**Atmospheric
Chemistry
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Discussions**

Interactive
Comment

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.

Y. Takahashi et al.

ytakaha@hiroshima-u.ac.jp

Replies to the comments kindly given by the Referee #2

We thank the referee for the constructive comments. Our replies to the comments are listed below.

All the corrections are shown in the pdf file uploaded as a supplement.

=== Comment 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.

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Reply 1: This is similar comment to the comment 1 by Referee #1. We toned down the effect of the mechanism in this study as shown in our reply 1 to the comment 1 by referee #1.

=== Comment 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.

Reply 2: Quantitative data were added to the abstract and conclusion as shown in our reply to the comment 2 by referee #1.

=== Comment 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 mea-surements, this manuscript can provide guidance on the important metrics that should be characterized to further refine our capability for solubility prediction.

Reply 3: As suggested, some previous studies only gave Fe(II)/Fe(III) ratio, which was used to discuss the reactions in the atmosphere and their solubilities in water. Based on more detailed Fe speciation and solubility experiments suggest that oxidation into Fe(III) can increase the solubility of Fe, whereas previous studies (Zhuang et al., 1992; Zhu et al., 1997) assumed that Fe(II) is much more soluble that Fe(III) species. However, Luo et al. (2005) suggested that there is not clear correlation between Fe(II) fraction and iron solubility, while Majestic et al. (2007) reported that Fe(III) contributed

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to a large fraction of soluble Fe. Our results also suggested that the oxidation of Fe(II) (mainly in chlorite) to Fe(III) as ferrihydrite can increase the Fe solubility. Thus, more information on Fe speciation data not limited to Fe(II)/Fe(III) ratio is needed to discuss the relationship between Fe species and Fe solubility in seawater.

New sentences added in Section 3.5: Some previous studies only gave Fe(II)/Fe(III) ratio (Zhuang et al., 1992; Zhu et al., 1997; Luo et al., 2005; Majectic et al., 2007), which was used to discuss the reactions in the atmosphere and their solubilities in water. Based on more detailed Fe speciation and solubility experiments in this study suggest that oxidation into Fe(III) can increase the solubility of Fe, whereas previous studies (Zhuang et al., 1992; Zhu et al., 1997) assumed that Fe(II) is much more soluble that Fe(III) species. However, Luo et al. (2005) suggested that there is not clear correlation between Fe(II) fraction and iron solubility, while Majestic et al. (2007) reported that Fe(III) contributed to a large fraction of soluble Fe. Our results also suggested that the oxidation of Fe(II) (mainly in chlorite) to Fe(III) as ferrihydrite can increase the Fe solubility, probably because Fe(II) in clay minerals is incorporated in the aluminosilicate structures. Thus, more information on Fe speciation data not limited to Fe(II)/Fe(III) ratio is needed to discuss more clearly the relationship between Fe species and Fe solubility in seawater.

=== Comment 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.

Reply 4: The discussion below was added into Section 3.1.

Revision (Section 3.1): There can be various particles classified into the coarse particle fractions such as sea salt, volcanic ash, and plant particle in addition to mineral aerosols (Finlayson-Pitts and Pitts, 1999). Due to the very large scale of the dust event in the period, it is most likely that contribution of aerosols from other sources can be

minor for iron supply compared with the mineral dust originating from western China.

=== Comment 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.

Reply 5: This comment is similar to the comments 3 and 7 by the referee #1. PCA analysis and fitting procedures were added, showing that the three components are needed to explain the spectra. All the fractions determined by the fitting were shown in Table S1 with R values.

=== Comment 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.

Reply 6: The English of the section was corrected by native English speaker.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C10764/2011/acpd-11-C10764-2011-supplement.pdf

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