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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Kinetic multi-layer model of gas-particle interactions in aerosols and clouds (KM-GAP): linking condensation, evaporation and chemical reactions of organics, oxidants and water" *by* M. Shiraiwa et al.

Anonymous Referee #2

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General Comments:

This manuscript presents a new model for describing gas-particle interactions in aerosols and clouds. The model further extends the Pöschl, Rudich, and Ammann (PRA, 2007) framework to include heat flux, evaporation and condensation of semi-volatile species. In addition to presenting the new model, the manuscript demonstrates how the new model can be used to analyze and interpret experimental results.

The new model represents substantial scientific progress in terms of methods used

to understand and interpret microphysical processes in atmospheric aerosol particles. Hence, the paper is well suited for publication in Atmospheric Chemistry and Physics but should first address adequately the following comments.

Major comments:

1) Abstract, line 10-11. The authors suggest an unlimited number of species, chemical reactions, and physical processes can be treated. As the number of species, reactions, etc. increase the computational cost must increase. The authors should give some indication how computationally expensive it is to carry out these calculations and discuss whether it is really practical to include a large or unlimited number of species.

2) When discussing the water condensation work, the authors state that they have confirmed that the accommodation coefficient of water at 270K is close to unity (see Abstract and Conclusions). This sounds like they have made an original contribution to our understanding of water accommodation. However, Winkler et al. (2006) have already come to this conclusion by comparing their experimental data with theory for droplet growth. The authors could do a better job of giving proper credit to the work of Winkler et al. (2006) while more clearly indicating their own contribution.

3) Page 33706, line 24-26. Here the authors are assuming a desorption lifetime of water based on molecular dynamic simulations that is uncertain. How sensitive are the calculations to this number? Because of the uncertainty associated with the desorption lifetime used, if the calculations are sensitive to this number, then I think it is too strong to conclude that the authors have confirmed that the mass accommodation of water is close to unity.

4) In the case of DOP evaporation, the desorption lifetime of DOP is assumed to be 10ˆ−6 s; however, the basis of this assumption is not stated nor is any citation given. Later the authors indicate that the desorption lifetime showed practically no effect. How much did the authors vary the desorption lifetime in their calculations? This information should be included.

ACPD

11, C15376–C15379, 2012

5) The authors could consider adding a shaded region to the modeling figures to represent some measure of the uncertainty associated with model results.

6) In the modeling of oleic acid ozonolysis, the bulk diffusion coefficient of oleic acid was 1.88x10ˆ-7 cmˆ2/s. In the previous KM-SUB model, the bulk diffusion coefficient of oleic acid used in fitting the experimental data was 10ˆ-10 cmˆ2/s, three orders of magnitude slower than the current model parameter. How do the two models compare when the same model parameters were used?

7) One of the novelties of the new KM-GAP model is that it considers gas-particle partitioning of reaction products. For ozonolysis of oleic acid, evaporation of nonanal appears to decrease the particle size. Is there any experimental evidence in the literature to support or contradict this conclusion? Are the results from Katrib et al. 2005 and Sage et al. 2009 consistent with the calculations?

8) The authors have chosen to model two single-component systems (condensation of water onto Ag nanoparticles, evaporation of DOP) that can be predicted with simple theory (such as droplet growth theory). For these systems the authors could do a better job of highlighting the benefits of their complex model compared with simple theory. Alternatively, the authors may want to model a more complex system (in addition to oleic acid ozonolysis) to illustrate the novelty of the KM-GAP model.

Technical corrections:

9) On page 33693, line 5, the authors state “all steps of mass transport and chemical reaction from the gas phase to the particle core are considered,” which is consistent with the rest of the text and figures. In the following paragraph (lines 11-13), however, when describing Figure 1, layers beyond the near-surface bulk are not included. The strata beyond the near surface bulk layer should be added here.

10) Figure 5 caption reads (a) ozone, (b) oleic acid, (b) nonanal, and (d) non-volatile products. This should read (a) ozone, (b) oleic acid, (c) nonanal, and (d) non-volatile

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11, C15376–C15379, 2012

products.

References:

1. Katrib, Y. et al. Density changes of aerosol particles as a result of chemical reaction. Atmospheric Chemistry and Physics 5, 275-291 (2005). 2. Sage, A. M., Weitkamp, E. A., Robinson, A. L. & Donahue, N. M. Reactivity of oleic acid in organic particles: changes in oxidant uptake and reaction stoichiometry with particle oxidation. Physical Chemistry Chemical Physics 11, 7951-7962 (2009).

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 33689, 2011.

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11, C15376–C15379, 2012