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Interactive comment on "Aerosol hygroscopic growth parameterization based on a solute specific coefficient" *by* S. Metzger et al.

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My interest in this paper comes from the fact that the resulting model (EQSAM) may potentially be used in atmospheric models. I think that for this to be de case, we need to know its added value, while I did not find the added value of the proposed parametrisation in the paper submitted here for comments.

I would first like to support the remarks made by the anonymous referee #2 and the comment by my colleague Ralph Lescroart. I personally regard the remark about equations (18) and (19) as one of the most important:

These equations define the functions A and B, which are a key part of the proposed parametrisation. But the derivation of A and B is deferred to another paper that we do not know about. It looks odd that the authors says that complex expressions such as (18) and (19) were derived on an "empirical basis". How did they come to such apparent complexity ? Why is it applicable to a range of conditions with only one parameter per solute, and can we trust that it is more generally applicable than just to two pure salts ?

The mysteries of this parametrisation set aside, we need to look at the benefits that it would need to have in order to be useful: it could be more accurate, more widely applicable, and/or faster to compute than existing parametrisations. However, I found no evidence for this, and would rather believe that the opposite may be true:

Faster computing

The authors propose to use equations 17(a...d) to compute solute molality (and subsequently obtain the wet diameter of droplets). However, only equations (17c) and (17d), which are said to apply to "flat surface" at RH < 95 or 98%, appear simple to solve (assuming that ν_i can be pre-computed, while this may raise problems already mentioned in previous comments). Equations (17a) and (17b) both contains μ_s (solute molality) on both sides, due to A and B, and do not appear easy to solve. Equations (17a) is said to have an analytical solution, but if so, I think that it should be given. For equation (17b), the authors rely on a numerical iterative method. First, I do not immediately see that given a value of RH, (17b) will only have one solution (value of μ_s). Associated with this, I am not convinced that a numerical method will easily find this solution. By contrast, other parametrisations summarised at the beginning of the paper, largely on the basis of Rose et al. (2008), are based on sets of equations simpler than the combination of 17b, 18 and 19. For these existing parametrisations, I can understand how to make the calculations : the technique involves varying a suitable independent variable, such as the mass fraction of solute in the droplet (see appendix of Rose et al., 2008), and the formulas directly provide corresponding relative humidities, growth factors, etc. The search for a given value of RH, for example, may involve iterations but does not seems to involve difficulties. Looking at (17b), (18) and (19), I can only conclude that the solution also needs to be searched with iterations, and suspect that it may be more difficult, thus potentially longer, than with the methods described in Rose et al. (2008). Increasing calculation speed by using alternatives to (17b) may well be possible, but such simplifications would likely also be possible with some other methods (at least, ignoring Ke is always possible). In conclusion, I found no evidence that this method could be faster - in the best case, it could be similar; if calculation speed is an argument, a proof should be provided.

Better accuracy and/or wider applicability

There are other methods that have only one parameter, and the paper under review interestingly presents comparisons to Petters and Kreidenweis (2007, hereafter PK2007). I think that to work well, a parametrisation that has only one parameter needs to be quite well designed, as there is limited potential for fitting to measurements or complex models. In this case, the parametrisation needs to work over the whole range of relative humidity, while, if I understand the paper well, the parameter (ν_i) is determined from a single point in the curve - namely deliquescence. If it works in various conditions, not just for the two investigated salts, and for the entire curve including supersaturation, this would be an achievement. However, the entire expressions A and B ((18) and (19)) have been "empirically determined" to match the reference model, E-AIM. In these circumstances, while it is useful to know that the parametrisation reproduces the results from E-AIM quite closely for the two salts under consideration, this result is much less impressive.

To show the accuracy of their parmetrisation, the authors then compare it to the representation presented in PK2007, referred to as "kappa". I have a serious objection to this comparison : as far as I know, the "kappa" parametrisation was not designed, and its parameter not set, to specifically match the results of E-AIM. The comparison is thus biased, as it is logical that the newly presented parametrisation will match the model from which it was calibrated better than other approaches that were not calibrated to achieve this specific objective.

In addition, I found in PK2007 that this paper not only investigated much wider conditions, involving combination of solutes in a droplet, but that it also considered uncertainties. It appears that the values for the κ coefficient considered in Rose(2008) and the paper under review are not the only ones that were presented in PK2007 : Table 1 from this paper presents values of κ for two types of determination of this parameter, as well as uncertainty ranges, for 25 compounds. In the reviewed paper, it is written (p 24834, line 6-8) that the method from PK2007 “is not optimally valid for concentrated solutions, as confirmed by figures 3 and 4”. These figures indeed show that the “kappa” line do not match the results of E-AIM as well as the proposed (17b). But is this a fundamental problem of the parametrisation or could this be due to uncertainties regarding the κ parameter, in relation either with the calibration method, or the reference measured / modelled values? My partial answer to this question is that the κ values reported in Rose(2008), and subsequently used in the reviewed paper, do not appear to be the choice, within KP2007, that provides the results best matching E-AIM. I checked this by reconstructing some of the results from the reviewed paper based on the alternative value κ= 0.53 (instead of 0.61), based on PK2007 table 1, for (NH4)2SO4. The results are shown in the file attached as supplementary material, overlaid on the reviewed paper figures 3 and 5. This example relates to dry particle size of 0.1 μm. Figure 1 shows that changing the parameter improved the comparability to E-AIM, so that I do not believe that in this case, it may be written that PK 2007 is “not valid for concentrated solutions”. Figure 2 shows that this change does not “deteriorate” the results in the supersaturated region, as one may have suspected.

In conclusion, I strongly have the impression that the proposed new parametrisation is not more accurate than existing ones. In addition, PK2007 is validated for a wide range of solutes as well as combination of solutes, and provided with estimates of its parameter that consider uncertainty (by contrast, the new parametrisation is said to be also applicable to mixed solutions, but I found no validation for this).

My general impression is that we have no indication that the proposed parametrisation is an improvement over the existing ones, and that its validation is much less complete. With the information that we have now, I would conclude that it is not ready to be used,

nor to be published.

References

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Rose et. al, 2008 : Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment. ACP, 8, 1153-1179