Review of

"Derivation of the stoichiometric coefficient of water (ν_w) to account for water uptake by atmospheric aerosols"

By Metzger et al.

The Xu et al. (2009) mentioned in this manuscript received strong criticisms from anonymous reviewers as well as several named researchers. Most of those criticisms can be classified into two categories: 1) criticisms on the construction and validity of the EQSAM3 model/concept, and 2) criticisms on the depth of application/evaluation of EQSAM3, specifically the lack of meaningful comparisons between EQSAM3 and EQUISOLV II and the conclusions drawn. The editor therefore suggested that this manuscript be resubmitted as two part papers. The first part would contain clarifications to all the issues pertinent to the construction and validity of EQSAM3, and the second part would contain a more in-depth and meaningful comparison of EQSAM3 with EQUISOLV II as well as AIM, which is considered a benchmark thermodynamic equilibrium model in the aerosol modeling community.

It is my understanding the present manuscript is "part one," which is intended to clarify the EQSAM3 concept, while the "part two" focused on comparing EQSAM3 concept has also been resubmitted to ACPD for review. My comments and recommendation on this manuscript are therefore based on how the authors have responded to all the criticisms that were made on the EQSAM3 concept as part of the Xu et al (2009) review process.

Unfortunately, the authors have only focused on the derivation of the stoichiometric coefficient of water in this manuscript while completely ignoring all other criticisms and concerns that were raised about EQSAM3 in the Xu et al (2009) paper. It was the responsibility of the authors to address all the issues that were raised, point by point. The authors may not choose which issues they would like to address or ignore. I have to therefore recommend rejection of this manuscript.

Here, I highlight just one of the several outstanding questions on the validity of the overall EQSAM3 concept. In the series of EQSAM papers and rebuttals, the authors have repeatedly claimed that their model solves the multicomponent aerosol gas-liquid-solid equilibria accurately and analytically (non-iteratively). However, despite several attempts, they have been unsuccessful in clearly describing their method and proving their rather fantastic claims. If they 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. The material discussed in the present manuscript only touches upon the calculation of water content for known amounts of different salts present in an aerosol. However, no information is given on how the multicomponent gas-solid-liquid equilibria are solved. As a result, the overall EQSAM3 concept still remains as cryptic as before.

Coming back to the "stoichiometric coefficient of water" concept discussed in this manuscript. As such, the authors could try to repackage this concept as a "parameterization" as opposed to a legitimate thermodynamic model. While there is nothing wrong with such a strategy, the question is what is the real benefit of this new parameterization for calculating the equilibrium water content of aerosols when the available ZSR parameterizations (based on polynomial fits or tabulated values of single electrolyte molalities as a function of water activity) in the literature are already quite accurate and efficient? It is the calculation of the multicomponent gas-solid-liquid equilibria that is computationally expensive and fraught with inaccuracies, especially when solving dynamic gas-particle partitioning over size-distributed aerosols.

Other comments
1) What is the difference between figure 2c and 2d? They are both labeled as "NH4NO3," but the results look very different. The scales of Y-axis in 2c and 2d are also very different. Results of ISORROPIA are also very different between the two cases.

2) Both EQSAM3 and ISORROPIA have trouble reproducing water content at moderate RH values for mixed salt solutions. It is known that ISORROPIA does not solve the actual solid-liquid equilibrium problem at low and moderate RH, but instead makes several simplifying approximations, which are clearly acknowledged and explained in their papers. It is not clear why EQSAM3 also fails in this region despite the claim that it solves multicomponent solid-liquid equilibrium analytically (non-iteratively).