**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Kinetic model framework for aerosol and cloud surface chemistry and gas-particle interactions: Part 1 – general equations, parameters, and terminology" *by* U. Pöschl et al.

U. Pöschl et al.

Received and published: 3 August 2005

We thank Rolf Sander for his comments and suggestions, which are highly appreciated (Sander, 2005).

Response to General Comments

We are grateful for the remarks on the mass transfer coefficient k_{mt}, which confirm and exemplify the compatibility of the proposed kinetic model framework with earlier formalisms for the description of aerosol and cloud particle bulk chemistry. This com-

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

EGU

patibility is an essential aspect of the proposed model framework (Pöschl et al., 2005a: pp. 2124-2125, 2166-2170).

At the same time, however, we would like to emphasize that the diffusion correction factor C_g and equation (20) of Pöschl et al. (2005) are based on the uptake coefficient γ, which describes the net flux of the investigated gas onto or into the particle, whereas k_{mt} and equation (4) of Sander (2005) are based on the traditional (bulk) mass accommodation coefficient α, which describes the gross flux of the investigated gas into the particle bulk.

k_{mt} convolutes not only two but three elementary molecular processes, each of which can determine the overall rate and extent of reactive and non-reactive gas-particle interactions in the wide range of atmospherically relevant aerosol and cloud systems and meteorological conditions (liquid/solid, organic/inorganic, concentrated/dilute, dry/humid, warm/cold): gas phase diffusion, surface accommodation (adsorption), and surface-to-bulk transfer.

Convolution of these processes may be convenient for the description of systems which are most likely not significantly influenced by surface effects (liquid cloud droplets). It is, however, inappropriate for the investigation and description of systems where surface reactions and surface saturation can be important (mixed clouds and aerosols comprising ice crystals and other solid or highly viscous liquid particles). Surface processes and effects are not only relevant for local air pollution and aerosol health effects (e.g. adsorption and chemical modification of hazardous pollutants on urban aerosol particles). They can also influence regional and global atmosperic budgets and climate effects of reactive trace gases, aerosols, and clouds (e.g.: ozone depletion and denitrification in polar stratosperic clouds and cirrus clouds; for references see Pöschl et al., 2005a and Pöschl, 2005).

In order to avoid pitfalls of over-simplification and dead-ends in the development and application of atmospheric (chemistry) and climate models, we propose that model

formalisms should be based on terminologies and rate equations which are sufficiently general and consistent to allow both: (a) the deconvolution and description of individual elementary processes whenever it is necessary; and (b) the convolution of multiple elementary processes whenever it is possible and convenient.

For example, the transport of molecules from the gas phase into the particle bulk can be described by different but fully consistent parameters and rate equations, depending on the properties of the investigated aerosol or cloud system. Deconvolution of the overall process into surface accommodation and surface-bulk transfer enable the description of surface reaction and saturation effects. If such effects are insignificant, the rate coefficient of surface accommodation and surface-bulk transfer can be convoluted into a single bulk accommodation coefficient (Pöschl et al., 2005a, pp. 2150, 2154, 2168).

In both cases as well as under any other conditions, the effects of gas phase diffusion can be efficiently described by the gas phase diffusion correction factor, which depends only on the Knudsen number and uptake coefficient γ, regardless of the elementary processes and parameters determining the net uptake of the investigated gas onto or into the particle.

Response to Specific Comments

Comment on symbols: I think the readability of the text and especially of the equations could be improved substantially by simplifying the symbols throughout the manuscript. For example, instead of X_i and Y_j , simply X and Y would be sufficient.

Response: We are aware that the use of (multiple) indices and countin variables does not generally enhance the readability of symbols and equations, and from the beginning we had thought about alternatives. Unfortunately, however, the use of indices and counting variables is indispensable for the generalised equations presented in Part I of our model framework companion papers (Pöschl et al., 2005a), which are meant to be directly applicable for consistent description of multi-component and multi-reaction systems in mathematical models and numerical simulations. As illustrated in Part II

EGU

(Ammann et al., 2005), the counting variables i and j could be omitted only in cases where only single species and reactions are considered, but they are required for systems with multiple species and reactions.

Comment on equation (4): I find the symbol C_g for the newly introduced gas phase diffusion correction factor confusing. In the literature, C_g is often used for the gas-phase concentration. What about using f_g for the factor, instead?

Response: Thanks - we are planning to implement this suggestion upon revision of the manuscript.

Comment on page 2118, line 13: The unit should be "molecules per unit volume per time" and not "molecules per unit volume".

Response: Thanks - we are planning to perform this correction upon revision of the manuscript.

Comment on page 2120, line 9: The Knudsen number should be defined when it is introduced: $Kn = \lambda/r$.

Response: Thanks - we are planning to implement this suggestion upon revision of the manuscript.

Comment on typos on page 2123, line 16 and page 2183, line 21: Change "occurr" to "occur". Change "cloiud" to "cloud".

Response: Thanks - we are planning to perform these corrections upon revision of the manuscript.

Comments on Appendix A: The unit of $[Y_j]$ should be m^{-3} and not m^{-2}. The unit of the fluxes J should be $m^{-2}\ s^{-1}$ and not $m^2\ s^{-1}$.

Response: Thanks - we are planning to perform these corrections upon revision of the manuscript. The latter had already been pointed out in an earlier Author Comment on technical corrections (Pöschl et al., 2005b).

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

EGU

Comment on Figure 1: In the caption, it is said that both the symbols and the dotted lines are calculated according to equation (20). However, they are different. Please clarify.

Response: The calculations which are illustrated by symbols (FS) and refer to Fuchs and Sutugin (1971) are indeed based on Equation (19), not (20). We are planning to correct the erroneous figure caption upon revision of the manuscript.

References

Ammann, M. and Pöschl, U., Atmos. Chem. Phys. Discuss., 5, 2191-2246, 2005.

Pöschl, U., Angew. Chem. Int. Ed., in press, 2005.

Pöschl, U., Rudich, Y., and Ammann, M., Atmos. Chem. Phys. Discuss., 5, 2111-2191, 2005a.

Pöschl, U., Rudich, Y., and Ammann, M., Atmos. Chem. Phys. Discuss., 5, S455-S456, 2005b.

Sander, R., Atmos. Chem. Phys. Discuss., 5, S1568-S1570, 2005.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 2111, 2005.

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

EGU