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Interactive comment on “Understanding and constraining global secondary organic aerosol amount and size-resolved condensational behavior” by S. D. D’Andrea et al.

Anonymous Referee #2

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In this work, the authors examine methods of secondary organic aerosol (SOA) condensation in the global chemical transport model, GEOS-Chem, and the implications for particle size distributions. The base model forms SOA by assuming a 10% yield of aerosol from monoterpene emissions. The condensational behavior of that aerosol was assumed to be either mass (thermodynamic, MASS) or surface area (kinetic, SURF) based. A second set of simulations added 100 Tg/yr to the preexisting 19 Tg/yr of SOA and reexamined the impacts of mass and surface area based behavior on particle size distributions. A third set of simulations added corrections to the surface area uptake and did not significantly affect aerosol size distributions compared to the

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second set of simulations. Adding 100 Tg/yr of SOA in both the mass and surface area based parameterizations generally significantly improved model-measurement agreement for particle number distributions. Best agreement with observations was found for the surface area condensational behavior with an extra 100 Tg/yr of SOA. The paper is well written and it is easy to understand the author's method. The manuscript should be suitable for publication after addressing the following comments.

1. The authors discuss the idea of thermodynamic vs. kinetic condensational behavior on page 18974. Could the authors better clarify/distinguish between equilibrium/kinetic uptake and nonvolatile/semivolatile aerosol? The definition of thermodynamic behavior used here seems to assume equilibrium of semivolatile SOA. The kinetic behavior assumes nonvolatile SOA and diffusion-limited growth. Is it possible to examine deviations from equilibrium and volatility separately? Is it possible to get good model agreement if semivolatile aerosol (or aerosol spanning a continuum of volatility) is taken up by a parameterization accounting for diffusion and volatility (presumably the diffusion term is always present, it is just more or less important at times)?

2. Except for one metric in one size range (LMB, N40), the MASS-XSOA simulation is significantly improved compared to MASS-BASE. Does that indicate the assumption of equilibrium condensational behavior is less of a problem than getting the overall rate of SOA formation correct (in terms of implications for size distributions)? Is the observational agreement with MASS vs SURF specific to the amount of mass added? (What happens if you add 150 or 200 Tg/yr?)

3. Figure 6 of Riipinen et al. 2011 indicates the kinetic model significantly increases the number of particles in the 10^{-8} to about 3×10^{-8} m diameter range. Figure 8 of this work indicates that the SURF-BASE and MASS-BASE are very similar with SURF-BASE often being lower. Can the authors comment on why they are different?

4. Page 18982, Line 1-14, can more explanation of why these changes are seen be added? In particular, why are some decreases in N3 seen in Figure 5?

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Minor comments:

1. Why does sulfuric acid decrease when switching from SURF-BASE to SURF-XSOA (Figure 4)?
2. Page 18973, line 2-4, awkward sentence.

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