Atmos. Chem. Phys. Discuss., 13, C12304–C12311, 2014
www.atmos-chem-phys-discuss.net/13/C12304/2014/

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Response of acid mobilization of iron-containing mineral dust to improvement of air quality projected in the future" *by* A. Ito and L. Xu

A. Ito and L. Xu

akinorii@jamstec.go.jp

Received and published: 17 February 2014

We would like to thank the reviewer for his/her constructive comments for the improvement of our manuscript. Our responses to the comments from Reviewer 1 are included in the followings.

Major Comments

Comment: 1. Page 28175 line 26-30. If the authors choose to evaluate other model's SO2 uptake rates then the authors should provide an in depth evaluation of this topic.

Reply: We do not intend to evaluate other model's SO2 uptake in our paper, but to explain the differences from previous modeling approach. This is reflected in the revised paper, p.4, l.65.

A chemical transport model that implemented an explicit iron dissolution scheme of hematite in dust aerosols suggests that doubling SO2 emissions can induce a significant increase in acid mobilization of iron and deposition to the remote Pacific (13% on average) (Meskhidze et al., 2005; Solmon et al., 2009). In our atmospheric chemistry transport model simulations, the inclusion of the alkaline minerals (e.g., CaCO3 and MgCO3) in aqueous chemistry substantially limits the iron dissolution of hematite in dust aerosols during the long-range transport to the North Pacific Ocean (Ito and Feng, 2010).

Comment: 2. Page 28180 line 22-25. The total amount of dust emitted is one of the controlling factors on the magnitude of filterable/soluble Fe deposited to the global oceans, therefore, the authors should consider stating what dust emission and source schemes are used in the IMPACT model.

Reply: A thorough description of the dust emission treatments is available in the Ito et al. (2012) paper and its auxiliary material. Briefly, this is stated in the revised paper, p.11, l.200.

The size-resolved dust emission scheme is based on that developed by Ginoux et al. (2001) and is revised using the size distribution at emission of Kok (2011).

Comment: 3. Page 28182 line 25-27. Is dust aerosol pH calculated separately for each size bin? If so does the model calculate aerosol thermodynamics, heterogeneous chemistry, mineralogy, etc. separately for each size bin? Can the authors please clarify this at this point in the manuscript?

Reply: The dust aerosol pH is calculated separately for each size bin. The model calculates thermodynamic equilibrium and heterogeneous chemistry for each size bin separately. The model carries separate mineralogical tracers for each size bin. This is stated in the revised paper, p.12, l.212, p.13, l.234, and p.15, l.281.

Aerosol pH is calculated from the internal particle composition (H+ and H2O) for each size bin by the thermodynamic equilibrium module (Jacobson, 1999). The heterogeneous uptake of nitrate (NO3–) and ammonium (NH4+) by each aerosol for each size bin is interactively simulated in the model following a hybrid dynamical approach (Feng and Penner, 2007).

We use the same effective mineral fractions for all sizes of aerosol particles, but these species (e.g., illite, smectite, calcite, hematite, and gypsum) are represented by specific tracers for each size bin. Since the dust pH is calculated for each bin separately, the dissolution rates are different among different size bins of mineral dust.

Comment: 4. Page 28190. In the paragraph discussing the emission changes between present day and the year 2100 (using the RCP4.5 emissions) can the authors show maps of emission or column concentration changes of the main pollutants? The authors do a good job describing the ratios between emission from present day and 2100 but I feel maps/figures would add to this discussions.

Reply: We show maps of the emission changes in a new Figure S1. This is stated in the revised paper, p.25, l.475.

The ratios of the emissions from fossil fuel combustion in 2100 to 2000 were 0.19 for SO2 (Figure S1a), 0.43 for particulate organic matter, and 0.48 for black carbon in RCP4.5. The ratios of total emissions in 2100 to 2000 were 0.64 for NO2 (Figure S1b) and 1.1 for NH3 (Figure S1c).

Comment: 5. When reading the manuscript it becomes difficult to remember differences between Scenarios 1, 2, and 3. Could the authors add a simple table (similar to Table 4) which reminds the reader what the differences are between these three scenarios?

Reply: The summary of emission scenarios is shown in a new Table 5.

Comment: 6. Page 28192 Section 3.1. Could the authors provide a value of the annual total dust emitted globally and compare that to other commonly used models (see Huneeus et al. [2011])? This would assist the reader in understanding if the dust emissions in the model are comparable to other models.

Reply: The total dust emission is stated in the revised paper, p.28, l.530.

The total emission rate of global mineral dust (2480 Tg yr–1 from Exp5) is higher than that of the median (1123 Tg yr–1) used in other simulations (Huneeus et al., 2011), partly due to different dust size distributions adopted in the emission mechanism (Ito et al., 2012).

Comment: 7. Figure 6. Can the authors add a plot (d) that shows the spatial distribution of the model predicted filterable (bioavailable) Fe percentage ((bioavailable Fe / total Fe) x 100) upon deposition and discuss this in the manuscript? This would be useful to the reader so the model can be compared to other bioavailable Fe model predictions.

Reply: We show the model-predicted (filterable Fe / total Fe) (%) in deposited mineral dust (Figure 6(d) is now Figure 7(d)). When the reader compares bioavailable Fe model predictions, one should pay attention to different definitions of potentially bioavailable iron between different models. This is stated in the revised paper, p.33, l.635.

To quantify the importance of the atmospheric chemical transformation of iron in mineral aerosols, we show the model-predicted (filterable Fe / total Fe) (%) in deposited mineral dust (Fig. 7d). In general, Fe solubility values remain low near the dust source regions but increase downwind as acidic trace gases enhance iron mobilization. The high Fe solubility values are predicted over the regions characterized by low concentrations of dust and high amounts of anthropogenic pollution in the Northern Hemisphere. On the other hand, Fe solubility values remain low over regions in the Southern Hemisphere. Our iron solubility for mineral dust is generally higher than the dissolved Fe fraction for hematite in dust aerosols predicted by Johnson and Meskhidze (2013), which partly reflects the different definitions of potentially bioavailable iron between filterable iron and dissolved iron.

Comment: 8. Figure 7. Could the authors increase the contouring in this figure? The authors state that due to improved air quality in the future that an increased amount of filterable Fe will be deposited to the northeastern Pacific Ocean, however, on Fig. 7d it is hard to see this. The ratio values of 1.0 to 1.1 have the same color value which greatly reduces the information that can be seen in this figure. The same comment applies to Fig. 7c also. 9. Same comment as #8 but for Fig. 8a.

Reply: The contouring is revised. We mean "due to continuing growth in global shipping and no regulations regarding particles emissions over the open ocean" by "due to improved air quality for human health projected in the future". The sentence is revised in p.36, l. 682, as follows.

Consequently, the improvement of air quality projected in the future will lead to a decrease of the total filterable iron deposition to the northwestern Pacific from 5.2 to 4.7 Gg yr–1 (–10%) due to less acidification in Asian dust (Fig. S2). At the same time, continuing growth in global shipping and no regulations regarding particles emissions over the open ocean led to the increase of the total filterable iron deposition to the northeastern Pacific from 2.7 to 3.0 Gg yr–1 (11%) (Ito, 2013).

Comment: 10. Page 28197 line 13-15. The authors state that the solution saturation state or equilibrium has an important effect on the decrease in readily released Fe. Could the authors produce and present plots with show the spatial distribution of dust aerosol pH (in the fine and coarse fraction if possible) for the present day and Scenario 1. This model-predicted variable will have a large influence on soluble Fe(III) equilibrium and I think these plots would be very beneficial to this portion of the manuscript and the discussions on page 28197.

Reply: We show the spatial distribution of pH weighted by dust load for fine (radius: <1.25 μm) and coarse fraction (radius: 1.25–10 μm) in a new FigureS2 for the present day and the ratio of (Scenario 1)/(Present). This is stated in the revised paper, p.34,

l.645 and p.36, l.682.

Our modeled Fe solubility values are highly dependent on the predicted pH of mineral aerosols (Fig. S2). In our model, the aerosol pH in the coarse particles (radius: 1.25–10 μm) is higher than that in the fine particles (radius: <1.25 μm).

Consequently, the improvement of air quality projected in the future will lead to a decrease of the total filterable iron deposition to the northwestern Pacific from 5.2 to 4.7 Gg yr–1 (–10%) due to less acidification in Asian dust (Fig. S2).

Minor Comments Comment: 1. Page 28174 line 23. The authors state that small particles are a key source of bioavailable Fe. Do the authors mean dust particles in the smaller size fractions or just any small particle? Please revise to clarify.

Reply: This is corrected in p.3, l.32.

Mineral dust and combustion aerosols are a key external source of bioavailable iron to surface waters in open oceans.

Comment: 2. Page 28174 line 24-26. The authors state that model estimations of dust aerosols are consistent with available observations. There are many differing dust models displaying a wide range in total dust-Fe deposition to the global oceans. Some of these models predict dust-Fe deposition rates which compare well to in situ data while some models have very poor comparisons. The authors need to remove this statement or expand upon it to clarify.

Reply: This is removed.

Comment: 3. Page 28176 line 19. Missing period.

Reply: This is correct in our uploading file. We will check it when printed.

Comment: 4. Page 28184 line 24. Missing period.

Reply: This is correct in our uploading file. We will check it when printed.

References

Feng, Y. and Penner, J. E.: Global modeling of nitrate and ammonium: Interaction of aerosols and tropospheric chemistry, J. Geophys. Res., 112, D01304, doi:10.1029/2005JD006404, 2007.

Ginoux, P., Chin, M., Tegen, I., Prospero, J. M., Holben, B., Dubovik, O., and Lin, S.-J.: Sources and distributions of dust aerosols simulated with the GOCART model, J. Geophys. Res., 106, 20255–20273, 2001.

Huneeus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Prospero, J., Kinne, S., Bauer, S., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Fillmore, D., Ghan, S., Ginoux, P., Grini, A., Horowitz, L., Koch, D., Krol, M. C., Landing, W., Liu, X., Mahowald, N., Miller, R., Morcrette, J.-J., Myhre, G., Penner, J., Perlwitz, J., Stier, P., Takemura, T., and Zender, C. S.: Global dust model intercomparison in AeroCom phase I, Atmos. Chem. Phys., 11, 7781-7816, doi:10.5194/acp-11-7781-2011, 2011.

Ito, A., Kok, J., Feng, Y., and Penner, J.: Does a theoretical estimation of the dust size distribution at emission suggest more bioavailable iron deposition?, Geophys. Res. Lett., 39, L05807, doi:10.1029/2011GL050455, 2012.

Jacobson, M. Z.: Studying the effects of calcium and magnesium on size-distributed nitrate and ammonium with EQUISOLV II, Atmos. Environ., 33, 3635–3649, 1999.

Johnson, M. S., and Meskhidze, N.: Atmospheric dissolved iron deposition to the global oceans: effects of oxalate-promoted Fe dissolution, photochemical redox cycling, and dust mineralogy, Geosci. Model Dev., 6, 1137–1155, 2013.

Kok, J. F.: A scaling theory for the size distribution of emitted dust aerosols suggests climate models underestimate the size of the global dust cycle, P. Natl. Acad. Sci. USA, 108, 1016–1021, doi:10.1073/pnas.1014798108, 2011.

Meskhidze, N., Chameides, W. L., and Nenes, A.: Dust and pollution: A recipe for enhanced ocean fertilization?, J. Geophys. Res., 110, D03301,

doi:10.1029/2004JD005082, 2005.

Solmon, F., Chuang, P. Y., Meskhidze, N., and Chen, Y.: Acidic processing of mineral dust iron by anthropogenic compounds over the north Pacific Ocean, J. Geophys. Res., 114, D02305, doi:10.1029/2008JD010417, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 28173, 2013.