

Interactive comment on “Aqueous phase oxidation of sulphur dioxide by ozone in cloud droplets” by C. R. Hoyle et al.

Anonymous Referee #2

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The authors present results from a combined experimental and model study simulating the oxidation of S(IV) by ozone in cloud droplets. I agree that this study is clearly unique and innovative since they perform experiments in individual samples and over a temperature range of -10C to +10C, i.e. conditions that were not accessible in previous (bulk) experiments. The manuscript describes very carefully the experiments and discusses possible uncertainties and biases. Given the thorough characterization of the CLOUD chamber using this rather simple chemical system, it can be expected that this study might be used as a reference for the investigation of more complicated aqueous phase systems in the future. I have only some minor comments that should be addressed prior to the acceptance of the paper.

Comments

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1) Throughout the manuscript: I think the authors confuse the term ‘reaction rate’ and ‘(reaction) rate constant (or ‘rate coefficient’). A reaction rate dc/dt is the product of the rate constant (k) and reactant concentrations (e.g., $[A]$ and $[B]$), i.e. $dc/dt = k [A] [B]$ (cf e.g. Seinfeld and Pandis, 2006) Therefore, a model includes rate constants that are based on laboratory studies. Reaction rates could only be compared between different studies if the same solute concentrations were present. The text should be carefully checked and revised for the correct use of ‘reaction rates’ and ‘rate constants’.

2) p. 33850, l. 11: Did any ice form in any of the experiments? If so, might that be differentiated from liquid droplets? Related to this comment: p. 33852, l. 16: Does ‘phase transition’ only refer here to gas-to-liquid transition or is any ice phase involved?

3) p. 33857, l. 21: Would it be possible to use E-AIM at pressures other than 1 atm? How much difference in the results could be expected if the variable pressure as in the chamber were considered?

4) p. 33858/9: Add units to J_c , D_g , J_k in the equations or text before.

5) What are the assumptions of the mass accommodation coefficients for H_2SO_4 and NH_3 ? What is the limit between ‘high’ and ‘low’ accommodation coefficient?

6) p. 33869, l. 2ff: Is the increased evaporation of ammonia from unactivated aerosol greater because of the larger curvature effect or because of differences in water activity? Does E-AIM take into account both of these effects?

7) p. 33869: The section about glyoxal gets a bit lost in this section and should be rather made to a separate section. A few more details should be given in terms of the comparison to NH_3 , e.g. how comparable are the solubilities (Henry’s law constants) and vapor pressure of these gases? Which parameter determines their uptake/evaporation behavior?

8) Table 1: It should be added which equilibria K_1 , K_2 , K_3 refer to.

Technical comments

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- p. 33845, l. 3: CLOUD and CERN should be spelled out here
- p. 33858, l. 17: P_{vap} should have lower case p
- p. 33864, l. 19: either 'all aerosol is activated' or 'all aerosols are activated'
- p. 33867, l. 27/8: 'concentration' misspelled
- p. 33873, l. 13: Spell out CIGAR
- p. 33874, l. 29: received

All references end with numbers (Page numbers?). I assume that this happened during copy editing but should be removed. Table 3: Is there no T dependence of k_0 ? If so, the minus sign should be removed and '0' added instead.

Figure 1 (and some other figures) 'shows that' can be omitted

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