Amended responses to the Review comments by Reviewer 2

After email contact and two long phone discussions with Nicholas Meskhidze from University of North Carolina who wrote one of the first papers on modelling Fe solubilisation in dust (Meskhidze et al., 2005), I have clarified some of the issues. As a result, we have made small changes to the manuscript and we amended our responses to the reviewer comments by Reviewer 2 as well.

Reviewer comment 8:
p. 26624, l.1: Fe dissolution calculated from rate constants used in Meskhidze et al. depends on pH, so the figure should be separated into two (i.e., pH 1 and pH 2). Please show all variables (i.e., K, T, a(H+), f(_G), and W) in the calculation of the Fe dissolution using rate constants in Meskhidze et al., which may correspond to some portions of the slow Fe pool. Presumably, you used the rate constant for the first 0-0.8% of the total Fe in the dust, but it should depend on the total amount of the Fe dissolved. The modelling studies also assume an extractable Fe pool, which may correspond to FeA. What if the three-stage kinetic process is considered for specification of the dissolution constant (Meskhidze et al., 2005), using FeA as the initial condition for the soluble iron fraction?

Also, what if the dissolution rate of illite is used for the dissolution (Ito and Feng, 2010), using FeA as the initial condition for the soluble iron fraction?

Response: In our original response, we stated that "Using the parameters from Meskhidze et al. (2005), less than 0.8% of Fe would be dissolved in 72 hours and therefore for this figure, only the first stage dissolution rate was used." We re-considered the reviewer comments. Although Meskhidze et al. (2005) did not consider an initially soluble Fe pool, their later paper (Solmon et al., 2009) did consider an initial Fe pool. We therefore assumed that there is an initially soluble Fe pool (equals to FeA) and used the second stage dissolution rate for hematite as recommended by the reviewer and Nicholas Meskhidze in final Fig. 8. We have clearly stated this in the figure caption. This change resulted in slightly more Fe release. However, this does not change the interpretation of figure 8.

Comment: p. 26624, l.10: If you mean the effect of the solution saturation state on dissolution rates (p. 26621), the model uses a function of Gibbs free energy change of a particular mineral dissolution reaction. However, it is unlikely to reach true thermodynamic equilibrium in the atmosphere, as you mentioned in p. 26611. Could you specify which effect was not considered in the model?

Response: After clarification with Nicholas Meskhidze, I confirmed that Meskhidze et al. (2005) did not assume f(ΔGr) to be 1 as what Ito and Feng (2010) did. Therefore Meskhidze et al. (2005) and Solmon et al. (2009) did consider solution saturation effect. In our revised manuscript, we have changed the sentence to: "***Finally, the parameterization in some of the models (e.g., Fan et al., 2006; Ito and Feng, 2010) may also lead to an overestimation at later stages of***

dissolution (e.g., >40 h, Fig. 8). Therefore, it is important to consider the solution saturation effect in dust aerosol water, e.g., by applying a function of Gibbs free energy, ƒ(ΔGr) in the models as did by Meskhidze et al. (2005) and Solmon et al. (2009). "

This revision does not change the interpretation of figure 8 because at the low dust/liquid ratio (60 mg L-1) used for comparison in this figure), f is close to 1 (>0.99) at the first 72 h of dissolution.

Furthermore, in responses to these comments and those from Nicholas Meskhidze, we have now revised the first paragraph of section 3.6 with added explanations why the predicted Fe concentrations based on Meskhidze et al. (2005) and Solmon et al. (2009)'s parameterizations are so low compared to our experimental data. The paragraph has now been changed to:

Several modelling studies have simulated the acidic processing of mineral dust in transforming insoluble Fe into labile Fe (e.g., Meskhidze et al., 2005; Luo et al., 2005; Fan et al., 2006; Solmon et al., 2009). These authors assumed that Fe existed only as hematite and applied one or a series of dissolution rates measured on laboratory-made or commercial ferric oxides. Our results demonstrated that the Fe dissolution in two representative dust samples (Saharan and Asian) did not follow such dissolution behaviour (Fig. 1-3, Fig. 8). In particular the selected slow dissolution rate for the first 0-0.8% compared to that for the 0.8-40% of hematite dissolved used in some atmospheric models (Meskhidze et al., 2005; Luo et al., 2005; Solmon et al., 2009; Ito and Feng, 2010) was in contrast to our data (e.g., Fig. 1) as well as others (Spokes et al., 1994, 1996; Desboeufs et al., 1999, 2005; Mackie et al., 2005; Cwiertny et al., 2008; Fu et al. 2010; Deguillaume et al. 2010). We attribute this as the dissolution of highly reactive first Fe pool. In addition to this, there are at least two additional reasons that lead to the significant under-estimation in the beginning of the Fe dissolution (i.e., <~40 h, Fig. 8): (i) The parameterizations adopted by the present models were based on experiments on laboratory-made or commercial ferric oxides particles (e.g., Azuma and Kametani, 1964; Zinder et al., 1986), which are fundamentally different from those in natural dust particles in terms of size distribution, surface area, crystallnity, and purity (Shi et al., 2010 and reference therein); and (ii) the choice of hematite as the only Fe mineral could not represent the complex Fe mineralogy (amorphous and poorly crystalline Fe, hematite, goethite, and clay minerals) in natural dust (e.g., Lafon et al., 2006; Mackie et al., 2008; Shi et al., 2010)…..