Atmospheric Chemistry and Physics Discussions

Interactive
Comment

Interactive comment on "Reformulating atmospheric aerosol thermodynamics and hygroscopic growth into haze and clouds" *by* S. Metzger and J. Lelieveld

Anonymous Referee #3

In the paper "Reformulating atmospheric aerosol thermodynamics and hygroscopic growth in to haze and clouds" by S. Metzger and J. Lelieveld, the authors propose a new approach to formulate the thermodynamics of atmospheric aerosol allowing an efficient solution of the multi-phase and multi-component equilibrium. In this approach, the stoichiometric coefficients for water and efficient stoichiometric coefficients for solutes are directly derived from the solute solubility; also aerosol molality is explicitly calculated from the solute solubility instead of using polynomial fits to water activity measurements. Realised within the EQSAM3 model, the algorithm was to some extent tested against the thermodynamic equilibrium models ISORROPIA and SCAPE2 and the results were compared with limited observation data from MINOS campaign. EQSAM3

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has been implemented in the GCM ECHAM5 and some preliminary calculation results of cloud cover are presented. This is definitely an interesting work, addressing topical issues. It will certainly be of interest among others for the chemical transport modelling community, as there is a need for computationally efficient parameterisations for gas-aerosol partitioning with improved accuracy. The work certainly falls within the scope of the ACP journal. However, the manuscript is excessively lengthy due to multiple repetitions and plentiful details and is rambling, which make it very heavy to read the paper through and difficult follow its logics. I would strongly recommend a significant shortening through the whole of the manuscript (by e.g. cutting back the repetitious sentences and undue explanations) and making it more focused. It should also be made more transparent which of the approaches/algorithms are really new (as it is hidden within an extensive description of thermodynamic basics). Furthermore, the manuscript should provide a more solid validation of the new calculation algorithms for a range of atmospheric chemical and meteorological regimes (or otherwise to avoid using sentences like e.g. "EQSAM3 accurately calculates aerosol chemical composition and aerosol water"). After these revisions (and corrections of some errors in equations as pointed out in the Short Comments) and attending to the specific points below, the manuscript could be recommended for publication in ACP.

Specific comments:

1). Abstract: p.850 (25-30) are those really main results/findings/conclusions? in particular (c)?

2). Introduction should be tidied up and made more "to the point" (for ex. lines 10-13, p.851 are redundant, lines 7-18, p. 852 can be moved to Sect.2, etc.); p. 853 (8-11): I do not really agree that the new concept will necessarily help to refine lumped categories like mineral dust, biomass burning.. as long as there is now better knowledge about the chemical composition of the emissions. 3).

Sect. 2 and 3 should be considerably shortened, cleaned from excessive details and

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made more transparent. It can be suggested to: - consider to move a larger part of Sect. 2 to Supplement; - use more references rather than trying to explain all thermodynamics theory; - clean up for various distractions, like repetitions, For ex. "At equilibrium the solution is saturated.." p.860(1), p.863 (21), p.864 (15), 868 (2), p.888(14-15); relative humidity definitions p. 854(3) and in Summary p. 882 (21), mss=ns/nw55.51 on p.863 (10), p. 865 (17), p. 868 (17); etc. - and excessive unit explanation (like, in Pascal [N/m2] p.854 (26), p. 855 (26) and further p.863 (14-16); l.20 with units gram per mol [g/mol]; p. 866 (19): with units in gram per cubic centimetre [g/cm3], p. 878 (22): parts per billion by volume, ppb; etc.)

4). Sect. 4.1 p. 870 (13): what does that mean "the - measured - solubility"? p. 871 (9-14) why not move comments to eq. 23 to the eq. 23. p. 871, point 6: why not use (a) for solubility determined and (b) prescribed reaction order.

5). Sect. 4.2 (p. 875) In the performed (box?) model calculations, what determined the day-to-day concentration variability? Is it only due to T and Rh, or advection was also accounted for? p. 875 (12-20) confusing text, recommended to re-formulate lines 24-26 recommended re-formulating ("reference model"? which is definitely not associated with "its computational costs"). p. 875 (23) and 876 (1): edit "relatively largest" ; re-write more clear paragraph starting from "Focusing further on the fine mode" (line 26); p. 876 (12-13) check if the sentence makes sense p. 876, lines 19-21 and 24-25: re-formulation is recommended;

6). Sect. 4.3: p. 877 (6-7): Only under humid conditions? (13-14) : Explain "focus on relative differences (of what?) because they provide indications of sensitivity" What happens with predicted visibility for 7-9 November (also Fig.4)? I do not understand the basis of the conclusion on lines 23-24. p. 878 (5-7) I think the stated here aim appears far more ambitious than the work presented in the section, as no verification has been presented which proves accurate simulation by EQSAM3 of the aerosol chemical composition and water; p.878 (from line 23): I do not understand why distribution/transport of nitrates and ammonium are so different, or it is just because of the scale cut at 1ppb?

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Formation of both ammonium sulphate and ammonium nitrate are dependent on the presence of NH3 emissions; and what about NaNO3 over sea it cannot be zero. p. 880 (14-15): should not it be all way around, that high aerosol load and low T are favourable conditions for haze formation? p. 881 (22-23): what the authors mean by E"QSAM3 compares qualitatively, though remarkably well with E5M1"? p. 882 (1-5); I think these are strange conclusions drawn from the comparison.

6). Sect. 5: I'd strongly recommended revising the section making a concise and clear summary of the work; in the discussion it is suggest avoiding exact repetitions of parts from Sect. 2-3 and excessive use of hardly relevant information to make it more reader-friendly.

7). Conclusions: p. 890 (19-21) compared with rather limited observation data. It should be stressed that the EQSAM3 still needs thorough validation (comparison with reference EQMs and with measurements for different meteorological conditions and chemical regimes.

Technical comments:

p. 852 (14): should be i.e. RH $>=1$

p. 899, 900: "scheme of" instead of "schematic of"

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