This paper discusses the correlation of atmospheric mineral dust, and heterogeneous chemistry with the lifetime of SO_2 and would give a closer prediction of the formation of sulfate aerosols. Therefore this will be a good addition to our current knowledge in this field of atmospheric chemistry. I have a few comments and suggestions.

1. Abstract: I think we shouldn't say accurately predict - This is because it is still a prediction. Maybe with near accuracy, some rewording would be good.

Answer:

Thanks for your comments.

We have rewritten the sentence in the revised manuscript (line 13-14).

"Regression models were then developed to make a reliable prediction for the heterogeneous reactivity based on the particle chemical compositions."

2. Authors discuss at several points that the differences of dust surfaces can affect their heterogeneous reactivities. (Under driving factors of dust surface, line 265). Another addition to this section: TiO_2 and other titanium-bearing minerals such as ilmenite showed night-time thermal reactions that involve redox cycling with nitrates, as well as pH dependency.

Answer:

We have reconsidered the different heterogeneous performances of Ti-bearing constituent under the dark and light conditions. The following explanation has been added into the revised manuscript (line 279-281).

"In addition, the heterogeneous reaction on the surface of TiO_2 or Ti-bearing mineral is thermodynamically favored during nighttime but largely dependent on photocatalysis under solar irradiation (Chen et al., 2012)."

3. Figure 2: why do some minerals have lower sulfates in the daytime while the others have lower sulfates in the nighttime? Can this be explained with the change of particle pH? What is the pH of each sample before the reaction? Fig 2b shows only the pH after reactions.

Answer:

All of the studied clay minerals, with the exception of NAu, present greater sulfate production during daytime than nighttime, which can be explained by their photocatalytic performances (mainly attributed to the Al- and Ti-bearing components as discussed by Section 3.1). The opposite situation of NAu can be explained by its rich abundance of iron oxides that would undergo photoreductive dissolution in acidic media. We have emphasized this point in the

manuscript (line 331-335).

"The diurnal uptake coefficients of CCa, SWy, KGa, IMt are respectively 1.52, 1.01, 2.94, 2.30 times greater than the corresponding nocturnal ones, reflecting the distinct photocatalytic performances of the clay minerals. Conversely, NAu presents the decreased heterogeneous uptake capacity under light irradiation than dark condition due to its rich abundance of Fe, whose oxide species may occur photoreductive dissolution in acidic media (Fu et al., 2010; Shi et al., 2012)."

In addition, the association between the uptake coefficient for sulfate formation and the corresponding particle acidity has been discussed in the revised manuscript (line 355-359).

"For one clay mineral, its different acidities after dark and light reactions correspond to the distinct heterogeneous kinetics. All of the studied clays, with the exception of NAu, become more acidic after the photoreaction than the dark reaction, which can be explained by the photoinduced SO_2 adsorption and sulfate production on these samples. The opposite situation of NAu coincides to the decreased heterogeneous kinetics over its surface by the presence of solar irradiation."

The current methodology cannot estimate the acidity of the unreacted sample. The acidity of the reacted sample was derived from the relative abundance of $SO_2 \cdot H_2O$ and SO_3^{2-}, as assumed to be equivalent to the relative integral areas of their characteristic peaks. By contrast, the particle sample before reaction was scanned by infrared as a background spectrum, and therefore no infrared signals can be obtained for the unreacted particles.

4. Is chemistries a real word?

Answer:

As a real word, it was widely utilized in the research papers of this field (e.g. doi.org/10.1021/cr500501m; doi.org/10.1021/cr5003485).

5. Figure 3: why does the reactive uptake coefficient drastically increases after particle acidity reached ~4.5/ 5?

Answer:

Figure 3 displays the particle-acidity-dependent reactive uptake coefficients (γ) for the dust-mediated heterogeneous chemistry. We have discussed the dramatically increased γ when pH exceeds 4.5 or 5.0, in the revised manuscript (line 367-369).

"The γ of NO_2, O_3, HOCl, HOBr and CH_3COOOH presents positive dependence toward pH, in accordance with the evolution of the effective Henry's law constant for the studied sulfur species, as theoretically illustrated by Eq. (5)."

6. For daytime chemistry, did you consider the possibility of the formation of sulfate radicals on surfaces with high Ti content?

Answer:

We have considered the impact of sulfate radical on the heterogeneous formation of sulfate. Necessary discussions are added into the revised manuscript (line 266-267).

"Sulfate radical ($SO_4^{\cdot-}$) is generated by the presence of abundant $\cdot OH$ and participates into the oxidation events (Antoniou et al., 2018; Kim et al., 2019; Li et al., 2020b)."

7. Figure 5: Why the kinetics has increased drastically after pH 5? Within the statistical error, do you see any difference between the aq phase and heterogeneous chemistry?

Answer:

We have explained the pH dependences of the aqueous-phase and dust-mediated heterogeneous kinetics in the revised manuscript (line 449-452).

"In more detail, the significant sulfate formation in the acidic environment is primarily contributed by the TMI-O_2 pathway where the concentrations of TMIs and H^+ keep increasing as the aerosol water becomes more acidic. When pH>5.0, the formation rate of sulfate increases with the elevated pH, in step with the evolution of the concentration of dissolved sulfur species."

Moreover, we have explained the kinetic difference between the aqueous-phase and dust-mediated chemistries. The addition to the revised manuscript (line 452-453) is shown as follows.

"Within the entire pH range, the aqueous-phase chemistry is more efficient than the dust-mediated heterogeneous chemistry in contributing to secondary sulfate."

8. Figure 7: Can you better label the y-axes to explain what % contribution? Panel b: why does the heterogeneous sulfate contribution for a daytime peak at a certain dust concentration and lowers, whereas it reaches a plateau at night time?

Answer:

We have re-labelled the y-axes of Fig. 7 as "Secondary sulfate contribution (%)".

In the revised manuscript (line 518-525), we have additionally explained the different responses to the concentration of dust, as shown follows.

"The increased dust concentration facilitates the aqueous-phase TMI-O_2 pathway and the dust-mediated and dust-

driven heterogeneous reactions, whereas presents negative impacts on the others by the removal of gaseous oxidants over dust surface. The unsusceptible response to dust concentration, as shown by Fig. 7b, can be related to the increased sulfate contributions from the TMI-O$_2$ pathway. Unlike the TMI-O$_2$ pathway and dust-driven chemistry receiving constant positive feedbacks from the increased dust concentration, the dust-mediated chemistry is somewhat affected by the decreased oxidants adsorbed on dust surface. Since the importance of dust-mediated chemistry is more significant during daytime than nighttime, there is a negative correlation between the high dust concentration and heterogeneous contribution proportion under solar irradiation."

9. Why is more SSA formed during nighttime, but more sulfate is formed during the daytime?

Answer:

We have noticed that the observed sulfate concentration during nighttime may be comparable to or even exceed that measured during daytime, in contrast to the calculated results derived from this work. Such phenomenon can be explained by the different meteorological conditions during nighttime and daytime in the real atmosphere. In the revised manuscript (line 396-401), we have discussed these uncertainties and emphasized the current comparisons through kinetic regime. The added discussion is shown as follows.

"It is worthwhile to mention that, in the real atmosphere, the observed sulfate concentration during nighttime may be comparable to or exceed that during daytime, which can be explained by the higher nocturnal humidity facilitating the liquid oxidations or the lower boundary layer causing the adverse diffusion conditions (Liu et al., 2017c; Tutsak and Koçak, 2019; Li et al., 2020c). The relevant meteorological factors were not considered by this comparison model, and the current results emphasized the different sulfate formation potentials through kinetic regime.