

Anonymous Referee #2

General comments

The authors investigated heterogeneous uptake of sulfur dioxide on iron-containing mineral particles. They found enhanced heterogeneous uptake of sulfur dioxide on the mineral particles through modification of iron speciation. The results shown in this paper are very interesting. This study may provide additional pathway to promote the formation of sulfate in iron-containing aerosols. The manuscript is worthy of publication for ACP after some improvements to the readability.

We appreciate the positive comments and suggestion about the manuscript. We agree with the reviewer's comments, and have updated the manuscript on the basis of these suggestion.

Specific comments

p.4, l.90: The reference of Ito et al. (2019) should be moved to previous sentence after Luo et al. (2008).

Response: The reference of Ito et al. (2019) has been moved to previous sentence after Luo et al. (2008) in line 91.

p.9, l.205: The formula of geometric area should be corrected.

Response: The formula of geometric area has been corrected as $A_{\text{geo}} = \text{mass} \times S_{\text{geo}}$ in line 209.

Table 1: The BET specific surface area is shown in Table S1. The unit and values of the total surface area should be corrected.

Response: Thanks for the reviewer's correction. The unit and values of A_{BET} has been corrected in Table 1.

p.16, l.373 and Figure 3: Is the trend for ATD statistically significant? Please show the statistics.

Response: Although the simulated cloud processing experiment on each mineral was conducted three times to explore the change of Fe speciation after each pH cycle, the SO₂ uptake experiment was carried out only twice. At present, it was a pity that we don't have a trend for ATD statistically significant. Next, we'll do experiments more detailed.

Please discuss the results in subsection 3.2 with those in subsection 3.6 as is described in p.21, l.501. Subsection 3.2 may be moved at the end of section 3.

Response: Thanks for the suggestion. Because Figure 1 in subsection 3.2 demonstrated that the characteristic peaks for the active OH sites and the sulfite/sulfate products on the mineral particles after CP were significantly higher than those on the ones before CP, indicating the higher hygroscopicity and more SO₂ uptake on the particles after CP, which is the most direct evidence that CP could potentially promote the transformation of SO₂ on the particle surfaces. In order to emphasize the results and significance of this article, we tend to put subsection 3.2 at the first of section 3.

Subsection 3.4 may be combined with subsection 3.1.

Response: We agree with the reviewer's comment. We've combined subsection 3.4 with subsection 3.1 in line 273-308. Thus, the name of every subsection in section 3 has been correspondingly changed.

p.18, 1.430: This has been already suggested by previous studies. Please cite references and rephrase the sentence. Subsection 3.5 may be moved to supplementary materials or before subsection 3.2.

Response: We've cited reference (Shi et al., 2009) and rephrased this sentence in line 432-436. "Previous research had indicated that Fe_A increased as a result of the simulated CP (Shi et al., 2009). Herein, we further proposed that the increased fractions of Fe_A could be mostly transformed from the "structural-Fe" in the aluminosilicate phase of the particles during CP, which is in good agreement with the TEM observation."

Subsection 3.5 is one of the most important content to inspect the Fe speciation before and after CP. We want to keep it in the manuscript.

p.20, 1.490: This has been already suggested by previous studies. Please cite references and rephrase the sentence. Subsection 3.6 may be moved to supplementary materials or before subsection 3.2.

Response: Herein, we've cited references and rephrased the sentence in line 494-495. "The fast Fe release was due to the redissolution of the Fe-rich precipitates, which was proposed to be reactive Fe phases (Shi et al., 2009; Shi et al., 2015)."

Subsection 3.6 "The dissolution-precipitation cycle of the mineral Fe during CP" is also one of the most important content to inspect the Fe speciation before and after CP. We tend to keep it in the manuscript.

p.22, 1.531: How did you know the particles were coated by reactive Fe? Please specify the relationship between the higher hygroscopicity and reactive Fe coating. Please show this

evidence or rephrase the sentence.

Response: The dissolution-precipitation cycle of the mineral Fe was happened on the surface of particles. Additionally, the TEM observation confirmed that the nanosized Fe-rich crystal were attached onto the surface of ATD particles. These results help to confirm that the particles after CP were coated by reactive Fe.

To the best of our knowledge, there was no reports about the direct relationship between the higher hygroscopicity and reactive Fe coating. Previous studies have indicated that the reactive Fe could provide more surface hydroxyl species (OH) to participate in chemical reaction (Fu et al., 2007). In the study, the results of DRIFTS experiment demonstrated that the H₂O and OH groups on the surface of mineral particles significantly increased after CP, indicating the higher hygroscopicity of the particles after CP.

Therefore, we've rephrased the sentence in line 535-537. "The particle surfaces after CP were coated by these reactive Fe to provide more surface OH species, resulting in enhanced SO₂ uptake."