

**Review of “An overview of current issues in the uptake of atmospheric trace gases by aerosols and clouds, by Kolb et al.**

This manuscript describes the state-of-the-art in the field of atmospheric heterogeneous kinetics. The authors, from a variety of US, Canadian and European research groups bring together a wealth of expertise and experience to generate a (justifiably) lengthy text. Being too general for the knowledgeable practitioner in this field yet too detailed for those hoping for a quick insight it is not always clear for which audience this was written. The manuscript has a “white paper” feeling to it and whilst demonstrating that significant progress has been made over the last decade it also indicates where improvements in data quality are required and also possible areas of future research. The inconsistent levels of detail presented for different chemical systems / surfaces presumably reflects different authors and is probably unavoidable in a document of this type.

One area where I feel that the authors have missed an opportunity is the lack of linkage to the recent IUPAC publication covering heterogeneous reactions on solid surfaces in the same journal. In many ways these two articles would both gain significantly in impact if close links were built in. The IUPAC document lacks the general overview, which Kolb et al provide, yet does give a high level of detail for individual trace gas – surface interactions that is not available here. As several of the authors are common to both papers, linking them can surely be accomplished without too much effort, most easily in section 5.2 but also in the introductory text.

Overall the paper of Kolb et al. is a valuable contribution to the field, and it should be published. The authors should consider revision in line with some of the comments below.

In section 2.2 terminology is discussed. Please indicate whether the recommendations are entirely consistent with the PRA or IUPAC usage and definitions, otherwise further confusion will result. In this context, define  $S$ ,  $C_g$ ,  $\alpha_m$  and  $\beta(\alpha_m)$  in section 2.3

Section 3. This deals with the various methods used for study of heterogeneous processes, with four selected as the most “successful”. I feel that if figures are warranted then so is a better and more detailed analysis of the range and limitation of each method. The droplet train, Knudsen reactor, coated wall tube and aerosol flow tube all have certain drawbacks. For solid surfaces, I am aware of only few examples where quantitative data from a Knudsen reactor has found its way into a model, whereas coated wall tubes have contributed substantially to the study of heterogeneous processes on e.g. ice and  $H_2SO_4$ , and droplet train and aerosol flow tube to study of aqueous surfaces.

P11159: In the equation on top of page:  $c$  should be  $\bar{c}$

P11159, L11: “An alternative approach” is a bad way to start a new section (alternative to what ?)

P11180, L8: “on the face of it” is too colloquial

P11184, L14: I’m not sure why IUPAC is cited here ?

P11186, L5: An atmospheric trace gas is not (neither strictly nor remotely) a surfactant film, Please re-phrase.

P11188, L18: Solubilities of HCl and HNO<sub>3</sub> are low in ice.....please provide reference(s).

P11191, L1: Have experiments at atmospherically relevant HNO<sub>3</sub> concentrations observed a decrease in the ice evaporation rate ?

P11191, L20-25: There are several theoretical studies of the bonding of trace gases to ice surfaces which reveal interesting aspects of the interaction and also provide adsorption enthalpies with which to compare experimental data. Perhaps some of these deserve mention

P11192, L10:  $\alpha$  has a small but significant.....what trace gas is the text referring to ?

P11194, L9: How are nitrates and sulphates formed from non-reactive uptake onto mineral dust ?

P11194, N<sub>2</sub>O<sub>5</sub> and SO<sub>2</sub> uptake to mineral dust. How does the uptake coefficient depend on relative humidity ? This should be mentioned.

P11196, L17: A similar example.....differ by three orders of magnitude. Which approach provides the larger value of  $\gamma$  ?

P11196, L22: ...”the insoluble/soluble transition should be taken into account”. Not sure what this is trying to say.

P11196-11197. There is (too) much discussion of BET versus geometric surfaces. In a nutshell, experiments using bulk porous samples, whether Knudsen or coated wall flow tube are problematic. The aerosol flow tube solves some but not all the problems. How does e.g. the AFT experiment cope with non-spherical particles ?

P11198, L18: The role of co-adsorption of traces gases is largely unexplored..... Co-adsorption might not be the main issue. Mineral dust can be rapidly chemically aged in polluted air masses. The role of aging (and loss of reactivity) due to exposure to especially, O<sub>3</sub>, HNO<sub>3</sub> and SO<sub>2</sub> are important questions as is the rate of re-activation by H<sub>2</sub>O. The rate of bulk dissolution of basic components of mineral dust is also of great importance (Sullivan et al, Phys. Chem. Chem. Phys, 2009).

P11199, L20-21. How about H<sub>2</sub>O<sub>2</sub> ? There is a recent study in ACPD (Pradhan et al, 2010).

P11200, L11: characterised should replace governed.

P11200, L30: Do not start the sentence with “it” (what affects soot reactivity ?)

P11201, L15: define PAH

P11201, L17-18: ...it appears that the RATE OF trace gas uptake .....faster than the RATE of reaction of the adsorbed PAH.

Figure 3. Poor quality labelling. Why acid gas and not trace gas ?

Figure 4. Poor quality labelling. Appears to be specific for  $\text{N}_2\text{O}_5$  uptake rather than a general experimental design.

Figure 5. Diagram does not really help to understand the principal of operation.