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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Derivation of the stoichiometric coefficient of water (ν_w) to account for water uptake by atmospheric aerosols" *by* S. Metzger et al.

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The current manuscript must be rejected in line with the recommendation of all three referees. The physical basis of the derivation of the stoichiometric coefficient in the manuscript did not convince the referees nor myself. By the authors' admission in the responses, they were unable to provide a physical explanation for the concept and it is necessary to recast the stoichiometric coefficient concept as a parameterisation. In this case, it is not necessary to attempt to derive the empirical coefficient from first principles and the paper is redundant. I therefore do not recommend resubmission of a revised version of the paper.

In trying to force a derivation from first principles, the presentation of the concept is too confusing and the terminology used is too unconventional to allow clear interpretation of the relationship between ν_w and other established theoretical approaches. Finally, no comparison of the performance of the relationship with respect to established concepts (e.g. Gibbs-Duhem) has been made in response to the reviews.

It is unclear whether some aspects of the EQSAM concept are not useful. Efficient parameterisations of the gas-aerosol equilibrium problem are required for large-scale use, and a clearly described framework satisfying this criterion should be well-received. For the EQSAM framework to be accepted as a useful parameterisation, I feel that the clarification of both ν_w and the approach to the analytical solution of the multiphase equilibria would necessitate a careful rewrite of ML07, stating exactly which aspects are parameterised and which have a clearly expressible and well-established physical basis. Such an effort could be worthwhile, serving to place future use of EQSAM on a less shaky foundation, allowing insightful critical evaluation of its performance in the light of the physical processes that it represents. Its most recent incarnation clearly can provide results that are comparable with other approaches in some conditions and it is to the detriment of a potential user community that the clarity of its description in the press is unconvincing.

I would like to thank all concerned for restraint and patience in the difficult process of evaluating this manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8165, 2010.