

Topical editor review of Crooks et al. "The co-condensation of semi-volatile organics into multiple aerosol modes".

The authors responses and associated revisions have addressed the issues identified by the two reviewers, and the paper is now close to being acceptable to proceed to publication.

However, despite the manuscript being much improved, my opinion as Topical Editor, is that there are a few key issues that need clarifying and parts of the manuscript are not well written, even in the revised version. The paper is not yet of a high enough standard to warrant publication.

For that reason, I am recommending the authors make a further revised version responding to the points I make in this Topical Editor review.

After reading the revised manuscript, it was clear that there are a considerable number of minor revisions required and I list these below.

Provided these issues can be addressed, the paper could be suitable for publication. However, I will need to see the paper again after the authors have revised it, to consider whether or not it can be signed off for publication in GMD.

In addition to the minor revisions, there are several major issues.

First, both reviewer 2, and my initial review identified an issue with the term "co-condensation".

There seems to have been some confusion during the reviews, which I believe to be mostly caused by the use of the term "co-condensation".

In one of their responses to reviewer 2 (the one numbered 1), the authors explain they decided to change an occurrence of the term "co-condensation" (on line 85) with the term "equilibrium absorptive partitioning".

In my initial Topical Editor review I suggested the authors consider replacing the term "co-condensation" with the more general term "partitioning". My view there was that the term "co-condensation" was not really appropriate terminology for semi-volatile species (because it implies only one-way gas-to-particle transfer). I suggested the term partitioning to avoid any potential confusion whereby the reader might initially assume the method follows a kinetic approach.

I agree that using the longer and more descriptive term "equilibrium absorptive partitioning" represents a further improvement over the single-word term "partitioning".

However, although the authors have revised that particular instance of the use of the term, the title of the paper still uses the term "co-condensation".

For the reasons given above, I strongly recommend that the authors avoid using that term "co-condensation".

In response to my querying the term in my initial editor review, the authors did not defend its use, instead stating

"It wasn't clear whether "co-condensation" in the title was ok or not."

When considering that response, I chose not to press the authors to revise the term, instead leaving the term in to be considered

later during the main review process.

Since reviewer 2 has also requested that change, I am here requesting the authors replace the term with "equilibrium absorption partitioning" in all instances in the text, including the title.

The 2nd major comment I have is regarding the two restrictions that are placed on the method as it is being tested here. See my two points 40 and 43 -- there needs to be stated clearly some discussion of how these two restrictions limit the method's applicability in a global model. How dependent is the method on these two restrictions?

I need to be convinced this is properly discussed and considered before allowing the paper to proceed to publication in GMD.

The 3rd point is regarding the section 7.3.2 which requires quite some improvement (see my points 46 to 49)

The final point I make is about the conclusions which need to be improved considerably with some discussion about the limitations of the method and its applicability in a global model (see my point 50).

It is for these latter 3 reasons that mean that I need to ask to see the paper again after these revisions have been made before I can decide whether or not it can proceed to publication.

Specific minor revisions

- 1) Abstract, Page 1 line 1 -- insert the word "absorptive" between "equilibrium" and "partitioning" to match the terminology used in response to reviewer 2's comment (see above). Also replace "gas/particle concentrations" with "gas and particle concentrations". In this instance the partitioning is determining both the gas and the particle concentrations, so both terms should be stated -- it is not a case of one or the other.
- 2) Abstract, Page 1, line 4 -- insert comma between "problem" and "the"
- 3) Abstract, Page 1, line 5 -- replace "on the aerosol" with "of the aerosol"
- 4) Abstract, Page 1, line 7 -- I found the term "pivotal" here not quite appropriate -- to me the other assumption about the organic mole fraction in the partitioning coefficient being across all modes seemed a more pivotal assumption that enabled the equations to be approximated by a simpler solution. In fact I recommend to re-order the sentences with the sentence beginning "The resulting coupled non-linear system" being moved up to follow after the sentence ending "on each involatile mode". The text seemed to flow well with that ordering, and it seemed to me that the primary assumption was the organic mole fraction -- with the part about each mode containing some involatile core (so never encountering complete evaporation) being secondary. I strongly suggest to replace the words "The pivotal" with "Another key" and move that entire sentence to come after the sentence ending "is set to be equal across all modes." In that sentence referring to the involatile constituent I also recommend to delete "to create a monodisperse aerosol at equilibrium" and instead finish that sentence as "thereby avoiding associated numerical difficulties" or similar. That "monodisperse aerosol" didn't make sense to me -- but replacing to something like that referring to the numerical issues avoided would work I think there.

- 5) Introduction, page 2, line 27 -- referencing style is incorrect here (and in similar instances of several refs in brackets). Need to revise to avoid double-brackets -- use /citep if in LaTeX rather than /citet.
- 6) Introduction, page 2, line 40 -- Insert "than smaller ones" after "water vapour more quickly".
- 7) Introduction, page 2, line 40 -- Replace "Consequently, this can suppress..." with "Consequently, the presence of a greater number of larger CCN can suppress..." or similar (be specific). Also replace "prevents the smaller particles from activating" with "causing fewer smaller particles to activate" or similar.
- 8) Introduction, page 2, lines 42-43 -- delete "the" between "alter" and "precipitation" and change "rate" to "rates".
- 9) Introduction, page 2, line 44 -- avoid the term "direct effect" here as that tends to be used in association with aerosol-radiation interactions -- suggest to instead use "main effect" or other term.
- 10) Introduction, page 2, line 47 -- "an approximate -0.7 W/m² increase" change the phrasing to avoid negative increase -- also please give the uncertainty range given by the radiative forcing chapter of the IPCC report. You have "which is on the order of" but it's not clear whether you are referring to the best-estimate value or the uncertainty range itself. Please clarify. Also use the recommended method to cite chapters of the IPCC report -- you should cite the chapter authors rather than the entire report -- i.e. Myhre et al. (2013) for the radiative forcing chapter.
- 11) Introduction, page 2, line 52 -- You have "primary and secondary aerosol" but I think you mean "primary and secondary particles" (i.e. you mean to differentiate between particles that are directly emitted (primary) and those which are generated separately via new particle formation. Also suggest to replace "Primary particles are..." with "For example, primary particles are...." at the sentence after.
- 12) Introduction, page 2, lines 55-56 --- whereas the preceding sentence describes primary particles, here the text is explaining the production of secondary aerosol *mass* rather than the presence of secondary particles (i.e. those which are nucleated). The way this para is worded currently could confuse the reader re: the distinction between secondary aerosol particles and secondary aerosol mass. The sentences after can still be used in relation to the secondary particles because the growth of the nucleated (secondary) particles to climate-relevant sizes is strongly influenced by the production of secondary organic aerosol (mass). So it's fine to mention that the oxidation of VOCs to form SOA is an important uncertainty, as you have done, but make it clear that the issue is how the condensation of SOA causes growth of secondary particles (which are only initially at nanometre sizes).
- 13) Introduction, page 3, lines 68-71 -- The first part of the sentence beginning "It is therefore impractical..." (up to where you have "...each compound individually") seems to be building up to making a point about how models need to lump organic species together into a small number of species representative of compounds with similar properties. Whereas the 2nd part (beginning "two popular methods") then seems to refer to the separate issue of how to treat the partitioning of the species to the particle phase. I know these issues are dependent on each other but in my mind they are separate issues. I'd advise to finish off the sentence finishing the point about the first part -- suggest to give a

reference to models that have lumped the organics in different ways -- you could cite (for example) Oâ\200\231Donnell et al. (2011), who have implemented a relatively complex representation of organics compared to other global models -- suggest you also cite the AeroCom organics intercomparison paper (Tsigaridis et al., 2014) in terms of the typical (lack of) sophistication or organic aerosol schemes in global aerosol models.

Then go on to make the points in that 2nd part with a separate sentence or sentences explaining the issues around difficulties & advantages of the different approaches to gas/particle partitioning.

14) Introduction, page 3, lines 74-75 -- re-word the excerpt of text "The first noteworthy approach (Odum et al., 1996) proposes an..." as "The first (Odum et al., 1996) involves an...". The "noteworthy approach" seemed a little unscientific language.

15) Introduction, page 3, line 77 -- replace "these models are found to be..." with "the approach has been found to be..." -- I think it's better to refer to the approach used rather than a particular set of models.

16) Introduction, page 3, line 88 -- you have "particles above above 50%" please correct -- do you mean "occurs only for relative humidity above 50%"? Also -- at the end of the sentence you say "which is typically of atmospheric relevance" -- re-word that -- what exactly is typically of atmospheric relevance -- the rh value? Or do you mean this is always achieved everywhere in the boundary layer or similar? Say why it's of relevance -- do you mean that therefore this condition is always satisfied in most atmospheric conditions?

17) Introduction, page 3, lines 92-95 -- re-word this sentence -- you say the existing particles are assumed to be involatile -- but is this a general statement about what most models do? Or the model you are using -- please clarify and improve the wording.

18) Introduction, page 3, lines 97-100 -- insert commas between "without it" and "a polydisperse" -- and also between "particle case" and "the smaller..."

19) Introduction, page 4, lines 108-110 -- insert commas between "as a result" and "the equilibrium" -- and between "organic mole fraction" and "a method..."

20) Section 2, page 4, lines 116-119 -- re-word this sentence that currently begins "Multiple organic species" and delete the citation to Donahue et al. (2006) in the first sentence (refer to it in the 2nd sentence when you actually introduce the VBS approach). I suggest to move the text "volatility basis set" to the start of the sentence so that it begins something like "The volatility basis approach (Donahue et al., 2006) involves binning multiple organic species into a limited number of notional organic species with representative saturation concentrations (volaility)...." Just needs rewording.

21) Section 2, page 4, lines 119 -- delete the word "calligraphic".

22) Section 2, page 5, line 139 -- change notation to avoid use of the term "C_OA" -- that notation is confusing beacuse the reader will expect the OA to refer to organic aerosol whereas in fact it is representing the total concentration of all compounds in the condensed phase.

23) Section 3, page 6, line 163 -- replace "size of the aerosol" with "size of the aerosol particle" or just "size of the particle".

24) Section 4, page 7, line 194 -- replace "the only the molecules" with "the only molecules".

25) Section 4, page 7, line 200 -- after "It is this quantity" suggest

to add in brackets exactly what you are referring to -- i.e. something like "(the remaining concentration in the vapour phase)"

26) Section 4, page 7, caption to Figure 1 -- there seems to be a word "equilibrate" on its own underneath the main caption -- presumably this is a typo in the LaTeX or similar? Please correct.

27) Section 4, page 8, line 209 -- Re-word start of sentence beginning "Dummy indices" -- they're not really dummy indices as they are presumably chosen to provide a particular distinctive information compared to the main indices, i and j . Suggest to re-word this as "We choose to use a separate pair of indices r and k for summations, to make clear..." -- and change "restricted" to "applied".

28) Section 4, page 9, lines 254-258 -- insert commas between "such problems" and "but require" and between "of the paper" and "we present" -- also replace "equations" with "equation" when referring to (24) (there's only 1).

29) Section 5, page 12, title of section 5.2 -- replace "with an Average Mass Fraction" with "with a common (average) organic mass fraction". Also -- it occurred to me here that I don't think you state that the common organic mass fraction to be assumed is actually the average value -- implicitly that's the case but I suggest you state that up front -- and in the earlier instances where you mention this is one of the key assumptions -- having the word "average" makes it seem a bit more of a reasonable assumption to take I think.

30) Section 5, page 12, line 284 -- insert "organic" between "common" and "mole fraction" -- perhaps also add "average" as well for the same reason as above?

31) Section 5, page 12, line 309 -- suggest to replace "solved for" with "solved by" and also replace "root find algorithm" with "root-finder algorithm" or "root-finding algorithm".

32) Section 5, page 13, line 314 -- insert comma between "been removed" and "it is sufficiently".

33) Section 5, page 13, line 320 -- insert comma between "parameter space" and "values of the".

34) Section 5, page 13, caption to Figure 4 -- "are distinguished by the shape of the points" -- say exactly what you mean here by adding (circles are xx , squares are yy).

35) Section 5, page 13, caption to Figure 4 -- the sentence saying "Equality of the solutions..." can be easily shortened substantially, Just say that the dashed lines show the 2:1 lines. Also it is very difficult to distinguish between the dashed blue and the dashed green (to my eye at least) -- suggest to make the 1:1 line a solid line and the 2:1 and 1:2 lines dashed -- I think here the colours are not needed (in fact they detract) and it's best just to use the black lines.

36) Section 5, page 13 -- Figure 4 -- there seems to be an error in Figure 4 -- the dashed lines should be indicating the 2:1 line -- which they are at the top --- but at the bottom they are much narrower than that -- presumably this is just a typo in the script used to generate the graph right? Please correct this. And add the legend to the plot to make the Figure easier for the reader to understand. This is also the case for Figure 6 on page 17.

37) Section 5, page 13 -- caption to Figure 4 -- please state in the caption what the initial concentration of the sVOC is in these

integrations. Also add this to the caption to Figure 5.

38) Section 5, page 14 -- line 328 -- insert "organic" between "common" and "mole fraction".

39) Section 5, page 14 -- line 343 -- replace "there increased water contend" with "the increased water content".

40) Section 7, page 19 -- lines 417-418 -- the sentence beginning "We derive a more accurate initial condition..." This seems like quite a strong limitation for a method intended to be generally applicable for global models. Please give a caveat here about the fact that in a global model, this restriction would not be possible.

41) Section 7, page 19 -- caption to Table 4 -- state what the initial sVOC concentration is here (summed over the vapour and particle phases).

42) section 7, page 20 -- the equation on line 461 has no number -- please make sure it has a number (even though it may not be referenced in the paper, that's still important). Also please change the lower-case "d" to upper-case "D" -- I asked this already in my initial submission review to make this change. Using lower case here is doubly confusing because the lower-case d is used in the derivative term. I suggest also to use upper case N rather than lower case n as the usual way of presenting a size distribution is "dN/dln D" (capital N and caption D). Please make this change throughout.

43) section 7, page 21, lines 474-475 -- sentence beginning "To speed up" --- this again seems rather a severe restriction to make on the method -- see also point 40 -- there needs to be some mention of how this restriction may limit the method's applicability in a global model. How dependent is the method on this and the restriction in point 40?

44) section 7, page 22, lines 497-500 -- "over predict" to "over-predict" and "under estimate" to "underestimate".

45) section 7, page 22, line 500 -- the errors mentioned here in the text sound potentially rather large. Are the authors saying the "cost" of these magnitude errors are a good "investment" since they are small compared to the "benefit" of being able to represent the partitioning of the semi-volatile organics in a global model? Please add a sentence clarifying this.

46) section 7.3.2 -- page 24 -- this section needs to be improved considerably -- the text on line 570 is unclear -- "two methods of approximating this size" -- what size? You mean the overall size distribution? Also the end of the sentence (line 571) says "that represent two regimes". What are the two regimes here -- need to be much more specific here about what you're referring to.

47) section 7.3.2 -- page 24, lines 574-576. The text "This method.... maintains a constant arithmetic standard deviation and decreases the geometric standard deviation in order to maintain mass within the system". I don't understand this method at all -- particle size distributions tend to follow lognormal size distributions which means that the geometric standard deviation is the appropriate

measure for the width of the modes. In any case the two measures (geometric stdev and arithmetic stdev) are related and if one is kept constant the other should stay constant too. Or am I missing something here? There needs to be better explanation of this method or the reader is left puzzled.

48) Section 7.3.2 -- page 24, line 590 -- What is the change in conditions that is being simulated here? What are the 2 hour and 24 hours indicating a change from -- the experiments being carried out are not sufficiently described here -- this needs to be further clarified before the paper can proceed to publication.

49) Section 7.3.2 -- page 24, lines 592-593 -- "This is in agreement with the initial proposal put forward by Connolly et al. (2014)" -- need to state explicitly what part of the proposal you're referring to here -- what is the point you are making here -- extend the length of the sentence to allow the issue to be properly communicated to the reader.

50) Conclusions, page 25 -- there needs to be included much more prominently in the conclusions mention of the simplifications applied in the method -- in particular the techniques used in the box model re: the initialisation of the simulations. It simply would not be possible to do that in a global model and I am not yet convinced therefore that the applicability of the method is as straightforward as the authors are making out.

51) Conclusions, page 25, line 625 -- add comma after "of the involatile compounds".

52) Page 35, caption to Figure 9 -- change "pcc" to "cm⁻³" (assuming in LaTeX form). Also in other Figures.

53) Page 35, caption to Figure 9 -- need to add somewhere in the caption that the 2nd mode has diameter at 125nm and its number concentration given -- the reader needs that info to be at hand (in the caption) in order to be able to effectively interpret what is shown. Also needs to be stated in Figures 10 and 11 and in Figures 14, 15, 16 and 18.

54) Page 36, caption to Figure 10 -- "models" --> "modes".

55) Page 46, caption to Figure 20 -- state what the initial conditions are here that are being integrated.

References

Myhre, G., and Coauthors, 2013: Anthropogenic and natural radiative forcing. Climate Change 2013: The Physical Science Basis, T. F. Stocker et al., Eds., Cambridge University Press, 659â\200\223740

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Tsigaridis, K. Daskalakis, N., Kanakidou, M. et al., Atmos. Chem. Phys., 14, 10845â\200\22310895, 2014.