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Interactive comment on "The Secondary Organic Aerosol Processor (SOAP v1.0) model: a unified model with different ranges of complexity based on the molecular surrogate approach" *by* F. Couvidat and K. Sartelet

Anonymous Referee #2

Received and published: 11 March 2014

Comment on "The Secondary Organic Aerosol Processor (SOAP v1.0) model: a unified model with different ranges of complexity based on the molecular surrogate approach"

The authors have addressed current known challenges in organic aerosol modeling with a detailed module that treats partitioning and mass transfer limitations between many phases, not just the bulk particle and gas phases as is typical. The SOAP module bases its predictions on a surrogate compound approach, choosing the same detailed compound mixture as was used in Couvidat et al. (2012), while developing the

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capability of the model to handle variable particle internal composition and morphology. The organic aerosol community could certainly benefit from such a module, but I have serious reservations about this particular implementation and the discussion presented in the manuscript. I echo the other referee's concerns about language issues throughout the text. While many of these issues are surface or stylistic, several (as pointed out in the other review) compromise and confuse scientific meaning. In addition, I find fundamental problems with the methodology applied in the model as it is presented in the manuscript. These problems (detailed below) must be addressed through revision or proper citation before I can recommend publication in GMD.

Major Issues:

1. The authors' treatment of particle-phase diffusion is unclear or inconsistent throughout the text. The root of the confusion can be traced to equation (26), where the authors refer to the transport equation inside a spherical particle, and assume D_{org}, the organic particle-phase diffusivity, is independent of radial distance from the center of the particle. This assumption holds nominally for their discussion in section 2.3.1, which focuses on diffusion of organic compounds in organic particles, but breaks down immediately when considering particles of any greater complexity, whether they be mixed organic/aqueous particles or organic particles with multiple phases. The authors never mention the applicability of this assumption, and instead automatically extend the calculation for the mixing timescale (eq 29) to very complicated systems. For example, the test cases reported in Figs 4-7 involve aqueous-phase equilibration for variable D_{org}, yet it is not discussed whether or not molecules equilibrating in the aqueous phase must travel through the organic phase (or vice-versa). The authors only remotely address this issue in section 2.3.8 with the discussion of mass transfer limitation due to surface area coverage (also problematic, see issue 4). In regards to the variable diffusion in simultaneous organic phases, the authors avoid the issue by assuming one single diffusion coefficient for all of the phases, but if the goal is to apply this module directly to 3-D chemical transport models, then the possibility of variable diffusion

coefficients should be addressed or at least discussed in a revised manuscript. If it cannot be addressed, this inconsistency would seem to invalidate most of the derived equations.

2. The radial dependence issue again shows up in another major inconsistency embodied in equation (34). It seems inaccurate that the "evolution of concentrations in a bin due to condensation limited by the diffusion of organic compounds" (Page 392, line 7-9) can be described by equation (34), which is only dependent on concentrations in the layer of interest, the gas phase concentration, and a diffusion rate constant of the layer of interest. How can the concentration in other areas of the particle (i.e. closer to the surface) not be significant? For example, if a net driving force towards condensation causes an enhancement one quarter of the distance from the particle surface to the center just before the particle is exposed to decreased gas-phase concentrations, then the inner regions of the particle are susceptible to the elevated concentration near the surface until this material evaporates out. Interactions like this are the underpinning in complex models like KM-GAP (Shiraiwa et al., 2012), but seem to have been simplified out without acknowledgement in this SOAP module.

3. I was likewise confused by the discussion of morphological effects (section 2.3.2). I have never encountered the specific approach that the authors put forward, and yet there are no references whatsoever to attest to its validity. It seems remarkable that the overwhelmingly complicated phenomena at play when particles are made up of variable amounts of liquid, semisolid, and solid phases each with variable composition and mixing times can be reduced to a four parameter polynomial. Assuming that the application of equation (26) to the fitting procedure is valid (which I would argue against), would not any user of this model would have to provide fit parameters for every possible morphology (and composition, since composition and liquid/solid behavior are likely coupled) that might be encountered in the simulated atmosphere? This would seem to seriously jeopardize the module's flexibility and applicability in a host model. Figure 2, Table 2 and their presentation are extremely lacking in detail and discussion. It's not

clear to me how to connect this to the solid core setup. In fact, when after section 2.3.2 is a solid core even discussed?

4. The presentation of surface area contribution to limit mass transfer to simultaneous organic and aqueous phases (section 2.3.8) is problematic as well. There is no discussion of the validity of this approach or why the authors necessarily expect the aqueous and organic phases to be simultaneously at the surface but in different regions. If, as the authors suggest, one needs to take into account the fact that a gas molecule may collide with the "wrong" phase and not condense, and these phases exist in different concentrations, then I would expect the assumption of angular symmetry in the fundamental transport equation to break down. It is one thing to assume that the vapor pressure of a component over a well-mixed surface phase is affected by its mole fraction in that phase. It is quite another to suggest that there are separate, unmixed regions where the molecule does and does not exist in the particle phase. Depending on the vapor pressure and/or solubility of the compound, an angular dependent concentration gradient could develop with, for example, a shallow gas-phase gradient (in the radial direction) over the aqueous phase and a steep gradient (in the radial direction) over the organic phase. Again, any references would be helpful for this sparse area of discussion.

5. Page 404, Section 2.3.10, Lines 8-14: Given the issues discussed above, I am not surprised that some kind of numerical approximation like this is necessary, and this seems like a completely arbitrary, unexplained, and really undefended solution to the problem. How are the concentrations redistributed? What are the uncertainties? Were there other options?

6. I find the lack of model result characterization quite surprising. Admittedly, there are very few if any measurements existing to compare to. However, the authors could have compared to a fully complex model like KM-GAP that explicitly tracks fluxes to and from particle layers. They could have further introduced their phase separation algorithm and then determined how much accuracy is sacrificed when moving to the

SOAP model under various configurations. I would also like to see, in agreement with the other referee, reporting of model computation times under varying configuration options. How long does the user have to wait after introducing phase separation versus introducing multiple layers or multiple bins?

Examples of Minor Issues:

Page 398, Line 10: This is an example of how the authors use the word "equilibrium" throughout the manuscript with quite imprecise context. For instance, is this equilibrium referring to mixing or evaporation, or both. I strongly urge the authors to take more care in the use of equilibrium as they are dealing (intentionally) with several complicated physical phenomena and driving forces at once.

Page 400, Equation (66): The font size in the numerator on the right side is not consistent.

Page 404, Lines 1-2: This sentence is incomprehensible.

Pages 405-406, Lines 25-1: This sentence should be rewritten.

Figure 3: It is confusing to say the least. How does the equilibrium approach feed into the dynamic approach, with another equilibrium approach inside. It is worth noting that the authors do attempt to describe the model execution in the manuscript; however, I was not able to follow it. It is also worth noting that Fig. 3 does not include an output arrow. When do the calculations stop?

I have a large number of other minor and technical recommendations but would like to see the authors first address the major concerns above.

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Interactive comment on Geosci. Model Dev. Discuss., 7, 379, 2014.