

Review of Holmes ACPD

Holmes provides a method to account for entrainment-limited kinetics in second-order aqueous-phase reactions in clouds in large-scale models of the atmosphere that do not resolve clouds. This builds upon his prior work in developing a method for first-order reactions. He uses the example of an important second-order aqueous-phase reaction in the atmosphere, oxidation of S(IV) by H₂O₂. He quantifies the numerical errors in this method by comparing it with results from a two-box model that explicitly represents clouds and finds that the errors are relatively small (typically $\ll 1\%$).

This paper is an important advance, and I am left very curious how this will impact sulfate formation, and other important aqueous-phase reactions, in large scale models of atmospheric chemistry. I hope a paper on this is forthcoming. My suggestion for improvement of this paper is to be clear on whether the concentrations and rate constants are for the gas-phase or the aqueous-phase. In Holmes et al., GRL, 2019, this was clear, but it is not clear here. This could be done by explicitly stating this in the text, as was done in Holmes et al., GRL, 2019, and/or by providing units for each variable. On a related topic, I wonder why the terminology has changed from the 2019 paper. In the 2019 paper, the variable c was used for gas-phase concentrations. In this paper, [A] and [B] are used instead. Often, square brackets signify aqueous-phase concentrations. Perhaps use c_A and c_B for consistency?