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Interactive comment on “Secondary Organic Aerosol formation from isoprene photooxidation during cloud condensation–evaporation cycles” by L. Brégonzio-Rozier et al.

Anonymous Referee #1

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This paper describes chamber-based experiments on aqueous SOA (aqSOA) formation. Products of isoprene photooxidation are exposed to “cloud events” that last several minutes. The subsequent droplet evaporation leads to production of organic particles (if there were none present before the cloud event) or to an enhancement of particle mass concentration (if particles were already present before the event). The amount of aqSOA produced in these experiments suggests that aqueous processing of oxidized organics may serve as an efficient particle generation and growth mechanism. This is an important result, and the experiments described in the paper are uniquely different from the traditional chamber experiments.

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My most significant concern about this paper is related to the statements on P20573, L15 and P20578, L15 about mass decay of particles observed after the cloud events. I am not at all convinced by the authors' assumption that particles will not similarly shrink without the walls. In addition to the explanation involving particle-to-wall repartitioning, there is also a possibility that particle evaporation is kinetically constrained. Evaporation from particles is not instantaneous (e.g., see Vaden et al. (2011), Evaporation kinetics and phase of laboratory and ambient secondary organic aerosol. Proc. Nat. Acad. Sci. 108, 2190-2195) so it is reasonable that gas-particle re-equilibration should take some time after a cloud event. If this is the case, the walls have nothing to do with the particle mass loss, and particles will shrink in the actual atmosphere in the same way as they do in these chamber experiments. Therefore, it is misleading to use the maximum mass concentration measured in the experiments because it will lead to an overestimation of the yield of aqSOA. The authors should use the difference between the stabilized particle mass concentration after the particle mass decay stops and the mass concentration before the cloud event to estimate the effect of cloud processing on aqSOA production.

The rest of the comments are minor:

P20562, L20: forcing -> forcing on climate

P20563, L15: volatile -> volatility

P20568, L25: the normalization of PTR-MS signals with respect to hydronium ion + hydronium ion-water complex is not common (to the best of my knowledge), and should be better explained/justified.

P20574, L8: seem not to be -> did not seem to be

P20574, L18: what is so special about 32%? The authors should provide a range of % decrease in concentrations from Table 4 instead of a comparison to a randomly chosen threshold of 32%.

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P20576: please describe how the particle density was measured

P20576: when discussing Fig. 3, I would mention what the O/C and H/C ratios were for the background aerosol present before the cloud event in diphasic experiments (if particle were detectable by AMS)

P20577, L11 and L22: Tang and Thompson (2012) discuss photochemistry of nitroaromatic compounds (specifically, nitrophenols), which are not expected to be produced in the experiments described in this paper. Photooxidation of isoprene under high-NO_x conditions results in organic nitrates (RONO₂), not nitro compounds (RNO₂). Furthermore, there is probably not enough time for photochemistry to produce any significant damage (e.g., see Nguyen et al. (2012), Direct aqueous photochemistry of isoprene high-NO_x secondary organic aerosol, Phys. Chem. Chem. Phys. 14, 9702–9714)

P20577, discussion of hydrolysis: papers by the Elrod group should probably be mentioned here: Darer et al. (2011), Formation and stability of atmospherically relevant isoprene-derived organosulfates and organonitrates, Environ. Sci. Technol., 45, 1895–1902; Hu et al. (2011), Thermodynamics and kinetics of the hydrolysis of atmospherically relevant organonitrates and organosulfates, Atm. Chem. Phys., 11, 8307–8320. As mentioned above, the Tang and Thompson (2012) paper is not relevant in this case as it deals with a different class of nitrogen-containing organics.

P20579, L7: initial seed wet particles -> pre-existing wet seed particles

Table 1: Mean diameter of droplets in mass -> Mean mass-weighted diameter
Table 1: Mean diameter of droplets in number -> Mean number-weighted diameter

Table 2: Corrected from -> Corrected for

Figure 4: It appears that peaks above m/z 60 are reduced after the cloud events (although it could be an illusion created by the different aspect ratios of the mass spectra). Is there any significance to this?

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