

## ***Interactive comment on “Prompt deliquescence and efflorescence of aerosol nanoparticles” by G. Biskos et al.***

**G. Biskos et al.**

Received and published: 29 September 2006

We thank Kaarle Hämeri and Ari Laaksonen for their useful comments on our manuscript. Below are detailed responses to their comments.

**1) Assuming that the explanation given by the authors is a correct one, one would expect qualitatively similar behaviour for all hygroscopic materials. However, the picture is rather different for sodium chloride nanoparticles (Hämeri et al., 2001; Biskos et al., 2006a,b). For NaCl nanoparticles the deliquescence behaviour is much more prompt with the experimental setup giving non-prompt results for ammonium sulphate, even if some intermediate points were obtained.**

It is correct that all hygroscopic materials should behave similarly for the two modes of operation of the H TDMA systems. To test our explanation of nonprompt deliquescence

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we performed additional measurement with 10-nm NaCl nanoparticles using the two experimental protocols (i.e.,  $RH_s = RH_a$ , and  $RH_s = RH_a + 3\%$ ) (see Figure S2). The new results with NaCl nanoparticles are very similar to those with ammonium sulfate, i.e., an apparent nonprompt deliquescence is observed when  $RH_s = RH_a + 3\%$ , whereas a prompt phase transition is shown for  $RH_s = RH_a$ . The new results are included as supplementary material to the manuscript and reference is made in the main text.

**2) The gradual deliquescence was previously obtained only for particles with diameter 30 nm or smaller. Qualitative change in behaviour takes place roughly between 30 and 50 nm. This may owe to different trajectories and consequently different residence times of particles in various regions inside the DMA or different type of DMA.**

Indeed, the different behavior on the promptness of the particles with diameter smaller than 30 nm can be explained by the different mixing conditions within the second DMA. This is mentioned in the original manuscript, and also it is reflected by the additional numerical calculations provided as a response to the 5th comment of reviewer 2.

**3) The effect, that aerosol flow RH is smaller than the sheath flow RH should influence, not only the deliquescence behaviour, but also adsorption of water onto the particle surface at RH below deliquescence value. The effect should be seen as somewhat smaller growth due to adsorption. However, careful look at Figure 4 at RH between about 40% and 80% shows systematically opposite behaviour. This can be seen both for the datasets by Hämeri et al (2000) and the data by the authors. This is very confusing. Such systematic difference is not likely to be just coincidental and is larger than the uncertainty as reported both in this paper and by Hämeri et al.**

Following our communication with Kaarle Hämeri we realized that the RH value of the excess flow ( $RH_e$ ) was used to plot the data of Hämeri et al. (2000). For a better

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comparison of the data provided by Hämeri et al. with our data when the RHs = RHa + 3% protocol is used (as shown in Figure 4), we have now plotted the growth factor vs. RHe, rather than vs. RHa as was originally plotted. In doing so, all the data points for the experiment where RHs = RHa + 3% are shifted to the right by almost 3%. The growth of the particles due to water adsorption is within experimental agreement when either of the two protocols (i.e., RHs = RHa, or RHs = RHa + 3%) are used in our system. Differences between our data and those reported by Hämeri et al. could be explained by uncertainties in the calibration of the HTDMA and/or the particle generation method used.

**4) We note also, that while operating both aerosol flow and sheath flow RHs at same value may reduce some problems, it also reduces the accuracy of the obtained DRH value. If the RH in aerosol line is lower than the final RH inside the DMA it is not likely that aerosol population has experienced higher RH conditions within the instrument than that is measured for the excess air of DMA and thus the accuracy of DRH is roughly as good as your RH-sensor measuring the excess air. However, if RH set point for aerosol line is the same as that of sheath air the RH experienced within the aerosol line may be higher than that measured in the excess air (within the accuracy of the aerosol line RH-sensor).**

As shown in Figure 1 of the manuscript, the two humidity sensors are located in the monodisperse flow (RHa) and the sheath flow in (RHs) just before they both enter DMA-2. We do not measure the RH of the excess flow because this can be calculated from the RHa and RHs measurements assuming full mixing in DMA-2. When the standard experimental protocol is used (i.e., RHs = RHa), the RH value used in Figures 2-6 are the average values of the aerosol and the sheath flow RH measurements. Because the two sensors are identical (and therefore their uncertainty,  $u$ , is the same), the combined uncertainty (or standard error) from the two measurements is given by  $U = (1/\sqrt{2}) \cdot u$ . The standard error from the two measurements is smaller than the uncertainty of a single sensor. This reflects the fact that the uncertainty of a variable gets smaller when

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we use a larger number of measurements. Therefore, the uncertainty of the measured DRH and ERH values is lower compared to the case where only one sensor was used to measure the RH of the excess flow (i.e., the final RH of the aerosol).

**5) Finally, as a minor point, the authors refer to two of their earlier papers (Biskos et al., 2006a,b) discussing possibility of contamination as an explanation for non-prompt experimental results. The role of possible contamination was actually first discussed in a paper by Russell and Ming (2002).**

Please see response to comment 6 of Reviewer 1.

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 7051, 2006.

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