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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Change of iron species and iron solubility in Asian dust during the long-range transport from western China to Japan" *by* Y. Takahashi et al.

Anonymous Referee #1

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General Comments:

This paper presents interesting results on the relationship between iron solubility and speciation of Asian dust during long-range transport, specifically from an arid region in western China to eastern China and Japan. In particular, the authors used a combination of advanced analytical tools to show unique changes in iron speciation of Asian dust during atmospheric transport as a result of anthropogenic chemical processing. These changes appeared to be related to iron solubility. The results of this study build on similar results observed in laboratory experiments (Shi et. al., 2009) and will ultimately improve scientific understanding of bioavailable iron dust deposition to the

ocean for phytoplankton productivity. Overall, these results are well constructed, and the manuscript is easy to follow. I recommend this paper to be published in ACP with revisions listed below.

Specific comments:

Although the results of this paper are undoubtedly profound, the authors tend to overstate the significance of their findings. For instance, the estimated difference between ferrihydrite content in Asku (desert) and Tsukuba (Japan) samples is 10±5% and 24±5%, respectively, which is attributed to a ∼7% increase in iron aerosol solubility (in rainwater, Table 1). This increase in iron solubility of Asian dust due to anthropogenic chemical processing is minor compared to results reported in other studies (1-80% increase) (Mahowald et. al., 2005). Therefore, it may be difficult to argue that changes in iron speciation of Asian dust during long-range transport are a "significant" source of bioavailable iron to the ocean. The authors should ultimately tone down the conclusions concerning the significance of this mechanism throughout the manuscript.

Abstract and conclusion: Quantitative iron solubility and clay mineral and ferrihydite content should be reported in both sections.

Pg. 19551, Methods, The authors use XAFS techniques to probe iron mineralogy of dust. In the method section it is specifically noted: "The samples appearing on the filter as dark spots (spot size: 0.5-2mm) were directly exposed to the incident X-ray beam." Throughout the manuscript, it is not clear whether the authors took several "single particle" measurements of iron mineralogy or bulk iron mineralogy using XAFS techniques. If the former, what did the authors do to ensure that single particle XAFS spectra were representative of the entire sample?

Pg. 19552, Line 1-5, In this study, XAFS energy spectra were fit to a number of iron standard spectra to determine mineralogy. A few details on XAFS fitting were left out of the discussion paper. For example, what XAFS analysis software was used for linear combination fittings? Did the authors do an initial principal component analysis

(PCA) to infer which standards were most appropriate for the linear combination fitting analysis? A table including the PCA and multiple LCF results (with R values) would greatly support their XAFS fits.

Pg. 19558, Line 12-15, Can you clarify why physical weathering decreases the Fe(III) oxide content?

Pg 19559, Line 12-14, The authors mention that the dissolution rate of chlorite is far greater than that of illite at pH 4. However, aerosol acidity appears to be mostly basic (pH ∼9 in Asku) to neutral (pH ∼6 in Takusba) during transport. As most iron minerals have variable solubility properties at different pH values, there may be reported differences in solubility properties of chlorite reported at pH 6. If so, what is the expected amount of chlorite that will dissolve at the pH 6? A short discussion on the amount of chlorite that will dissolve and subsequently form ferrihydrite under these atmospheric conditions (e.g. aerosol acidity and atmospheric transport time) would be useful.

Pg. 19560, Line 20-23. Once again, a principal component analysis coupled to linear combination fits would support your CEY-XAFS results. A table either in the manuscript or supplement showing these results would support their results.

Pg 19561, Line 1-13. This argument needs some clarification. It is not clear why these results point to formation of ferrihydrite nanoparticles. Please add a few sentences to further explain this argument.

Technical comments:

Pg. 19548, Line 11, Change to 2.1. Sample collection and characterization

Pg. 19560, Line 16, Change to (ii) alteration of chlorite

Figure 1 and Figure 9, Increase the font size for axis label and legend

References:

Mahowald, N., Baker, A.R., Bergametti, G., Brooks, N., Duce, R.A., Jickells, T.D., Kubi-

lay, N., Prospero, J.M. and Tegen, I.: Atmospheric global dust cycle and iron inputs to the ocean. Glob. Biogeochem. Cycle. 19, GB4025, 2005.

Shi, Z., Krom, M.D. and Bonneville, S.: Formation of iron nanoparticles and increase in iron reactivity in mineral dust during simulated cloud processing. Environ. Sci. Technol. 43, 6592-6596, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 19545, 2011.