Dear professor Thorsten Bartels-Rausch,

Thank you for your decision to allow us to revise our manuscript for publication in ***Atmospheric Chemistry and Physics***. Below is a point by point response to the comments.

Referee #1：

Thanks for the detailed response from the authors. The authors have clarified most of my comments and made significant improvement in this revision. There is still one concern related to the possible contamination or impurity of trace photocatalytic components in these non-photoactive mineral dust samples, which is NOT yet ruled out but is indeed my main worry in my comment (1).

The authors reported the purities of different mineral substances to be 95%–98%. They argued that if their observation is driven by photoactive impurity, then SO_2 uptake on photoactive substances would be a factor of 20-50 higher than 1E-7 to 1E-6, while SO_2 uptake coefficient on photoactive substances was normally reported to be 1E-7 to 1E-6 in previous literatures.

This is clearly not a good argument because some studies did not use pure photoactive substances, e.g. Arizona Test Dust (ATD) as cited in Park et al., 2017, ES&T. I found it is also a bit contradictory in their subsequent reply to my comment (7): "Does this mean both photoactive and non-photoactive mineral oxides showing equal/comparable ability of SO_2 photochemical uptake, and those photocatalytic components (such as TiO_2, GDD, ATD) do not actually play much role", where they explained due to different specific experiment conditions. Such explanation is always true, but again, it is still necessary to rule out the possible photoactive impurity used in their samples through detailed chemical characterization.

To be rigorous and cautious, I would encourage the authors to provide a table containing chemical analysis and elemental distribution of their samples, if applicable prior published in ACP. A good example is shown in the SI Table S1 in Dupart et al., 2012, PNAS. Another paper cited by the authors, e.g., Park et al., 2017, ES&T, also provides mineral dust chemical characterization in SI Section S1. In addition, the newly added text in both comments (1) and (7) should be rephrased to minimize such logical contradicts. The new data (SO_2 uptake on SiO_2 at pH=9) in Figure S11 is good, which can be implemented in the main text Figure 4.

Refs

Dupart Y, King S M, Nekat B, et al. Mineral dust photochemistry induces nucleation events in the presence of SO_2[J]. Proceedings of the National Academy of Sciences, 2012, 109(51): 20842-20847.

Park J, Jang M, Yu Z. Heterogeneous photo-oxidation of SO_2 in the presence of two different mineral dust particles: Gobi and Arizona dust[J]. Environmental science & technology, 2017, 51(17): 9605-9613.

Re: Thank you for your comments. According to your suggestion, the chemical composition of SiO_2 has been analyzed, as shown in Table S1 of the revised Supporting Information. Table S1 shows that the fraction of SiO_2 in the sample was 99.02%, accompanied by a small amount of Al_2O_3, K_2O, Fe_2O_3 and CaO. Photoactive substances (Fe_2O_3) was very few in the sample, and they should not be the main contributor to the photochemical uptake of SO_2. This description has been added in Lines 218-221 in the revised manuscript, which avoided a bit contradictory in original comment (1) and (7).

The new data (SO_2 uptake on SiO_2 at pH=9) in Figure S11 has been implemented in Figure 4 in the revised manuscript.

Referee #3：

Despite decades of research on the oxidation of SO_2 to sulfate in the atmosphere, it is surprising that some potential pathways have still not been studied in sufficient detail. Yang et al. provide such a study with detailed experiments shedding light on the heterogeneous oxidation of SO_2 on non-photoactive dust. They focus on SiO_2, but also present results for other dust types, which together actually constitute by far the largest fraction of mineral dust in the atmosphere. This topic is, thus, of interest for the readers of ACP. As acknowledged by the other reviewers the study is sound and clearly structured. It now also contains all relevant technical information and I recommend publishing it. Nevertheless, I add below some comments, which only concern editorial or technical corrections or improvements, which should be considered before publication.

Re: Thank you for your comments.

Major comment

I would like to come back to one of the comments by reviewer 2, who wondered if other studies on SO_2 oxidation on non-photoactive minerals exists. In their response the authors admit that such studies can be found mentioning the study by Zhang et al.. Other studies can also be cited like Xu et al. (Sulfur isotope composition during heterogeneous oxidation of SO_2 on mineral dust: The effect of temperature, relative humidity, and light intensity, Atmospheric Research, 254, 2021, 105513, https://doi.org/10.1016/j.atmosres.2021.105513) and should be mentioned in the manuscript. Taking this into account, I still think that statements like (L. 347) "Nevertheless, it was very surprised that the light can greatly promote the formation of sulfates via the SO_2 uptake process on mineral oxides without photocatalytic activity, which was strongly suggested to be a new and important finding for atmospheric sulfate sources." or (L. 449) "This suggests that the SO_2 on non-photoactive surfaces is a newly identified sulfate formation pathway in some dust-rich conditions." should still be toned down.

Re: Thank you. Few studies observed the photochemical uptake of SO_2 on non-photoactive minerals (Xu et al., 2021; Zhang et al., 2022). This description has been added and the related studies were cited in Lines 196-197 in the revised manuscript. The original sentences in Lines 347-350 and 449 have been toned down. Sentences have been revised into "Nevertheless, the light can greatly promote the formation of sulfates via the SO_2 uptake process on mineral oxides without photocatalytic activity", and "This suggests that the SO_2 uptake on non-photoactive surfaces may be an important sulfate formation pathway under irradiation in some dust-rich conditions." in Lines 342-344 and 442-443 in the revised manuscript, respectively.

In my opinion the manuscript requires some serious proofreading and editing since it still contains a high number of language errors or confusing or incomplete sentences. Some examples from the introduction are:

L. 35: "Typical mixing ratios of SO_2…"

L. 36: "…for a clean weather in remote areas,…"

L. 38: "…which is one of the most significant compositions in fine particles.

L. 38: "The mass fraction of sulfates in $PM_{2.5}$ is high up to 30% …"

L. 43: "…causing respiratory illness and cardiovascular (…"

I realized that one of the sentences was recommended by one of the reviewers (L. 38), but still it should be re-written.

Re: Thank you. These original sentences have been modified, as shown in Lines 35, 36, 38-39 and 42-43 in the revised manuscript. We also carefully revised the full text to avoid other mistakes.

L. 327ff: The uptake coefficients on SiO_2 with a pH adjusted to 9 are mentioned. It would be good to include these numbers as further bars in the Figures 4A and 4B.

Re: Thank you. According to your suggestion, the data (SO_2 uptake on SiO_2 at pH=9) in Figure S11 has been implemented in Figure 4A and 4B in the revised manuscript.

Fig. 5: Wouldn't it be better to arrange all experiments in the dark in the top row and all experiments with irradiation in the bottom row?

Re: Thank you. According to your suggestion, the experimental results (Figure 5) in the dark and with irradiation have been arranged in the top and bottom rows, respectively.

Thank you very much for your help.

Sincerely yours,

Chong Han

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