

Interactive comment on “Water activity and activation diameters from hygroscopicity data – Part I: Theory and application to inorganic salts” by S. M. Kreidenweis et al.

S. M. Kreidenweis et al.

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General comments:

I strongly support the idea that HTDMA data can be used to predict critical supersaturations and thus also CCN concentrations, for both laboratory and ambient aerosols. This paper would help to further promote this way of thinking. I would therefore like to see this paper published in ACP. Having said that, it deserves to be noted that this concept is not new. It has been used in several previous studies. Recently, it has been extensively elaborated upon along the same principles as outlined in the submitted paper in Rissler et al. (2004) for ambient aerosols,

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and in Svenningsson et al. (2005) for laboratory aerosols. Nearly all equations have been derived and thoroughly discussed elsewhere, also of course in some of the references that are already cited.

We appreciate the comments by the reviewer. We do not wish to imply the concept is new. In fact, we cite Brechtel and Kreidenweis (2000a,b) which to our knowledge is the first published attempt to formalize this relationship. This work builds on those ideas. We were unaware of the interesting work by Rissler et al. (2004) and thank the reviewer for pointing this out; we will reference it in our revision. However, we note that the method for predicting CCN properties from the HTDMA is notably different between that work and this paper. We are unable to find the Svenningsson et al. (2005) reference on the ACPD website at this point. We are interested in reading it and can include this reference if the reviewer can provide the correct information.

Equation (17) appears to be very useful in order to provide simple parameterizations of the hygroscopic behaviour of both single compound aerosol particles and ambient aerosols. Since it can be derived from the Köhler equation (as shown in this paper) it is more appealing than some other parameterizations previously used, such as the “gamma” function, where $GF(RH)=(1-RH/100)$ raised to “gamma”, or similar. “Gamma” is then fitted to the HTDMA data. Equation (17) has been used previously, as mentioned, but deserves more attention.

We agree with this remark and appreciate the positive feedback.

The parameterization of critical supersaturation (section 5) is not new. Equation (30) is identical to equation 15.34 in the text book of Seinfeld and Pandis (1998).

We agree; these equations are all rearrangements of standard Kohler theory, also found in the text by Pruppacher and Klett. In Equation (30), we treat 61537; as a best-fit parameter from our calculated CCN values, although it could be computed from first principles. The intent is to show how a parameterization could be built for unknown compounds and HTDMA data, following the ideas in this paper.

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As noted in Rissler et al. (2004), this relationship can be used to estimate critical supersaturation from HTDMA data. Anyway, I believe it can be a very useful way to link HTDMA data to critical supersaturation, and also deserves more attention.

We include reference to this work in the revised paper.

Specific comments

Abstract: The H-TDMA is most often referred to as “Hygroscopic” TDMA, not “Humidified”.

Our group has used this terminology consistently over the years. The terms are similar enough there should be no confusion.

I would prefer to leave out the reference to Part II.

We omit this in the revised version.

Introduction: A 2% uncertainty in RH is not very good, at least not for laboratory conditions. Dew point hygrometers are more precise. The problem is then knowing the temperature inside the second DMA.

The 2% uncertainty in RH corresponds to the manufacturer-listed accuracy. We periodically calibrate the RH sensors using a dew point hygrometer.

Section 2.1: It is good to be reminded once in a while about the origin of the Köhler equation, and that it is actually the partial molar volume of water that enters the equation. But since the assumptions normally made are quite valid at the point of activation, it is perhaps not necessary to note this.

We note that it is the partial molar volume of water in the Kohler equation because many studies still erroneously use solution density instead of the density of water in the Kohler equation. However, we agree that at the point of activation, for most previously-studied substances, this difference is generally negligible.

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Section 2.2: The use of SI units is recommended (not dyne/cm).

We will change this in the revision if the Editor so suggests.

Section 2.3: The definition of shape factor also includes the Cunningham slip correction factor, so equation (25) does not comply with the normal definition of the dynamic shape factor.

The reviewer is quite correct and we appreciate that s/he has brought this error to our attention. We have corrected the revised text.

In this section, I would also rather see that the authors do not use “true diameter” or “dry diameter”, but use “volume equivalent diameter” and clearly distinguishes this from the measured dry mobility diameter. For the wetted particle, the measured mobility diameter equals the volume equivalent diameter since the shape factor is unity.

These are also good points and we have adopted the suggestions.

Section 3: Reference to Fig. 3 should be Fig. 2.

We have corrected this.

I wonder if it is really necessary to carry out such an elaborate sensitivity study for two of the most common inorganic salts. These are often used to calibrate CCN counters, since it is believed that their activation can be modelled accurately.

The sensitivity studies done were included in Part I for these well-known species to demonstrate how well the proposed method applies to experimental data (i.e., we had to do a case for which we know the “right” answer), and to show how errors in shape factor, assumed dry density and surface tension would propagate through the calculations to the predicted CCN activity. Of course shape factor, density and surface tension are known for ammonium sulfate and sodium chloride, but are not as well quantified

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for other species, particularly the organics which will be presented in Part II. We also believe that the insensitivity of predicted CCN activity to the assumed dry density is a powerful result that may be surprising to many readers. Finally, it may be of interest for scientists who are not very familiar with this field to compare the various theoretical estimates of critical diameter as shown in Tables 2 and 3. We believe the range in the predictions expresses a measure of how well we can constrain observations.

Section 5: Reference to Equation (28) should be (27).

The reviewer is correct that the reference should be Equation (27). This should have been fixed in the online version, but we will ensure that it is corrected in the final version.

Before equation (31) it is stated that the only thing that is not a constant in the equation is “beta”. The surface tension (σ) [note from authors: σ_w ?] should actually be the surface tension of the solution at the point of activation, which may actually differ significantly from that of pure water.

We have only used the pure-water surface tension to deduce ν for comparison with other estimates of ν . While composition-dependent surface tension could be used for this purpose, the final equations are based on simplified Kohler theory which assumes that surface tension of the drop is equal to the surface tension of pure water. In general, the dependencies of solution surface tension on composition at the critical point can be captured in the beta parameter when it is fit using equation (29), so the assumption of pure-water surface tension is not required.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 287, 2005.

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