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ACPD

8, S6005–S6007, 2008

Interactive Comment

Interactive comment on "SO₂ oxidation products other than H₂SO₄ as a trigger of new particle formation – Part 1: Laboratory investigations" by T. Berndt et al.

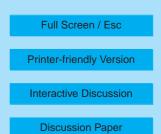
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I comment on two ACPD papers, T. Berndt, et al. (ACPD, 8, 9761–9782, 2008) and its companion paper A. Laaksonen, et al. (ACPD 8, 9673–9695, 2008), based on our recent results of H2SO4-H2O BHN laboratory kinetics observations (Benson et al., JRL, 2008; Young et al., ACP in press) which have utilized a similar approach to produce H2SO4 (via the SO2 + OH reaction) as in Berndt et al. studies:

(1) In the introduction of Part I paper, the authors have summarized the Berndt et al. 2004, 2005, 2006 results and came to conclusion that the SO2 + OH reaction provides lower threshold of H2SO4 than those in liquid experiment (Ball et al., 1999; Zhang et





al., 2004). We have done experiments with SO2+ OH reaction to produce H2SO4, but our threshold is not as low as Berndt et al. (that is, e8-e9 cm-3, as opposed to e6-e7 in Berndt al.). Our findings are summarized in David R. Benson, Li-Hao Young, F. Rifkha Kameel, Shan-Hu Lee, Laboratory-Measured Sulfuric Acid and Water Homogeneous Nucleation Rates from the SO2 + OH Reaction, Geophys. Res. Lett., 35, L11801, Doi:2008GL033387 and L.-H. Young, D. R. Benson, F. Rifkha Kameel, Jeffrey R. Pierce, Heikki Juninnen, Markku Kulmala, and Lee, S.-H., Laboratory studies of sulfuric acid and water binary homogeneous nucleation: Evaluation of laboratory setup and preliminary results, Atmos. Chem. Phys. Discuss. 8, 1-47, 2008 (in press for Atmos. Chem. Phys. now). In these two studies, we have used CIMS to directly measure H2SO4 and also took into account wall loss of H2SO4 for our threshold estimation – for some of resultspresented in Young et al. 2008, we even have used two CIMSs to simultaneously measure both the initial and residual H2SO4 at the beginning and the end of the nucleation reactor. This is so far one of the most constrained data on H2SO4 for BHN studies. I think it would be more appropriate if these two ACPD papers discuss our experimental results - as our results are not entirely supporting these authors' conclusion.

(2) While the main conclusion as well as the motivation of these two works are based on the difference in H2SO4 threshold, I am concerned that the Berndt et al.'s H2SO4 has never been measured directly. The kinetic calculation used in Berndt et al. – especially not proven by a calibration with H2SO4 measurements yet – is not a best approach for H2SO4 detection, as this indirect calculation method can introduce large uncertainties in their threshold estimation of H2SO4 and thus their conclusion. Without this issue resolved, any further discussions can be less convincing.

(3) Similarly, the Berndt et al. system especially with SO2, O3, UV, H2O, CO/hydrocarbon and NO/NO2 must be carefully reexamined. This system is in fact very similar to the LA smog chamber condition and the authors' discussion requires comprehensive reexamination to understand the byproducts of their system and their

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effects on nucleation. This discussion is very important and yet unclear to me in their three previous and current ACPD papers – note that tropospheric chemistry productions are non-linear and sensitive to different NO/NO2 and hydrocarbon concentration regimes.

(4) In Part I Figure 2, In this Figure 2, the slopes of Berndt et al. and Zhang et al., and Ball et al. are all in the range 5-8. Our SO2 + OH reaction experiments also show similar values – please include our SO2 + OH experimental results (Benson et al., 2008; and Young et al., 2008) in this figure. Please give explanations on these similar slopes from different experiments, especially with regard to the discussion on the authors' new pathway involving H2S2O8.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 9761, 2008.

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