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Interactive comment on "A comparison of water uptake by aerosols using two thermodynamic models" *by* L. Xu et al.

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I am writing this comment at the editor's request since I had also commented on the 2007 ACPD paper of Metzger and Lelieveld (hereafter referred to as ML). I agree with the assessments of Lescroart et al. and Referees 1 and 3, and would like to make a few additional comments.

In the 2007 paper, ML claimed that EQSAM3 could solve the multicomponent aerosol gas-liquid-solid equilibria accurately and analytically (non-iteratively). However, despite several attempts, ML have been unsuccessful in clearly describing their method and proving their rather fantastic claims. If ML have indeed discovered an analytical solution to such a complex non-linear system, then they should very clearly demonstrate and report it with all the simplifying assumptions explicitly detailed. So far it has been very

difficult to follow their methodology, which also contains several conceptual errors.

Secondly, ML have repeatedly claimed that other aerosol thermodynamics models (e.g., AIM, MESA, etc.) calculate activity coefficients in a manner that is only applicable under "laboratory conditions," and do not apply the "necessary transformations" for atmospheric modeling applications. While it is not clear what these transformations are, this claim basically implies that all other aerosol thermodynamics models (including EQUISOLV II whose predictions agree quite well with AIM and MESA) are incorrect. If this is indeed true, then Xu et al. should not have even attempted to compare EQSAM3 with EQUISOLV II or other similar models in the hope of evaluating and verifying EQSAM3's accuracy.

In any case, the comparison between EQUISOLV II and EQSAM3 was done at a rather superficial level in the present paper and the conclusions drawn are misleading. For example, under sulfate-rich conditions it is rather trivial to get a relatively good agreement between different thermodynamics models for NH4 (which is mostly associated with SO4), Cl (which is mostly associated with Na or displaced by SO4), total PM (which is dominated by SO4, NH4, Na, Cl), and water (dictated by total PM). The difficulty lies mainly in sulfate poor conditions when HNO3 and NH3 are present at moderate to appreciable levels. From the comparisons shown in the paper, it is clear that EQSAM3 and EQUISOLV II differ drastically for nitrate (see Figures 1), and EQSAM3 consistently and significantly overpredicts nitrate compared to observations (see Figures 5 and 6). On the other hand, the two models compare quite poorly with each other for pH under sulfate-rich and sulfate neutral conditions.

Second, a clear evaluation of solid salt formation in both models for the 20 test cases is shown only at 30% RH where most salts exist as solid (again quite trivial). The major differences between the two models would be seen at moderate RH where deliquescence occurs and solid/liquid typically co-exist. Evidence of this can be seen from the relative difference in the growth factor shown in Figure 4.

However, despite very poor performances by EQSAM3 for nitrate and pH, Xu et al. recommend its inclusion in global aerosol models. They also express their intention to use EQSAM3 in their hybrid dynamic gas-particle partitioning model, which again is of interest mainly for nitrate (and the associated NH4) and chloride partitioning. The hybrid approach itself has some fundamental problems, but with EQSAM3 at its core, the authors should not expect reliable results for size-distribution of nitrate and possibly other species as well.

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