Atmos. Chem. Phys. Discuss., 11, C11395–C11400, 2011
www.atmos-chem-phys-discuss.net/11/C11395/2011/

Atmospheric Chemistry and Physics Discussions

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

S. Metzger et al.

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Received and published: 3 November 2011

Reply to Referee #1.

We thank the anonymous Referee #1 for the constructive and helpful review.

We hope to have clarified the issues raised and satisfactorily addressed all comments with the point-to-point reply below. The suggested corrections and additions will be included in the revised manuscript.

The results from the presented method to determine the HGF for inorganic salt particles seems promising since the results for single salt particles compare fairly well with the E-AIM model. However, as the authors mention there are other simplifying methods to determine the HGF for particles. Hence, if this article should be published in ACP the authors need to show clear evidence that this method is superior the other methods in some important way (e.g. computation time or accuracy). For this to be the case there are a few important results and clarifications which I think needs to be included in the article before I can completely evaluated if the article could be published in ACP or not.

Clarification: We provide evidence, e.g. with figures 3 and 4, that the ν_i method has advantages compared to the recently emerging κ method as it is applicable to the whole range of a_w, i.e. from the compound's RHD up to to its critical supersaturation S_c. The κ method, which is to our knowledge the only other analytical method that is comparable to the ν_i method in terms of computation time, instead cannot be used to cover this range with only one coefficient. Note that both methods are implemented in a test program that is available in the supplementary material, so that the results and the computation time needed can be easily verified. Even if other κ parameters can be determined for the salt $(NH_4)_2SO_4$ to yield a better compromise over the entire a_w range, as suggested in the comment of Philippe Marbaix, the result is still less satisfactory than those of the ν_i method, while for NaCl it is not possible to cover the whole range of a_w with a single parameter for the κ method. As far as we know, this is the first time that the gap is closed in modeling the a_w from the compound's RHD until to its critical supersaturation S_c with only one parameter, while ν_i can be easily predetermined from the compound's RHD using the analytical equations presented.

Major comments:

1. *As I understand it, one of the main advantages of this method compared to earlier methods (e.g. the kappa-method) is that it should be more computationally efficient because the water activity correction coefficients are independent of the water activity. Could you please demonstrate this by giving values of the CPU-time for the new parameterizations, the kappa-method and E-AIM.*

   Clarification: As mentioned above the main advantage of this method compared to earlier methods is not only that it is more computationally efficient. This is the first time that the gap is closed in modeling the a_w from the compound's RHD until to its critical supersaturation S_c with only one parameter. As far as we know, all other methods detailed in Sect. 2.1.1 require a_w-dependent parameters (e.g. activity coefficients). This makes these methods not only more comprehensive but also computationally more expensive, or, if they are applied with only one parameter (e.g. kappa-method) less accurate as demonstrated with Fig. 3 and 4. We will provide additional clarification on this point in the revised manuscript.

2. *Page 24827, line 13-15: You write that: "Here, A and B have been empirically determined to best match the reference results of E-AIM – the explicit derivation is beyond the scope of this work and will be presented separately." As referee you want to check the accuracy of the empirically determined A and B coefficients. Hence, you need to describe how these coefficients have been derived e.g. in a supplementary material.*

   The A and B functions have been empirically determined such that (a) the a_w parameterization well fits the E-AIM reference data using the least square method, (b) ν_i can be easily predetermined from a single data point, i.e. the compound's RHD using the analytical equations presented, and (c) so that comprehensive fitting methods, which require the knowledge of the complete Köhler curve, are

   not needed. Since this approach is empirical ν_i has to be validated for each salt. We will clarify this in the revised manuscript.

3. *Page 24828, line 15-19, point 4. You write that when you derive ν_i you assume that K_e=1. I guess that this assumption introduces errors for the smallest particles? You need to clarify this in the text and illustrate this with some results.*

   For the chosen procedure our assumption K_e = 1 does not introduce errors for the smallest particles as shown by Figure 3 and for the reason explained on page 24831, line 11-20. Our results agree well with those of E-AIM also for submicron size particles e.g. with a diameter of 0.05 µm. We will make this more clear in the revision.

4. *You present results for single solute solutions but if the method should be useful for atmospheric aerosols it need to work for mixed solute solutions as well. Hence, I suggest that you include a figure which compares the HGF calculations from E-AIM and the simplified parameterizations for a mixed solution of ammonium sulfate and sodium chloride.*

   We agree, mixed solute solutions are crucial for atmospheric aerosol modeling. Actually this is the subject of the companion paper, i.e. the description of version 4 of the EQuilibrium Simplified Aerosol Model (EQSAM4). There we show a comparison of EQSAM4 with E-AIM (and ISORROPIA2) for various mixed solutions. The companion paper has been published in the meantime for discussion at Geosci. Model Dev. Discuss., 4, 2791-2847; http://www.geosci-model-dev-discuss.net/4/2791/2011/gmdd-4-2791-2011.html.

5. *For most atmospheric conditions the model also need to consider nitrate (e.g. NH_4NO_3 and $NaNO_3$). I want to know why this is not included in the model. You should at least ad a short discussion about this and the limitations with a model which do not consider nitrate.*

Nitrates, and various other compounds, are included in the EQSAM4 companion paper. Since the GMD paper about EQSAM4 is now online, we will add the reference in the revised manuscript so that the readers can verify this more easily.

6. *Line 24-25 page 24817. You should not refer to a publication which is not published. Either you simply refer to the ACPD manuscript from Xu et al. (2009) or you actually submit a revised manuscript which then will be referred to as Xu et al. (2011). As referee I want to have access to this paper. Is this the paper you call the companion paper or is it the Metzger et al., 2010 paper which is the companion paper? I cannot find any reference in the text to Metzger et al., 2010. If this is the companion paper you should refer to this paper in the text.*

   Clarification: We hope to submit a revised manuscript of Xu et al. (2009) once this and the EQSAM4 companion paper are accepted, providing a justification for the model inter-comparison. We will correct the cross-referencing in the revised manuscript.

Minor comments:

1. *Page 24816, line 6-10. Consider reformulating the sentence "The aerosol HG can be determined for certain solutes from laboratory a_w measurements (e.g. Tang and Munkelwitz, 1994), or calculated by considering the vapor pressure reduction that occurs by dissolving a salt solute in water – known as Raoult's law (Raoult, 1888) – if non-idealities of solution are taken into account (e.g. Warneck,1988; Pruppacher and Klett, 2007)."*

   We will change the sentence to: "The aerosol HG can be determined for certain solutes from laboratory a_w measurements (e.g. Tang and Munkelwitz, 1994), or calculated from Raoult's law (Raoult, 1888), if non-idealities of solution are taken into account (e.g. Warneck,1988; Pruppacher and Klett, 2007)."

2. *Line 1, first two words on page 24817. Change from "both methods" to e.g. none of the methods.*

   This will be changed to: "none of the methods accounts for"

3. *Line 11, page 24817. A reference to the companion paper is missing. It is not clear to me which the companion paper is.*

   The companion paper has been published just recently for discussion at GMDD: http://www.geosci-model-dev-discuss.net/4/2791/2011/gmdd-4-2791-2011.html.

4. *In eq. 16b and on line 11 and line 18 on page 24825: Should w_s be X_s?*

   We assume you refer to χ_s, and not to the mole fraction of solute X_s. For the RHD we always refer to saturation and hence to the mass fraction solubility, w_s, i.e. the solute's dry mass required for saturation. χ_s denotes the solute's mass fraction, which equals w_s only at saturation. We will make this more clear in the revision.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 24813, 2011.