

Interactive comment on “Reformulating the atmospheric lifecycle of SOA based on new field and laboratory data” by M. Shrivastava et al.

Anonymous Referee #2

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This manuscript addresses the potential broader implications of the results of the Vaden et al. (2011) PNAS paper. The paper presents a simple correction to the traditional Volatility Bases Set (VBS) SOA modelling approach that the authors feel is consistent with the results of Vaden et al. (2011). This correction is “irreversible partitioning” of the SOA (net condensation is allowed, but net evaporation is not). The authors use a box model and a 3D model to show some possible implications and compare to common SOA assumptions.

However, I agree with reviewer #1 that there are several major issues in the interpretation of the data. There are also several other issues that I am uncomfortable with in the paper. Therefore, I too feel that the paper is unpublishable in its current form and I recommend at least major revisions to the paper.

Major issues

- I agree with the first three major points of reviewer #1 regarding the interpretation of the Vaden et al. (2011) results. These appear to be potential misinterpretations of the results. I feel that reviewer #1 has done a thorough job of detailing these results, and thus I will not go into further detail. If the paper is to be published, I will need to be convinced that the points made by reviewer #1 are not correct, or the points made by reviewer 1 will need to be thoroughly incorporated into the revised paper.

- Irreversible partitioning: The conceptual framework of irreversible partitioning is physically impossible. It may be a useful means of getting behaviour that may be realistic in models (although the model results were not evaluated); however, I am uncomfortable with the concept of it.

Here is why: Lets say that reversible partition predicts that 50% of the SOM mass in one of your volatility bins should be in the aerosol phase with the rest in the gas phase. At this gas-phase concentration, the condensation and evaporation are equal, so there is no net condensation/evaporation. In the proposed irreversible partitioning construct proposed in this paper, you follow the reversible partitioning solution if this solution is larger than the previous aerosol amount. However, if the reversible partitioning solution is lower than previous aerosol amount, the aerosols do not net evaporate to the reversible partitioning solution. This implies that there is no evaporation at all (not even just no net evaporation).

If there is no evaporation when the reversible partitioning solution is lower than the previous aerosol amount, there should be no evaporation from the aerosol in other cases too (why would it be any different now?). Yet, we know that there IS condensation to the particle as long as there are condensible particles in the gas phase (e.g. 50% of the SOM in this scenario). Why does this SOM not stay stuck in the particle? Why does the amount of SOA in the reversible solution get “stuck” to the particle but any SOM that impinges onto the aerosol above this not get stuck?

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The above does not make physical sense. I realize that the authors intended the irreversible partitioning construct to be a rough means of representing the experiments (i.e. the last paragraph of the paper), but I feel that the oddness of physics should be described better. It is awkward to put down 100% functionalization approaches as unrealistic while this approach is also unrealistic. They both might work at giving better model solutions, but they certainly can not be describing the chemistry/physics that are occurring.

Other comments:

(Page 20124, Line 16) What is the basis for 75% fragmentation. Is this more realistic than 100% functionalization?

(Page 20124, Line 20) How do results change if POA and SOA were allowed to mix?

Lower activity coefficient: This is saying that when in solution, the aerosols have a lower volatility (you are lowering the volatility by a factor of 5). As the other reviewer said, the VBS approach has the activity coefficients implicit in the C^* s (and assumes they are constant), so you are essentially just reducing your C^* values by a factor of 5.

How do the SOA properties depend on RH? Would you expect them to still be highly viscous at high RHs?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 20107, 2011.

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