

Interactive comment on “Influence of aerosol copper on HO₂ uptake: A novel parameterized equation” by Huan Song et al.

Anonymous Referee #1

Received and published: 4 April 2020

Review of “Influence of aerosol copper on HO₂ uptake: A novel parameterized equation” by Song et al.

The authors present a modeling study of the HO₂ uptake coefficient as a function of aerosol copper concentration and relative humidity. Their methods seem thorough and robust for liquid particles containing ammonium sulfate and copper. They are able to explain the previous large discrepancy between experimental measurements and models, which is extremely useful in helping to push our understanding of HO₂ uptake coefficients forwards. They also provide a parameterization, based on their more complex model, which could easily be implemented in regional and global models. They are able to show that a wide range of HO₂ uptake coefficients are likely to be required for atmospheric aerosols, rather than the fixed value which is often currently used in

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models. However, I feel that there are many places throughout the manuscript where clarifications and additional details would be helpful. The authors could also state the limitations of their model and their implications more clearly. The manuscript would also benefit from being proof read. Overall, this is a really useful study, which should be published after the comments below have been addressed.

Comments: 1. Please clarify how the ionic strength in Equation 4 is calculated. Is this calculated in the MARK model and what would a typical value be?

2. For all tables please add units where these are missing.

3. In Table 4 what are the values of k_{mt} or how are these calculated?

4. Are all reactions in the tables included in the model? If so how does this relate to k_{eff} in Equation 11?

5. It's stated that in the model it is assumed that the surface concentration and the bulk concentration equal each other. It is also stated that this is only valid for particles with a radius less than 200 nm. However, the model and resulting parameterization are then applied to particles which are larger than this and many particles in the atmosphere are larger than this. I wonder why the authors don't seem to have used the correction in equations 10 and 11 and what impact this will have on their final results and the applicability of their parameterization to future studies?

6. In Figure 2 what is the main cause for the decrease in the uptake coefficients between the original parameterization and the new model results. Is the difference mainly due to the different rate coefficient being used, the use of activity coefficients or something else?

7. For Figure 2 what is the aerosol pH and how is it calculated?

8. Figure 2 seems to be missing some previously published data point(s) from Lahey et al., JPCA (2016). It seems that the point at the highest copper concentration in that work (which is not shown in Figure 2) would not fit the modeled line. The authors

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should include any previously published missing points for completeness. Are they able to model or at least speculate as to why this data point does not fit their model.

9. George et al. PCCP (2013) noticed higher uptake coefficients for lower HO₂ concentrations for copper doped particles. Did the authors do any sensitivity tests with different HO₂ concentrations and do they see any difference?

10. Figure 3: Is lg on y axis log₁₀?

11. Figure 3: Please explain this figure in a more detailed fashion. It is unclear to me what the markers are and why there is a range of values. Why does there seem to be a larger difference between the model and the parameterization at low relative humidity?

12. Figure 4: Is the data shown measurements or a simulation?

13. Figure 5: This figure is not mentioned at all in the text and as such I don't know what the difference is between Figures 4 and 5.

14. Why not combine Figures 4 and 5 for better comparisons?

15. Can the authors speculate as to why the HO₂ uptake coefficient is higher at night (Figure 6)?

16. In Figure 6 what is the main cause of the distribution in HO₂ uptake coefficients? Is it due to different copper concentrations in the particles or something else?

17. Were any HO₂ measurements made during the Wangdu field campaign and if so was any box modeling of the Wangdu campaign performed to determine whether there was a discrepancy between measured and modeled HO₂ uptake coefficients? Were predicted HO₂ uptake coefficients in the range that was expected? If no HO₂ measurements were made, could the authors clarify why they chose this particular field campaign to apply their model to?

18. Line 313: The authors may want to clarify that 'aerosol properties' may include phase state and that previous measurements have shown lower uptake coefficients

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for semi-solid and solid particles (e.g. Lakey et al. ACP (2016)). The authors should also clarify somewhere that one of the major limitations of their model is that they assume steady-state concentrations and do not consider concentration gradients which will occur and could change over time for semi-solid particles.

19. The authors fix the solubility of copper at 25%. In reality solubility can vary considerably. How sensitive is this parameter in their model?

20. Another limitation of the model is that they don't consider reactions between different metal ions (such as Reaction 4 in Mao et al. ACP (2013)) which they have stated. However, could they also speculate how this could impact the estimated uptake coefficients for atmospheric aerosols (e.g. is it expected that this would increase the uptake coefficient)?

21. Please check the references carefully as many seem to be wrong (e.g. Schwartz and Meyer 1986 line 108 and references in Figure 2).

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-218>, 2020.

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