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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 #2**

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The authors use XANES, EXAFS, XRD, ICP-AES, ICP-MS among other analytical techniques to characterize atmospheric aerosols of predominantly mineral composition along a Lagrangian trajectory in Asia and report on the change in chemical composition and solubility along this path. The ambient aerosol samples are not only characterized for chemical composition but also subject to extraction and dissolution experiments in the laboratory, which are ultimate properties desired for assessment of ecosystem impacts. The results of this manuscript is important to the atmospheric science community and should proceed to publication after these comments are addressed.

1. While the suite of analytical methods applied are impressive, the sample size is



small. The conclusions, relevance, and implications of this study are worded too strongly for statements based on a few days worth of observations.

2. It may be advised that the abstract and conclusions report the quantitative changes in solubility of aerosols observed under the specific conditions studied here, as the amount of change is equally as relevant as the mechanism.

3. One important conclusion from this manuscript is the relationship between composition, transformation in composition, and solubility, which the authors have addressed using a combination of measurements in the Lagrangian configuration. Much of the past dicussion on composition and solubility of ambient iron aerosols have focused on measurement of iron oxidation state (FeII vs. FeIII) to characterize iron speciation (e.g., Zuang et al. 1992, Luo et al. 2005, Majestic et al. 2007, Takahama et al., 2008), or direct measurement of solubility (e.g., Zhu et al. 1997). Could the authors place comment on the solubility implied from (many) past studies where iron solubility was inferred from measured composition and vice versa? Given the interest in the community to predict iron solubility/bioavailability from aerosol composition measurements, this manuscript can provide guidance on the important metrics that should be characterized to further refine our capability for solubility prediction.

Baker, A. R., T. D. Jickells, M. Witt, and K. L. Linge (2006). Trends in the solubility of iron, aluminium, manganese and phosphorus in aerosol collected over the Atlantic ocean, Mar. Chem., 98(1), 43– 58.

Luo, C., N. M. Mahowald, N. Meskhidze, Y. Chen, R. L. Siefert, A. R. Baker, and A. M. Johansen (2005). Estimation of iron solubility from observations and a global aerosol model, *J. Geophys. Res.*, 110, D23307, doi:10.1029/2005JD006059.

Majestic, B. J., J. J. Schauer, and M. M. Shafer (2007). Application of synchrotron radiation for measurement of iron red-ox speciation in atmospherically processed aerosols, *Atmos. Chem. Phys.*, 7(10), 2475– 2487.

Takahama, S., S. Gilardoni, and L. M. Russell (2008). Single-particle oxidation state and morphology of atmospheric iron aerosols, *J. Geophys. Res.*, 113, D22202, doi:10.1029/2008JD009810.

Zhu, X. R., J. M. Prospero, and F. J. Millero (1997). Diel variability of soluble Fe(II) and soluble total Fe in North African dust in the trade winds at Barbados, *J. Geophys. Res.*, 102(D17), 21,297– 21,305.

Zhuang, G. S., Z. Yi, R. A. Duce, and P. R. Brown (1992), Link between iron and sulfur cycles suggested by detection of Fe(II) in remote marine aerosols, *Nature*, 355(6360), 537–539.

4. The authors make assumptions which are perhaps not entirely incorrect, but lack a priori rationalization in the manuscript. For instance, supermicron particles are called "mineral aerosols" – such statements should be justified if this equivalence in terminology is to be assumed.

5. The spectra used as regressors in the XANES LCF appear to be highly collinear, in which case the estimates of their contribution may come with substantial error (and possibly fail to support differentiation among reported composition by tests of statistical significance). Could the authors comment on this observation.

6. Some paragraphs of section 3.2 are written poorly with respect to English usage, while the presentation of the rest of the manuscript is reasonably good.



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