

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

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### **Reply to P. Marbaix**

We thank P. Marbaix for his Interactive Comment.

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.

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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.*

We would like to especially point out that for concentrated solutions our approach provides advantages. For relative humidity below 98% Eq. 17(d) can be easily inverted (and analytically solved), so that either molality (or aerosol diameter) can be directly calculated for a given RH, or the water activity (or RH) can be directly calculated for given aerosol diameter (or molality). This covers most applications of aerosol models in GCMs, i.e. applications that do not intend to calculate CCN. Well established approaches like the Osmotic coefficient (OS) model or Van't Hoff factor (VH) model often can not be directly inverted and solved for molality or water activity (a numerical root finding method has to be used), since the osmotic coefficient or the van't Hoff factor is in practical applications not a single parameter but a polynomial. Note also that we wrongly stated (page 24821) that the Van't Hoff factor  $i_s$  is a constant, in fact it is usually represented by a polynomial e.g. a second order polynomial that is a function of molality  $\mu_s$  or of  $\log \mu_s$  with three coefficients (parameters), see e.g. Rose et al. 2008. As a result, our approach has the potential for significant computational speedup. Furthermore our approach is a single parameter description that applies to both single and multi-charged solutions. This parameter stays constant over the entire RH range and we suggest a simple method to estimate it at RHD.

The added value further is the accuracy of our parameterization and its applicability to a wide range of relative humidities for use in regional and global models. We will emphasize these points more strongly in the revised manuscript.

*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*

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equations (18) and (19) as one of the most important:

Please also see our answer to R. Lescroart.

Our A and B terms depend on solute molality for  $RH > 98\%$ . This is the only variable, whereas  $\nu_i$  is the single parameter which is determined at RHD and stays constant over the entire range of RH. So Eqns 17(a,b) can be calculated given a single parameter,  $\nu_i$ . For our two simplified water activity-solute molality parameterizations, i.e, Eqns (17c,d), they do not have this dependency, and nevertheless lead to accurate results, e.g. for a concentrated NaCl solution.

*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 ?*

The expressions are empirically derived based on our experience – summarized as follows:

#### 1. The combined fitting function

In section 2.1.1 we have presented widely used water activity representations, e.g. the Osmotic coefficient model (OS, Eq.4), the Van't Hoff factor model (VH, Eq.5), the Activity coefficient model (AC, Eq.9) and the Effective hygroscopicity parameter model (EH, Eq. 7). A closer inspection of the numerics used by these models shows, that

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they have in common the use of one class of fitting function type that is combined with a parameter, i.e.: the VH, AC and EH model use a rational function approach, whereas OS uses an exponential fit. The parameter itself is usually (with the exception of the EH-model) a multi parameter function that is more complex than the basic fitting function type. The basic idea behind our new approach is to combine two types of fitting functions: a rational function with an exponential function. This has been motivated by several aspects: a more accurate representation since the combined fitting function might better adopt the water activity curve; to cover a wider range of the Köhler curve; a simpler and more robust parameter, i.e ideally a single number which allows the new function to represent the entire Köhler curve without dealing with a parameter function; and possibly a simple method to obtain the single parameter.

We choose to combine the rational function of the VH-model (Eq. 5, p24821) with an exponential function. A straightforward approach to do so, is:

$$a_w = (1 + M_w \cdot i_s \cdot \mu_s)^{-1} = \left(1 + M_w \cdot \nu_i \cdot \mu_s^o \cdot \exp\left(\nu_i \frac{\mu_s}{\mu_s^o}\right)\right)^{-1}$$

where we have introduced  $\nu_i$  as the new single parameter and the constant  $\mu_s^o = 1$  [mol/kg] to correct for the units, (p.24824, 15). Numerically this is simply a combination of two types of functional fits, physically this means that the molality  $\mu_s$  is now replaced by two factors:  $\mu_s^o \cdot \exp\left(\nu_i \frac{\mu_s}{\mu_s^o}\right)$ , a constant and an exponential term. But is this the best approach to use? Any water activity representation has to fulfill this criteria for the limit of a dilute solution:

$$\lim_{\mu_s \rightarrow 0} a_w = 1$$

which is clearly not the case for this approach.

$$\lim_{\mu_s \rightarrow 0} \left(1 + M_w \cdot \nu_i \cdot \mu_s^o \cdot \exp\left(\nu_i \frac{\mu_s}{\mu_s^o}\right)\right)^{-1} = (1 + M_w \cdot \nu_i \cdot \mu_s^o)^{-1}$$

A slight modification in the exponential term, the change towards a logarithmic dependence, will give the right result for the dilute solution limit.

$$a_w = \left(1 + M_w \cdot \nu_i \cdot \mu_s^o \cdot \exp\left(\nu_i \ln\left(\frac{\mu_s}{\mu_s^o}\right)\right)\right)^{-1}$$

$$= \left(1 + M_w \cdot \nu_i \cdot \mu_s^o \cdot \left(\frac{\mu_s}{\mu_s^o}\right)^{\nu_i}\right)^{-1}$$

This approach is exactly Eq. (17c) or Eq. (15) with  $A=1$  and  $B=0$ , and it provides good results for the relative humidity range:  $RHD < RH < 95\%$ . Furthermore this approach, which can be regarded as a modified Van't Hoff factor model, is the type of fitting function we have introduced in this paper. The Eq. 17(a,b,d) are just extensions of this fundamental equation, which provides the basic relation between  $a_w(RH)$  and  $\mu_s$  in our framework. Additionally this also sheds some light on the physical interpretation of the new parameter  $\nu_i$ . Similar to the Van't Hoff factor  $i_s$ , it can be interpreted as a measure of solution non-idealities. Setting Eq.5, p24821, equal to Eq. 17c reveals this aspect. Note that the Van't Hoff-factor is a function of molality (e.g. a second order polynomial with three parameters, see Rose et. al., 2008 for an overview) whereas our approach uses a single parameter.

## 2. The A and B-terms

As we have shown in Fig. 2, p24851, the new approach fails to represent the water activity beyond 95%. Thus this first straightforward approach has to be modified. The modifications have been chosen so that the fundamental structure of our fitting function is preserved: The numerator should be unchanged. The denominator should preserve its form, a sum with two addends. For simplicity the exponent of the second

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addent shouldn't be modified. The modifications should be only functions of molality, but are allowed to include the parameter  $\nu_i$ . The modifications shouldn't be allowed to dominate the basal mathematical characteristics of the original functional fit, i.e. the deviation from using  $A=1$  and  $B=0$  should be small. Eq. 15, p. 24824, represents our choice, where the additional correction terms  $A$  and  $B$  are introduced:

$$a_w = \left( A + \mu_s^o \cdot M_w \cdot \nu_i \cdot \left[ \frac{1}{\mu_s^o} \cdot \mu_s + B \right]^{\nu_i} \right)^{-1}$$

with

$$A = A_{\nu_i}(\mu_s); \lim_{\mu_s \rightarrow 0} A = 1)$$

$$B = B_{\nu_i}(\mu_s); \lim_{\mu_s \rightarrow 0} B = 0)$$

where the dilution limit  $a_w = 1$  puts another constraint on the  $A$ ,  $B$ -terms.

The functions we found for the  $A, B$ -terms are presented as Eq.18,19,p.24827. These functions might look complicated, but in fact they are just products of very basic functions. The  $A$ -term, Eq.18, consists of two factors. The first one is simply a linear function in  $\mu_s$ ,  $1 + M_w \cdot \nu_i \cdot \mu_s$ , which equals 1 for  $\mu_s = 0$ . The second factor is of the well known Gaussian or bell curve type, which also equals 1 for a dilute solution. Since the bell curve is the dominant factor, the  $A$ -term could be interpreted as a bell curve with a slight linear correction. The first factor of the  $B$ -term, Eq.19, is a basic rational function of the type  $\frac{x}{1+x}$ , which equals 0 for  $\mu_s = 0$ . During our numerical experiments we realized that this rational function converges too rapidly to 0 for small  $\mu_s$ . This has been corrected by the multiplication with a moderate root function. The pole at  $\mu_s = 0$  of the root function is compensated by the fast conversion of the rational function term and the combined result gives 0 for  $\mu_s = 0$ . In practical

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applications the B-term has to be treated with some caution, i.e. very small molalities close to 0 have to be excluded. Additionally, numerical experiments revealed that in the range of  $RHD < RH < 98\%$  a B-term independent of molality can be obtained,  $B = B_{98}$ ;  $A = 1$  Eq. 20 p.24827, which provides good results. We included this as Eq. 17(d), which covers most applications of aerosol models in GCMs, i.e. applications that do not intend to calculate CCN. The figures 1,2 showing the A,B-terms for the range  $0 < \mu_s < \mu_s^{sat}$  illustrate the above arguments.

We will include this explanation in the revised manuscript.

*Why is it applicable to a range of conditions with only one parameter per solute,*

Because of the special form  $x a^x$ , as stated on page 24824, line 17-22. It results from the combined approach with two types of fitting functions.

*and can we trust that it is more generally applicable than just to two pure salts ?*

Yes. Eq. (17b) yields results similar to those of Eq. (17c) for a wide range of water activity, i.e. from the compound's RHD until a fractional  $RH=0.95$ , for many compounds that are important for atmospheric aerosol modeling and that are applied in the EQSAM4 concept. These additional compounds are the focus of the companion paper; published for discussion at GMDD: <http://www.geosci-model-dev-discuss.net/4/2791/2011/gmdd-4-2791-2011.html>. The validation of Eq. (17b) for additional compounds at supersaturation (Köhler theory) is, however, the subject of ongoing work; the required  $S_c$  measurements are currently lacking, or unavailable to us.

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*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:*

Complementary to the benefits listed on the first page of this reply and on the pages C13563, C13564 of the reply to R. Lescroart, we would like to mention that especially the potential to directly invert Eq. 17c,d (these Eq. are valid for  $RH < 98\%$ , which covers most aerosol applications in GCMs) allows one to construct an efficient algorithm for mixed solutions. We tried to do so within the EQSAM4 concept, which is the focus of the companion paper. Furthermore as discussed above the direct solution (and inversion) of Eq. 17(d) for  $RH < 98\%$  has the potential to significantly decrease the computation time of aerosol model applications with GCMs. Note that we call the calculation of  $\mu_s$  for a given RH inversion. This is motivated by the fact that with our approach, but also with the VH, AC, OS and the EH model, the RH can be directly calculated for a given  $\mu_s$ . Furthermore, the Köhler-curve describes a function, if  $\mu_s$  is chosen as the independent variable and RH as the dependent variable. Whereas for the inversion, the calculation of  $\mu_s$  for a given RH often requires a numerical root finding. In the case of supersaturation the Köhler-curve is not a function, if RH describes the abscissa and  $\mu_s$  the ordinate.

To our knowledge, the  $\nu_i$  method is so far the only single parameter concept that can be applied in chemistry-transport and -climate models to solve the entire gas-liquid-solid aerosol partitioning, requiring only one measurement per compound.

### *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  $\nu_i$  can be pre-computed, while this may raise problems already mentioned in previous comments).

$\nu_i$  can be pre-computed and applied (e.g. via look-table) without problems as noted before. In our reply to R. Lescroart we provided additional figures that show the range of  $\nu_i$  values for Eq. 17(a-d), pre-computed with the bisection method. The figures demonstrate that a solution can easily be found.

*Equations (17a) and (17b) both contains  $\mu_s$  (solute molality) on both sides, due to A and B, and do not appear easy to solve. Equation (17a) is said to have an analytical solution, but if so, I think that it should be given.*

Thank you for the hint, the sentences at lines 9,10,11 on page 24826 are misleading. What we intended to say is that for given  $\mu_s$  the relative humidity can always be directly (analytically) calculated with our equations. This goes beyond concepts provided in other parameterizations, which require water activity dependent coefficients to calculate the water activity. Or, their functionality is not applicable to the whole range of water activity for various single and multiple charged solute ion-pairs, e.g. the  $\kappa$ -method which has difficulties with concentrated NaCl solutions. The Eqs. (17a,...,d) are the re-arranged Eq.(16a) with special choices for  $K_e$ , A and B, see table on page C13566 in the reply to Lescroart. For a given  $\mu_s$  and after the determination of  $\nu_i$  at the the point of deliquescence, Eq. (16b), RH can always be analytically calculated with the Eqs. (17a,...,d). We are aware that the re-arrangement of Eq. (16a) might possibly be misleading and for the sake of clarity we may omit Eqs.(17a,...,d) and simply present Eq.(16a) with the table for different cases of A,B and  $K_e$  as indicated in the reply to Lescroart (p. C13566). Note the figures remain unchanged, only the labelling has to be adjusted.

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  $\mu_s$ ). Associated with this, I am not convinced that a numerical method will easily find this solution.

- Iteration, convergence and computational effort of the determination of molality in the case of given RH

For the sake of consistency we would like to summarize the relevant arguments here:

a) Determination of  $\nu_i$

The single parameter  $\nu_i$  is determined at the point of deliquescence with Eq.(16b). As shown in the reply to Lescroart (page C13567, and figures 1,2) for all four cases:

1.  $RHD < RH < 95\%$  :  $K_e = 1$ ,  $A=1$ ,  $B=0$
2.  $RHD < RH < 98\%$  :  $K_e = 1$ ,  $A=1$ ,  $B_{98}Eq.(20)$
3.  $RHD < RH < 99.9\%$  :  $K_e = 1$ , A Eq. (18), B Eq. (19)
4.  $RHD < RH < RH_{Scrit}\%$  :  $K_e$  Eq. (1), A Eq. (18), B Eq. (19)

There is always a unique solution  $\nu_i$  for a given pair of RHD and  $\mu_{s,sat}$ . Note that especially for the cases with A Eq. (18) and B Eq. (19) the curves are extremely good natured, which is of no surprise since the A and B-terms are very smooth, as shown in Fig. 1,2 of this reply. So a root finding algorithm always converges. When  $\nu_i$  is determined, it stays constant for  $RH > RHD$ .

b) Inversion of Eq. 16(a)

For cases 1,2 above, 16(a) can be inverted (and analytically solved), so that either molality (or aerosol diameter) can be directly calculated for a given RH, or the water activity (or RH) can be directly calculated for given aerosol diameter (or molality). These cases cover most applications of aerosol models in GCMs. As discussed above in these cases our single parameter representation is more efficient than the OS, VH and AC-models, since no iterative root finding is involved. Here the EH-model is not considered, because it is not meant to be applied for concentrated solutions.

The cases 3,4 involve a numerical root finding method to invert Eq. (16a). There is always a unique  $\mu_s$  for a given RH. This follows from a basic argument: As shown in the figures, the  $\nu_i$  parametrization is able to represent the Köhler-curve. So the question of inverting Eq.(16a) is similar to the problem of solving the Köhler-curve for either solute molality or water activity as discussed above, which obviously is possible. Note that for  $RH > 100\%$  the inversion of the Köhler-curve has two solutions and one has to state whether the aerosol has been activated or not. But this holds for all activity representations. For the cases 3,4 with iterative root-finding our parametrization is of comparable efficiency as the OS, VH and AC-models and slower than the Kappa-method. Note that because of the smooth functional characteristics of the A,B-term, a root-finding algorithm converges quite effectively. For practical applications, which have to be computationally fast, one could also either combine case 1,2 with the Kappa-method, or determine  $\nu_i$  for different RH regimes, e.g. for concentrated solutions ( $RH < 98$ ) and dilute solutions ( $RH > 98$ ), by using Eq. (17d) with the  $K_e$ -term.

*Associated with this, I am not convinced that a numerical method will easily find this solution.*

The bisection method, which is not the most sophisticated numerical method, easily finds the solution within a few iterations (approx. 8). So this is not of concern.

*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.*

We don't agree. The parametrizations (with the exception of the EH-model) involve multi-parameter functions often combined with lookup tables and may lead to non-linear equations and more complex internal logic, which may be numerically more difficult (e.g. multiple roots). They are also more restricted in their application. Furthermore only the parameters RHD,  $\mu_{s,sat}$  have to be known to apply our algorithm.

*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 seem to involve difficulties.*

As outlined above the search for a given value of RH doesn't involve difficulties. Furthermore it is often not easy to obtain the appropriate parameters for other representations, e.g.  $\kappa$  or activity coefficients. For instance,  $\kappa$  values are known only for a few compounds and particle sizes, while activity coefficients are rather difficult to calculate and are also not without problems and uncertainty regarding the numerics. Especially around the compound's relative humidity of deliquescence (RHD), the iterative methods usually applied in thermodynamic equilibrium models need many iterations to converge, and sometimes a solution is not found. The  $\nu_i$  method, instead, can be applied to calculate the water activity or solute molality of single or mixed

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solutions without iterations around the mutual RHD regime as demonstrated in the EQSAM4 companion paper.

*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).*

This conclusion does not hold for the reasons outlined above.

*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  $K_e$  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.*

The question is actually not relevant, i.e. whether the solution of Eq. (17a), (17b), (17c) or (17d) is faster than e.g. the  $\kappa$ -method, which is also fast. The difference between methods is important when applying the method to mixed solutions, e.g. to solve the gas-liquid-solid partitioning of single or multi-component size segregated aerosols. In these cases, the  $\nu_i$  method will have its largest benefit, since its application requires much fewer parameters or coefficients as the other methods listed in Sect. 2.1.1. The  $\nu_i$  method is the only method that covers the whole range of water activity with only one coefficient for various single or multiple charged ion-pairs, including sodium and sulfate salts, while it can be applied to parameterize the water activity or solute molality of single or mixed solutions, as demonstrated in the EQSAM4 companion paper.

*Better accuracy and/or wider applicability*

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*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 ( $\nu_i$ ) 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.*

Indeed, this is the case for the  $\nu_i$  method, as demonstrated by Fig. 1-6.

*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.*

Eq. (17c) does not depend on the A and B terms and still can reproduce the E-AIM results for a wide range of RH, i.e. from the RHD until 95%, depending only on  $\nu_i$ . The E-AIM results are used for testing, and Eq. (17c) agrees well with Eq. (17a,b,d). Only the expressions A and B have been "empirically determined" (see comment included above) such that the E-AIM results can be reproduced for the remaining RH range, i.e. from 95% until supersaturation. However, we do not claim that our choice of the A and B terms can not be improved, but we are quite confident from our modeling application with EQSAM4 (RH up to 98%) that the A and B terms can be used for NaCl and  $(\text{NH}_4)_2\text{SO}_4$  and many other compounds. But to prove this for supersaturation further

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measurements are required for validation, which is therefore the subject of ongoing work.

*To show the accuracy of their parametrisation, 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.*

We have chosen the same "kappa" ( $\kappa$ ) values only for consistency with Rose et al. (2008). The comparison is not biased in the sense that it is showing results from AIM,  $\kappa$  (as presented in the literature), and our method.

*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  $\kappa$  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  $\kappa$  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  $\kappa$  parameter, in relation either with the calibration method, or the reference measured / modelled values? My partial answer to this question is that the  $\kappa$  values reported in Rose(2008), and subsequently used in the*

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*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  $\kappa = 0.53$  (instead of 0.61), based on PK2007 table 1, for  $(\text{NH}_4)_2\text{SO}_4$ . 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  $0.1 \mu\text{m}$ . 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.*

There are two main limitations of the  $\kappa$  method:

(1)  $\kappa$  values depend on the particle size, so its is not surprising that a  $\kappa = 0.53$  (instead of 0.61) fits better for a dry particle size of  $0.1 \mu\text{m}$ , which is not the case for other particles sizes, which were of interest to Rose et al. (2008) – which we also haven chosen as a reference to compare the  $\nu_i$  method, since Rose et al. (2008) give to our knowledge the most complete review on this subject.

(2) The  $\kappa$  method is not accurate for concentrated sodium solutions, as shown in the Addendum of S. Kreidenweis (Referee).

Instead, the  $\nu_i$  method does not have these limitations:  $\nu_i$  does not depend on particle size, if  $\text{RHD} = \text{RHD}_o \cdot K_{e_s}$ , and  $\nu_i$  applies also to sodium salts (and not only to NaCl). Note that our results of Eq.17b depend on particle size, only because of the Kelvin-term. The same applies to the results of the  $\kappa$  method and E-AIM. These limitations of the  $\kappa$  method will be more clearly discussed in the revised paper.

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*In conclusion, I strongly have the impression that the proposed new parametrisation is not more accurate than existing ones.*

We do not agree for the reasons given above.

*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).*

The  $\nu_i$  method is applied to mixed solutions in the EQSAM4 companion paper, which has been published for discussion at GMDD: <http://www.geosci-model-dev-discuss.net/4/2791/2011/gmdd-4-2791-2011.html>.

*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.*

We do not share this impression for the reasons given above.

### References

*Petters, M. D. and Kreidenweis, S. M. (2007) : A single parameter representation of hygroscopic growth and cloud condensation nucleus activity : ACP, 7, 1961-1971*

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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*

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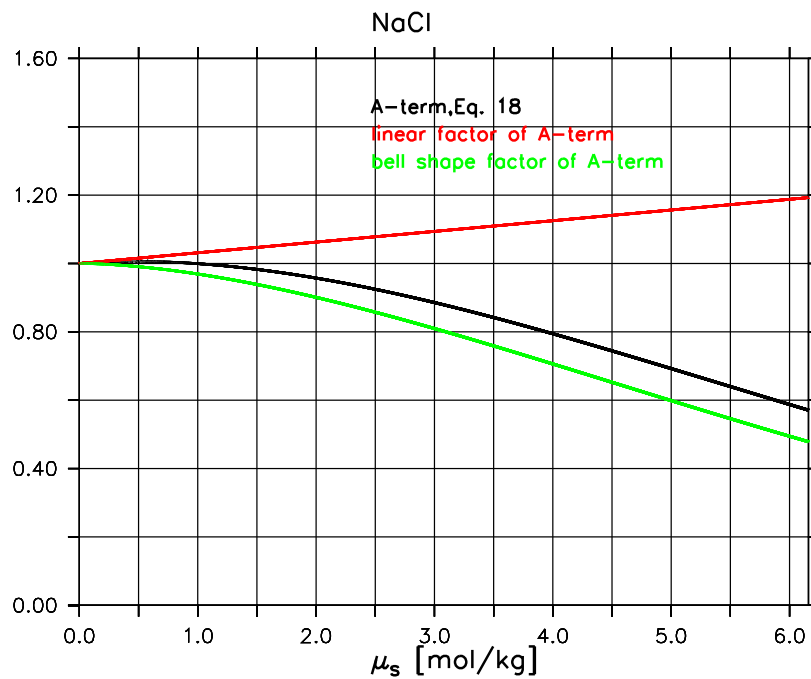
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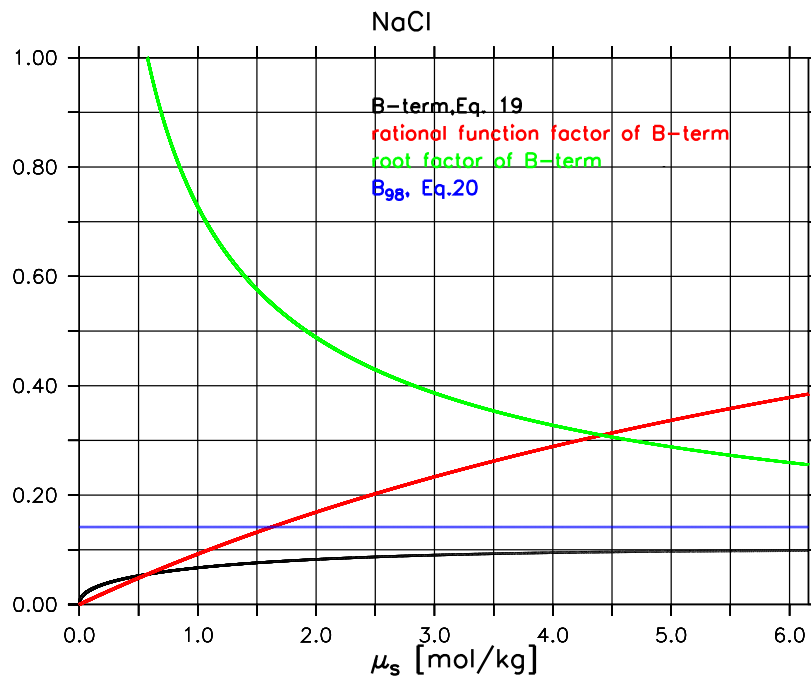
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**Fig. 1.** A-term and its linear and bell (or Gaussian) shape term for NaCl.

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**Fig. 2.** B-term and its rational function and root term for NaCl. The B<sub>98</sub> term is also plotted for comparison.

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