

## **Anonymous Referee #1**

The authors focused on the heterogeneous transformation of SO<sub>2</sub> on mineral dust during cloud processing by the simulated experiment. They characterized Fe morphology using the combined methods including TEM, Mossbauer, and CBD extraction method. The iron mobilized from mineral particle was also measured. Generally, heterogeneous transformation on the surface of mineral particle play a vital role on second particle formation. The result shown in this manuscript shed a light on heterogeneous chemistry, and it is helpful to further understand the fog and haze formation in China. Thus, this manuscript made some new contribution to atmosphere chemistry. The manuscript was well written and organized. The topic is of interest to the journal's readers. I thus recommended that this manuscript could be published on ACP. However, the manuscript also suffered from some flaws, and I listed as follows.

Thank the reviewer's chariness and suggestion. 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 suggestions.

### **General questions/comments/suggestions:**

1 The four examined clays were purchased from the clay mineral depository. The author should discuss why they were selected, particularly why they are representative of clays in aerosols.

**Response:** Clay is a more important component of mineral dusts. It was well documented that a long range transport would result in a decrease of quartz relative to the clay fraction because of the more rapid removal of quartz, which has the relatively larger mass median diameter of quartz (Mahowald et al., 2005). Dust deposition modeling showed that the clay fraction becomes more and more dominant on the downwind of the sources from Asia. In the African dust collected over Atlantic Ocean and Mediterranean, the clay group represented between 48 and 82% of the total aerosol mass, illite and kaolinite were the dominant minerals, smectite and chlorite were detected but in low concentrations (Journet et al., 2008). To explore the linkage between iron speciation in the clay minerals and properties of iron solubility, the typical Fe-containing clays, including the Fe-rich nontronite, illite and smectite were selected. Arizona test dust (ATD) is a commercially available material that has also

been widely studied as surrogate in the field of atmospheric chemistry and Fe chemistry.

The sentence has been added in line 55-58. “A long range transport would result in a decrease of quartz relative to the clay fraction because of the more rapid removal of quartz, hence clay is an important component of mineral dusts (Mahowald et al., 2005; Journet et al., 2008).”

The sentence in line 128 has been rewritten as “In this study, we employed four typical Fe-containing mineral samples as surrogates to perform simulated CP experiments.”

2 Line 159: “TEM observation”. The methods of TEM sample preparation will change the aggregation state of such aggregates. Some researchers have extensively worked on this and gave detailed information how they did it. Nothing is reported here on sample preparation. Therefore, please supply the detail about TEM sample.

**Response:** Thanks for the suggestion. We describe the TEM method in detail in line 214-221.

“Suspensions ( $0.2 \text{ g L}^{-1}$ ) of each particle were prepared in methanol and sonicated for at least 1 h. A drop of this suspension was then applied to a carbon-coated Cu TEM grid (400 mesh; EMS) and allowed to air-dry. The operation was conducted in bright field mode at 120 kV. The Fe content of the typical individual mineral particle were calculated from the values of 50 typical particles. To obviously observe the morphological changes, high-resolution TEM (HRTEM) images were also collected to observe nanoscale structural features, e.g., surface roughness and lattice fringes.”

3 The Fe<sub>A</sub> and Fe<sub>D</sub> content of Arizona Test Dust was “ $0.067 \pm 0.005\%$  for Fe<sub>A</sub> and  $0.41 \pm 0.04\%$  for Fe<sub>D</sub>” in Shi et al. (2011), which was not completely in accordance with the values in Figure 4. The author should try to explain the discrepancy of Fe<sub>A</sub> and Fe<sub>D</sub> content.

**Response:** In the published paper of Shi et al., the Fe<sub>A</sub> content ( $0.067 \pm 0.005\%$ ) and Fe<sub>D</sub> content ( $0.41 \pm 0.04\%$ ) were the proportion of Fe<sub>A</sub> and Fe<sub>D</sub> per mass of dust, respectively. The total Fe content (Fe<sub>T</sub>) of Arizona Test Dust (ATD) was 1.48%. The Fe<sub>A</sub>/Fe<sub>T</sub> and Fe<sub>D</sub>/Fe<sub>T</sub> was calculated to be  $4.53 \pm 0.34\%$  and  $27.70 \pm 2.70\%$ , respectively, which were comparable to the values in this study,  $3.8 \pm 0.3\%$  and  $35.5 \pm 3.7\%$ .

Further specific comments/suggestions: In Figure 3, please change the label “2 cycle” and “3 cycle” to “2 cycles” and “3 cycles”, respectively.

**Response:** We’ve changed the label “2 cycle” and “3 cycle” to “2 cycles” and “3 cycles” in Figure 3.

Remove some “the”, such as in Line 151.

**Response:** We’ve removed “the” in line 155.

Line 149, please comment on the rationale of the 1 g/L concentrations used in this experiment.

**Response:** The mineral particle samples are available in large amounts (g-kg) compared to atmospheric dust ( $\mu\text{g-mg}$ ). These samples are clearly the precursor of atmospheric dusts prior to uplifting and thus have not been subject to changes that can happen to mineral dust in the atmosphere, allowing them to be used to investigate how these properties are modified during simulated atmospheric processing. Previous studies examined the iron dissolution over the range of dust loadings from 0.05 to 5 g/L in solutions. The solution with 1 g/L is usually representative for a cloud water solution.

Table 1 can be reported by both confirmation of sulfate ions origin (uptake vs  $\text{H}_2\text{SO}_4$ ) and uptake coefficient (before vs after cloud processing).

**Response:** The IC measurements on sulfate ions released from mineral surface after CP were performed as blank experiments. The sulfate formation rates and uptake coefficients of  $\text{SO}_2$  on particle samples after CP in Table 1 were recalculated by deduction of the blank value.

Line 381: Please replace “were” by “was”.

**Response:** The “were” has been replaced by “was” in line 412.

Line 376-377: This sentence contains partly the same data with the above sentences.

**Response:** Thanks for the reviewer’s suggestion. This sentence has been deleted in the revised manuscript.

Line 539: Please replace “during” by “by”.

**Response:** The “during” has been replaced by “by” in line 543.

Line 550-552: Is this a new result? or already known from other studies (then provide proper references)?

**Response:** This is a new result of our study.

In the References, Please exchange “Global Biogeochem, Cycles” by “Global Biogeochem. Cycles”.

**Response:** We’ve replaced “Global Biogeochem, Cycles” by “Global Biogeochem. Cycles”.