Atmos. Chem. Phys. Discuss., 13, C7141–C7143, 2013
www.atmos-chem-phys-discuss.net/13/C7141/2013/

Atmospheric Chemistry and Physics Discussions

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 #3

Received and published: 19 September 2013

The authors present a model study exploring sensitivities of SOA characteristics (amount and condensational behavior) to changes in CCN number concentrations. Specifically, they explore differences in the number concentration of particles with cutoff diameters of 3, 10, 40, and 80 nm with the latter two being proxies for CCN. They find that a surface-based distribution of SOA mass leads to best results in terms of statistical parameters if model results are compared to measurements at various locations in the US and Europe. Other parameterizations such as a SOA mass distribution based on preexisting aerosol mass or size-dependent growth rates for small particles lead either to a bias in predicted CCN number concentration or have little effect, respectively.

Overall, this model study provides an important step towards a better representation of SOA formation and particle growth in global (and likely smaller scale) models. The paper is very clearly written and easy to follow. I have only some minor comments that should be taken into account before the manuscript can be accepted for publication in ACP.

1) The conclusions are mostly a summary of the model results. There is nothing wrong with this fact; however, I am missing somewhat a conclusive recommendation for future model studies. Do the authors recommend that only surface-controlled ('kinetic condensation') should be used in models? In the beginning, they say that the truth is likely a combination of both thermodynamic and kinetic condensation. How should that transition be handled in models?

2) One proxy for CCN is N40, i.e. the number concentration of particles with diameter < 40 nm. Some information might be useful under what circumstances these particles might indeed be activated not cloud droplets. Using a typical hygroscopicity for SOA of ïĄń = 0.1, the critical supersaturation is >1% for a 40 nm particle. How frequent are such relatively high supersaturations around the globe?

3) Overall, the authors should carefully go through the manuscript and define all symbols and terms, even if they are common within the model community (e.g., BL, Fuchs surface area, C*, NH, SH, etc). In addition, while it is clear what the authors mean, some of the rather colloquial expressions should be corrected, e.g.: p. 18972, l. 4: CCN number concentration (not simply 'CCN') p. 18979, l. 8: "Each site measures. . ." should be replaced by "At each site, it is measured. . ." (or similar – the sites do not perform the measurements) p. 18982, l. 2 (and other places): "the annual-average BL percent change in N3, N10, N40 and N80" might imply that BL changes. It might be better to use " the annual-average percent change in N3, N10, N40 and N80 throughout the BL" (or similar) p. 18983, l. 4: Be more specific and explain what is meant here by 'aerosol microphysics'

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4) Some more explanation of the sensitivity of the CCN number concentration to SOA amount (Sect. 3.1) should be given: - Can the decrease of sulfuric acid with an added SOA condensation sink be explained by a faster particle removal? - The fact that fewer ultrafine particles exist in the SURF-XSOA case might appear somewhat counterintuitive. Is it because growth rates are so quick and thus all ultrafine particles are nearly immediately converted into N40 or larger? Please clarify.

5) End of Section 3.4: In order to better evaluate the order of magnitude of additional effects that impact CCN number concentration, could you give some approximate (percentage) uncertainties introduced by effects such as uncertainties in nucleation, primary emission etc?

Minor/technical comments

p. 18977, l. 3: What is meant by 'emission size'?

p. 18977, l. 24: is this a completely empirical factor without any units?

p. 18978, l. 10: In the previous section, it is 18 Tg year-1

p. 18979, l. 22: Why are N3 excluded from this analysis? Are there fewer measurements to compare with available? If so, say so.

p. 18980, l. 19: '. . .are the highest' (or 'sensitivity. . .is the highest')

p. 18985, l. 5: . . .are biased low

Figure 9, caption: It might be useful to refer here to Table 3.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 18969, 2013.