactive comment on “Water uptake and chemical composition of fresh aerosols generated in open burning of biomass” by C. M. Carrico et al.***

**C. M. Carrico et al.**

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Interactive comment on “Water uptake and chemical composition of fresh aerosols generated in open burning of biomass” by C. M. Carrico et al.

Anonymous Referee #2 Received and published: 4 April 2010

Comment: This manuscript describes laboratory experiments measuring and parameterizing the hygroscopicity of aerosols generated from burning various biomass. These hygroscopicity measurements are related to the measured composition of the aerosols. This manuscript provides new information that is helpful in elucidating the water uptake of aerosols. I recommend that this manuscript be accepted with very minor changes; most of the changes are in regards to expanding existing sections to provide more

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context.

Reply: We have made a number of changes thanks to the reviewer's input.

Comment: Page 3634, lines 11-13, please explain briefly what the three fitted parameters mean rather than just listing what they are so that the reader can fully understand the fitting.

Reply: We have added the following sentence in this spot: "The growth factor dispersion factor gives a measure of particle chemical heterogeneity at a given size and it is discussed in relation to cloud condensation nucleus activation in Petters et al. (2009). For burns with multiple modes we also computed the volume-weighted average of the GF for the two modes, which we term the ensemble hygroscopic growth factor (Carrico et al., 2005). The ensemble GF uses the mean GF values from each mode and converts the number fraction in the select mode to volume fraction to determine a volume weighted ensemble growth factor. "

Comment: When the different types of biomass are introduced in Section 4.1 the authors discuss how different types of biomass burn differently, in general the smoldering flames produced particles with smaller GFs than those with a strong flaming phase. This paper goes on to show that composition is also important component that impacts GF. How do these two variables fit together?

Reply: Yes, it does seem that there are relationships as the reviewer proposes. This concept was also suggested by Hopkins et al. (GRL, 2007), who studied single particles produced in our 2003 series of burns. Hopkins et al. (2007) write: "It is generally accepted that flaming conditions produce more BC and less OC while smoldering fires result in higher OC content [Ward et al., 1992]. This is consistent with the strong flaming phase noted and the sp<sup>2</sup> hybridization and optical properties measured for category 3 fuels. However, several category 2 fuels exhibited a strong flaming phase and produced significant salts during the burning process, resulting in less carbonaceous particulate matter and optical properties inconsistent with BC. Category 1 fuels

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exhibited a range of burning conditions, including flaming, yet produced particulates more characteristic of OC with the corresponding optical properties. Hence, it appears that even when a flaming phase occurs, high salt contents and/or other fuel properties may influence the chemical and optical properties of particulate matter produced.”

We similarly conclude that both the fuel type and combustion conditions play a role in the aerosol properties. It is difficult to state any relationships more conclusively, in part because we recognize that the lab studies cannot access all of the combustion conditions experienced in an actual fire.

In section 4.1 we have added the following statement: “The connection between fuel type and its combustion conditions, described in detail in (Hopkins et al., 2007), ultimately relates to the hygroscopicity of the smoke which is explored in more detail below.”

In the discussion and conclusion section the following statement has also been added: “The type of fuel, combustion phase, smoke chemical composition and resultant aerosol hygroscopic properties are intricately linked. Moreover, combustion products likely also depend on fuel moisture content, though we did not study that variable. Here we can state that the general trend observed is that fuels that burned primarily in a smoldering manner in our studies produced aerosols with a large organic carbon mass fraction and low  $\kappa$  values.”

Comment: Does biomass that generally produces a smoldering flame also produce large amounts of organic matter when burned with a strong flaming phase? Which is more important to GF, the particles’ composition or how the biomass burns? As this manuscript does a great job in detailing the impact of composition on GF it would be helpful to the readers to understand how that relationship may change with different types of burning.

Reply: Please see the reply above. We also added the following statement in the end of section 4.3 to emphasize what we know: “Based on the relatively narrow range of

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hygroscopic response with the variation in combustion conditions as seen in Figure 6 vs. the wide range shown in Figure 5 as a function of fuel type, our experiments suggest that the fuel combusted is the more important determinant of smoke hygroscopic response.”

Comment: Perhaps it would be helpful if the data shown from different types of fires were shown on a graph in different colors; that would make it easier for the reader to understand how these different variables fit together. Figures 4-6 are very hard to read as-is. Will they be a full-sized page in final manuscript? That might be helpful, but still, these are very busy graphs. In Figure 5, what is the line that connects some of the dots (e.g., sagebrush, ponderosa pine)? In the caption to Figure 5 it would be helpful to explain what the different sizes of dots represents.

Reply: We believe the figures 4-6, though admittedly busy, provide the best picture of the complexities of the hygroscopic response of smoke based on our experimental results. We will work with the production staff to make these figures larger and more decipherable. The following statement has been added to the caption in Figure 5: “A range (shown as a range bar) of  $\kappa$  over multiple experiments is shown above for two fuels, ponderosa pine and sagebrush. A breakout by individual experiment for these two fuels is shown in more detail below in Figure 6.” The following statement has been added to the caption in both Figures 5 and 6: “The bubbles’ position on the x axis represents the arithmetic mean value of the GF for a given mode, and the area of the bubbles is proportional to the number fraction in each individual mode.”

Comment: Section 4.3, page 3637, starting line 21. The explanation of heading vs. backing is a little confusing in this section as how it relates to the actual experimental set-up (difficult to understand what uphill and downhill means in a controlled burning chamber). Does it relate to where the sampling was in relation to the fire? A better explanation here would be helpful.

Reply: The following has been added to clarify: “In order to create heading or backing

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conditions, the fuel bed was angled upward and the fire started at the bottom or top of the fuel bed, respectively.”

Comment: Section 4.4 and Figure 8 show a relationship on a log-log scale. There is still quite a bit of scatter around the line even on this log-log scale. Thus, I would not agree with the statement on page 3640 that the measured and predicted  $\kappa$  were in reasonable agreement. The trend was definitely in agreement, but only from Figure 8 it looks like there is still plenty of scatter. This “reasonable agreement” should be quantified (e.g., all measured vs predicted were within XX% of each other). Additionally, error bars on Figure 8 would be helpful.

Reply: The authors agree, and this section has been revised to include a revised figure shown below and the following: “An ordinary least-squares regression gives a slope = 0.89, intercept = 0.03, and  $R^2 = 0.62$  for the regression of measured vs. calculated  $\kappa$ . Overall, 26 out of 32 data points are within a factor of 2 in  $\kappa$  envelope shown in Figure 8. Based on estimated uncertainties in measured GF and  $a_w$ , error bars for measured  $\kappa$  are shown in Figure 8. With these uncertainty bars, 19 of 32 data points overlap the 1:1 line.”

Comment: Section 4.5. In this section it seems as if the authors put a piece of biomass in water, atomized that and then tested its GF and the GF of this solution was similar to when it was combusted. To me, this seems incredible and seems as if combustion thus is not important, only the composition that is water-soluble. Or is it that combustion does not change the composition of those that are water soluble? This section needs to be expanded more to put these findings into context and answer the above questions.

Reply: We agree these results are remarkable. However, we only tested a few samples, and our limited experiments cannot indicate clearly why the smoke aerosol and the extracts appeared so similar. The following statements have been added to further clarify: “The hygroscopicity data for the water extracts, together with the correspondence seen between ionic species content and hygroscopicity, suggest that the water-soluble com-

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position of the biomass fuel is likely to play a role in determining the hygroscopicity of the resultant smoke from its combustion. Based on our studies, the relative fractions of inorganic vs. carbonaceous material in the aerosol are the primary driver of smoke hygroscopicity. Furthermore, laboratory evidence from several studies has shown that the oxidation process with certain organic carbon compounds only modestly increases the hygroscopic response (Petters et al., 2006; George et al., 2009; George et al., 2008; George et al., 2007). A more comprehensive study with many fuels and integrating chemical analyses of both the smoke and fuel extracts would be necessary to attribute a more general relationship between fuel and smoke properties.”

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