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Interactive comment on "On the hygroscopic growth of ammoniated sulfate particles of non-stoichiometric composition" *by* H. Kokkola et al.

Anonymous Referee #3

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While the topic of this study, the hygroscopic growth of aerosol particles with a range of ammonium to sulfate ratios, is highly relevant to our field, substantial additional work must be done before I can recommend this manuscript for publication. Reasons for this assessment are described below.

First, the manuscript is overall disorganized to the extent that it is even sometimes unclear whether model results or experimental measurements are being discussed. There are significant typos, including use of the wrong formula for ammonium sulfate and no formula for letovicite, at one point. The most interesting result is that the author's

modeling fits the experimental results better when a model was used which assumed that dry particles have a letovicite core surrounded by an ammonium sulfate shell, instead of the equilibrium model picture. Unfortunately, this result gets somewhat lost in the quagmire of poorly written pages.

On a scientific note regarding agreement between the modeling and theoretical note, from the results in Figure 3 it is clear that the letovicite core model does a bit better, it is not clear whether or not the equilibrium model might also fit the data, considering uncertainties in both the modeling and the experiment. How many experimental runs were done? What is the uncertainty in RH and in GF? Without careful attention paid to collective uncertainties, we cannot be assured that the authors' main conclusion is correct, and therefore whether or not the manuscript contains a result worthy of publication.

Other points- The authors refer to the compositions studied here by the weight percent of the solids originally put into solution. Since these are then solubilized, where dissociation occurs forming ammonium, sulfate, etc., ions in solution, then the solution is aerosoled and then dried out once again. During the hydration, we then have solids and solution all in equilibrium (or in the alternative situation levocovite core suggested by the authors.) The solids present may be ammonium sulfate, letovicite or ammonium bisulfate. So, throughout the experiment, it is not certain that we have the mass ratios of ammonium sulfate to ammonium bisulfate. Is there not a more appropriate way to refer to these solutions, than the wt% annotation used here?

The caption on figure 2 says the schematic is a setup for measuring ORGANIC composition of aerosol particles. What ORGANIC!?

Page 3- "Ĕ the TDMA can be applied to obtain information on atmospheric particles which have mass too small for conventional chemical analysis" What chemical analysis is being referred to? The TDMA is a sizing instrument (which indirectly might tell us about chemical composition), but it is not a chemical composition measurement.

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Page 3- Please define DMPS and SMPS.

Page 4. Studies of mixed salt particles show gradual growth with increasing relative humidityĚ. There are numerous studies on this- the authors should review the literature and include these.

Page 7. Including curvature effects for nano-sized particles is first deemed very important. Then in the same paragraph, we are told that “in practice, the effect is only noticeable for particles of dry diameters of 30 nm or less". It is not precisely clear what the authors mean. Do they mean that theoretically curvature is important over a larger size range than the experiments indicate?

Page 10. “the equilibrium model predicts much higher hygroscopic growth than observed at relative humidities about 60%...” It would really help the authors’ case to demonstrate that the equilibrium model is successful in predicting GF in simpler cases, such as ammonium sulfate alone, and levovicite alone. How do we know the model isn’t just “off” in some other way?

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 1, 2006.