

## ***Interactive comment on “Iron dissolution kinetics of mineral dust at low pH during simulated atmospheric processing” by Z. Shi et al.***

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Comment: (1) Page 26609, lines 25–28. "or measured in fine marine aerosols ..." seems to be irrelevant to the acidity of aqueous solution coating dust particles. These aerosol and cloud observations do not establish the acidity of dust particles.

Response: This part has been removed.

Comment: (2) Page 26610, line 10. "Measures et al, 2010" is missing from the reference.

Response: This reference has now been added.

Comment: (3) Page 26610, line 19. Is the iron dissolution kinetics "zero-order" or "first

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order"?

Response: These models quoted in the text assumed "zero order" iron dissolution kinetics. The results in our study showed that the iron dissolution does not follow zero-order dissolution kinetics but is better described by first order kinetics.

Comment: (4) Page 26612, line 11. Do you mean "Table 1"?

Response: Yes. It is table 1. We have changed the text accordingly.

Comment: (5) Page 26620, lines 21–23. It is not clear what it is meant by "is much faster" and by "much larger reactive surface area". Fig. 3 shows that a smaller iron dissolution rate coefficient is obtained at a higher load of dust suspension in the solution. At a dust loading of 1000 mg/L, Fe(III) concentrations resulting from dissolution of FeT (total reducible Fe) are below the equilibrium solubility of ferrihydrite and nanogoethite and near the equilibrium solubility of hematite shown in Fig. 7. Combine results shown in Fig. 3 and Fig. 7, could you conclude that the smaller rate coefficient at higher dust loading is caused by Fe(III) concentration exceeding the solubility of hematite (or the largest crystals of hematite suspended in the solution)? Alternatively, the results shown in Fig. 3 are indicating slower detachment of Fe(III) from the mineral surface when Fe(III) concentration is high in the aqueous phase?

Response: We have changed this sentence to "In terms of dissolved Fe concentration (in  $\mu\text{mol L}^{-1}$ ), it is much higher in a 1000 mg L<sup>-1</sup> dust suspension than 60 mg L<sup>-1</sup> and a fortiori 10 mg L<sup>-1</sup> suspensions at a particular time".

We believe that the major reason for the smaller apparent rate coefficient at higher dust loading in Fig. 3 is caused by Fe(III) concentration exceeding the solubility of hematite (or the largest crystals of hematite suspended in the solution), leading to cessation of the dissolution of these minerals. But we cannot rule out the possibility that higher Fe concentration in the aqueous phase would slow down the detachment of Fe(III) from the mineral surface. To fully address the importance of the later effect, further long-term

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experiments on Fe dissolution kinetics at high dust/liquid ratios would be required.

Comment: (6) Page 26623, line 7. Perhaps the "slow" iron pool represents crystalline Fe oxides and a fraction of Fe-containing clay minerals? Lafon et al. (2004) suggest that iron contained in clay minerals is not "free". However, previous measurements show clay minerals also dissolve at low pH but at much slower rates,  $< 1 \times 10^{-12} \text{ mol m}^{-2} \text{ s}^{-1}$ , than hematite and goethite (Ganor et al., 1995; Köhler et al., 2003; Amram and Ganor, 2005).

Response: We agree that the "slow" iron pool probably represents crystalline Fe oxides and a fraction of Fe-containing clay minerals. Iron contained in clay minerals was defined as "structural Fe" and was assumed to be not "free" (Lafon et al., 2004). However, this terminological description does not mean that Fe in clay minerals cannot dissolve at low pH as the references listed in the comments showed. Our dissolution of "cleaned" illite at pH 2 showed that it had a dissolution rate constant (first order) close to pool III described in this study.

We have responded to this comment by adding a sentence "This agrees with the results that clay minerals like smectite, illite and kaolinite dissolve at low pH conditions although at a slow rate, i.e.,  $< 1 \times 10^{-12} \text{ mol m}^{-2} \text{ s}^{-1}$  (Ganor et al., 1995; Köhler et al., 2003; Amram and Ganor, 2005)" after the sentence "This suggests that illite and potentially other clay minerals may be an important component of the "slow" Fe pool." (line Page 26623, line 7)

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