

Previous referee comments are displayed in green and preceded by 'R', previous author responses are displayed in blue and start by 'A'. They are followed by editor comments. At the end, there are numerous additional editor comments that need to be addressed before the paper can be considered for acceptance.

## Referee #2

R4: Is chemistries a real word?

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

**Editor comment:** 'Chemistry' is defined as a branch of science that studies the composition and transformation of molecules and materials (cf e.g. Wikipedia). Therefore, in this context, it should not be used at all, but should be replaced by 'chemical processes' (or 'chemical composition' where appropriate) throughout the manuscript.

R5. Figure 3: why does the reactive uptake coefficient drastically increases after particle acidity reached ~4.5/ 5?

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

*"The  $\gamma$  of NO<sub>2</sub>, O<sub>3</sub>, HOCl, HOBr and CH<sub>3</sub>COOOH 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)."*

**Editor comment:** I do not understand this response.

A few lines above you state that "the dust mediated heterogeneous oxidation can be assumed to primarily proceed in the surface water layers" but yet you ascribe a significant impact on  $\gamma$  by the term that describes the aqueous phase diffusion, i.e. the second term in the right-hand side in Eq. -5. What are the contributions to the total  $\gamma$  of the mass accommodation term ( $1/\alpha$ ) and the aqueous phase diffusion and reaction term  $[v/(4 H RT (D_a k_{chem})^{-1}) \times 1/f_r]$ ? A figure should be included to demonstrate that indeed the second term increases significantly at increasing pH.

R6. For daytime chemistry, did you consider the possibility of the formation of sulfate radicals on surfaces with high Ti content?

A: 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<sub>4</sub>•-) is generated by the presence of abundant •OH and participates into the oxidation events (Antonίου et al., 2018; Kim et al., 2019; Li et al., 2020b)."*

**Editor comment:** This response and revision is not sufficient to discuss the potential role of sulfate radicals. A discussion on possible chemical mechanisms of the observed processes should be added including the possibility of oxidation by the sulfate radical. You can either add this to section 3.3. or dedicate a full new section to this discussion.

### **Referee #3:**

R: Line 373-374: Earlier authors assert that the sulfate formation is more during nighttime and less during the daytime, but this statement is opposite and confusing. Can the author clarify this?

A: In the real atmosphere, the sulfate concentration during nighttime could be comparable to or even exceed that during daytime, as normally explained by the different meteorological conditions. We have explained the relevant details in the revised manuscript (line 396-401) to make our calculation results more reasonable.

*"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."*

**Editor comment:** I still got confused what you compare in this section vs to the discussion earlier in the manuscript. Please re-organize Section 3.3. such that it becomes clear that these are estimates based on a model for which you assume ambient conditions whereas previously you discussed experimental lab data.

It might help if you refer in the discussion of your experimental data to 'illuminated' and 'dark' conditions only and in the discussion of model results for atmospheric conditions to 'day' and 'night'.

R: Line 380-390: In addition to the temperature dependence, did the authors consider the change of relative humidity during the day and night can also contribute to the heterogeneous chemistry reaction with sulfate?

A: We attempted to carry out the heterogeneous laboratory experiments under the higher relative humidity (RH) relative to the current setting. However, the presence of surface adsorbed water makes the Gaussian/Lorentzian deconvolution difficult to be performed, as reflected by the large uncertainties in determining the relative abundance of SO<sub>2</sub>·H<sub>2</sub>O and SO<sub>3</sub><sup>2-</sup>. Accordingly, the RH of 50% was utilized for the experiments. In the revised manuscript (line 654-656), we have emphasized the importance of various meteorological factors when discussing the atmospheric relevance of heterogeneous chemistry, as shown follows.

*"In addition, the influence of meteorological factors, like temperature, humidity and irradiance, on the atmospheric relevance of heterogeneous chemistry warrants further research."*

**Editor comment:** I understand that you cannot do any quantitative estimates on the influence of humidity. However, a statement on the extent to which relative humidity would change particle radius and aqueous volume and therefore sulfate formation should be added.

### **Additional editor comments**

I. 32: What do you mean by 'in mainstream'?

I. 35: What do you mean by 'aimed liquid reaction' as opposed to 'aqueous phase reaction'? Do you imply reactions in liquid non-aqueous phases?

I. 37: What do you mean by 'gas-solid interactions'? What solid phase are you referring to? In the lines above, you refer to reactions on or in liquid phases.

I. 64 – 66: This sentence is too convoluted and mixes multiple processes and effects. The study by Clegg and Abbatt was performed on ice, Wu et al explored uptake on calcium carbonate and Wang et al. reactivity on hematite. Either split this sentence into three describing the essential information from each study or remove.

I. 68: What do you mean by 'negative light effect'?

I. 73-74: 'In terms of...' – what does this sentence say?

I. 75: 'is more efficient' for what?

I. 77: What do you mean by 'dust community'?

I. 108: correct 'derisble'

I. 119: replace 'Technical Route' by 'Experimental methodology' or something similar.

I. 125: In the sentence starting 'In the dust-mediated...' is a verb missing.

I. 140: Please clarify this sentence. Why don't you state simply the relevant temperature range as being representative for the warm season?

I. 178/179: Which 'gas and aqueous phase parameters' are you referring here to?

I. 195: The reference to Hennigan et al. 2015 would be much better placed in this context here than in I. 190 as it was shown in their study that HSO<sub>4</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> are not sufficient to calculate acidity.

I. 212: what do you mean by 'standard item'? Is it the 'standard mixture' or 'standard solution'?

I. 255: replace 'corrects' with 'correlates'

I. 259: Why only 'nocturnal heterogeneous reaction'? Why does this not happen during the day? Could 'nocturnal' be omitted in this sentence?

I. 263 – 291: This text should not be in the result section but belongs in the introduction where it is partially already cited. Please condense this paragraph and either move relevant parts into the discussion of your results (which are not presented at this place of the paper yet) or to the introduction.

I. 350: This sentence is not clear. Please clarify what you want to say here.

I. 350/351: 'no significant correlations can be found between  $\gamma$  and pH' contradicts your later discussion and findings in Figure 3. Please clarify.

I. 359/360: Are three digits of the pH value significant figures? What is the variability in this value?

I. 430: replace 'great heterogeneous performances' by 'significant contributions by heterogeneous sulfate formation' (or similar)

I. 446 – 448: The sentence starting ‘As proved by ...’ seems out of place here. Either remove or clarify its connection to the text in this paragraph.

I. 454-461: 1) Refer to Figure 6 at the beginning of this text , 2) Be more specific in the text, rather than just saying longest, second longest etc lifetime.

Figure 4: 1) Make labels on panels b, c, d, f, g, h larger. 2) Explain the colors in panels a and e. Are they same as in panels b and f?

I. 526: should ‘merely’ be ‘only’ here?

I. 547: These dust concentrations seem only representative for some regions of the globe (e.g. <https://agupubs.onlinelibrary.wiley.com/doi/epdf/10.1029/2021MS002845>). Please tone down your statement and state that your conclusions are only relevant for specific regions.

I. 587: What do you mean by gas-solid interface? Aerosol particles are never completely dry and thus, gases are always exposed to aqueous surfaces.

I. 648: I do not understand the expression ‘plausible influencer’. I suggest removing it.

I. 674: Please add all data into a suitable repository that can be accessed freely by the readers, corresponding to the journals’ data policy [https://www.atmospheric-chemistry-and-physics.net/policies/data\\_policy.html](https://www.atmospheric-chemistry-and-physics.net/policies/data_policy.html) .