Atmos. Chem. Phys. Discuss., 6, S2107–S2110, 2006
www.atmos-chem-phys-discuss.net/6/S2107/2006/

Atmospheric
Chemistry
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Discussions

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

Anonymous Referee #1

Received and published: 28 July 2006

The paper by Biskos et al., presents some very careful measurements of the hygroscopic behaviour of ammonium sulphate nanoparticles using a tandem differential mobility analyser. The paper sheds light on a previous finding that the phase transition is 'so called' non-prompt, that is to say there is a finite range of humidities over which the equilibrium water associated with the (NH4)2SO4 increases from its sub-deliquescence value to that of a completely deliquesced one. The paper demonstrates that no such behaviour occurs and that by repeating their methodologies shows that previous findings can be explained by relative humidity gradients in the DMA leading caused by differences in RH A further significant result is to corroborate the lack of size dependence in the relative humidity at the onset of deliquescence as has been reported by Hameri et al. This is important as the models of Ming and Russell and

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Topping et al, do not predict this behavior. The paper is well written, clearly presented and has clear, well annotated figures. It is certainly worth publishing in ACP.

I do have some comments that the authors should address.

The most general of these is the size calibration of the two DMAs and the RH calibrations. These are central to the success of the experiments and as far as I can tell have been performed very thoroughly. The data do appear to be of very high quality and the authors should be congratulated. However, the only place the accuracy with which the DMA sizing is carried out and the RH repeatability is mentioned is in the final summary comments. Though this may have been stated in the authors' earlier paper on NaCl it should be included here. The authors should detail how the DMAs were size calibrated and the second DMA referenced to the first. For what sizes of particle were these characterisations conducted? Does this vary with time? How repeatable were the measurements? The repeatability of the RH is mentioned but also not discussed. How was the primary RH calibrated, with what accuracy and how were all other RH measurements referenced to it? How often was the calibration repeated and how repeatable were the measurements? The day-to-day variability is mentioned but it is not discussed. What effect do the size and RH uncertainties have on your measurements?

In the introduction, the authors state that “once particles are large enough to be optically active they influence the radiative properties of the atmosphere” but then they say “The larger particles also indirectly affect climate through their action on cloud formation” This is a little loose and should be tightened up. The activation of aerosol to form cloud droplets can occur for particles as small as 50 nm diameter, smaller than those that are optically active. In fact such activation can be an important route for growing particles to optically active sizes through gas-particle conversion in cloud which for many species, including sulfate is far more efficient than in the gas phase.

Introduction: pg 7054 line 9, ‘Ě.refers to continuous growth’ the use of the word growth when discussing equilibrium behaviour is a little misleading.

Page 7056: lines 19-20 This means that the transfer function and absolute sizing of the DMAs will change. Does this matter? Furthermore, in earlier lines it is clear that the flow through DMA2 is actively controlled via a feedback loop to maintain the total flow. This is not done in DMA1, does volumetric flow variation in this DMA affect the results.

Pg 7057 line 5: It is worth stressing that these particles are an external mixture of two populations.

Pg 7057-8 A discussion of the numerical modelling would be very useful. It may detract from this paper but I would like to see such calculations somewhere, possibly in a technical note.

Pg 7058 line 10: The authors state that this observation rules out their earlier hypothesis. However, this could be a cause if the solutions used in the previous wok were not as pure as those used in this study.

Pg 7059 line 12-13: Do the relative populations of the two fitted functions at the different RHs tell you something about the probability of crystallisation at different RH assuming that it is not due to the RH gradients mentioned.

Pg 7060 lines 7-9 Are the flow rates available for the DMAs in the Hameri study? This would help to confirm your hypothesis.

Pg 7060 line 24: ‘Ěappears to be a systematic bias of 1-2%..’ in what? , you need to say. Taking as an example, there is a measured GF of 1.2 at 80% for 8 nm particles and a modelled prediction of 1.25 at the same conditions. This would give rise to wet particles of 9.6 and 10 nm diameter respectively. Can the DMAs resolve these difference, they are less than half the FWHM of the transfer function for a 10:1 flow. Are these statistically significant? These discussions need the measurement accuracies reporting as requested above.

Pg The prediction of size dependent deliquescence RH for ultrafine $(NH_4)_2SO_4$ particles by Ming and Russell and Topping et al is not mentioned anywhere in the text. This

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is an important confirmatory result of that presented by Hameri et al. The previous work should be included in the introduction and the authors' contribution highlighted in the main results section as well as the result summarised at the end.

Figure 1: The sheath air is left without a grey coloring. This is a minor point but it is confusing when tracing the diagrams given the comments about coloring at the bottom of the figure caption.

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

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