

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 #2

This review on heterogeneous reactions of mineral dust aerosols (desert dusts, SiO₂, Al₂O₃, TiO₂) is very well written, useful for atmospheric chemists and recommended to be published after some revisions commented below.

Reply: We would like to thank ref #2 very much for recommending our manuscript for publication. His/her comments, which largely helped us improve our manuscript, have been properly addressed in our revised manuscript, as detailed below.

1. In this manuscript, logical uncertainty resides around the experimentally obtained values of uptake coefficients of benchmark sample particles based on assumed surface areas, and uptake coefficients of real ambient dust particles to be used for the calculation of their heterogeneous reaction rates and lifetimes in the atmosphere. Experimentally obtained values of γ depends on the assumed surface area, and those based on BET area and geometrical area are in general different in more than a couple of orders of magnitude as reported in this review. It is recommended to clarify the idea which area is more relevant to the heterogeneous reaction on the ambient aerosol particles. How is the real surface area of ambient dust particles, and how to calculate the best heterogeneous reaction rates based on reported γ values? In p.23 (line 486-487), although it is described “The surface area actually available for heterogeneous uptake falls between two extreme cases and varies for different studies,” this description is not enough to give a solid idea to readers.

Reply: As pointed out by ref #2 as well as in our initial manuscript, uptake coefficients based on geometrical area and the BET area can differ by a few orders of magnitude. Nevertheless, up to now there is no universally accepted method to estimate the surface area actually available for heterogeneous uptake, and the statement “The surface area actually available for heterogeneous uptake falls between two extreme cases and varies for different studies” is the only one we can make.

In fact in our initial manuscript we discussed a few methods (such as the KML model and the LMD model) which have been proposed in order to better estimate the surface area available for heterogeneous uptake. In the revised manuscript, as suggested by ref #2, we have

expanded our discussion on the KML model, the LMD model, and etc.

It is suggested to add a new section on “characteristics of ambient mineral dust particles” for example at the beginning of “2 Background” to describe; 1) their chemical characteristics such as relevance of desert dust and benchmark minerals, SiO₂, Al₂O₃, and TiO₂, and 2) their physical characteristics such as surface area, porosity, etc. which are relevant to estimate the heterogeneous reaction rates. Quantitative descriptions on these topics are very useful for readers.

Reply: In the revised manuscript (line 75-77) we have provided additional information on mineralogy of tropospheric mineral dust aerosols: “According to a recent global modeling study (Scanza et al., 2015), major minerals contained by tropospheric mineral dust particles include quartz, illite, montmorillonite, feldspar, kaolinite, calcite, hematite, and gypsum.”

In addition, we have also provided further information on physical characteristics of mineral dust particles. For examples, in line 77-80 of the revised manuscript, we have included the following sentence for particle size: “Formenti et al. (2011) summarized published measurements of tropospheric mineral dust particles, and the size of mineral dust particles depends dust sources and transport, with typical volume median diameters being a few micrometers or larger.” In line 495-497 of the revised manuscript, we have included the following sentence for particle shape: “It has been reported that the median aspect ratios are in the range of 1.6-17 for Saharan dust particles (Chou et al., 2008; Kandler et al., 2009) and 1.4-1.5 for Asian dust particles (Okada et al., 2001).”

Some detailed description of the methodology to obtain surface area of dust particles, how to obtain geometrical area of non-spherical particles and BET area, would be helpful.

Reply: In line 512-513 of the revised manuscript, we have expanded the sentence to make it more clear how to obtain geometrical area of non-spherical particles: “In these experiments the surface area available for heterogeneous uptake is assumed to be either the projected area of dust particles (usually referred to the geometrical area of dust particles, equal to the geometrical surface area of the sample holder) or the BET surface area of the dust sample.”

BET surface area measurements are widely used in characterization of solid particles. Instead of providing a detailed description on how to measure BET surface area, in line 514-515 of the revised manuscript, we have included a sentence to refer interested readers to proper

references: “Description of methods used in measuring BET surface area of solid particles can be found elsewhere (Sing, 2014; Naderi, 2015).”

Also some detailed description on transport of gaseous molecules within interior space of the powdered sample, and KML model would be helpful.

Reply: As suggested, in the revised manuscript (line 520-525) we have provided a description on reaction and transport of gas molecules within powdered samples: “When gas molecules are transported towards the top layer of the powdered sample, they may collide with the surface of particles on the top layer, be adsorbed, and undergo heterogeneous reaction; they may also be transported within the interior space and then collide and react with particles in the underlying layers. The depth gas molecules can reach depends on the microstructure of the powdered sample (e.g., how compactly particles are stacked) as well as their reactivity towards the surface.”

Furthermore, in line 536-538 of the revised manuscript, we have included another sentence to further introduce the concept of the KML model: “An “effectiveness factor” was determined and used in the KML model to account for the contribution of underlying layers to the observed heterogeneous uptake.”

2. It is interesting to compare the uptake coefficients of dust with more commonly available soil and ambient aerosol. Although some description is available for HCHO in p. 72 (line 1309-1310), further comparison of γ for HO₂ by ambient common aerosols, and others should be described whenever the data are available.

Reply: Studies on heterogeneous reactions of reactive trace gases with ambient mineral dust aerosol are very limited, and most of them (if not all) have been discussed in our manuscript. There are many studies on heterogeneous reactions of reactive trace gases (mainly HO₂, H₂O₂ and N₂O₅) with other types of ambient aerosol particles, but their chemical composition and thus heterogeneous reactivity may be very different from these of mineral dust; therefore, such comparison may not be proper.

In addition to heterogeneous reaction of HCHO with soil, in our original manuscript (line 1403-1408) we have also discussed heterogeneous uptake of HONO onto soil samples.

A very recent study (Moon et al., 2017) examined heterogeneous uptake of HO₂ radicals by TiO₂ particles. In the revised manuscript (line 784-780), the study by Moon et al. have been

discussed, and Table 4 has also been expanded to include the major result reported by Moon et al. (2017).

3. p. 47 (line 889): Specific wavelengths should be described for “UV illumination”.

Reply: The wavelength range is 315-400 nm, and this information has been included in the revised manuscript.