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Interactive comment on “Laboratory studies of $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ binary homogeneous nucleation from the SO_2+OH reaction: evaluation of the experimental setup and preliminary results” by L.-H. Young et al.

Anonymous Referee #1

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General comments:

The manuscript is dealing with experimental findings from laboratory regarding new particle formation by oxidizing SO_2 . This topic is very important for atmospheric science and at the moment it is still open what the most important nucleation process is. The authors present a couple of experimental results for different conditions, e.g. residence time, relative humidity, precursor concentrations. These data are worth to publish. But, while reading this manuscript few questions arose, mainly technical points. Before publication a major revision is necessary.

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Specific comments:

1) In text, figures and tables it is stated that residual H₂SO₄ concentration at the end of the flow tube was in the range 10(8) - 10(10) molecule cm⁻³. At page 5 and figure 2b OH concentration in the photolysis zone is given with about 10(9) molecule cm⁻³. Downstream the photolysis zone this gas stream is diluted with N₂/SO₂ before entering the nucleation reactor, see figure 1. That means that after dilution maximum OH is < 10(9) molecule cm⁻³ and after total conversion via OH + SO₂ maximum H₂SO₄ is < 10(9) molecule cm⁻³. Given wall loss factors are in the range 2 to 12 resulting in maximum H₂SO₄ at the end of the tube < (0.83 - 5) x 10(8) molecule cm⁻³ neglecting any contribution from condensation. The authors should clearly explain the discrepancy between maximum OH level and residual H₂SO₄ measured. Do they assume additional channels for H₂SO₄ generation other than OH + SO₂? What is the accuracy of H₂SO₄ determination?

2) What is the photolysis time, page 5? What is the dilution factor for the OH/N₂ gas stream in the mixing region, figure 1?

3) The WLFs depicted in figures 3a and b are incorrect, log WLF is not about 10 or in the range 10(1) to 10(6) !

4) H₂SO₄ was produced via R1 - R3. For chosen conditions, i.e. SO₂, O₂ and H₂O concentrations, the time scales should be given needed for complete conversion of OH, HOSO₂, and SO₃. What was the residence time in the mixing zone? E.g. the data for a 4 sec nucleation experiment given in figure 6b: Using k(OH+SO₂) = 1.5 x 10⁻¹² for 0.03 ppm SO₂ (7.4 x 10¹¹ molecule cm⁻³), for a 90% OH conversion 2.1 sec are needed and for a 99% OH conversion 4.2 sec!!!

5) What is the reason for the very low O₂ mixing ratio in N₂ of 0.001/(1.7 - 5.3)?

6) Total residence time in the nucleation region was taken for the nucleation time for J-calculations. From my point of view, total residence time stands only for maximum

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nucleation time. For a wall loss factor of 2 (neglecting condensation) and a power equation, Eq.7, with $n = 5$ the ratio $J(\text{start})/J(\text{end}) = 1 / 0.03$. And for a wall loss factor of 12 the ratio $J(\text{start})/J(\text{end})$ is $1 / 0.000004$. That shows that nucleation is inhomogeneous with time in this experiment. Ball et al. (1999) defined only a part of the tube as the "nucleation zone". The authors should discuss this topic and the consequences for J vs. H_2SO_4 .

7) Page 11: The authors did not observe clear effects regarding H_2SO_4 and produced particles switching off O_2 in the carrier gas. What does it mean "without O_2 "? What is the residual O_2 concentration in the carrier gas under conditions "without O_2 "? Is it enough for converting HOSO_2 via R_2 ?

8) Page 12 and figure 5: It is confusing to see the particle number depending on the SO_2 concentration in the bottle. Was the purity of the 100 ppm SO_2 bottle checked?

9) In the figures in some cases there is an increase of particle number with time for constant H_2SO_4 visible. What is a possible explanation for this behavior? Are there saturation effects regarding the wall loss of H_2SO_4 ?

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 6903, 2008.

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