

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.

Anonymous Referee #1

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This paper is a valuable contribution to the understanding of SO₂ oxidation by a careful experimental study, though soliciting further studies. As part 1 of a combined laboratory and field study it prepares the ground for part 2, accessible in ACPD 8, 9673-9695, 2008. The language and the statements are sometimes strong (possibly calling for copy editing). On the other hand, this will promote further discussion after the final version has appeared in ACP.

The key to the different behaviour of H₂SO₄ formed by evaporation from that formed by oxidation is clearly the chemistry, where the liquid-phase reactions (so-called heterogeneous reactions) or real surface reactions most probably play a decisive role.

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Even though the chemistry is discussed in detail, the reviewer feels that the reference to the excellent chapters by Finlayson-Pitts and Pitts (2000) should be separated for gas-phase and liquid-phase chemistry and that the original publications and even earlier work might be discussed in addition. Aerosol formation is a known impediment in industrial chemistry of sulphuric acid production, where the rapid uptake of SO_3 by concentrated H_2SO_4 has been explained by the intermediate $\text{H}_2\text{S}_2\text{O}_7$. The books by Warneck (Chemistry of the Natural Atmosphere, 1988 and 2000) and Heicklen (Atmospheric Chemistry, Academic Press, 1976) and the original work cited therein might be consulted.

Since the purities of the gases and chemicals are major concern of such study on H_2SO_4 nucleation, the stated (and possibly unwise, since its definition may be based upon the hydrocarbon traces detectable by a GC with flame ionisation detector) extreme purity of synthetic air should be supplemented by the principle and properties of the Gate Keeper. The purity of the SO_2 and the ppm-mixtures of SO_2 in N_2 then need to be discussed as well. Would it be feasible to generate SO_2 by reduction of H_2SO_4 vapour from the same reservoir? This argument of limited purities holds for the O_3 as well, that would react with any wall contaminants and polymer tubing and plasticizers/additives of Teflon tubing and gaskets to bring volatile organics into the system.

In figure 2, the particle number density from H_2SO_4 vapour shows a constant, exponential increase, not having reached the limiting value on the time scale of the illustration. On the other hand, the particles disappear immediately after switching H_2SO_4 off. This indicates rapid loss processes acting against a slow saturation of surfaces. So it might be interesting to monitor the increase of particle number density over several days or even weeks. What is the explanation for this exponential(!) increase ("2 to 3 particles cm^{-3} after more than 5 h") that must be quite far from steady state?

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

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