

Overall quality and significance:

The submitted article represents an enormous undertaking on the part of the authors to catalog, sort and evaluate a huge number of studies related to heterogeneous uptake of important atmospheric trace gas species onto relevant aerosol surfaces. It will certainly serve as the definitive reference for many, if not all, future laboratory studies of heterogeneous chemistry of significance to atmospheric processes. Hopefully, the results presented here become of interest and use to scientists involved in climate and chemistry modeling. The detailed comparisons and expert commentary may facilitate the incorporation of these laboratory results into more sophisticated models of atmospheric chemistry.

The quality and detail of the work are excellent and the presentation, especially with the detailed evaluative notes, is very good.

Specific comments:

There were a few issues I thought might have been addressed more thoroughly, or perhaps a little differently, in the background material of section 3.

The presence and amount of atmospheric water is one of the most critical environmental determinants in uptake kinetics and partitioning. In some systems, RH acts to facilitate uptake and surface reaction and in other systems water adsorption is competitive with trace gas uptake. Many of the results in the tables are for low pressure experiments with negligible or very low RH, conditions which are not relevant for tropospheric processes. For example, the preferred value for O₃ uptake is for RH values below 5% which is not relevant for the troposphere. I thought this might have been discussed in a little more detail. I do see a lot of information about RH and some parameterizations in the appendices, however.

The discussion of mineral dust was confusing in the presentation. At first, I thought the contention was that only studies on authentic dusts were to be included which I found questionable from the standpoint of heterogeneous chemistry. A number of atmospheric chemistry models account for a degree of heterogeneity in the aerosol population. There was also no discussion of the potential differences in the “authentic” dust samples, unlike the discussion of the details of ice formation. As far as I can tell there is no accepted protocol for collecting, storing and preparing these samples and from experience, the methods employed are sometimes pretty crude (perhaps necessarily so). On the other hand, commercial samples of mineral dust components are generally reproducible and well characterized. There have been at least some attempts to model the reactivity of actual dusts from a consideration of the reactivity of the individual components. Again, I do see many studies in the appendices that use model dust surfaces.

While surface saturation is discussed, I think it warrants a little more attention. Many of the systems exhibit irreversible uptake that either saturates or manifests a strong coverage dependence. Many of the values quoted are initial (clean surface) uptake coefficients. To put this into perspective, an uptake coefficient of 0.01 with 1 ppb of trace gas will yield coverages near a monolayer ($5 \times 10^{14} \text{ cm}^{-2}$) in only a few minutes. Perhaps the discussion in 3.3 is sufficient but I thought some more context would be useful.

Potential technical corrections (in no particular order):

Page 5247; “Equation (5) may also be used to parameterise the temperature dependence of the uptake coefficient observed at a given time or averaged over a given time interval.” Do the authors mean time dependence?

Page 5246 and 5249, eqn. 3, eqn. 11 and maybe some of the others should be formatted like eqn. 13 so that numerator and denominator are clearly defined.

I don’t understand the symbol “=c” for thermal velocity. It is very confusing in the context of some of the equations. Maybe this is an html formatting issue.

The discussion of units in 3.4 is a little confusing. Page 5248, eqn. 6 is given in terms of the fractional coverage (unitless) but the discussion of the use of eqn. 6 and the subsequent discussion of the linear regime partition coefficients is in terms of surface coverage ($\# \text{ cm}^{-2}$). It is not until eqn. 15 that θ is defined (as N/N_{max}). Perhaps that should be made explicit in eqn. 6.

I don’t think there should be a factor of “ $k_{\text{B}}T$ ” in the “ K_{linP} ” term of eqn. 13. As it is rendered, the term is not unitless.

Table on page 5250 is missing right parenthesis on some partition coefficients in the “To” column. Also a superscript “a” appears on the “T” in the third row and I am not sure why.

What is “[X]” in eqn. 15? Should this be “[X]_g”? Also, this notation seems to take different forms (e.g. “[X]_g” appears in eqn. 13, the only place it is used).

I assume notation like “ $K_{\text{LangC}}(X)$ ”, in eqn. 16, and “ $\gamma_{\text{gs}}(X)$ ”, in eqn. 17, refer to parameters for species “X” but this should be made explicit.

I didn’t understand this from page 5253; “Equation (16) demonstrates that γ depends on the gas phase concentration of X, if $K_{\text{LangC}}(X)[X]_{\text{g}}$ is similar to or larger than 1 (i.e., at high coverage).” Do they mean at high pressures of X? Similarly, for the next sentence; “This is especially important when interpreting data from laboratory experiments performed using gas-phase reactant concentrations, which lead to significant surface coverage.” Do they mean “high gas-phase reactant concentrations”?

It may be useful to point out that, in practice, it is very difficult to experimentally differentiate between LH and ER surface mechanisms. The uptake must be evaluated in the high pressure/high coverage regime for both reactants and the data must be of high quality.

On page 5255, they may want to describe eqn. 21 as; “For trace gas reactive uptake to the bulk...” to distinguish from eqn. 20.

On page 5256, in eqn 23, I presume “r” is the drop radius. That should be defined.

Also on page 5256, I am not sure about the validity of eqn. 24. The text implies that it is the effective Henry’s law constant for an acid but the formula is not right (it implies that solubility is enhanced in acidic solutions). If the equation is explicitly for an acidic species, then that should be made clear.

On page 5256. There are two Hanson et al. 1994 references. Please indicate which is which.

On page 5257, there is no Hu 1995 reference.

In datasheet, V.A2.4, the γ value maybe should be “ 10^{-6} ”

On page 5489, footnote (c), should maybe be “ 0.25 m^2 ” for surface area.

There is no appendix 3.