

Interactive comment on “Novel Pathway of SO₂ Oxidation in the Atmosphere: Reactions with Monoterpene Ozonolysis Intermediates and Secondary Organic Aerosol” by Jianhuai Ye et al.

Anonymous Referee #1

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This is a very nice investigation of the interactions between SO₂ and reactive intermediates from monoterpene ozonolysis under different humidity conditions (10% vs. 50%). Increasing humidity seems to change the chemical regime in the system, shifting from a stabilized Criegee Intermediates (sCI) produced from ozonolysis, to peroxy-based particle phase chemistry leading to organosulfates production. I really enjoyed reading this contribution, which is nicely and convincingly presented. Therefore I do recommend its publication subject to very minor changes.

Experiments were made in presence of excess ozone to ensure the complete consumption of the terpenes. Have you performed tests experiments where this was not

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the case? Would expect any difference in the chemical regimes to SO₂ oxidation at lower ozone concentration? For instance due to reduced peroxy-functions productions, concomitant presence of unsaturations, etc. . .

Maybe you could add in the experimental section 2.2, the ozone levels used in the flow tube experiments with, maybe, some indications how this compares to the chamber ones?

You are making clear that limonene is needed to induce a SO₂ loss (line 225), but does this fully exclude that loss of SO₂ is not firstly physically driven by solubilization into the nascent aerosols? (Adding limonene would also affect such an equilibrium due to the growth rate of the particles).

The paragraph starting at line 230 is slightly confusing to me. How do you conclude/affect to the growth effect to sulfuric acid? How would that acid be produced efficiently in your system? OH reaction can be excluded due to the presence of an OH scavenger and ozone reacts quite slowly under acidic conditions. Would all this just be linked to sCl chemistry? Maybe adding a few words of explanation would be helpful.

Humidity seems to reduce the SOA enhancement, as measured in the chamber. But what about the products distribution is humidity reducing the amount of organosulfate being produced? Also as noted, an increased humidity leads to an increased particle phase pH and hence an increased reactivity of ozone toward SO₂ leading to sulfate production that may explain partly the observed enhanced SO₂ loss under humid conditions. Would that be a sign of a competition between ozone reactivity and peroxy-type chemistry leading to organosulfate production, as this exist for cloud processing of SO₂ (which is strongly pH dependent)?

Line 76 : please add the reference for the statement : “The reaction rate of particle-phase SO₂ + RO₂” Line 88: add also the reference to: Passananti, M.; Kong, L. D.; Shang, J.; Dupart, Y.; Perrier, S.; Chen, J. M.; Donaldson, D. J.; George, C. Organosulfate Formation through the Heterogeneous Reaction of Sulfur Dioxide with Unsaturated

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