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Interactive comment on "Influence of measurement uncertainties on soluble aerosol iron over the oceans" *by* N. Meskhidze et al.

Anonymous Referee #1

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Influence of measurement uncertainties on soluble aerosol iron over the oceans N. Meskhidze, M. S. Johnson, D. Hurley, and K. Dawson Biogeosciences Discuss., 12, 14377–14400, 2015 www.biogeosciences-discuss.net/12/14377/2015/ doi:10.5194/bgd-12-14377-2015

Summary: The authors suggest that the wide range of aerosol iron fractional solubility estimates in the literature derive from inconsistencies in the operational definition of soluble iron used in observational studies. This manuscript reports the results of a comparison of chemical transport model results using different aerosol fractional solubilities for particles sized between 0.2 and 0.45 μm. A fractional solubility of 50% is used to represent the results of observational studies employing flow-through leaching techniques while much lower solubilities of 1% and 4% are used in the model. Differences in the results are predicted to be most significant (~30%) in regions distant from the dust sources.

Reaching an accurate estimate of aerosol iron fractional solubility, as well as the factors affecting the solubility, is an ongoing challenge for the research community and the modeling community will play a role in furthering our understanding. An important point that I feel is missing from this paper's discussion is the ultimate fate of aerosol particles within the ocean. While understanding dissolution immediately upon deposition is vitally important subsequent dissolution and/or scavenging in the upper ocean will also play an equally important role in the marine iron cycle.

Notes:

The terms in Equation 1 need to be defined.

The authors argue that particles between 0.2-0.45 μm make up 1.6% of dust particles between 0.2-2.0 μm. Is this value true regardless of aerosol source region? Transport time? Are there any caveats that need to be included?

Why was the DI Method fractional solubility set at 50% for one of the model simulations? This value is not representative of most bulk aerosol fractional solubilities from the literature (e.g. Sholkovitz et al., 2012). If derived from size-fractionated aerosol data (<0.45 μm) then the sources should be cited. R would be considerably lower and closer to 1 if a more realistic fractional solubility was set for Fed1. In two other simulations, the fractional solubility of aerosol iron is set at 1% and 4% because "the range in fractional solubility of Fe is the value required by global ocean biogeochemical Fe cycle models to enter the ocean as bioavailable Fe…" Are these values relevant for <0.45μm particles?

Page 14386: The authors mischaracterize observational efforts. "A large number of studies have attempted to measure sol-Fe concentrations in mineral dust particles present over the ocean." Oceanographers are interested in the fractional solubility of all aerosols that deposit to the oceans because they are all potential sources of iron. Why have the authors chosen to only focus on mineral dust when combustion emissions are significant sources of soluble iron in many regions?

Page 14388: "To explain these findings, we propose possible changes in dust size distribution caused by wetting of the mineral dust grains and long term acidification involved in the DI Method." It appears that the authors misunderstand the flow through extraction technique. During collection using cascade impactor or slotted impactor, the bulk aerosol is divided into prescribed size classes which are collected on a filter substrate. Therefore, a filter would only collect particles within a certain diameter range e.g. 0.2-0.4μm. The subsequent soluble fraction extraction would only include those particles so no change in size distribution outside of that range is possible. If in the short time that the particles were wetted (10-20 seconds in most cases) particles did break apart then it is reasonable to expect that these particles would also break apart upon deposition to the sea surface. The dissolution of salts and release of "finest" particles is desired as these processes are part of environmentally relevant aerosol dissolution. The comparisons to wet and dry sieving are not apt.

Page 14389: The role of nanoparticles in iron cycling is an area of current study. Might these very small particles that could be introducing variability in fractional solubility observations ultimately be dissolved iron sources in the photic zone? Baker and Croot (2010) offer extensive discussion of this topic. They are likely a source of dissolved iron in the surface ocean dependent on a number of factors reviewed by Baker and Croot (2010) among others. Analytical methodology will dictate whether nanoparticle bound iron is included in the soluble pool. It is unlikely that bound iron would be misattributed to the soluble pool by any methodology using a pre-concentration column or which relies on reaction chemistry such as DPD catalysis.

"The long-term exposure of Fe-laden leachate solution to low pH conditions can then cause release of Fe from the pool that is not expected to be soluble under typical atmospheric conditions. . ." Chemical oceanographer are not particularly interested the

fraction of iron soluble under atmospheric conditions. The purpose of flow through extraction, or any extraction method, is to mimic dissolution under marine conditions. The low pH conditions are a necessary preservation method to prevent loss of iron to the walls of the container and to prevent precipitation during storage.

Page 14390: Wet deposition is the dominant pathway in many areas. Why then is the wetting of aerosols during leaching not representative?

Page 14398: None of the citations appear in the References.

1. Does the paper address relevant scientific questions within the scope of BG? Yes.

2. Does the paper present novel concepts, ideas, tools or data? Yes.

3. Are substantial conclusions reached? The authors use comparisons of simulation results to conclude that extraction methodology may produce variability in soluble iron estimates. This outcome is not new (e.g. Morton et al., 2013) but to my knowledge their analysis is novel.

4. Are the scientific methods and assumptions valid and clearly outlined? Yes.

5. Are the results sufficient to support the interpretations and conclusions? Yes.

6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists? No, added discussion of Equation 1 and calculation of Dust0.45 is required.

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution? Generally yes but additional literature is indicated in this review.

8. Does the title clearly reflect the contents of the paper? Yes.

9. Does the abstract provide a clear and concise summary? Yes.

10. Is the overall presentation well-structured and clear? Yes.

11. Is the language fluent and precise? No, the submission would benefit from careful proofreading.

12. Are mathematical formulae, symbols, abbreviations and units correctly defined and used? No, Equation 1 needs additional explanation.

13. Should any parts of the paper be clarified, reduced, combined or eliminated? No.

14. Are the number and quality of references appropriate? The authors should seek out additional literature beyond those suggested in this review which discuss the role of nanoparticle iron in the surface ocean as well as reviews of aerosol solubility.

15. Is the amount and quality of supplementary material appropriate? N/A

Baker, A.R. and P.L. Croot. Atmospheric and marine controls on aerosol iron solubility in seawater. Mar. Chem. 120, 4-13, 2010.

Morton, P. et al. Methods for the sampling and analysis of marine aerosols: results from the 2008 GEOTRACES aerosol intercalibration experiment. Limnol. Oceanogr. Methods, 11, 62-78, 2013.

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