

**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"**

**General Comments:**

*It is still not apparent to me why assuming the independence of the relationship between each layer and the gas-phase is acceptable. The authors assert this fact and express it in plain form (Eqn. 52 and 53), while also stating that overcoming it will be the subject of future work. There may be something to the underlying mathematics and scaling that makes the assumption reasonable, but, if so, I don't see it. More work with the model and other applications will bear this out. In the meantime, assumptions like this one make the methods section difficult to follow, as described below. For instance, it is hard to swallow the uniform application of the Kelvin effect term on each individual layer in Eqn. 53; it is equations like this that give the reader pause and interrupt the flow of the manuscript.*

*I appreciate the extra work the authors have put in to improve the presentation quality, model description, and discussion of results. Unfortunately, I think it is still a generally confusing manuscript (specifically parts of section 2.3). The language used is rather wordy in places and somewhat imprecise in others. The level here would normally be acceptable, but the assumptions and complexities of this particular model application make it extremely important that every word is as precise as possible. I am willing to recommend this paper for publication after the issues below are addressed.*

It is not quite right to say the relationship between each layer and the gas-phase is independent. There is no **direct** exchange between layers. Eq. 52 and 53 do not represent the flux of condensation/evaporation/diffusion (which is computed with Eq. 48) but the concentrations at equilibrium, which are used to compute the flux. The relationship between layers and the gas-phase depends on the other layers indirectly via the computation of the condensation/evaporation/diffusion flux. More specifically, this dependence is expressed by the factors  $f_i$  which are used in equation 52 to compute the concentrations at equilibrium and then the total condensation/evaporation flux. The factors  $f_i$  are also used to redistribute the total flux between layers. However, the model cannot simulate with this approach the entrapment of a compound inside a layer by the compounds inside another layer due to lack of affinity. It is this phenomenon that needs further work.

For the Kelvin effect, layers are just part of the particle. The Kelvin effect is a phenomenon acting at the surface of the particle but it impacts the condensation/evaporation over the whole particle. Therefore, the Kelvin effect is uniform over the whole particle.

*To make our paper clearer and easier to follow, we added 2 figures to detail the algorithm of section 2.3 (see answer to comments below).*

**Presentation Issues:**

*1) Section 2.3.3 is extremely difficult to understand. After reading it three times, I am still having a hard time following the subscripts and pluses and minuses. If there is any way to clarify the presentation, maybe with the addition of a cartoon illustrating the relationships among the various fluxes and factors, I would recommend adopting it. Part of the problem, I think, is that the authors are suspending, as they state, some laws of physics in order to make the simplifications they desire for efficiency. It then becomes hard to remember, for instance, which summations are happening over bins (Page 21, line 6) and which are over layers (Eqn. 52). The authors attempt to explain why local fluxes for specific layers can be ignored when they are opposite in sign to the global flux (Pages 22-23), but this explanation is also somewhat lost in*

*nomenclature and subscripts. I repeatedly missed the essence of the argument. Sentences like that on Page 23 (Lines 2-5) are generally confusing, "The total flux of... is separated into two fluxes..." yet there are three fluxes on the right hand side of equation 56.*

Two figures (figures 3 and 4) were added to present the approach for the computation of fluxes. A paragraph was also added at the beginning of the section :

"In this part, the methodology to compute condensation/evaporation/diffusion inside a viscous particle is described. The algorithm to compute condensation/evaporation/diffusion fluxes is divided into two steps shown in Figures 3 and 4. In the first step, the total flux of condensation/evaporation/diffusion over the whole particle is computed whereas in the second step, this flux is redistributed among layers. The detailed methodology to compute fluxes is described hereafter. »

As suggested by the reviewer, the sentence on page 23 needed clarification. Therefore,

"The total flux of condensation/evaporation/diffusion  $J_{\text{tot},i}^{\text{bin}}$  is separated into two fluxes  $J+$  (equal to the sum of diffusion fluxes which are positive) and  $J-$  (equal to the sum of diffusion fluxes which are negative)" is replaced by:

"The total flux of condensation/evaporation/diffusion  $J_{\text{tot},i}^{\text{bin}}$  is separated into three fluxes  $J+$  (equal to the sum of diffusion fluxes which are positive),  $J-$  (equal to the sum of diffusion fluxes which are negative) and the flux at the interface  $J_{\text{interface}}$ "

*Why is the statement on Page 23 (Lines 13-17) true?*

"compounds and layers for which diffusion acts in a different directions as condensation/evaporation (fluxes of different signs) should not be affected by the kinetic of condensation/evaporation." Is replaced by "compounds and layers for which diffusion acts in a different direction as condensation/evaporation (fluxes of different signs) should not be affected by the kinetic of condensation/evaporation (because the compounds are transferred from one phase to another without exchange with the gas phase)."

*2) The equilibrium timescale,  $\tau_{\text{eq}}$ , is defined with respect to a specific bin, layer, and species (Eq 75), but then in Sections 2.3.9 and 2.3.10, it is applied with reference to 'cases'. Are these 'cases' just specific combinations of bin, layer, and species volatility, or are the equilibrium/dynamic option applied globally. How much control does the user have? What error is introduced in Eq. 83 when only the mass at equilibrium is counted in the 'ratio' variable? Why isn't all of the material (equilibrium and dynamic) needed to get the driving force correct?*

A case corresponds to one bin/layer/species.

For clarity, the sentence "it is necessary to solve separately cases at equilibrium from other cases, which are solved dynamically." is replaced by "it is necessary to solve separately cases (combinations of one layer, one bin, one species) at equilibrium from other cases, which are solved dynamically."

Only the mass at equilibrium should be taken into account in Eq. 83 because only the mass at equilibrium is available for very quick condensation/evaporation. Equilibrium is here seen at a very fast (almost instantaneous) condensation/evaporation rate. Therefore for a compound to be able to be at equilibrium with some mass, the characteristic time of condensation/evaporation has to be quick enough (lower than  $\tau_{\text{eq}}$ ). If the characteristic time is higher, the compound cannot be at equilibrium with this mass.

There is no error introduced when only the mass at equilibrium is taken into account for the computation of equilibrium (as the mass in which condensation/equilibrium is dynamic does not change instantaneously).

Equation 81 is obtained by generalizing Equation 2 for several bins, layers and phases.

3) Page 29, Lines 16-20: How can the mass of layers stay constant if the layers are absorbing mass from the gas phase or releasing mass to the gas phase? I think a cartoon would help a lot here as well, but I leave it to the authors' discretion. It's difficult to understand how material can be transferred between layers instantaneously due to numerical inconsistencies and yet fidelity with respect to internal gradients can still be maintained.

It was a typo.

"the mass of layers must stay constant" changed to "the mass fraction (ratio of the mass of the layer over the mass of the particle) of layers must stay constant"

4) Does the 'explicit' case make the same assumptions about equilibrium between the internal layers and the gas phase that the 'implicit' case does? Is the major feature of explicit-ness the fact that there are 100 layers instead of 4 or 5? If so, are there important differences between this explicit case and another model like KM-GAP? Specifically, how are the fluxes in Eq. 85 calculated? I recommend moving the explanation of the explicit model to the methods section as well.

There are no equilibrium assumptions in the explicit representation. The condensation/evaporation/diffusion is solved dynamically.

The explicit representation is not just the implicit representation with 100 layers. Like in KM-GAP, the condensation/evaporation/diffusion was solved explicitly by discretizing the particle, and condensation/evaporation is only on the first layer.

The text was modified to emphasize those points.

The fluxes of Eq. 85 are computed with the equations given in the supplementary materials.

#### **Technical Issues:**

Page 2, Line 25-26: Can the authors add a quantitative measure here of what good agreement means?

Added (no significant differences after a few hours of condensation)

Page 3, Line 12: Please consider rephrasing "species need to be lumped" to "it can be advantageous to lump species"

Lumping species in 3D models is a necessity. Such models cannot represent all the species due to the computational cost.

Page 4, Line 26: What minimum solubility is implied by a species that completely dissolves in the aqueous phase at  $0.01 \mu\text{g m}^{-3}$  of water? My guess is one with a solubility of  $1 \times 10^{-4} \mu\text{g m}^{-3}$ ? Please state this somewhere.

"which condense only into an aqueous phase when the concentration of water in aerosol is higher than  $0.01 \mu\text{g m}^{-3}$ " changed to "which condense only into an aqueous phase when an aqueous phase is present, i.e, when the concentration of water in aerosol is higher than  $0.01 \mu\text{g m}^{-3}$ "

Hydrophilic compounds condense on the aqueous phase when there is water. If there is no water, hydrophilic compounds condense on the organic phase.

Page 9. Lines 9-10: The authors should state why they bother using parameters from UNIFAC at all, instead of just using parameters from AIOMFAC. The text reads as if AIOMFAC can give all the parameters, but the authors have chosen to use both methods. This could be a point of inconsistency.

As stated in the text, AIOMFAC computes only the activity coefficients for long range and medium range interactions and UNIFAC computes the short range interactions.

AIOMFAC can be used to compute short range interactions but it use the parameterization of UNIFAC with the parameters of UNIFAC.

*Page 31, Line 8: "...concentration and of organic particulate-phase..."*

No it is the organic phase concentration (as the organic phase can include water which is not an organic compound).

*Page 34, Lines 16-20: Can't see this from figure 4.*

*Figure 4: It is difficult to see most of the action in these plots. It does not seem like the low vicinity of the x-axis ( $10^{-12}$  to  $10^{-5}$ ) is necessary. Please consider zooming in.*

*Section 3.2: Should be moved to before the results discussion.*

Figure 4 was modified to be more readable.

We think it is better to let this section in results to focus on the model description in the other sections. The explicit representation is different from the implicit model presented in this paper.

### **Typos:**

*Page 3, Line 19: "strongly impact predicted OA formation."*

Replaced by simulation OA formation

*Page 3, Lines 13-16: Perhaps replace "the surrogate based methodology" with "surrogate-based methodologies" to emphasize their use throughout the field.*

Done

*Page 5, Line 24: Please remove the second occurrence of "dynamic". i.e. "In the dynamic approach, the condensation and particle diffusion of organic compounds"*

Done.

*Table 4 Caption: Reverse the last comparison. "...very high for compounds with  $K_p \leq 0.01$ ". The last sentence should read "A and B types are, respectively, hydrophilic..."*

Done.