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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An overview of current issues in the uptake of atmospheric trace gases by aerosols and clouds" *by* C. E. Kolb et al.

Anonymous Referee #2

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This overview manuscript by Kolb et al., is an important service to the community that studies interactions of gases with atmospheric liquid and solid particles. A great deal of information has been processed in a systematic way that will be very useful, particularly to people starting research in these areas. A gas molecule colliding with a surface can undergo multiple interactions, such as for example adsorption, surface absorption or uptake into the bulk phase. Although these processes have been studied for several decades, an accepted uniform nomenclature describing the various processes has so far not emerged. Further, often the same symbol is used to represent different processes and at times even the processes get confused. For example, some researchers have confused surface adsorption with mass accommodation. A very important and immediately useful part of the manuscript is the chapter devoted to laying out the full scope of possible gas liquid interactions and presenting suggestions for un-

ambiguous nomenclature. In this field, most experiments measure gas uptake in one form or another. However, experimental measurements seldom provide directly values for the basic physical parameters such as αb (mass accommodation coefficient). The actual measurement contains that information but is also affected by experimental conditions such as diffusion of the trace gas of interest to the liquid or solid surface, the density of the trace gas, the shape of the surface and the gas-liquid interaction time, etc. The measured uptake coefficient has to be de-convoluted to provide the unambiguous basic parameter independent on experimental conditions. Kolb et al. present clearly and discuss the complexities of gas surface studies and review ways that these complexities can be treated. The article treats with particular care and at length interaction studies of gas phase species with water and aqueous surfaces in general. This is an appropriate choice because of the atmospheric importance of such interactions. The results of several experimental studies as well as computer simulations disagree with each other. The authors do not take sides with any set of results or experimental and computational techniques. Rather, they discuss the complexity of interactions and measurements and in most cases they discuss possible sources of error. I agree with this approach. The manuscript presents all the relevant references and an interested reader can decide what reliability to attach to a given result. (The article contains nearly 400 references.) This approach is applied also to the second section of the manuscript dealing with interaction of gases with surfaces of ice, minerals, organic compounds and soot where likewise many conflicting results are encountered. In connection with gas-mineral interactions, the authors provide a section discussing "Outstanding issues and future work". I think such a discussion is very useful and will help to formulate future studies. In a few cases (for example the effect of films on N2O5 uptake) the authors speculate about the mechanism responsible for the observed results. I find this an interesting and useful change of pace from the rhythm of the usual review article. The final Section 6 oriented toward suggested future work, includes a summary of funding available for gas uptake research. In summary, this is a very important contribution that merits publication. I present a few suggestions that the authors might want to consider.

Suggestions: 1. The uptake processes with their suggested coefficient designations are presented in Section 2.2. It would be helpful if a simple schematic diagram illustrating the processes and coefficients were introduced to accompany this section. 2. It is noted in Section 2.2 that the coefficients Γ can have values greater than unity. This could be confusing. It might be helpful to add a paragraph something like: "Gas uptake into a liquid is generally described by differential equations that couple the various processes affecting uptake. Important simplifications result if these processes are decoupled. In many cases this is possible without significant loss of accuracy. The Γ factors are obtained by decoupling some of the specific processes. These factors are most often employed in the resistive gas uptake model where $1/\Gamma$ is the effective resistance due to the related factor limiting the uptake. These equivalent resistors are always in series with other effective resistors for example $1/\alpha$b. If Γ is large say, greater than 1 that simply means that the resistance to uptake due to that process is negligible compared to the bulk uptake coefficient." 3. This survey article is written in the context of atmospheric chemistry. It would be helpful to discuss briefly how to determine which coefficients and under what circumstances dominate atmospheric gas uptake. Section 2.3.1 alludes to this issue by referring the reader to other articles. However, in this case I think two or three paragraphs discussing specifics would be helpful. 4. The treatment of surface thermodynamics is out of keeping with the rest of the manuscript. No other topic receives the type of detailed mathematical coverage as is presented in this section. For example, the resistor model of uptake is described only a qualitatively (as I think is appropriate). I suggest that surface thermodynamics be likewise described only qualitatively and only as it pertains to gas uptake especially since this thermodynamic formalism seems to have no bearing on the rest of the text. If the authors decide to keep the section as is, they should point out that Γ in this section is not the same as the coefficient Γ presented in connection with gas uptake in section 2.2.2. 5. In Section 5.2.1 it is stated "The mass accommodation coefficient α of H2O vapor on ice is..." It should be pointed out that when dealing with solids it is not always possible to make the distinction between adsorption and mass accommodation. Discussion of uptake

terminology applied to solids would fit well into Section 2.2. 6. It should be pointed out in the Introduction that although the workshop motivating this overview manuscript took place in April 2007, the information and references are brought up to 2010.

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