

***Interactive comment on “Laboratory investigation of photochemical oxidation of organic aerosol from wood fires – Part 1: Measurement and simulation of organic aerosol evolution” by A. P. Grieshop et al.***

**Anonymous Referee #2**

Received and published: 20 September 2008

Review of [Laboratory investigation of photochemical oxidation of organic aerosol from wood fires-Part 1: Measurement and simulation of organic aerosol species](#); By A. P. Grieshop et al. for ACPD.

Overview: First I would like to point out how pleased I am that this work was done. With out a doubt, this is important research that merits publication. This vastly moves the field forward, if taken in the right context. That said, my tardiness in submission has also allowed me to go through the other two reviews. I agree with everything that was stated in these. I will not repeat what was said, unless I think it needs highlighting.

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First and foremost, I agree with other statements regarding the applicability issue. For example, to add to the other criticisms consider the size distributions in figure 2. In all of my years of biomass burning research, I have never seen a size distribution this small. Currently, the record for smallest VMD for fresh smoke is on the order of 0.22  $\mu\text{m}$ , whereas here you have 0.06 and 0.16  $\mu\text{m}$ . Such a small size will result in massive coagulation and wall loss. Hence with time an issue is that the smaller particles will quickly go to the walls and result in a bias in measurements. I think this bias is small, but it is likely there. Given the experiment design, this small size is no doubt to the use of a wood stove-considerably more efficient than a real fire. This coupled by the very quick dilution, results in the abnormally small size. This also makes me wonder about the particle emission factors. Are they much less than what is normally reported in the literature? These criticisms said, I do not think that this weakens the paper. You have given considerable evidence regarding the mechanisms of particle mass growth. I do not think it is necessary that you show an exact match to the environment, but as the reviewers point out this point of view must be clearly expressed. In fact, some effort should be taken to look at air pollution wood stove emissions papers which technically are closer in line with the measurements here than any actual biomass wild fire.

Second (in the emphasize category) is the issue of measuring black carbon with an aethelometer. To be clear one cannot measure black carbon optically, especially given the massive change in size experienced during growth (Reid et al., 1998 on BC). Single scattering albedo and absorption cross-section will undoubtedly change rapidly with size growth as these particles are moving out of the near Rayleigh into the Mie regime. This should be on the order of 20-50%-relatively small compared to the mass changes. Again, as long as this is treated only semi-quantitative to show the likely chemical mechanism, I am ok. Given that an AMS is available, you should consider more traditional tracers. Perhaps potassium, or levoglucosan.

Another fundamental issue not discussed is that of the control run. The assumption here is that mass growth only occurs photochemically. Because the authors assumed that a "dark case" can be used to subtract out wall loss and such, we don't know anything about what dark reactions are occurring. Indeed, there are a few limited datasets I have seen that suggest mass growth even without photons. I would really like to see more data on their dark control runs. Even if no data is really easily available (because it was a blank), you can say you are looking at only the photochemical part. But, again, this needs to be made clear.

I was also fairly impressed about the rapid increase in particle mass growth followed by the fast roll off after an hour or so. This matches very well what was observed in Hobbs's 1996 paper in Levines book.

Other minor comments: 15708 line 5: To compare to more "real data"; the authors may want to look at Blake's and Ferek's data in the literature.

15710 line 4: The authors should also consider that with the small particle size in this experiment, heterogeneous ozone loss to the particles is also likely more limited.

15710 line 21: That's what Reid et al., 1998 and Trentemann et al., 2003 said;

15711 line 14: This definition holds for large wildfires that have both flaming and smoldering combustion and is for "predominately flaming"; or "predominately smoldering"; I do not think that is the issue here. Next time perhaps the combustion state should be logged on film to help separate things.

15712 line 18: Yep-and it is almost certainly NO<sub>x</sub> limited;

15715 line 26+: I understand the point that is being made, but the way it is said "which are based on diesel exhaust data, to represent wood smoke"; is a total non-sequitor. That is like saying, "which are based on a car to repre-

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sent a boat. You may want to rephrase this.

15720 line 7: Reid et al., 1998 included this hypothesis, as well as many others. Besides, I do not think the case has been made that it is photochemistry that is the most active in the first 30 minutes. You guys see the mass growth (e.g., figure 3), but you don't account for dark reactions.

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 15699, 2008.

## ACPD

8, S7321–S7324, 2008

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