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Kinetic modeling of nucleation experiments involving SO_2 and OH: new insights into the underlying nucleation mechanisms

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Abstract. Nucleation is an important source of atmospheric aerosols which have significant climatic and health implications. Despite intensive theoretical and field studies over the past decades, the dominant nucleation mechanism in the lower troposphere remains to be mysterious. Several recent laboratory studies on atmospheric nucleation may shed light on this important problem. However, the most interesting finding from those studies was based on the derived H_2SO_4 concentration whose accuracy has not yet been evaluated by any other means. Moreover, the threshold H_2SO_4 concentration needed to reach the same degree of nucleation reported by two separate nucleation studies varies by about one order of magnitude. In this study, we apply a recently updated kinetic nucleation model to study the nucleation phenomena observed in those recent experiments. We show that the H_2SO_4 concentration can be estimated with a higher level of accuracy with the kinetic model by constraining the simulated particle size distributions with observed ones. We find that the required H_2SO_4 concentrations to achieve the best agreement between modeling and measurements are a factor of ~2 to 4 higher than reported in those experiments. More importantly, by comparing the derived thermodynamic properties associated with the nucleation process, we conclude that different unknown species may participate in the two separate nucleation experimental studies, which may explain the large difference in the reported threshold H_2SO_4 concentration. Although the unknown species involved has yet to be identified, the derived values of thermodynamic properties can serve as a valuable guideline for the search of their chemical identities using advanced quantum-chemical approaches.

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