The authors have developed a new single-parameter equation that is demonstrated to predict particle hygroscopic growth from sub- through super-saturated (CCN activation) ambient conditions. As such, it can be used to close the “hygroscopicity gap” that has been documented in the literature for organic compounds and mixtures, namely, the observation of very low water uptake under subsaturated conditions, implying very low hygroscopicity, but much larger implied hygroscopic growth from measured critical supersaturations required to activate into a droplet. This was achieved by conceptualizing water uptake as a two-step process as relative humidity is increased: initially, the water uptake thermodynamics are described using adsorption theory (using the FHH model); when sufficient water has been incorporated into the droplet to reach the solubility limit of the compound, then the water content of the dissolved phase is treated as obeying Raoult’s Law. The model interpolates to fill in between these extremes. Interestingly, during the phase transition, the remaining solid phase is allowed to contribute additional water according to FHH predictions until full dissolution occurs.

The authors present a nice series of laboratory studies that demonstrate the fitting procedures and the superior performance of the combined model in predicting the full spectrum of water uptake. For their experimental studies, they have cleverly chosen to use three compounds with identical molecular weights, but different structures and hence solubilities. Since standard Koehler theory assumes similar behaviors based on molecular weight, the contrasts between compounds can illustrate the impacts of solubility on water uptake behaviors.

Finally, the combined model is reduced to a useful single-parameter estimate that can be fit using experimental data and that parallels prior simplified treatments for ambient aerosol hygroscopic behavior. This is a very nice feature of this work.

Overall, this paper is an interesting read and presents stimulating ideas for furthering...
discussion of mechanisms of water uptake in atmospheric aerosols, and how to adequately parameterize these for inclusion in models on a variety of scales.

I offer the following comments for the authors’ consideration prior to finalizing this manuscript.

- A nice review of relevant literature is provided in the manuscript, including Riipinen et al. (2015). That reference seems especially relevant in setting up the theoretical equilibria (Figure 1 in Riipinen et al.) that also apply to the cases considered in the manuscript. I am not entirely clear how a corresponding model for the mechanisms proposed here would modify this figure – can the authors please comment? Are there more than two phases conceptualized in the aerosol – undissolved organic, one associated with the surface and one that is an aqueous solution of organic(s)?
- Following on point 1 above, the theoretical basis for equation 6 (including equation 6b) was not clear to me. Is the assumption that the total water content is the sum of each independent water content (one from dissolution, one from adsorption)? The authors propose that \( \text{aw}_{\text{HAM}} = \text{aw}_{\text{KT}} \times \text{aw}_{\text{FHH}} \). How was this derived?
- As discussed in the manuscript, Shulman et al. (1996) early on suggested that partial dissolution of sparingly soluble compounds could generate more complex shapes for the curves expressing the equilibrium between ambient relative humidity and the particle water content. It is good that this possibility was explored here, although somewhat surprising that agreement was not better. Considering that the experiments showed the measured CCN activity to indicate higher hygroscopicity than expected (Figure S4), is it possible that small levels of impurities could affect the results? The sensitivity has been discussed previously, e.g., by Bilde and Svenningsson (2004), among others. Note that Hori et al. (2003) also measured CCN activity of phthalic acid and found it to exhibit higher hygroscopicity than expected from its limited solubility. They tentatively attributed this to incomplete drying of the particles.
- Surface tension plays a role in CCN activation; less so in modulating growth factors. Although Shulman et al. (1996) found that phthalic acid did not affect the water surface tension, it seems worthwhile to at least mention the possibility of surface tension affecting CCN activity and discuss whether it applies in the systems studied, as it has been raised as one of the potential drivers of the “hygroscopicity gap” (Wex et al., 2009). In agreement with the behavior reported in this manuscript, those authors also showed low hygroscopic growth factors up to 95% relative humidity (RH), but were able to map out rapid changes in water uptake (corresponding to enhanced hygroscopicity) as RH was increased above 98%.
- In a companion paper to Wex et al. (Petters et al., 2009), those authors described two alternative explanations for the hygroscopicity gap. These included (1) gradual dissolution of multicomponent particles and (2) nonideality of aqueous organic solutions (Amundson et al., 2007). For the discussion of (2), equation (12) in Petters et al. (2009) is analogous to equation (6) in the manuscript (the HAM model equation), where in HAM the activity coefficient has been replaced by the water activity expression according to the FHH model (equation 6b). Similar to the FHH model, the activity coefficient is based on an exponential relationship. Therefore, is it possible that HAM can fit the experimental data because, as found for models for activity coefficient, the functional form selected can effectively estimate the variation of the activity coefficient over the large range of solution compositions? Can the authors comment on any parallels in the two approaches, and whether one may have a sounder first-principles basis (e.g., surface coverage data can better constrain HAM)?
- Line 295: These statements are not completely accurate. Because the kappa parameter
obeys volume mixing rules, that treatment can be applied to mixtures of soluble and insoluble (but wettable) compounds. The latter are assigned \( \kappa = 0 \).

- The authors did a nice job reporting on calibrations and shape factor considerations in the Supplement, thank you for the documentation.


Bilde, Merete & Birgitta Svenningsson (2004) CCN activation of slightly soluble organics: the importance of small amounts of inorganic salt and particle phase, Tellus B: Chemical and Physical Meteorology, 56:2, 128-134, DOI: 10.3402/tellusb.v56i2.16406


