

## ***Interactive comment on “Reformulating atmospheric aerosol thermodynamics and hygroscopic growth into haze and clouds” by S. Metzger and J. Lelieveld***

**S. Metzger and J. Lelieveld**

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We thank Dr. Wexler for his comments, and will try to remove or reduce his concerns with this reply.

### General remarks

One objective of this work is to facilitate the modelling of aerosol-cloud coupling, by providing a method that is (a) based on physical and chemical principles and (b) suited for meso-scale and global modeling applications with a focus on aerosol effects on weather and climate. The simulation results clearly indicate that our method works. Our method does not replace nor competes with comprehensive thermodynamics models, but aims to calculate aerosol hygroscopic growth more explicitly than is customary in

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present models. We hope to stimulate additional measurements to further test our method.

#### Specific remarks

A. Wexler: In eqns K1, the additions should be multiplies.

Reply. p861, Eq. (K1); the additions should not be multiplies, but multiplied by the stoichiometric coefficients which appeared here by accident as exponents.

A. Wexler: Eqn K2 is wrong, but seems to form the basis for all that follows. For instance, for pure water  $a_w=1$  and  $a_s=0$ . How does this product (to any power) give you 1? The eqns 11-14 all are somehow justifying K2, but just rehash it.

Reply. p861, Eq. (K2); the equality to unity should only refer to eqs. (11-14), and is not needed for Eq. (K2). This will be changed. Eqs. (11-14) are appropriate.

A. Wexler: The source of Eqn 19 is not clear, although it is key to the single solute activity model ñ how as this derived? Note that  $\nu_w$  is a function of temperature because the solubility is; this was never discussed by the authors. And what is the value of  $\nu_w,0$  and where does it come from?

Reply. p864: Eqn 19 is newly introduced based on what has been said before (p865). The value and meaning of  $\nu_w,0$  is given at p865 line 10.  $\nu_w$  is a function of temperature because the solubility is, as discussed at page 870-871, 884, and 889.

A. Wexler: In any case, the  $\nu_w$  values appear to be derived from the solubility of the compound? What is the solubility of sulfuric acid? I can't even define it at ambient  $T,p$  but somehow the authors have and find a  $\nu_w$  value of 1.67 whereas the value for ammonium nitrate is 1.84. Since the authors state that higher solubility gives higher  $\nu_w$  values, we have an apparent inconsistency (or I continue to not understand what they're doing).

Reply. p864: Yes, values given in Table 1 have been derived from measured solubility

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values by Eq. (19). This can be easily verified. The values of  $\nu_w$  therefore depend on the (accuracy) of the measured solubility values used (listed in Table 1). These solubility data might be subject to discussion. Any constructive contribution will be highly appreciated.

A. Wexler: The authors finally wind up with eqn 20 which is an approximate equation for molality as a function of RH with a single (temperature dependent) parameter fit. This is sort of like Kusik and Meissner (single parameter fit) but more directly applicable to atmospheric situations in that it is a function of RH. Why do I say it is approximate? Well, it doesn't perfectly match the data (but that could be the fault of the data) and they then derive ZSR (eqn 23) which we know is not exact (osmotic pressures do not necessarily add). Since it was so hard to follow the derivation of eqn 20, I do not understand the approximations that went into it but these should be identified by the authors.

Reply. p865: The fact that Eq. (19) and (20) can be applied to a wide range of inorganic and organic salt compounds makes our concept not only very useful, but also shows that our method extends beyond present concepts. For the first time, equations for molality are not fitted or approximated but explicitly derived from the solute properties, i.e. the solute solubility. Thus, if applied to atmospheric conditions - where the water uptake compensates the vapor pressure reduction associated with e.g. the dissolution of a salt compound -  $\nu_w$  and  $\nu_e$  fully determine the aerosol water uptake. Comparisons with observations should show if these theoretical considerations fail or not. One aspect not addressed by our method relates to the humidity range over which a solid salt compound deliquesces, i.e. changes its physical state from solid to liquid. Although this RH range is rather narrow for most salt compounds, certain atmospheric aerosol mixtures might behave differently, especially if organic compounds are involved. These issues should be discussed and require additional measurement and modelling efforts. Our method could provide a starting point for such applications, because it not only allows an explicit formulation of molality, but also of the deliquescence

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relative humidities. We hope for recognition of this aspect.

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