**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 #3

Received and published: 16 February 2012

This paper describes KM-GAP, a multiphase model of gas-particle interactions that treats explicitly mass transport and chemical reaction of semi-volatile substances, and that resolves concentration gradients within the particle. KM-GAP is the latest addition to a series of models with increasing complexity (K2-SURF, K2-SUB, KM-SUB) that are all based on the formalism put forward by Poeschl, Rudich, Ammann. The model development that distinguishes KM-GAP from the previous model versions is that it includes condensation and evaporation (and therefore particle growth and shrinking) and associated heat fluxes. The authors present the model description and apply it to

model experimental data from three case studies.

The paper fits well within the scope of ACP, and addresses an important topic. This model represents a very useful tool to investigate processes such as the aging of organic substances on a very detailed level, with less simplifying assumptions than traditional models typically have and suffer from. I recommend this paper for publication after the following questions and comments have been addressed.

1. Since it is the focus of this paper, the model description needs some improvement in organization and precision. Specifically, from section 2 it is not clear what system of coupled differential equations is actually solved. What are the dynamic variables? Clearly, eqs. 1-5 are part of the system, but there are more, such as eq. 15, and also the equations for particle temperature and environmental temperature, which appear much later. It would help this paper if these were clarified.

2. Then, after the governing equations are defined, there needs to be a section on the numerical implementation. The authors mention much later that the Matlab solver ode23tb is used. Does this apply to all cases or only the one presented in 3.3?

3. I echo the comment by Referee #2 about the phrase in the abstract that "unlimited numbers of species" can be simulated. Clearly this is not the case in practice, and should be rephrased.

4. It looks like the number of layers is predefined in this model. Consequently, when the particle grows, the thickness of the layers becomes larger. Does this introduce problems when the particle grows by large amounts? Could it be necessary to introduce more layers?

5. Section 3.2: What is n for these model runs? Also, I would like to know more about the wall losses mentioned in this section. Are these particle wall losses, gas phase wall losses or both? The expression how these wall losses are incorporated should be added to the equations.

6. Furthermore, the calculations for the DOP experiment are conducted assuming that the particles stay monodisperse during the experiment, this means assuming no coagulation. While for this particular experiment, this assumption may be appropriate due to the low number concentrations and relatively short time scales, it would be good to point out this assumption since it could be a limitation when applying the model to other conditions.

7. Sensitivity studies with respect to number of bulk layers. The authors mention that varying the number of layers between 5 and 200 does not impact the results of oleic acid degradation. I am curious to know how few layers would be sufficient?

Minor points:

p. 33693, l. 18: Strictly speaking, these equations do not describe a mass balance, but a number balance.

Equations 1-5: Define n.

Equation 6: Over what quantity is this summation?

Do we need eq. 14?

Eq. 15: add "m=1"

Something needs to happen with the symbol for the Knudsen number in eq. 19 and 20, the typesetting looks like K is multiplied with n_{Z_i}

Page 33703, l. 9: typo: surface tension

Eq. 58: What is the summation index in this equation?

Figure 1(b), the fluxes below J_{abs} and J_{des}, should these read $J_{s,ss}$ and $J_{ss,s}$?

Table A1: Line with $J_{ads,Zi}$ says flux of adsorption and desorption (need to mention $J_{des,Zi}$ somewhere).

C15402

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