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Interactive comment on “Introducing the concept of Potential Aerosol Mass (PAM)” by E. Kang et al.

E. Kang et al.

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General comments

We are pleased that all the referees think that this work should be published after we address their concerns. We agree with many of their suggestions and have modified the manuscript to meet many of their suggestions. The greatest change is the inclusion of a section on the comparison of a SOA mass spectrum generated in the PAM chamber to one generated in a large environmental chamber. Since Dr. Darin Toohey, University of Colorado, provided the AMS and worked with us on interpreting the results, we would like to include him as a co-author.

We respond to the comments of each referee separately. Their comments are included in italics, followed by our responses. Since some of the referees have some of the same comments, we repeat our responses.

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Specific comments.

1 - *The data-set is very rich and I think the reader would risk coming into difficulties to digest the whole data-set in a single research paper. Therefore I would suggest moving some material to a future publication before the final peer-review publication. Most suitable to remove from this first paper would be parts of the large set of different VOCs studied.*

We understand the introduced data-set is rich. However, Section 3.3. SOA yields for various hydrocarbons is important to keep in this first paper. This section describes the PAM concept could really work for various range of hydrocarbons, not only for α -pinene. It also shows how the PAM chamber could work and be compared with large environmental chambers. If we really need to take out some data, we prefer to take out the SOA yield tests for various factors with α -pinene in the Section 3.2.

2 *The paper nicely illustrates fair to good agreement between the SOA yield in the PAM chamber and the presently used large environmental chambers. It also illustrates some divergence between seemingly similar experiments in different large environmental chambers. Another important point of the paper is the direct measurement of atmospherically relevant oxidants generated in the chamber.*

- When it comes to the characterization of the PAM chamber the arguments about losses and conversion efficiency would have been sharper if some particle size dependent analysis would have been included (for example using mobility analyzers) For example the occurrence of particle losses and potential non complete conversion efficiency is derived from a rather complicated procedure which involves calculating the amount of water present on the TEOM filter. Another type of control experiment would have been to compare the yield with and without a pre-existing non-acid seed aerosol.

This first paper about the PAM concept demonstrates the feasibility of the PAM concept; therefore future studies in many ways are planned. We plan to do the future experiments about particle size dependent analysis. If other research groups learn about the PAM concept and either collaborate with us or build their own, the viability of the PAM concept will be more rapidly assessed. The first step is to introduce that concept in the peer-reviewed literature, as we are trying to do here. There is clearly much to do and we intend, along with colleagues in other laboratories, to do it.

3 - *An important finding is the strong dependence of the yield on the total particle mass concentration. Especially the fact that the yield is a factor of 2-3 lower at typical atmospheric concentrations of 10-30 $\mu\text{g}/\text{m}^3$ compared to more often used higher concentrations in the large experimental chambers. During PAM experiments at low total mass concentrations the particles are expected to be smaller. These smaller particles have a higher deposition probability due to brownian diffusion. Is it possible that size dependent losses contribute to the dependence of yield on the total mass concentration?*

Yes. At low particle mass concentrations, the particles were measured to be smaller. And it is possible that size dependent loss can contribute to the dependence of yield on the particle mass concentration. The size dependent experiments and the particle loss test will be performed in a future study. While there is an issue with the size cut-offs of the instruments that are attached to the PAM chamber, the results from the SO₂ to sulfate tests suggest that sticky sulfate aerosol particles are not lost in the PAM chamber. More tests are needed, to be sure, but these results are quite encouraging.

4 - *Robinson et al. (2007), recently suggested that a potentially large fraction of*

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the SOA detected in the atmosphere comes from relatively low vapor pressure gas phase organics, which are oxidised only the first couple of generations. Trusting the reference, this could be a very important mechanism within and downstream urban environments. It would be interesting if the authors could elaborate whether the yield and speciation of such compounds could be expected to be similar in the PAM chamber and the atmosphere. Could any rough estimation be made of what time-scales in the atmosphere the conditions in the PAM chamber would translate to?

Table 1 in the Section 3.3 shows a general agreement between SOA yields in the PAM chamber and in the large environmental chambers for limited hydrocarbon species. Additionally we observed a similar chemical speciation in the PAM chamber and in the large environmental chamber (see the response for the next comment).

The oxidation time-scale in the atmosphere and in the PAM chamber is roughly calculated with the amount of OH and the exposure time. For instance, the amount of OH and the exposure time in the PAM chamber is roughly 200 pptv and 180 s, respectively. If the atmosphere has about 0.2 pptv of the OH mixing ratio, 50 hours are needed to complete the same oxidation in the PAM chamber. Therefore, roughly it can be told that about 3 days of the atmospheric time-scale is reproduced in the PAM chamber in about 3 min. The next generation PAM chamber will be capable of producing equivalent atmospheric OH exposure times of ~ 1 hour to several days.

5 - *In future publications the chemical composition of the potential aerosol mass formed in large environmental chambers and the PAM chamber needs to be carefully compared, for example using aerosol mass spectrometry, filter analysis or indirect measurements such as hygroscopicity. Only then do we now for certain whether measurements with the PAM-chamber can be safely extrapolated to atmospheric conditions.*

Another referee also requests the chemical composition data in the PAM chamber; therefore we provide the mass spectra of m-xylene in the PAM chamber taken by the Aerosol Mass Spectrometer. The mass spectra for m-xylene photo-oxidation in the PAM chamber are quite similar to the large environmental chamber from Bahreini et al. (2005).

6 - *Throughout the introductory section secondary organic aerosol particles and secondary sulfate particles are described as being separate classes of particles. I would argue that the different classes of compounds are to a large extent internally mixed with other sources in many relevant environments.*

The mixture of inorganic and organic secondary aerosols are more realistic in the atmosphere, and its effect can be studied in the SOA formation with and without neutral or acidic seed particles. Thus, we mentioned that a mixture of inorganic and organic secondary aerosol in Page 9927, line 12 and line 25.

7 - *Page 9927. Add “Quantifying the particle organic components from oxidation of precursor gasses...”*

- *Page 9931: change from “how much precursor gas is oxidized” to “the amount of precursor gas that becomes...”*

- *Page 9933: Move “the Rupprecht and Patashnick” to just give the company name in parenthesis “(TEOM, Model X, R&P...)”*

We agree to modify the sentences in page 9927, line 11, page 9931, line 9, and page 9933, line 11 according to the reviewer’s suggestion.

8 - *Page 9933: Doesn’ t delta M_{filter} mean mass increment rather than mass*

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ΔM_{filter} means the collected particle mass on the filter at a given collecting time. We replace line 18 in Page 9933: “where ΔM_{filter} is the collected particle mass on the TEOM filter at a given time,”

9 - Page 9936: Add “from knowledge of solely the UV light intensity and the relative humidity”.

We agree to modify the sentence in page 9936, line 3 as reviewer’s suggestion.

10 - Page 9938: *In arriving at the “absolute uncertainty in conversion efficiency of 16 %” assumptions are made of the water uptake on the TEOM filter and SO₂ and particle deposition on the chamber walls. Taking into account error propagation, are these 16% still a valid number? What uncertainties does this translate into when determining the SOA yields? Is the SO₂ losses to the walls dependent on RH?*

The absolute uncertainty is calculated from the error propagation analysis based on the measurements of SO₂ mixing ratio, flow rate, relative humidity, and TEOM mass. The large contribution comes from the measurement of SO₂ mixing ratio and TEOM mass, but they are still quite small. We performed the SO₂ loss test at constant RH condition, and the SO₂ loss test dependent on RH could be performed in a future study.

We modified the manuscript as following:

Page 9937, line 28: “The calculated and measured conversion ratios agree well for both of 30°C and 50°C.”

Page 9938, line 12: “For 4 SO₂ experiments, the measured conversion ratio was $92 \pm 4\%$ of the calculated conversion ratio, well within the absolute uncertainty of $\sim 13\%$ and $\sim 12\%$ for the calculated conversion ratio and for the measured conversion ratio, respectively.”

11 - Page 9940: Replace "containing in purified dry" air with "using purified dry air"
- Page 9940: Add to "achieve maximum aerosol mass concentration..."

We agree to modify the sentence as reviewer's suggestion.

12 - Page 9941-9942: *For the temperature dependence in the PAM-TEOM system to reflect the physical reality in the PAM-chamber, there needs to be a particle/gas phase equilibrium in the PAM chamber which is not affected by the heating in the TEOM cell before reaching the filter. Then the particle phase material needs to stay on the filter at 30C and gas-phase material needs to pass the filter. Is it likely that the system works in this ideal way so that for example temperature effects can be quantified and reflect the physical reality?*

The issue of the TEOM temperature and the loss of semi-volatile gases applies to filters and other particle measuring devices. We set the TEOM temperature to 30°C and then observe the loss of mass from the TEOM filter after we turn off the UV lamps. The integrated loss is less than a few percent of the observed mass, indicating the little of the semi-volatile mass is lost. It is interesting that by turning the UV lamps on and off, we can examine the loss of semi-volatile potential aerosol mass on the TEOM filter. However, this issue is important for determining an operational definition of PAM. Creating that operational definition, we hope, will be a collaborative effort of the aerosol community.

13 - Page 9952: *What is the likely reason for getting only 50% conversion efficiency? Is it due to only a single UV lamp used or is it due to some interaction with other trace gases in the complex sample in outdoor air?*

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It is because we used a single UV lamp. We constantly observed 50% of sulfate conversion in the laboratory with only one lamp. If we use this amount of UV for SOA formation, we find that the SOA mass is reduced from its peak value. This aging phenomenon is currently being investigated.

14 - *Figure 4a: The term "Expected aerosol mass" is misleading in this graph since it refers to SO₂ measured in the gas phase. I would suggest something like Sulphate equivalent SO₂ concentration.*

We have corrected the legend in Figure to be consistent with the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 9925, 2007.

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