

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.

S. Lee

slee19@kent.edu

Received and published: 15 August 2008

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

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al., 2004). We have done experiments with $\text{SO}_2 + \text{OH}$ reaction to produce H_2SO_4 , but our threshold is not as low as Berndt et al. (that is, $e8\text{-}e9 \text{ cm}^{-3}$, as opposed to $e6\text{-}e7$ 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 $\text{SO}_2 + \text{OH}$ Reaction, *Geophys. Res. Lett.*, 35, L11801, Doi:2008GL033387 and L.-H. Young, D. R. Benson, F. Rifkha Kameel, Jeffrey R. Pierce, Heikki Junninen, 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 H_2SO_4 and also took into account wall loss of H_2SO_4 for our threshold estimation – for some of results presented in Young et al. 2008, we even have used two CIMSs to simultaneously measure both the initial and residual H_2SO_4 at the beginning and the end of the nucleation reactor. This is so far one of the most constrained data on H_2SO_4 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 H_2SO_4 threshold, I am concerned that the Berndt et al.'s H_2SO_4 has never been measured directly. The kinetic calculation used in Berndt et al. – especially not proven by a calibration with H_2SO_4 measurements yet – is not a best approach for H_2SO_4 detection, as this indirect calculation method can introduce large uncertainties in their threshold estimation of H_2SO_4 and thus their conclusion. Without this issue resolved, any further discussions can be less convincing.

(3) Similarly, the Berndt et al. system especially with SO_2 , O_3 , UV, H_2O , 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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