The manuscript was modified to take into account the comments of reviewers. The parameterization of AIOMFAC used in the model was also corrected according to Corrigendum to "A thermodynamic model of mixed organic–inorganic aerosols to predict activity coefficients" (Zuend et al., 2012). Numbers were corrected but the conclusions of this study remain unchanged.

Anonymous Referee #1
Received and published: 11 March 2014
The Couvidat and Sartelet manuscript reports on a flexible model (SOAP) for predictions of secondary organic aerosols (SOA) intended for use in three-dimensional air quality models. The availability of a flexible SOA model is attractive and would represent a novel contribution to the field. However, it is recommended that the manuscript be significantly improved before being published in GMD. The manuscript has weaknesses in overall language and fluidity (presentation quality), as well as proper citation of related work (scientific quality). There are also a few specific scientific quality questions to be addressed. Details of these areas needing improvement are provided below.

Presentation Quality: It is recommended that the authors carefully review the language in the manuscript and if necessary, request the assistance of an additional reader/editor. In many places it seems errors may be due to carelessness and thus the latter would not be required. For example, condensation or absorptive partitioning describes dissolution of a gas-phase species into a particle; throughout the manuscript the authors discuss condensation "on" or "onto" aqueous and organic phases/particles.
In a few places the authors correctly describe condensation into the aqueous and organic phases/particles. It is important to distinguish these processes because partitioning on/onto a particle surface is also important in the atmosphere, but not the subject of this manuscript. A few more examples follow, which are relevant for interpretation and reproduction:
"Condense on or onto" was changed to "condense into". Furthermore, the language was carefully checked.

p. 385, line 10: The authors discuss the need to account for inorganic aerosol formation "previously". Do they instead mean separately? Or concurrently? Also, the authors should provide a complete list of the SOAP inputs from ISOROPIA, rather than a partial list as indicated by "such as".
"SOAP does not currently take into account formation of inorganic aerosol, which must be computed previously with an inorganic aerosol model like ISORROPIA to provide input for the model such as pH, concentrations of inorganic ions, ionic strength and the liquid water content of aerosols." Replaced by:
"SOAP does not currently take into account the formation of inorganic aerosols. An inorganic aerosol model like ISORROPIA must be called separately and prior to the call of SOAP to provide inputs to SOAP: pH, concentrations of inorganic ions, ionic strength and the liquid water content of aerosols."

p. 405, line 3-7: It is not clear what the authors mean when they write that "As in H2O, some species do not have a molecular structure attached: : :". "However, : : : is taken into account using a default molecular structure as in H2O." Do they mean that while molecular structures are specified for some surrogates, the others are defined by a single default structure? Is that structure the same for each surrogate (which seems problematic given the range of surrogates listed for which there is no molecular structure)? If it is not the same for each surrogate, how is a default structure for each surrogate different than a molecular structure?
More information about the default molecular structure was added:

"In H2O, the species that are representative of primary and aged SVOC (POAlP, POAmP, POAhP, SOAlP, SOAmP and SOAhP) do not have a molecular structure attached. Therefore processes depending on the molecular structure (like absorption on the aqueous phase) are

not estimated for those species. They are assumed to be hydrophobic and their influence on the activity coefficients of other species is taken into account by assigning to them a default molecular structure representative of primary aerosol and lowly oxidized compounds. This default structure is constituted of 40% of $C_{23}H_{47}COOH$, 5% of $C_8H_{17}CH=CHC_7H_{14}COOH$, 15% of 4-(2-propio)-syringone, 12% of $C_{29}H_{60}$ and 28% of 2-carboxybenzoic acid based on EPRI (1999). In SOAP, the same default structure as in H2O can be used if the user does not provide a molecular structure."
More details and discussions about the molecular structure can be found in the H2O paper [Couvidat et al. (2012)].

The abstract lacks discussion of results. Inclusion of some statistics on the relative performance and computational time of the various model configurations is recommended. A section "time analysis" was added to provide information about computation times. Result discussions are added to the abstract:
  "According to preliminary results, the non-ideality of the aerosol (including interactions with inorganic ions and separation of the organic phase into several phases) should be taken into account in models as it may affect SOA formation. Moreover, some compound should not be assumed only hydrophilic or only hydrophobic as they can condense on both the organic and the aqueous phases."

The authors may want to consider putting some of the developmental equations in supplementary material. For example, equations 1-7 are well known and may not be needed. Equations 17-19, 22, 24 describing partitioning to an aqueous phase are analogous to equations 14-16, 21, 23 for partitioning to an organic phase; one set could be moved to a supplement, with the differences described in one or two sentences.(e.g., "Analogously, partitioning to an aqueous phase can be represented by equations x, y, z, where Kp,i is replaced by Ka,i: : :".)
Equations 1 to 7 are well known but for text clarity, we think it is better to keep them in the text, because we refer to them several times in the text and they are necessary to understand this paper. Therefore, they need to be clearly stated with the notations used in the paper.
Equations 17-19, 22, 24 are analogous to 14-16, 21, 23 but we think that the paper may be difficult to read if they are removed, as they are also referred to.

There are multiple instances in which the authors mention "several" particulate phases; however, the manuscript details consideration of up to two phases only.
Yes, regardless of the aqueous phase, SOAP can be separated into several organic phases and the model can have more than 2 phases as described in the section "Saturation and separation of phases".
The following sentence has been added to this section to provide more explanation about phase separation:
"If compounds having a low affinity with each other coexist inside a single organic phase, the organic phase may become saturated by some compounds and may become unstable. In that case, the separation of the organic phase into several organic phases may occur."

Scientific Quality: The authors introduce a number of model approaches in which one or more of the considerations discussed in the manuscript have been taken into account. The authors miss an important publication in which phase separation and partitioning kinetics have been considered: Shiraiwa et al. (2013), Phys. Chem. Phys., 15: 1141. Relatively few references appear in the methodology, for example previous model approaches for phase separation (e.g., Zuend and Seinfeld, ACP, 2012; Erdakos and Pankow, 2004).
The references were added in the methodology.

The results are not put in the context of existing work considering non-ideality, phase separation, and kinetic partitioning (including the influence of RH on equilibrium timescales, see for example Saukko et al., 2012; Renbaum-Wolff et al., 2013).
The references were added in the introduction of the section about dynamic representation.

p. 381, line 6: Total particulate mass? Or total fine particulate mass?
Total fine particulate mass. Corrected.

p. 382, line 3-5: It is not clear what the authors mean when they discuss separation of organic matter into several phases due to saturation.
"the separation of organic matter into several phases due to saturation" was changed to "the phase separation of particulate organic matter into several organic phases"

p. 384, line 18: The authors state that saturation vapor pressure is usually determined experimentally. However, the manuscript is describing a molecular surrogate approach, thus experimentally determined vapor pressures would not be available. It would be more appropriate to describe the approach for obtaining the saturation vapor pressures for the molecular surrogates.
For clarity, "Moreover, P0i is generally determined experimentally at a temperature Tref" changed to "Moreover, P_i^0 or partitioning constants $K_{p,i}$ are the same as those used in H2O (they are generally determined by fitting to experimental results obtained in environmental chambers at a temperature Tref)."

p. 386, line 6: Does "AQ" include water?
Yes. Added in the text.

p. 386, line 16: The authors state that UNIFAC only accounts for short-range interactions. This is not accurate. See for example Erdakos et al., 2006.
"However, UNIFAC only compute the activity coefficients due to short-range interactions" changed to "However, the original UNIFAC (Fredenslundet al 1975) only computes the activity coefficients due to short-range interactions"
Furthermore, it isn't clear why the authors don't just use AIOMFAC for the short-range interactions as well?
AIOMFAC use the UNIFAC parameterization for short range interactions.

p. 387, line 18-21: It isn't exactly clear why SOAP would predict less water than the inorganic model, assuming that total water is "organic water" + "inorganic water", and the inorganic water is used as an input to Eq 13. The authors should expand/clarify this point.
The following discussion was added to the text:
"This problem arises because inorganics are not computed within SOAP (in that case, there would be no numerical issue) and water concentrations can be lower in SOAP than in the inorganic model ISORROPIA for several reasons. First, there can be numerical differences between SOAP and ISORROPIA because different parameterisations are used in SOAP and ISORROPIA to compute water. Second, the amount of water absorbed by the aerosol can be different from the sum of the water amount absorbed by inorganics and the water amount absorbed by organics. Choi and Chan (2002) found that organic species can either reduce or enhance the water absorption of inorganics compounds. Depending on the conditions, the amount of water computed by SOAP could be higher without organics than with."

p. 387-388: Given that each molecular surrogate has some fixed properties, it isn't clear why the user would want to specify hydrophilicity/hydrophobicity, unless there is an option to add structures, which is not discussed in the manuscript. The flexibility to consider partitioning to one or both phases is appealing, but when the "both" option is turned off, it seems that each structure would simply have an assigned preferred phase.

We moved the discussion about the specification of hydrophilicity/hydrophobicity to the overview section and we added more details.

"A molecular structure has to be assigned by the user to each surrogate compound. A default structure is provided for each surrogate, as in H_2O. This structure is used to compute the non-ideality of the aerosol via activity coefficients. If the user specifies that a compound is both hydrophilic and hydrophobic, the repartition between the phases is done according to the value of activity coefficients. However, the user can decide that a compound is only hydrophilic or only hydrophobic because the absorption of the compound in one of the two phases is negligible (for example alkane or lowly oxidised compounds are probably not absorbed by the aqueous phase of particles). Moreover, if there is no compound that is both hydrophilic and hydrophobic, the condensation into the organic phase can be solved separately from the condensation into the aqueous phase. The system is then uncoupled. On the opposite, if there is at least one compound which is hydrophilic and hydrophobic, the condensation into the organic phase and the condensation into the aqueous phase must be solved simultaneously. The system is then coupled and consumes more CPU time. The user may prefer an uncoupled system for 3D application due to higher time efficiency."

p. 394-395: The authors describe the use of Peq,i instead of saturation vapor pressure to account for the Kelvin effect (equations 41, 45); however, Peq,i is not the Kelvin corrected saturation vapor pressure (as suggested by Eq 41) which is the correct substitution in Eq 45. See for example Pierce et al., 2011.
Corrected.

p. 408, line 6-7: What is the rationale for chosen accommodation coefficient and gas phase diffusion coefficient?
For all the compounds, the gas-phase diffusion coefficient is taken equal to 0.1 cm2 /s which is the order of magnitude of this parameter (Seinfeld and Pandis, 1998) and the accommodation coefficient (value between 0 and 1) is taken equal to 0.5 so that condensation is mainly limited by diffusion in the organic phase, which is the main phenomenon that we want to study here.

Editorial (these are a few examples, many others exist): Section 2.1, p. 382, line 24/25: "has been developed" not needed p. 383, line 4: "process" should be "processes" p. 409, line 11: sp. "measuremeants"
Corrected.

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 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.

We want to thank the reviewer for his/her comments. We improved the paper by adding details about the assumptions made and citations. The dynamic approach of SOAP is deliberately a simplified representation of the particle phase diffusion and it does not aim at representing it as thoroughly as Km-GAP or the model ADCHAM newly published in ACPD. The SOAP dynamic approach presented here is a simplified approach to take into account particle phase diffusion with a low number of layers, and a computation time as low as possible to be used in 3D air quality models.
The introduction is modified to emphasize this point.

An introduction to the dynamic approach in section 2.3 was also added to describe and discuss properly the effect of the different assumptions made in the model:
"The dynamic approach, which is presented hereafter, is a simplified approach to take into account particle-phase diffusion with a low number of layers, and a computation time as low as possible to be used in 3D air quality models.
The first assumption is that the organic-phase diffusion coefficient is constant over the entire particle. It does not depend on the distance to the center of the particle. Although this assumption may not be valid, it is reasonable, because, currently, to our knowledge, there is no parameterization to evaluate the order of magnitude of organic-phase diffusion coefficients.
The second assumption is that the concentrations in one layer can be described independently from the concentrations in other layers (see the following section for more details). This hypothesis means that in the model there is no exchange of compounds between layers and that the compounds condense directly from the gas phase to the layer or that they evaporate directly from the layer to the gas phase by taking into account an equilibration time specific of the layer. Compounds condense into a layer or evaporate from a layer as if the other layers had the same affinity with the compounds. Effects of entrapment of compounds inside central layers by the layers closer to the interface with the gas phase (compounds inside the central layers having a low affinity with the compounds of the layers at the interface are not able to evaporate whereas compounds having a high affinity with the central layer but having a low affinity with the layers at the interface are not able to condense into the central layer) are not taken into account. The model should give however a good estimation of the capacity of the organic matter to absorb compounds.
The third assumption is that the organic matter and the aqueous phase evolve separately, i.e., there is no kinetic of transfer of compounds between the organic matter and the aqueous

phase due to the complexity of representing properly those transfers, which should be strongly depend of the morphology of the particle. If a compound tends to go from the aqueous phase to the organic matter, it has first to go from the aqueous phase to the gas phase and then from the gas phase to the organic phase. For example, in case of evaporation of the aqueous phase (that can be due to a strong change of the relative humidity), this assumption can create some evaporation/recondensation issue (compounds evaporate and recondense after some time into the organic matter according to condensation/evaporation fluxes) whereas a part of organic compounds should go directly from the aqueous phase to the organic matter. It could also be possible that if an aqueous phase and an organic phase coexist into the same particle that organic compounds do not condense directly from the gas phase into the organic phase (because the kinetic is too slow) but condense first into the aqueous phase and then go from the aqueous phase to the organic phase (if it is quicker for a compound to condense into the organic phase by this pathway). However, it can also be argued that if there is an aqueous phase (the relative humidity is too high), the organic phase may not be significantly viscous."

Conclusions are also modified.
"Future works will focus on improving the framework of this dynamic approach to take into account varying diffusion coefficients with layers, to represent layer exchanges and transfers between the organic and the aqueous phases. Simulations with SOAP and comparisons to measurements should be performed to validate the model and to test the influence of each process and parameters on organic aerosol formation. Comparison to a model like KM-GAP can also be useful for validation and in the development of layer exchange processes."

These modifications should address most of the comments below.

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 Dorg, 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 Dorg, 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.

These points are now addressed in the introduction of the dynamic approach. We now state and discuss the assumptions (constant diffusion coefficient and no exchange between aqueous and organic phases).The assumption on Dorg (particle-phase diffusion coefficient) for Eq. 27 is now written clearly in section "Diffusion of organic compounds in spherical organic particles"

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.

This point is now addressed in the introduction of the dynamic approach.

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?

The morphology factors approach is an approach that we designed to take into account the morphology of particles in a simple way. We chose this approach because 3D air quality models need simple parameterizations (which do not solve explicitly the diffusion into very complex morphology) with layers (the volume fraction needs to be constant like the diameter of size sections is constant in air quality models) and characteristic times of layers (which can change with the morphology).

As diffusion of organic compounds is affected by the presence of solid phases, morphology factors are designed to take into account this solid phase where diffusion may not occur. We assume that this solid phase is located at the core of particles.

The sections on "diffusion of organic compounds in more complex particles was partly rewritten for clarity, and more details on how to compute morphology factors are added in Appendix. Morphology factors were used for every simulation of this paper where a solid core occurs, as mentioned in the"Results" section:

"The volume of the solid core of each size section is computed from the concentrations of solid species (like dust, black carbon, solid inorganic given by ISORROPIA) to compute morphology factors."

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.

One of the assumptions of SOAP is that the organic matter and the aqueous phases evolve separately. This assumption has to be made, because of the difficulty to represent transfers between the aqueous and the organic phases within particles with coexisting organics/inorganics of various origins. This assumption is now described in the introduction of the dynamic approach.

" The third assumption is that the organic matter and the aqueous phases evolve separately, i.e., there is no kinetic of transfer between the organic phases and the aqueous phase, due to the complexity of representing properly those transfers, which should strongly depend on the morphology of particles. If a compound tends to go from the aqueous phase to the organic phase, it has first to go from the aqueous phase to the gas phase and then from the gas phase to the organic phase. For example, in case of evaporation of the aqueous phase (that can be due to a strong change of the relative humidity), this assumption can create some evaporation/recondensation issue (compounds evaporate and recondense after some time into the organic phase according to condensation/evaporation fluxes) whereas a part of organic compounds should go directly from the aqueous phase to the organic phase. It could also be possible that if an aqueous phase and an organic phase coexist into the same particle that organic compounds do not condense directly from the gas phase into the organic phase (because the kinetic is too slow) but condense first into the aqueous phase and then go from the aqueous phase to the organic phase (if it is quicker for a compound to condense into the organic phase by this pathway). However, it can also be argued that if there is an aqueous phase (the relative humidity is too high), the organic phase may not be significantly viscous."

Furthermore, because the organic phase and the aqueous phase evolve separately and because compounds can condense into the organic phases and the aqueous phase (if they are both hydrophilic and hydrophobic), the kinetic of condensation into the aqueous phase must be a fraction of the total condensation (organic+aqueous). Otherwise, the sum of the kinetic of the condensation into the aqueous phase and into the organic phases may be too high (because the kinetic of condensation is then counted twice).

As for angular symmetry, as we assumed that there is no transfer between the organic phases and the aqueous phase, there is no angular dependency of concentrations. However, if those transfers are added and if the aqueous and the organic phases are in two different region of the particle, a radial dependency of concentrations should be taken into account. For 3D air-quality modeling purposes, the gas-phase gradient (in the radial direction) the angular is not important, as particles are assumed to be fully mixed in a large volume (at least 1 km2 x 30m).

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?

A section about the redistribution was added in the text.

"To use this approach the mass of layers must stay constant and the mass of each layer must respect the condition given by Eq. (35), which specifies the mass of the layer with respect to the total mass. However, due to rapid condensation or evaporation of the layer near the interface, concentrations of organic compounds need to be redistributed over layers to respect this condition. If compounds are not redistributed some layers will become bigger (due to differences in fluxes) and the layer near the interface may for example become bigger than the layer in the center of the particle. Moreover, as the volume of the particle will change with the condensation/evaporation, the concentration of a layer can be transferred to another layer. If a particle grows due to condensation the layer at the interface, compounds that were previously in the layer will be "pushed" into more internal layers and the newly

condensed compounds will be at the interface layer. On the opposite, if a particle shrinks due to evaporation of the layer at the interface, the missing mass of the layer will be taken from more internal layers. For a case of evaporation, the mass of the layer at the interface may be too low (due to the more rapid evolution at the interface) and the missing mass of the layer is taken from layers at the inside of the particle, i.e. concentrations are redistributed from the outside to the inside. For a case of condensation, the mass of the layer at the interface may be too high, the exceeding mass of the layer is redistributed over the layers at the inside of the particle, i.e. concentrations are redistributed from the inside to the outside. The algorithm is detailed in Table 3.

The redistribution algorithm induces numerical diffusion as a small fraction of the mass of a layer is always transferred to other layers. This redistribution effect should decrease if the number of layers increases but it is necessary for 3D application to keep the number of layers low. However, the redistributed amount should be low compared to the absorbed amount of organic compounds. Similarly, in 3D air quality model, concentrations and number of particle in size sections have to be redistributed between sections so that the bounds diameters of sections are kept constant, which does also create some numerical issues. In SOAP, the diameters of sections evolve according to the mass that condense or evaporate (without changing the number in sections) and there is no size redistribution between sections. However, a size-section redistribution algorithm should be added to the code if coagulation is added (for modeling purposes) or in 3D models (in which SOAP would be implemented) after the call of SOAP."

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?
A section on time analysis was added. The usefulness of comparison of SOAP to KM-GAP is added in the conclusion for future work.

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.
The phases involved in the equilibrium (equilibrium between gas phase and a condensed phase or equilibrium between condensed phases) are added at each occurrence of the word equilibrium in the paper.

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

Page 404, Lines 1-2: This sentence is incomprehensible.
The paragraph was rewritten.

Pages 405-406, Lines 25-1: This sentence should be rewritten.
"1.20 µgm^{-3} for non-ideality and without long and medium ranges interactions but with short-range interactions." replaced by "1.20 µgm^{-3} for non-ideality with only short-range interactions."

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?

The text was modified to provide more information about the model structure. Output arrow was added to the figure.

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