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Interactive comment on “Rethinking the global secondary organic aerosol (SOA) budget: stronger production, faster removal, shorter lifetime” *by* A. Hodzic et al.

Anonymous Referee #1

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This manuscript presents another ’rethink’ concerning global secondary organic aerosol, and although ’rethinking’ has already been done and will certainly be required many times more in the SOA world, I like both the title and intention of this paper. The manuscript presents a number of new ideas and points the way to the use of new constraints (namely vertical profiles) for the evaluation of proposed SOA mechanisms.

The general approach is generally sound I think, but there are some confusing aspects which I think the authors could address. My main concerns are:

- Discrepancies between observed and modelled OA are explained or discussed through the lens of problems with SOA-production and loss mechanisms, but we

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are not told how well the model performs for simpler species such as sulphate or nitrate (for vertical profiles), or for example NO_2 or CO. Maybe the under or over predictions seen for OA simply reflect dispersion issues and can be diagnosed through other pollutants? (As an extreme example, the authors worry about factor of two changes in OA over urban areas. I wonder how appropriate GEOS-Chem is for NO2 in urban areas for example.)

- In fact, I don't understand why a model with such a coarse resolution (2 × 2.5 degrees!) is compared with urban data or used to evaluate population-weighted SOA concentrations. This model is only suitable for consideration of large-scale concentration fields. Although I know that previous GEOS-Chem papers have also made use of urban data, I don't see the point and think that the paper would be stronger if it stuck to scales where one would expect GEOS-Chem to have some validity. I would remove the sections dealing with urban concentrations and health effects.

  (The authors were not really consistent with this anyway. In Sect. 2.5.2 they exclude data from heavily polluted regions because of the coarse resolution, but elsewhere they make use of urban data.)

- I missed more use of supporting data, e.g. O/C ratios, 14C, etc.
- P32423, L24 The authors should explain why their estimate of J-SOA differs from that of Henry and Donahue (2012) by an order of magnitude, and why their's is to be preferred. Presumably, if the much higher rates of H& D were used the vertical profiles and budgets would look very different.
- I missed a sensitivity test to illustrate the importance of the enthalpy assumptions. Would a different set of ΔH change the vertical profiles in a significant way?
- I found Sections 2.1 and especially 2.2 to be rather confusing.

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Sect. 2.1:

Explain why you have no oxygen gain after OH oxidation. This sounds unrealistic, and has implications for the O/C ratios of your system I would have thought.

The sentence "we do not support in any case ad hoc aging of oxidation products", begs the question: why not? Although the phrase ‘ad hoc’ sounds bad, ageing should be expected for SOA and SVOC in the atmosphere, and is usually assumed to add oxygen.

Sec. 2.2:

There were several parts of this section which confused me, since the methods mix SOM, VBS and ageing approaches in a complex way.

- P32419, L7, The SOM method is said to provide yield curves. However, Cappa and Wilson stated that the SOM framework is not well suited for species with multiple double bonds, such as sesquiterpenes or isoprene. In this case, how were yield curves made for these species?
- P32421, L9. Related to this, SOM was run assuming 10 ug/m^3 OA background, which suggests that the yield curves are more suitable for polluted environments rather than free tropospheric. How does this affect the results of this paper?
- And further, yield-curves and fits are rather specific to the chamber data being modelled, with the implied restrictions on time-scales. How can a VBS system (which excludes ageing) derived from a SOM run over limited chamber data cope with multi-generational chemical ageing?
- I don’t understand why only low-NOx yield curves are used for BSOA, since such compounds may clearly undergo oxidation in urban atmospheres too. (Indeed, this might be one reason for anthropogenic enhancement of BSOA production.) In any case, the cited Jo et al. paper just refers back to Henze et al. 2008 for this assumption, so it is better to cite the original source too.

- Finally, I am curious, why didn't the authors just use the SOM model, since it seems to underlie their VBS schemes?

Other points:

1. P32416, line 3. Better to say 'assumed small' dry deposition velocities, since most such models also neglect the observations of fast particle deposition to forests (e.g. Pryor et al., 2008, below).
2. P32416, line 5. It can be noted that Hallquist et al (2009) estimated that vapour-phase deposition of OC was substantially greater than particulate phase (800 TgC/yr vs. 150 TgC/yr, c.f. Fig. 1), so this issue of gaseous deposition has been highlighted previously.
3. P32422, L12-13. I was puzzled that H^{eff} values for terpenes were used for terpene products. Why would this be a good assumption for compounds such as pinic acid for example?
4. Notation issues.
    - Eqns. (1,4) and elsewhere uses () to represent concentrations, but the normal practice in chemistry is to use [].
    - P32427, μgsm^{-3}! The use of 's' for STP and not seconds here is very unconventional, in fact downright misleading, and completely unnecessary! I was actually a little shocked to see experienced scientists redefine such a well-known symbol within an otherwise SI-conforming expression. And why? The abbreviation STP has been used in every text book I have seen since my school days. Should readers expect to check the meaning of "m" also, or "g" to see if these were also redefined somewhere in the text?!

5. P32425, L5. Explain what loses 10% by mass - presumably that means for one molecule of SOA?
6. P32426 Which sampling times are used for IMPROVE and EMEP, and does this matter for the comments about evaporation of the IMPROVE samples?
7. P32433, L27. The recent paper by Denier van der Gon et al. (2015, below) reinforces the lessons about wood-burning from these earlier studies.
8. P32434. As noted above, I don't think we learn much from comparisons against urban sites, and in order to learn anything at all I would have wanted more information on model performance for other pollutants. For SOA the non-linearities of the equilibrium assumptions also make comparison of large grid-cell data against observations in an urban area very questionable.
9. Given the importance of free-tropospheric concentrations, I wonder what the authors used for any "background" aerosol assumptions? (Most VBS or SOA models require some kind of background OA, and this is usually assumed to consist of OA from sources not explicitly modelled, e.g. possible marine or other biogenic sources.)

Additional references cited

Denier van der Gon, H.A.C., et al., Particulate emissions from residential wood combustion in Europe - revised estimates and an evaluation, Atmos. Chem. Phys., 2015, 15, 6503-6519

Pryor, S.C. et al., A review of measurement and modelling results of particle atmosphere-surface exchange Tellus, 2008, 60, 42-75

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 32413, 2015.

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