

Interactive comment on “Kinetic regimes and limiting cases of gas uptake and heterogeneous reactions in atmospheric aerosols and clouds: a general classification scheme” by T. Berkemeier et al.

Anonymous Referee #2

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This paper presents the conceptual framework and proof-of-concept-type applications of a systematic classification scheme for heterogeneous reactions. The authors defined different regimes based on three quantities: the reaction location (surface versus bulk), the supply of reactive gas (reaction-diffusion limited versus mass-transfer limited), and the “well-mixedness” of the system (well-mixed versus gradient-limited). These quantities can vary independently, hence form a three-dimensional space. Based on this, eight limiting cases were identified. After establishing this conceptual framework, the authors applied it to some idealized test cases and also the oleic

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acid/ozone system by performing numerical simulations with a detailed multiphase model (KM-SUB).

This work builds nicely on previous work done by several co-authors by effectively using the PRA-framework in combination with a state-of-the-art explicit multi-layer model for multiphase processes to explore these processes. I view this paper as a very valuable contribution to the knowledge in this field, since it provides a comprehensive framework of how to think about and categorize heterogeneous processes relevant for atmospheric aerosols and clouds. It fits within the scope of ACP, and I recommend it for publication. However, the challenge with this paper is how to present the content so that it is accessible to a wider audience. The first 4 sections are relatively straightforward to follow, and I appreciate the authors' efforts to define and stick with a consistent notation (consistent also with prior papers). However, in particular the presentation of the simulation results in sections 5 and 6 should be improved before publication. A concern about the practical usefulness of this framework is listed under point 3 below. Specifically, I suggest the following modifications:

1. Use of acronyms: In general it would be helpful to keep the use of acronyms in the text to a minimum. Examples for this are the use of "SR" and "MP" within the text of sections 2.3.2 and 2.3.3. Or, in sentences like the one on page 1006, line 16: "e.g. change in reactive uptake coefficient as a function of r_p or $[X]_g$ ", change this to: "e.g. change in reactive uptake coefficient as a function of particle radius r_p or gas phase concentration $[X]_g$ ". Small changes like this will help making this manuscript more readable.

2. Readability of figures: Figures 5 and 6 are way too small, hence hard to read. Is this possibly an artifact of the formatting for the ACPD version? For the revised manuscript, I definitely recommend to make sure that these figures are large enough. Also enlarge the boxes in Figures 2 and 3 so that the labels can be in a larger font.

3. Practical usefulness of the classification approach: From the example of the Zie-

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mann data set I conclude that for the same experimental dataset, using two different parameter sets, one can arrive at the assignment of two different regimes. So given that many of the parameters listed in Table 6 are not well constrained at all (for example kBR and kSLR), I wonder how useful this approach is in practice. The authors discuss this somewhat in the conclusions, but this shortcoming should also be clearly stated in the main body of the manuscript.

Minor remarks: - Page 1009, line 2: Should read $[X]_g$ s read $[X]_g$?

- Page 1010, lines 2-9: Should $S(r_p)$ read $S^n(r_p)$? Table 4 uses yet a different variant of notation for the sensitivity coefficients. These are admittedly small details, but for a paper like this consistent notation is key.

- “the oleic-acid–ozone reaction system”: hyphen between oleic and acid and en dash between acid and ozone.

- Table 5: Note in the caption that the Cases 1-5 refer to the cases in the original papers.

- Figure 6: Are there error bars available for the Ziemann data and the Lee&Chan data? Also, it seems strange to extend the time axis to 35 s, while there is no data beyond 15 s.

- Page 1015, line 14: Which one is the SB^α regime mentioned here and in Table 6. Shouldn't I expect finding one of the regimes listed in Table 3?

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