

Comments by Referees are in blue. Our replies are in black. Changes to the manuscript are highlighted in red both in here and in the revised manuscript.

Reply to Ref #3

This is a very interesting review focusing on the heterogeneous reactions of mineral dust aerosol with trace gases in the atmosphere. It presents a comprehensive and critical review of laboratory studies of heterogeneous uptake of OH, NO₃, O₃, and related species by mineral dust. The point of view which has been chosen here i.e., assessing the importance of the heterogeneous processes by comparing the associated lifetimes with other major loss processes and by discussing relevant field and modelling studies is very interesting and brings real added-value to already published reviews.

I really enjoyed reading this manuscript and would therefore recommend its publication in Atmospheric Chemistry and Physics. I have only one comment on which I would like to draw the authors' attention to.

Reply: We would like to thank ref #3 for his/her highly positive comments on our manuscript. All the comments have been properly addressed in the revised manuscript, as detailed below.

As one of the major target of that review is to derive lifetimes associated to heterogeneous processes, which is a valuable information, I would encourage the authors to put more emphasis on the needed/missing input information and more specifically on the need to use uptake coefficient derived under steady state conditions. Clearly this point is already more or less addressed in this review but without being properly emphasized. I believe that devoting a full paragraph to this issue in section 1.2 ("Introduction to heterogeneous kinetics") would be the way to go, and then for every targeted compound to highlight what is known and unknown with respect to long exposure times (i.e., steady state conditions), as initial uptake coefficients should not be used to derive atmospheric lifetimes for mineral dusts.

Reply: It is true that initial uptake coefficients should not be used to calculate atmospheric lifetimes due to heterogeneous uptake onto mineral dust. Nevertheless, as discussed in Section 2.2.2, steady-state uptake coefficients reported by laboratory studies depend largely on experimental conditions, such as trace gas concentrations and the mass of particle samples. Therefore, with the knowledge available up to now, we feel it very difficult to have definite

answers to the important question raised by ref #3.

Also, when the authors derive these lifetimes, they fix one gas phase concentration and then do the calculations. However, for many of the processes discussed here, the lifetime will change with concentration and therefore the associated lifetimes will be spatially different. For instance, an uptake process could be slow at high concentration (i.e., at ground level) but significantly faster under reduced concentration (i.e., at higher altitude). Maybe the authors could bear that in mind when assessing lifetimes for some of the compounds, and especially ozone.

Reply: We absolutely agree with ref #3 that a single uptake coefficient is not enough to describe the kinetics of a heterogeneous reaction. In the revised manuscript (line 643-649), we have included the following sentence to mention this caveat: “We also acknowledge that a single uptake coefficient may not always be enough to describe the kinetics of a heterogeneous reaction of mineral dust, because 1) uptake kinetics may change with reaction time, as discussed in Section 2.2; 2) uptake kinetics are also affected by particle mineralogy and composition, RH, temperature, the co-presence of other reactive trace gases, and etc.; and 3) for some reactive trace gases, such as O₃, the uptake coefficients may strongly depend on their concentrations.”

Minor points

The introduction, and justification of that review, is maybe a bit lengthy and could be reduced without loss of information.

Reply: Indeed the introduction section in our manuscript is quite long. Nevertheless, since we aim to provide a comprehensive review of this topic, we feel it is necessary to try our best to mention all the aspects in the introduction. In addition, because in the last two decades there have been a few excellent reviews in this field, we would like to emphasize why writing our current review paper is justified and what distinguish our review paper from previous ones.

The simplified figure 1 is still somewhat difficult to follow.

Reply: As suggested, we have simplified Figure 1 in the revised manuscript to make it easy to follow.

Why plotting the uptake coefficient versus the gas phase concentration on log-log scale (e.g., figure 13)? Is there a justifications for that? There are different ways of linearizing the adsorption isotherms and extract meaningful information.

Reply: This is because both O_3 concentrations and $\gamma(O_3)$ shown in this figure (and Figure 12 as well) span over a few orders of magnitude. In the revised manuscript (line 1244-1245) we have added one sentence to explain why we use log-log scale: “Both O_3 concentrations and $\gamma(O_3)$ are plotted on the logarithm scale because their values span over a few orders of magnitude.”