

Review of “Response of acid mobilization of iron-containing mineral dust to improvement of air quality projected in the future” by Ito and Xu, 2013

The manuscript by *Ito and Xu*, [2013] focuses on the detailed model simulations of how improved air quality conditions in the future (reduced acidic trace gas emissions) impact filterable/bioavailable iron (Fe) deposition rates to the Pacific Ocean. The authors spend the majority of the manuscript describing the dust mineralogy and aerosol chemistry in the chemical transport model and its comparison to in situ measurement data. Finally, the manuscript concludes with a relatively short section demonstrating the impacts of future air quality on mineral dust and filterable Fe deposition to the Pacific Ocean.

Overall, I think the manuscript is well written and presents interesting results on an important scientific topic. I feel the authors spend too much time discussing the model and spatial distributions of emissions and deposition ratios of each aerosol Fe specie compared to how future air quality impacts filterable Fe deposition to the oceans, however, with the revisions suggested below I believe this study should be published in Atmospheric Chemistry and Physics (ACP).

Major Comments

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. Past research has shown that measured SO₂ uptake rates are highly variable and dependent on experimental setups. For example some studies would suggest that SO₂ uptake rates on dust are better represented by studies that use authentic mineral dust [e.g., *Ullerstam et al.*, 2002, 2003; *Usher et al.*, 2002, 2003; *Adams et al.*, 2005; *Seisel et al.*, 2006; *Zhang et al.*, 2006] while others use mineral dust proxies (such as calcium carbonate (CaCO₃) powder) [e.g., *Al-Hosney and Grassian*, 2005; *Preszler-Prince et al.*, 2007]. If the authors choose to use statements such as this please review past literature and experimental methods and present a thorough analysis of SO₂ uptake rates used in dust-Fe chemistry models.
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.
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?
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.

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?

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.

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.

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.

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.

Minor Comments

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.

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.

3. Page 28176 line 19. Missing period.

4. Page 28184 line 24. Missing period.

References

Adams, J. W., D. Rodriguez, and R. A. Cox (2005), The uptake of SO₂ on Saharan dust: a flow tube study, *Atmos. Chem. Phys.*, 5, 2679-2689.

Al-Hosney, H. A., and V. H. Grassian (2005), Water, sulfur dioxide and nitric acid adsorption on calcium carbonate: A transmission and ATR-FTIR study, *Chem. Phys.*, 7, 1266-1276.

Huneeus, N., et al. (2011), Global dust model intercomparison in AeroCom phase I, *Atmos. Chem. Phys.*, 11, 7781-7816, doi:10.5194/acp-11-7781-2011.

Preszler Prince, A., P. Kleiber, V. H. Grassian, and M. A. Young (2007), Heterogeneous interactions of calcite aerosol with sulfur dioxide and sulfur dioxide nitric acid mixtures, *Chem. Phys.*, 9, 3432-3439.

Seisel, S., T. Thorsten, Y. Lian, and R. Zellner (2006), Kinetics of the uptake of SO₂ on mineral oxides: Improved initial uptake coefficients at 298 K from pulsed Knudsen cell experiments, *International Journal of Chemical Kinetics*, 38, 242-249.

Ullerstam, M., R. Vogt, S. Langer, and E. Ljungstrom (2002), The kinetics and mechanism of SO₂ oxidation by O₃ on mineral dust, *Phys. Chem.*, 4, 4694-4699.

Ullerstam, M., M. S. Johnson, R. Vogt, E. and Ljungstrom (2003), DRIFTS and Knudsen cell study of the heterogeneous reactivity of SO₂ and NO₂ on mineral dust, *Atmos. Chem. Phys.*, 3, 2043-2051.

Usher, C. R., H. Al-Hosney, S. Carlos-Cuellar, and V. H. Grassian (2002), A laboratory study of the heterogeneous uptake and oxidation of sulfur dioxide on mineral dust particles, *J. Geophys. Res.*, 107.

Usher, C. R., A. E. Michel, and V. H. Grassian (2003) Reactions on mineral dust, *Chem. Rev.*, 103, 4883-4939.

Zhang, X., G. Zhuang, J. Chen, Y. Wang, Z. An, and P. Zhang (2006), Heterogeneous Reactions of Sulfur Dioxide on Typical Mineral Particles, *J. Phys. Chem.*, 110, 12588-12596.