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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Air-to-sea flux of soluble iron: is it driven more by $HNO_3$ or $SO_2$? – an examination in the light of dust aging" *by* H. Yang and Y. Gao

**Anonymous Referee #2**

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Review of "Air-to-sea flux of soluble iron: is it driven more by HNO3 or SO2? - an examination in the light of dust aging" by H. Yang and Y. Gao. Submitted to ACP.

Summary

This paper shows the impact on the soluble iron content in dust when exposed to HNO3 as acidifying agent and compare with the effect of acidification by SO2. The authors use a global atmospheric transport and chemistry model (MOZART-2) to run three experiments. The impact of the individual effect of HNO3 and SO2 and their combined effect on iron solubility is estimated. They conclude that iron mobilization by HNO3 is significantly higher than that by SO2. They suggest that given the future

pollution scenarios, HNO3 fertilization can have a larger relative contribution to iron mobilization. This paper nicely shows an alternative pathway by which iron in dust can become soluble. However, there are some concerns that need to be addressed before recommending this paper for publication.

General comments

Many of my comments refer to missing information which ostensibly is included in another submitted manuscript by the same authors. Since the manuscript is not available, this reviewer is left with many pieces of information missing as noted. I leave it to the Editor to include my suggestion of additional information. Below I make some general comments and then I show some major (points regarding paragraphs 10048:20-10049:3, 10051, 10052,10054) and minor comments that should be addressed.

I think the authors should make an effort to provide more specifics on details of the processes explained from the viewpoint of the possible audience. This paper has implications related to marine biology, aerosol and gas transport modeling and detection (e.g. satellite monitoring) and aerosol-cloud interaction (Fe processing in cloud). Because the potential audience will include researchers with disparate background such as marine biology and aerosol remote sensing, the authors should explain more the setup and conditions of the simulations as well as provide more background on the explanations suggested. Specific points are detailed in the next section.

Although this paper deals exclusively the chemical aspects of this effect, it does not emphasize the importance of the right dynamics conditions that need to occur too. A major condition needed for dust transformation into a soluble form is the transport of the dust into an area of high humidity. Without the mixing with clouds and exposure to high levels of humidity, this phenomenon would not happen or be extremely minimized. However, I find that there is little discussion of the relevance of this fact and possible pathways through which it may occur. For example, many of the big dust sources in the NH are in large subsidence areas where dry air is common in the free troposphere.



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When the dust is lifted to the FT and advected away, it will have little contact with high humidity unless encounters a different weather pattern. Dry dust can travel thousands of km as demonstrated by the hygroscopicity measurements of Li-Jones et al., (1998) in Bermuda. So, for example, the explanation in 10049,5-10 , "little solubility of dust in the N tropical Atlantic is due to its proximity of the source", (also the explanation starting on 10049,21-23 "The smaller . . ." ) . However, what would happen if dust near the source is processed by a cloud? Would this near source dust become more soluble? In this scenario, one conceivable could have soluble Fe near the source. Another interesting pathway is the dust that travels in the oceanic boundary layer (and exposed by sea-salt) and then processed by clouds. Dust does travel long distances in the BL ( Gassó and Stein, 2007) and it can be impacted sea-salt (thus becoming more hygroscopic) as it was shown by Levin et al, (2005) .

Specific comments

Major Concerns:

10048, 20-10049,3: The description of data in Figure 1 is poor. It does not explain the origins of the data sets and some basics. Several questions and comments arise from the comparison offered in this plot: 1) Are the in-situ data collected in airborne (such on the ship's deck) or extracted from the ocean? 2) There is no explanation or descriptions of the time resolution (yearly or monthly) and spatial resolution (cruises or sites?) of the in-situ data. 3) Why comparing data taken in a period of 1988-2002 is comparable to a simulation run over year 2001? 4) In any case, I see little value of inclusion of Figure 1 as it is shown and explained does not make a convincing point. Given the few points for comparison, their scatter and the diverse nature of the data set in time and space, it is not clear how much random variability is being shown in the plot and how much of the actual solubility effect is being shown. In addition, this figure suggests a scaling between modeled and deposited solubility. But it is not clear from the in-situ data whether the HNO3 is responsible of the observed solubility which is the main point of this paper. My suggestion would be to remove the comparison altogether



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(and limit this study to a purely model study). 10051, 1-5.This statement refers to my previous comment regarding fig. 1. 10052, 24-25. I do not agree with this statement. F23 is markedly different in large area NW and SE of Australia encompass a significant portion of the South East Pacific. Please, address why since it is the only major feature observed in the SO. S10054,20-23. The emphasis of this summary should be changed. This is a modeling study and the effect of the relevance of HNO3 in dust solubility has not been validated by measurements. It does not show that Fe fertilization (implying a response from phytoplankton) occurs because this effect. This reference should be removed. However, it does suggest the existence of a viable alternative mechanism by which Fe can be mobilized.

Minor comments, clarifications and suggestions

Abstract 10044,10-15 : Why reporting an average as a range? not clear. 10044,10-15: Last sentence is a bit confusing. This is a matter of semantics but I think it is important to stress in this sentence the following points: the emission trends of NOx will increase according to future scenario and that this increase will be mostly anthropogenic. Introduction 10045,5-10: There should be an acknowledgement of the poor understanding of dust composition and mineralogy (with the consequently unknown hygroscopic affinity of the dust) in most of the deserts of the world, particularly those in the Southern hemisphere. 10045,5-18: What is the natural or background contribution of HNO3? or is it exclusively of anthropogenic origin? Please make it explicit. Anywhere in the introduction: this is a suggestion. Coating by sulfates and nitrates on dust is also relevant in radiative forcing and you may want to add comment on how it relates or complements your study a recent publication in JGR-Atmos (Bauer et al, 2007) which looks at this issue. Method 10046,5-10: Please expand and clarify your definitions of fresh, coated and dissolve. These criteria are used several times in the next sections. Furthermore, I found the definition in Fan et al, (2006) not clear either. For example, is it a "fresh" particle a particle that has not entered to a cloud or has not been exposed to high humidities? 10046,5-10: Model was run for year 2001. Is there a particular reason



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why? Was it just the modeled data was just available for that year? Because actual measurements are shown later, there should be a bit of discussion of worldwide dust activity and pollution in 2001. For example, was this year particularly dusty? Did pollution traveled more or less than past or following years? I think the point is that there should be more context added on the election of this year for the simulation. 10047,1-5: Equation 2), there must be a typo since there is a reference to N2 in the text but there is no N2 in the equation. 10048,5-10: This paragraph is the only one that discusses or mentions the fate of Fe in a cloud. I think more context should be added here and refrain to leave the explanations to the referenced study (Fan et al,2006). For example, these questions should help as a guide: does the model deal only with water phase clouds? What happens with dust in the presence of ice? How good is the MOZART-2 model in representing realistic wet removal processes and microphysics (if it has any)? Information on the strengths and weaknesses of the model on these points would be very useful. 10048, 10-20. It is not clear the definition of the experiments E2 and E3. Can you clarify more the nature of tests ran? For example, if E2 only deals with the effects of HNO3, does it mean that ks[SO2] is set to zero in eq 1? Results and discussion 10049, 3-4. Figure 2 depicts global distribution corresponding to E1,E2 or E3 ?. Please, clarify. 10049,15-16. Please provide range of values for this estimation and average solubility. These absolute values do not have much meaning without a sense how much variable they can be. 10050,20-21 . Same comment as before. Please provide range of values for this estimation and average solubility. These absolute values do not have much meaning without a sense how much variable they can be.

References

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 10043, 2007.